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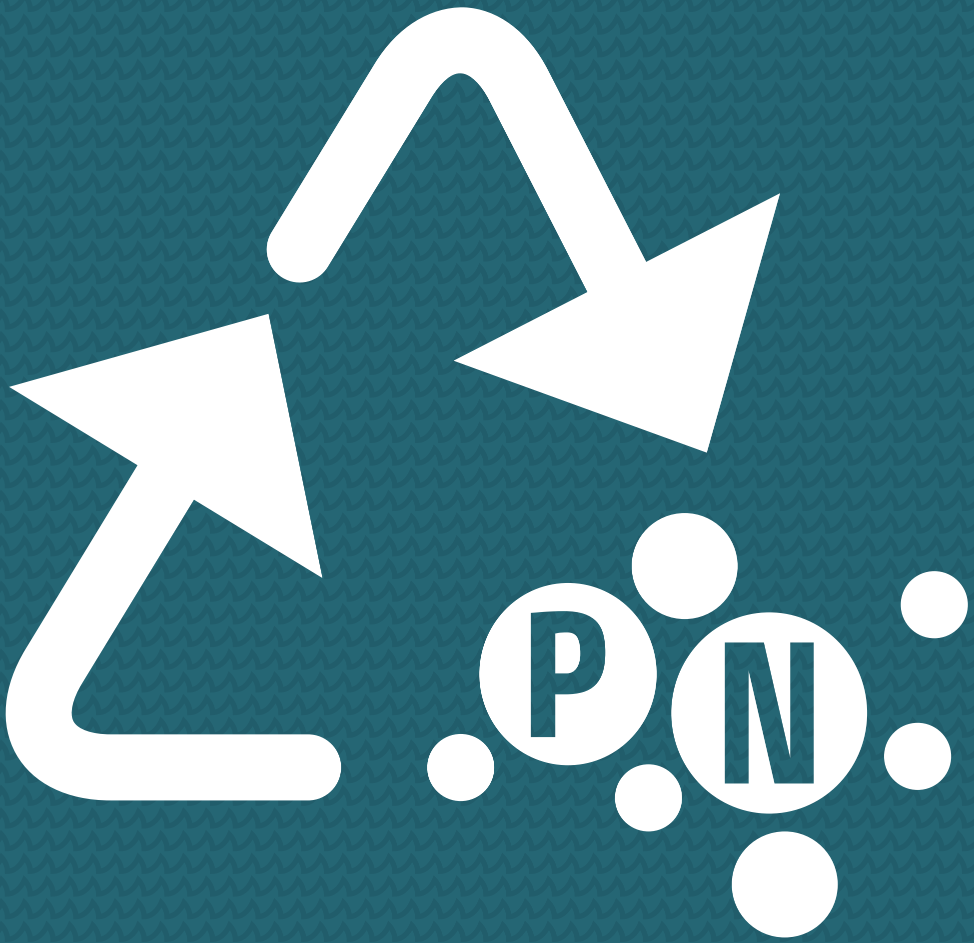
Palette of Solutions for Nutrient Recycling in the Baltic Sea Region

HELCOM

Nutrients

2021



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About BSR WATER
The EU co-funded project BSR WATER – Platform on Integrated Water Cooperation (2018–2021; www.bsrwater.eu) aims to enhance cross-sectoral cooperation in smart water management by providing a possibility for transnational experience exchange, sharing of good practices and solutions, as well as delivering comprehensive overview of the current and future regional policy. The platform brings together experts representing diverse projects that have generated through transnational cooperation many replicable as well as unique solutions, covering broad variety of water-related issues.

The platform cooperation is based on practical achievements and results of seven projects addressing a wide range of water management challenges. The outcomes and practical findings of the contributing projects (IWAMA, BEST, iWater, Manure Standards, Village Waters, Reviving Baltic Resilience, CliPLivE) support the long-term development of regional environmental policy and recommendations, which will further strengthen the policy-practice link in implementation of advanced water protection measures, including smart nutrient management and sludge handling, storm water management and the energy efficiency cycle at the national and municipal levels.

Contents

Foreword

Nutrients, especially phosphorus are vital for our society. As phosphorus is one of the main limiting nutrients in agriculture, phosphorus-based fertilisers have been used for centuries and will need to be used in the foreseeable future. Widespread use of fertilisers however has created one of large environmental issues we are facing – eutrophication. Similar to plants, phosphorus is also the main limiting nutrient for algae growth and the amount of phosphorus in the water systems has increased greatly due to human activities (runoff from fields, effluent from wastewater treatment plants (WWTPs), leakages). For past decades, many different actions have been undertaken to reduce the amounts of phosphorus entering the sea from different tributaries and the total loads of nutrients emitted to the Baltic Sea have been decreased significantly.

WWTPs have been one of the main point sources of phosphorus entering the environment. Today, advanced phosphorus treatment is mandatory for most medium sized and larger WWTPs in the region. Chemical precipitation and biological accumulation are used to take the phosphorus out of the wastewater streams and tie it into the sludge. This sludge is then treated in various ways and quite often the phosphorus potential there is lost – based on a key figure collection carried out in the IWAMA project almost 60% of the treated sludge is not used directly (Raudkivi *et al.*, 2018). Most of it is either incinerated or accumulated at the WWTP and finally landfilled.

Almost all of the phosphorus currently used in the EU is imported from countries outside of Europe (Morocco, the US and China) (European Sustainable Phosphorus Platform, 2017c). The only raw phosphorus available in the EU is in Finland and Estonia and the potential mining is environmentally very complicated. Therefore, due to increasing welfare in the exporting regions, world population growth and a growing demand, phosphorus is essential to the food security in Europe. Research has clearly shown that at the moment, phosphorus is not treated sustainably in the EU, disappearing from the food chain in three different areas: wastewater sludge, manure and organic waste.

This document explores feasible solutions for nutrients and phosphorus recovery primarily from wastewater and sludge. More concrete measures must be put in place in order to allow and demand WWTPs to invest into more feasible solutions. Phosphorus can be recovered from many different streams in the WWTP, from treated sludge, incineration ashes, sludge liquor etc, scientific research, pilots and full-scale applications have been concluded to show these options and test the efficacy and feasibility. The palette of solutions serves as a review of the different options and can be used as guidelines in the planning of national phosphorus recovery strategies, key technologies and potential applications by authorities, local practitioners and companies. Main focus is on the technologies already tested in larger scales.

Introduction

The overview of phosphorus (P) is often divided into two large categories – the essential element for organism growth versus a large source of environmental pollution and the main cause of eutrophication. As phosphorus (or by some researches the joint effect of nitrogen (N) and phosphorus (Elser *et al.*, 2007) is the main limiting nutrient in most marine environments, anthropogenic phosphorus additions have significantly altered the natural balance. This results in a multitude of issues – poor water quality, loss of habitats and biodiversity, dangers to human health, summing up to large environmental and financial losses (Schaum, 2018). In the US, the conservative estimate of financial loss due to eutrophication has been shown as over 2.2 billion dollars annually, taking into account drinking water, recovery of endangered species and losses of property and recreational value of waterfronts (Dodds *et al.*, 2009). There is no question that phosphorus as a pollutant is an essential issue in the Baltic Sea region (BSR) as well (HELCOM, 2018).

Phosphorus is part of many organic molecules, such as DNA and ATP and therefore is vital and irreplaceable for all lifeforms. The phosphorus demand and production in the world is steadily growing, by the latest report and outlook of the Food and Agriculture Organisation of the United Nations, the average yearly growth trend is around 2% (Food and Agriculture Organization of the United Nations, 2019). As phosphorus is extracted from sedimentary and igneous phosphate rock deposits, the resource is limited and non-renewable. Although the mass media has broadcasted news about the depletion of phosphorus reserves in the next 50 years, this has been mainly based on outdated production and resource data (Van Vuuren, Bouwman and Beusen, 2010). This however should not be taken as the lack of need for better solutions – historical phosphorus scarcity scares have been common, while the rebuttal with new information has commonly caused no implementation on P conservation policies or other practices (Ulrich and Frossard, 2014). Easily accessible and financially feasible phosphate rock deposits are depleting, although in current estimations depletion will become an issue in the next few centuries (rough estimation of current reserves and production show possible depletion in 370 years) (Desmidt *et al.*, 2015). Another major problem is the high Cd concentrations (average of 21 mg/kg, but up to 150 mg/kg) in many phosphate rock reserves, most of which depending on the fertiliser production process can stay in the finished products (Robertsa, 2014).

Whether P resources are ending or not, the production of phosphorus fertilisers cause multitude of environmental problems in the producing countries (waste rock piles, water stress). The production is also expected to become more expensive (from current 30-40 $/t up to 110-150 $/t based on different models), therefore rising the overall food prices and endangering our food security (Van Vuuren, Bouwman and Beusen, 2010). According to a study on phosphorus balances, losses and use in the EU (van Dijk, Lesschen and Oenema, 2016), around 1.7 kg/P was used annually by capita in the EU (2005), close to 80% of it as food. This gives even more emphasis on the fact how much the food security is tied to phosphorus use, with significantly higher phosphorus prices, the effect on everyday life will be tremendous.

As the European Union (EU) imports more than 90% of phosphorus from other countries (Russia, Morocco, Israel, Senegal, Algeria, Tunisia), we are dependent on their price policy (European Sustainable Phosphorus Platform, 2017c). These imported phosphorus streams are inefficiently used and large quantities of it are lost from the fertilized soil with run-offs or transported to urban areas as secondary P in food products, food additives and non-food products (detergents) (Schaum, 2018). This secondary P in urban areas is then transported to wastewater treatment plants and eventually to landfills or fields, with large percentages (around 17% of total system losses in 2005) finding their way to marine environments (van Dijk, Lesschen and Oenema, 2016). For the EU and BSR, the movement of phosphorus must be transformed from linear to circular in order to retain the safety of our economy and food. The efficient use of natural resources is also one of the main factors of becoming a sustainable society, therefore coupled with the environmental issues, reuse and recovery of phosphorus needs to be one of our main goals for the near future.

Looking more closely to the impact of phosphorus in the marine environments, soluble reactive P (SRP), dissolved hydrolysable P (DHP) and particulate P (PP) fractions need to be differentiated (Jarvie *et al.*, 2010). Although depending on the source these fractions can differ greatly, generally the SRP has highest bioavailability to aquatic biota (Schaum, 2018). Algae can also be efficient in assimilating DHP and even PP, especially with longer retention times in the system (lakes) and in conditions where other P fractions are limited (Heisler *et al.*, 2008). For WWTP effluents, SRP is often measured as the highest fraction, while rural contamination resulting more in high PP concentrations (Neal *et al.*, 2010). Therefore, WWTPs can result in the most significant phosphorus pollution in the receiving waterbodies as the emitted phosphorus is generally more bioavailable than the run-off from agriculture.

WWTPs in the BSR have been subject to strict effluent limit values for the past decade as in many countries the limit values for phosphorus in medium and large sized WWTPs is 0.5 mg/L and for nitrogen 10 mg/L. The enforcement of these limits has decreased the flow of nutrients to the marine environment significantly, already from 1995 to 2014 the total phosphorus load to the Baltic Sea has decreased from 41 000 t to 31 000 t (Sonesten SLU *et al.*, 2018). In the WWTPs however, this reduction in effluent phosphorus loads means higher P loads to waste activated sludge (WAS; 90-95% of inflow P (Cornel and Schaum, 2009)), which today could be one of the major sources for secondary phosphorus recovery. While a large proportion of the nitrogen in the wastewater from urban sources is transferred to dinitrogen gas via multiple biological processes, some of it (around 13-15% of inflow TN (van der Hoek, Duijff and Reinstra, 2018)) is kept in WAS and can be recovered as well.

From total losses of phosphorus in the EU, municipal sewage sludge is responsible for about 18.6% and the total for wastewater (WW) and sludge sector is around 30% (including effluents of centralised and decentralised WWTPs, untreated WW and uncollected WW) (van Dijk, Lesschen and Oenema, 2016). Together with organic wastes, 81.9% of total losses should be able to be recovered. As the total loss from imported phosphorus is estimated to be around 50%, good recovery operations can increase regional self-supply security significantly (Schaum, 2018).

In previous decades, a large proportion of the wastewater sludge was used directly in agriculture, in both treated and sometimes even untreated forms. In order to reduce environmental risk, this practice however is not feasible anymore due to increased knowledge of different inorganic and organic pollutants in the WW and subsequent sludge (Egle *et al.*, 2016). Treated and thoroughly analysed sludge can still be used directly today in many of the BSR countries, while a specific certification process is usually necessary to label waste into product. Still, as the list of different chemicals possibly absorbed or adsorbed into or onto sludge particles is growing constantly, the direct use of sludge either for agriculture or recultivation is becoming less and less popular and can even be phased out from legislation (Roskosch, 2017)) Direct use can also come with significant transportation costs (25-65% of total operation costs together with disposal), as the water content in sludge is often more than 70% (Li *et al.*, 2014; Cieślik and Konieczka, 2017).

The reduction of direct use for sludge means new technologies are needed to extract and recover phosphorus. As incineration is one of the growing practices for sludge treatment, phosphorus recovery from incineration ashes is one of the most important technologies. Other research has shown the possibility to recover phosphorus before incineration via precipitation from either sewage sludge or sludge liquor (Cornel and Schaum, 2009). The list of options and technologies currently being worked on is growing steadily, while technological readiness index (TRI) and financial feasibility are still the main barriers for widespread adoption.

The technologies for phosphorous recovery are currently under rapid development, with both pilot and full-scale demonstrations of existing ideas and new laboratory pilots of new approaches being built. There are more than 70 full-size phosphorous recovery plants in the world today (Ohtake, 2018). The technologies related to precipitation of phosphorous from the soluble phase at the wastewater stream are becoming more attractive for investment. However, technologies related to the recovery from incinerated sewage sludge are not as widespread and feasibility of those processes has to be demonstrated in large scale.

Different measures can be set up in regional and national levels to help with investments concerning phosphorus recovery or substitute operational fees (tax reduction, investment co-funding etc). It is however difficult to set up national supportive measures without specific goals, strategies and long-term understanding of the mechanisms (Egle *et al.*, 2016). Therefore, a unified strategy in phosphorus recovery is needed, preferably on the EU level. The Baltic Sea region can be a forerunner, with already established efficient intraregional cooperation and joint measures. Tested guidelines and solutions and legislative recommendations can later be transferred out to other EU sub-regions or even adapted in EU as a whole. The revised EC Circular Economy action plan promises to develop an Integrated Nutrient Management plan, to ensure more sustainable application of nutrients and stimulate the market for recovered nutrients (European Commission, 2020).

HELCOM represents an ideal platform for BSR internal cooperation and should be one of the main facilitators of the move towards more sustainable and circular nutrient management. In order to help this change, regional strategies and guidelines are needed for authorities, while both authorities and practitioners also need the understanding on possible good solutions, adaptable technologies and limitations.

Current document on the palette of solutions on nutrient recycling aims to be one of the potential foundations of such strategies and guidelines. This document focuses mainly on phosphorus recovery from wastewater, sludge and incineration ashes, with some information on possible nitrogen recovery as well. Other parts of the foundation are expected from different sources, such as a palette of solutions concerning manure and agricultural run-offs. Together these approaches to different streams of nutrients can hopefully be used to create a more comprehensive understanding on the issues and possibilities for nutrient recycling and serve as a strong base for future legislative needs.

1. Nutrient sources to the WWTP

When looking at the sources of nutrients in a WWTP, the source of the wastewater first needs to be considered. Comparing municipal and industrial WW, the concentration and load of nutrients can vary significantly. Therefore, these streams will be looked into separately in the following section, as the main sources of nutrients are different. The bulk of the solutions and technologies discussed in this document are traditionally meant for municipal WWTPs, while many of them can also be transferred to industrial WWTPs with high nutrient flows. Industrial WW is usually more predictable as well, which means it's easier to fit specific tailored solutions, while municipal WW can be greatly influenced by various factors, one of the main ones being stormwater.

In centralised treatment, municipal WW is collected throughout urban areas using a sewer network. While there is a great diurnal and seasonal difference in concentration and load profiles, the WW mainly has similar characteristics. The fluctuations in the total nutrient flows are quite small, especially the phosphorus load in the excess sludge (Schaum, 2018). The flow profile can also be influenced by the weather – the stormwater collection systems are often not separated from wastewater sewer and therefore pass through the WWTP. This can greatly increase the hydraulic peak and dilute the nutrient concentrations in the influent wastewater. Nowadays many municipal wastewater treatment plants have invested into a separated sewer system to bypass stormwater. Although stormwater and the run-off from non-permeable areas can also include phosphorus (Yang and Toor, 2018), the total load from the streams is miniscule compared to nutrients loads flowing into the WWTP.

The average phosphorus load in the raw wastewater has been shown as 1.8 g P per capita per day (g P/(C·d)) (Cornel and Schaum, 2009). This compared to average total imported phosphorus per capita (2005) of around 4.7 g P/(C·d) (van Dijk, Lesschen and Oenema, 2016) means that about 38% of the per capita import of P in the EU ends up in the WWTP. While most of it comes from the food supply (both animal- and plant-based food), about 13.1% of the total phosphorus consumption is from household chemicals, especially automatic dishwasher detergents (van Dijk, Lesschen and Oenema, 2016). The sources of phosphorus making up the per capita phosphorus load to WWTPs was more thoroughly shown in a 2009 study in the UK (Comber *et al.*, 2013), which is shown in Table 1.

Table 1 – *Estimation of domestic phosphorus load to UK sewer network (Comber et al., 2013), numbers rounded up; Food additives[1] – additives in drinks and processed meats, such as sodium and potassium diphosphates (E450a), calcium orthophosphates (E341), sodium and potassium triphosphates (E450b) and other traces emitted as urine.*

PHOSPHATE DISCHARGES	PER CAPITA DISCHARGE TO SEWER (G P/(C·D) (DATA FROM UK)	PERCENTAGE FROM TOTAL DOMESTIC LOAD
FOOD ADDITIVES[1]	0.59	29%
FAECES	0.21	10.3%
URINE	0.61	30%
WASHING MACHINES	0.275	13.5%
AUTO DISHWASHERS	0.176	8.7%
P DOSING OF TAP WATER	0.125	6.2%
FOOD SCRAPS	0.10	4.9%
PERSONAL CARE PRODUCTS	0.022	1%
TOTAL DOMESTIC LOAD TO SEWERS	2.03	

Faeces and urine (combined with food additives) are the main sources of phosphorus in the municipal wastewater, resulting to around 69% of total load. More than 22% of phosphorus in the study was resulting from detergents, while EU issued a prohibition of phosphorus-containing laundry detergents in 2013 (Official Journal of the European Union, 2012), therefore this load is decreased. This prohibition does not however apply to dishwasher detergents due to the lack of economically feasible alternatives (Schaum, 2018). Some other sources include P-dosing of tap water (to decrease lead transfer from pipes, also regional), food scraps discharged to sewers and personal care products (soaps, lotions, hairspray, toothpaste as main products containing phosphorus) (Comber *et al.*, 2013). In the past decade however, phosphate-free personal care products have become more popular, reducing this small load even more.

Besides phosphorus there are many other elements and substances in the municipal wastewater, many of them potentially dangerous and problematic. Persistent organic pollutants (POPs), heavy metals, pharmaceutical residues and microplastics are some of the more common substances that can be included in the municipal wastewater (Egle *et al.*, 2016). As the consumption patterns of people are very complex and difficult to predict or change, the loads of substances coming to the WWTP can have large diurnal and seasonal changes (flu season can increase pharmaceutical residue loads many times (OECD, 2019)). As these substances can be either absorbed into sludge particles or adsorbed on their surfaces, it is the main reason behind the decrease in the direct use of treated sludge. Analysing hundreds of different substances from the sludge can be costly (and there are so-called unknown dangers not evaluated yet) and therefore it is safer to view both municipal wastewater and resulting sludge as potential environmental risk factors.

Compared to municipal wastewater, the industrial effluents vary much more based on the specific sector, industry and process. The main industrial sources of phosphorus is the food industry (especially dairy), while many other sectors use phosphoric acid for cleaning (metal treatment and etching, electronics) (Schipper *et al.*, 2004). While industrial wastewaters differ greatly between each other, the composition of a specific wastewater stays very similar when no processes or substrates are changed. The specific concentrations can also be controlled by process parameters, which is not possible with municipal streams. Therefore, the concentrations of nutrients and hazardous substances in the industrial wastewater and sludge can stay constant and some can be used directly with lower risks.

The industrial effluents and sludge can however be emitted to centralised municipal WWTPs and mixed with municipal equivalents. This practice is often used by WWTPs trying to reach energy neutral or even positive internal energy balance. Accepting water and waste with high biodegradable organic fraction can significantly boost biogas production, therefore such streams are valuable and wanted by municipal operators. Although the industrial streams can be clean of hazardous substances and could have a potential direct use, mixing with municipal streams possibly contaminates them, making the phosphorus recovery more complicated. This could be managed with proper indirect discharge management, setting the limit values and monitoring systems. For WWTPs with a working phosphorus recovery technology this type of management might not be a problem, while currently the number of such plants in the BSR is small.

One of the main streams of secondary phosphates to the environment comes from the agriculture – both stormwater run-offs from fields and improper manure handling can cause significant pollution. The handling of slaughter waste is also deemed the field with the largest phosphorus losses (24.1% of total imported P losses) (van Dijk, Lesschen and Oenema, 2016). As the incineration of manure and animal waste might be necessary in the future, solutions for phosphorus recovery from the ashes get more important. Unfortunately, the ash from manure and animal waste is often considered unattractive for P recovery due to high concentrations of zinc and copper (1500 mg/kg and 500 mg/kg respectively) and low phosphate content (15-20% P_2O_5) (Schipper *et al.*, 2004). This document will however focus on the wastewater treatment and nutrient recovery from within sludge, sludge liquor and sludge incineration ashes.

2. Nutrient flows in a WWTP

Understanding the flows and pathways in the WWTPs is important in finding the key places for phosphorus recovery. The composition of the different fractions of phosphorus (soluble reactive P (SRP), dissolved hydrolysable P (DHP) and particulate P (PP)) can be different depending on if industrial WW is accepted or if phosphate-based detergents are used (Gu *et al.*, 2011). In average cases, roughly half of the influent phosphorus can be assumed to be in the particulate form (PP) and half as SRP (Schaum, 2018). In primary sedimentation about 11% of phosphorus is expected to be removed with primary sludge (Cornel and Schaum, 2009) depending on the settling time, this reduction comes from the sedimentation of some of the heavier PP.

The effluent from primary sedimentation consists mainly of SRP (57%) and different particulate P fractions (~40%) (Gu *et al.*, 2011). In regular biological wastewater treatment (w/o enhanced biological phosphorus removal (EBPR)) approximately 28% of the influent phosphorus is taken out with surplus sludge (Cornel and Schaum, 2009). All phosphorus fractions including PP are incorporated in the sludge and with EBPR over 90% of all phosphorus fractions are reduced. The remaining fractions depend on influent and treatment process, while specific tests (Gu *et al.*, 2011) have shown about half of it being in dissolved form (SRP and DHP) and half in particulate form. With additional chemical precipitation the dissolved form can be targeted, while reduction from particulate form is usually miniscule. In secondary clarification phosphate accumulating organisms (PAOs) can release very low amounts of SRP back to the dissolved phase. Throughout the treatment process around 90% of inflow phosphorus is incorporated into the sludge and the rest is emitted with the WWTP effluent (Cornel and Schaum, 2009). This fraction can be lowered by applying tertiary treatment such as membrane or sand filtering (with subsequent filtration and coagulation, concentrations as low as 0.02 mg/L P have been reached (Li and Brett, 2012)). A short overview of the phosphorus flows in the municipal WWTP can be seen from Figure 1.

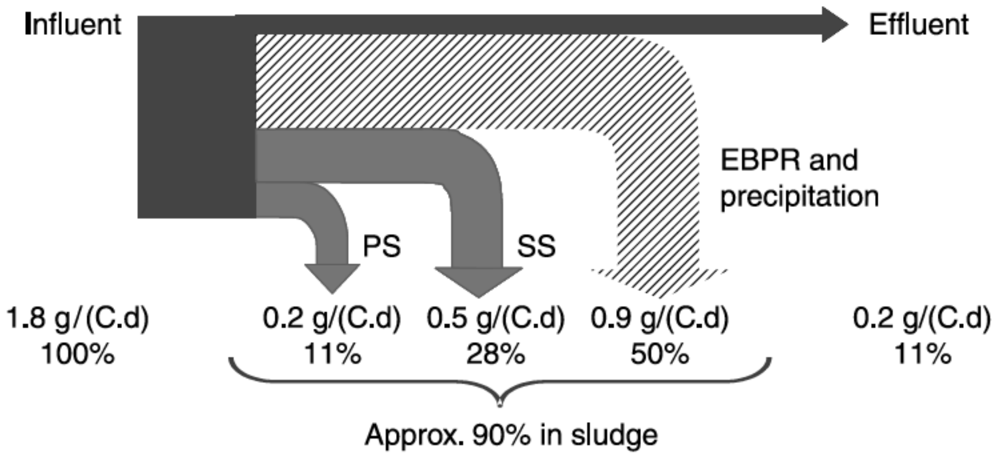


Figure 1 *– Phosphorus balance for a typical German municipal WWTP; PS – primary sludge, SS – secondary sludge, EBPR - enhanced biological phosphorus removal, g/C.d – grams per capita per day* (Cornel and Schaum, 2009)

As most of the incoming phosphorus to the WWTP ends up in sludge, majority of the phosphorus recycling technologies are targeted there as well. Both sludge liquor (effluent from dewatering) and incineration ashes are considered to be optimal places with good recovery potential. The specific approaches can vary based on the phosphorus removal process used, as chemically precipitated and biologically bound phosphorus have different properties. More than 30 different phosphorus recovery technologies from the sewage sludge have been created, with new research constantly developing. It has however been recommended that a phosphorus recovery method should be developed separately or modified accordingly for each treatment plant (Cieślik and Konieczka, 2017). Ready-to-use general marketable solutions might not be as effective in the P-recovery field as in many other WWTPs sectors.

There are many different factors, that can influence the choice of P-recovery technology for a WWTP. For example, one of the important factors is the phosphorus removal technology – studies have shown that EBPR can have a significant effect on the sludge treatment in general, especially sludge dewaterability. Practice has shown that EBPR can decrease dewatered cake solids up to 3-4% TS and require double or triple the amount of polymer dosage (Nicholson *et al.*, 2017). EBPR can also increase the risk of struvite precipitation throughout the anaerobic digestion. In such cases phosphorus recovery from mixed sludge before digestion can be feasible to consider, as it can also make subsequent treatment more efficient and bring savings on polymer use or pipe maintenance. Solutions such as thermal hydrolysis and sludge pre-treatment tanks for stored phosphorus release can be used before the digester (Schaum, 2018) with potential positive effects for following processes.

Struvite precipitation is another of the most wide-spread P-recovery technologies, which is mainly used to recover phosphorus from the sludge liquor. Different technical solutions can be applied either upstream or downstream from the dewatering process, potentially also mitigating some of the negative effects of EBPR to dewatering efficiency (AirPrex process, (Yoshida *et al.*, 2019)). Extracting phosphorus from the sludge liquor can also significantly reduce the recycle flow back to the biological process and reduce the amount of organic carbon or precipitation chemical needed. These processes will be explained and viewed more specifically in the next sections.

The third major source of phosphorus recovery in the WWTPs is from the incineration ashes. During thermal treatment the sludge volume is reduced significantly (70-90%), greatly increasing the phosphorus concentrations. While the phosphorus concentration in activated sludge is estimated as 1-5%, concentrations in ash can be 5-11%, even up to 20% in case of very specific technologies (Cieślik and Konieczka, 2017). The extraction can be done in various ways, one of easiest and least expensive technologies is acid (H_2SO_4 or HCl) extraction, while there are more complex and efficient processes (electro dialysis (Guedes *et al.*, 2014), sequential extraction SEPHOS (Cornel and Schaum, 2009), etc). It is however important to consider, that incineration as a technology is feasible only for large WWTPs (traditionally over 500 000 PE, though new smaller alternatives are being developed (Endev, 2017; HUBER SE & WTE, 2021)), therefore the option for P-recovery from ashes is reserved to a few.

Figure 2 outlines all the different points in a complete wastewater and sludge treatment scheme (for large WWTPs) where phosphorus recovery installations can be added. As mentioned before, ready-to-use commercial applications might be very inefficient and WWTP-specific modifications are recommended for feasible P-recovery solutions (Cieślik and Konieczka, 2017). Following chapters will go more into depth for each of these streams, with similar types of technologies grouped together.

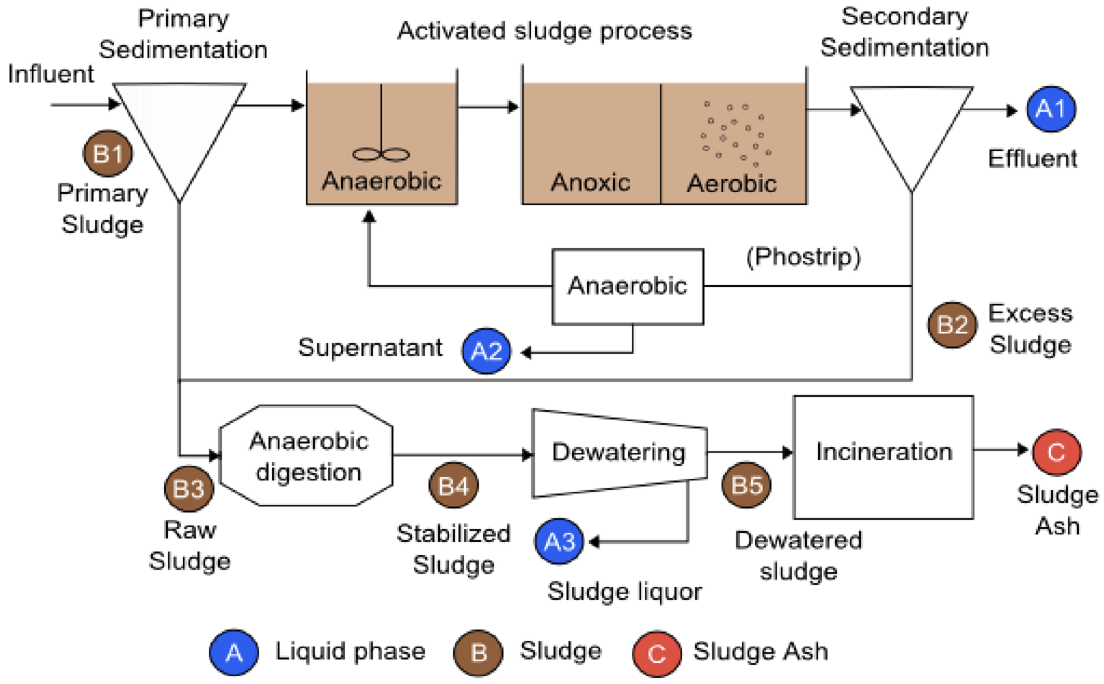


***Figure 2** – Points of installation of phosphorus recovery technologies for large WWTPs*
(Cornel and Schaum, 2009)

3. Nutrient recycling technologies

3.1 From reuse to recovery

Reusing nutrients from the waste stream is not in itself a new concept – in the past both wastewater and sludge have been used directly for various applications, including irrigation, fertilisation, landscaping, recultivation etc. During last decades however the environmental impact of such practices has come under question, as heavy metals, micropollutants, microplastics and pathogens can be transmitted to the environment from wastewater. For many countries around the world the direct use of untreated or treated wastewater and sludge is still in effect, while the main reason is due to the shortage of appropriate disposal facilities (Biswas *et al.*, 2009).

With many treatment methods, the sludge can be stabilised, reducing the odour, pathogens and in some cases heavy metal and micropollutant concentrations. Composting has become a standard method for small and large-scale WWTPs, as the quality of treated sludge is quite good in comparison with past standards. Compost products have been marketed around the BSR and through them significant nutrient streams from wastewater treatment have been reused. The main problem with this practice is the increase of trace elements (heavy metals, persistent organic pollutants (POPs), pharmaceuticals) in the treated soils as even treated sludge can still consist of both known and unknown pollutants. Some countries around the BSR still use the sludge directly, while more complex measurements and standards are needed for this process (certification system).

All in all the practice in the EU is currently moving towards the prohibition of the direct use of sludge, especially for larger WWTPs (Cieślik and Konieczka, 2017). While smaller WWTPs are not prohibited from direct usage, the required certification processes make it both financially unfeasible and complicated. Many smaller WWTPs using composting, humification or other process currently face an unknown future due to rapidly changing legislations. As reuse is being phased-out in the Baltic Sea region and in EU as well, the emphasis in nutrient management is moving towards nutrient recovery and recycling instead.

Compared to reuse, nutrient recovery and recycling needs more complex technological approaches, as valuable components have to be separated and extracted from the streams, while in reuse the whole stream was used. Extraction processes are very common in metallurgy and chemical manufacturing businesses, while often these mature processes can’t be directly applied for wastewater or waste activated sludge due to low recoverable material concentrations. For many processes the purity of input material is of vital importance and some elements in the material can inhibit the processes or greatly lower the efficiency. Changing the approach from reuse to recovery needs significant scientific research, laboratory, pilot and full-scale testing and development of new technologies, some of which are introduced in the following sections.

3.2 P-recovery from liquid phase

Liquid phase in the WWTP mainly refers to wastewater and reject water (often called sludge liquor), while the recoverable phosphorus form in these streams is the soluble phosphate ion (PO_4^{3-}). Recovering phosphorus from wastewater can be difficult to make feasible, as the concentrations of phosphorus are low and therefore the recovered amounts are low as well (Schaum, 2018). Phosphorus recovery from sludge liquor can be more feasible and has had more scientific interest. As anaerobic digestion is only feasible for medium and large scale WWTPs, most technologies are specifically targeted there. With larger WWTPs and subsequent larger production of recovered phosphorus, the financial feasibility concerning the initial investment is easier to offset as well.

There has been however significant scientific research towards applying liquid phase P-recovery in smaller WWTPs as well. Technologies aimed for anaerobically digested sludge liquor have been also tested for smaller WWTP (<10 000 PE) in the US using aerobic sludge digestion and potential financial feasibility has been shown (Hallas *et al.*, 2019). Similar methods can also be applied to wastewater in other special cases, such as for treating the high P concentration effluent from sewage sludge ozonation (instead of sludge treatment, sludge reduction is sometimes used in Asia (Saktaywin *et al.*, 2005; Zhang, Tian and Zhang, 2017)).

The most widespread methods used for phosphorus recovery from liquid phase is through crystallisation reaction, during which phosphor is precipitated as struvites, hydroxy apatites or calcium phosphates (Cieślik and Konieczka, 2017). These technologies have been applied in full scale worldwide, from Japan to Denmark, as the products can generally be used directly without an extraction process. Struvite also has a low solubility in natural conditions, therefore applications in agriculture have a smaller change of stormwater carry-off and leaching. Furthermore due to higher Mg concentrations, plant growth and phosphorus uptake can even be increased, which has been shown in various experiments with different plant cultures, such as maize, ryegrass, lettuce, broad bean plant, etc (Ahmed *et al.*, 2018).

Main processes applied for P-recovery from liquid media in pilot or full scale applications are MAP (struvite precipitation, details in section 3.5.1) process and HAP (hydroxyapatite precipitation, details in section 3.5.2) process. Different sorption processes (details in section 3.5.4), including ion exchange and inner layer adsorption are under large scientific interest as well, while full-scale applications are not very well documented yet.

3.3 P-recovery from sludge

As discussed in previous chapters, around 90% of inflow phosphorus to the WWTP is transferred to the activated sludge and precipitated minerals mixed with the sludge. In most cases, the phosphorus in the sludge is insoluble and needs to be extracted into more soluble forms for recovery, usually accomplished by adding acid, base and/or higher temperatures (Cornel and Schaum, 2009). The recovery itself uses similar precipitation techniques as discussed previously in the last sub-chapter (struvite, hydroxyapatite), with some extra minerals as calcium phosphates being used as well (Cieślik and Konieczka, 2017). Recovery potential has been shown for both undigested and digested sludge, with the former having lower phosphorus concentrations and efficiencies while the latter needs more chemical addition due to higher carbonate concentrations (Quist-Jensen *et al.*, 2019).

The main technological point of interest for P-recovery from sewage sludge is the extraction of insoluble phosphorus from the media with different chemical leaching and oxidation processes (Egle *et al.*, 2016). Phosphorus is present in sludge in various forms, such as chemically bound phosphorus (precipitated), organic bound phosphorus (sludge mass growth) and polyphosphates (from EBPR). Acidification for example can easily dissolve phosphorus bound with metal compounds, increasing soluble phosphorus concentration (Quist-Jensen *et al.*, 2019). One of the main problems with the different extractions, especially acid-based oxidation, is however the reintroducing of heavy metals and other potentially hazardous substances to the liquid or soluble form (Cieślik and Konieczka, 2017). Separating heavy metals from the phosphate solution is difficult, therefore additional separation technologies are often used in the technological applications (ion exchange, additional reagents, pH adjustments) (Schaum, 2018).

Phosphorus can be extracted from sewage sludge with wet-chemical leaching (section 3.5.5), while extracted phosphorus from sludge is usually precipitated either as MAP (struvite precipitation, details in section 3.5.1) or HAP (hydroxyapatite precipitation, details in section 3.5.2). New advances have also shown a possible magnetic separation of some phosphate minerals, such as vivianite (section 3.5.3). Thermal hydrolysis and other common technologies applied to disintegrate sludge are not discussed in depth in this document, as they might be considered more as a pre-treatment method before P-recovery.

3.4 P-recovery from incineration ashes

According to many recent reports, P-recovery should focus more on the ashes of the sewage sludge incineration. The statistics are quite clear – from 5 to 10 times more phosphorus could be recovered from the ashes compared to sludge or liquid forms. Unfortunately, incineration technology needed is feasible only for very large WWTPs, therefore the use of these technologies is severely limited (Cieślik and Konieczka, 2017).

During sewage sludge incineration the organic material in the sludge is destroyed, which reduces the volume of materials significantly (70-90%) (Schaum, 2018). Sewage sludge ash (SSA) has much higher phosphorus concentrations, usually from 5 to 11%, potentially up to 20% (Cieślik and Konieczka, 2017). The process disintegrates all organic pollutants and in contrast with sewage sludge, wet extraction is significantly easier, as all phosphorus in inorganic formations (Cornel and Schaum, 2009). In SSA the phosphorus is mainly as $Fe_4(P_4O_{12})_3$ or $Al(PO_3)_3$, as Fe and Al based agents are most common in chemical phosphorus precipitation. Based on the precipitation chemical, the SSA is often categorised as either Al-rich or Fe-rich (Petzet, Peplinski and Cornel, 2012).

Some approaches have aimed towards the direct us of the ashes as a fertiliser, due to reduction of heavy metals in high-temperature incineration (processes at 900-1050° C can evaporate up to 90% of Cu, Zn, Cd and Pb from the ashes). At the same time, other heavy metals are not removed (Cr, Ni), which can prove to be a problem for legal limits. Another big factor is the relative bio-unavailability of minerally bound phosphorus, therefore limiting the possible use of SSA as direct fertilisers (Cieślik and Konieczka, 2017).

The process for P-recovery from SSA can divided between two principle approaches (Petzet, Peplinski and Cornel, 2012):

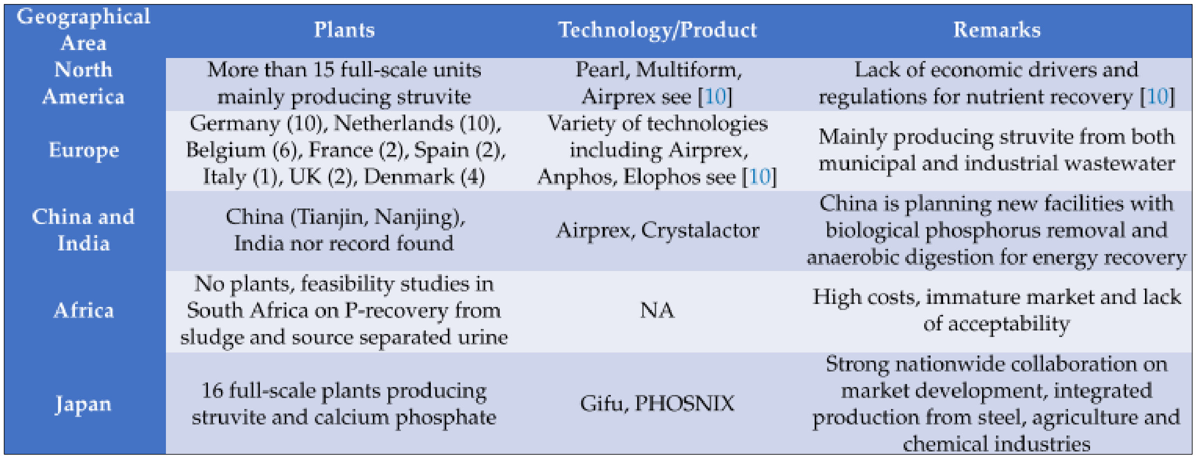
a. separation of heavy metals from P;
b. conversion of P into a plant-available or industrially applicable form.

The most common processes applied are wet chemical extraction (section 3.5.5) and thermo-chemical process (section 3.5.7), the latter of which is often aimed toward vaporizing heavy metals from the SSA (Cornel and Schaum, 2009). New approaches such as the thermo-electrical process (section 3.5.8) have also under high scientific interest (Egle *et al.*, 2016).

3.5 Summary of P-recovery technologies

By 2019 more than 80 WWTPs (60 of which were municipal WWTPs) recovered phosphorus in full-scale with different types of implementations and processes (Table 2). Most of the applied processes used struvite (MAP) or calcium phosphate (HAP) crystallisation to produce phosphorus in bio-available forms (Shaddel *et al.*, 2019). Additionally, many WWTPs and industries have shown interest in the P-recovery methods, especially the extraction of phosphorus from SSA to incorporate recovered P in the marketable products (Egle *et al.*, 2016). One of the main concerns at the moment is financial feasibility and realistic and comprehensive cost-analysis is often very difficult to produce. There are more than 50 different approaches to P-recovery, many of them still in laboratory scale and scientific publication of the technologies often lack financial information. This makes evaluating potential technologies and approaches difficult not only to operators and potential investors, but also decision makers (Egle *et al.*, 2016).

***Table 2** – Overview of the global P-recovery practices and projects* (Shaddel *et al.*, 2019)



Geographical Area	Plants	Technology/Product	Remarks
North America	More than 15 full-scale units mainly producing struvite	Pearl, Multiform, Airprex see [10]	Lack of economic drivers and regulations for nutrient recovery [10]
Europe	Germany (10), Netherlands (10), Belgium (6), France (2), Spain (2), Italy (1), UK (2), Denmark (4)	Variety of technologies including Airprex, Anphos, Elophos see [10]	Mainly producing struvite from both municipal and industrial wastewater
China and India	China (Tianjin, Nanjing), India nor record found	Airprex, Crystalactor	China is planning new facilities with biological phosphorus removal and anaerobic digestion for energy recovery
Africa	No plants, feasibility studies in South Africa on P-recovery from sludge and source separated urine	NA	High costs, immature market and lack of acceptability
Japan	16 full-scale plants producing struvite and calcium phosphate	Gifu, PHOSNIX	Strong nationwide collaboration on market development, integrated production from steel, agriculture and chemical industries

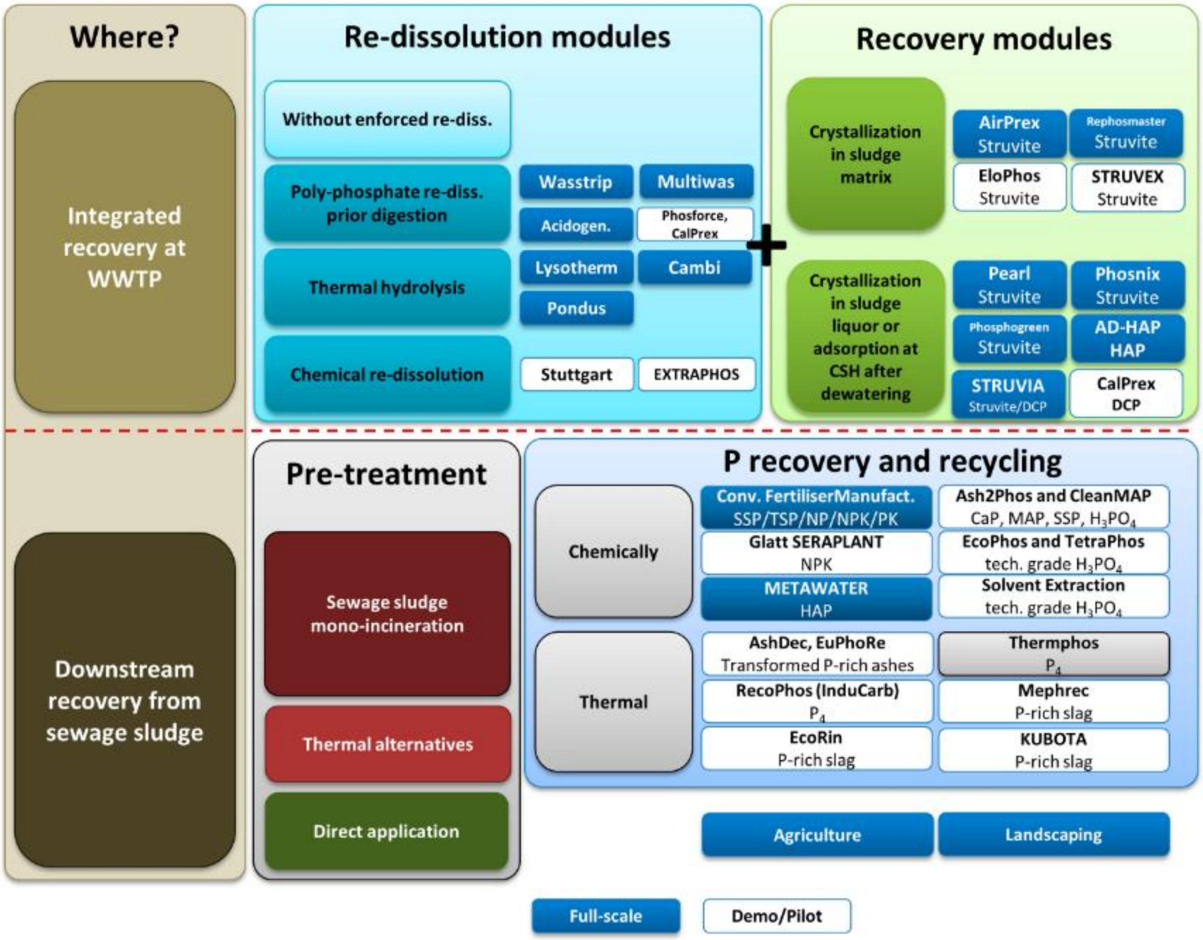


***Figure 3** – Technologically applied concepts of P-recovery in different WWTPs* ((Kabbe, 2019; Shaddel *et al.*, 2019)

In the context of this document, the main attention is given to full-scale applications of technologies and processes, while applications with published feasibility information are given extra emphasis. As many of the full-scale applications are based on similar technologies (Figure 3), the main processes have been identified and will be summarised in this section (MAP process, sorption processes, wet chemical extraction, thermo-chemical extraction, etc), with example cases shown in section 3.6.

3.5.1 Struvite crystallisation

MAP (magnesium-ammonium-phosphate, also known as struvite, $MgNH_4PO_4 \cdot 6(H_2O)$ precipitation is one of the most common P-recovery methods. The process can take place unintentionally in many WWTPs with digesters and high alkalinity as the two main parameters facilitating the crystallisation are magnesium (especially magnesium oxide MgO as a precipitation agent) and high pH (over 8.0 used in many technical applications). This process is feasible for different streams with high ammonium ion concentrations, such as sludge liquor, pig wastewater, landfill leachate and urine (Dai *et al.*, 2018). The struvite precipitation reaction can be seen on Equation 1.

$$\mathrm{Mg^{2+} + NH_4^+ + PO_4^{3-} + 6H_2O \rightarrow MgNH_4PO_4 * 6(H_2O)}$$

Equation 1 *- Struvite aka magnesium-ammonium-phosphate (MAP) precipitation*
(Doyle and Parsons, 2002)

Struvite grows as orthorhombic crystals (into all 3 axis directions under right angles, growths on different axis can differ in size). This formation of crystals is controlled by pH, degree of supersaturation, temperature and the presence of other ions (calcium) and it occurs in high magnesium, ammonium and phosphate ion concentrations based on equilibrium solubility. The link between struvite formation and the availability of ions are directly linked to pH values, as the solubility of struvite reduces and more precipitation occurs at higher pH values (Doyle and Parsons, 2002). As the ammonium ions are stripped due to the formation of free ammonia at higher pH values (starting from pH 8, while becoming limiting at pH values greater than 9.8), the most efficient struvite precipitation pH ranges are often considered between 8 and 9.5 (Booker, Priestley and Fraser, 1999).

Co-precipitation of other mineral components also happens during struvite precipitation and can cause negative impacts to product purity, heavy metal concentrations and phosphorus recovery rates. Main ions interfering with struvite formations are Al^{3+}, Ca^{2+} and Fe^{3+}. Main competitors for precipitation are brushite ($CaHPO_4 \cdot 2H_2O$) and brucite ($Mg(OH)_2$). Other minerals such as amorphous calcium phosphate (ACP, $Ca_3(PO_4)_2$), hydroxyapatite (HAP, $Ca_5(PO_4)_3OH$), magnesite ($MgCO_3$), newberyite ($MgHPO_4$), bobierrite ($Mg_3(PO_4)_2 \cdot 8H_2O$), dolomite ($CaMg(CO_3)_2$), octacalcium phosphate (OCP, $Ca_8H_2(PO_4)_6 \cdot 5H_2O$) and tricalcium phosphate (TCP, $Ca_3(PO_4)_2$) can all be co-precipitated in different conditions, while the precipitation rate should be lower than of struvite (Hallas *et al.*, 2019).

The formation of struvite crystals often needs a seed material, onto which the struvite crystallizes. The seed material purity can greatly influence the purity of the product, while crystals grow better on crystal structure, common materials include quartz, silica sands and anthracite (Doyle and Parsons, 2002). Technologically, the most common struvite precipitation reactors are fluidized bed reactors, specific processes and applications can be seen from the case study section of the document. Besides recovery from sludge liquor, the MAP process has been successfully applied to extract phosphorus from sludge (before dewatering), with extra washing step to separate the crystals from the sludge (Schaum, 2018).

MAP process can also result in significantly lower nutrient concentrations in return streams to biological treatment as both phosphorus and nitrogen are precipitated. At the same time, the need for high ammonium ion concentration makes the process unfeasible for municipal wastewater (Dai *et al.*, 2018).

High profile case studies about the use of MAP process can be read in sections 4.2.1 and 4.2.2.

3.5.2 HAP crystallisation

The HAP (hydroxyapatite, aka calcium phosphate - $Ca_5(PO_4)_3OH$) method is a suitable method for P-recovery from wastewaters, as relatively low P concentrations are needed (2-100 mg/L) (Schaum, 2018) and no ammonium ions are needed (Dai *et al.*, 2018). Similarly to MAP precipitation, seed crystal is needed for effective crystallisation and materials such as sands, calcite, apatite and cow bone have been used (Chen *et al.*, 2009). The precipitation process to seed crystals is described in Equation 2.

$$3\mathrm{HPO}_4^{2-} + 5\mathrm{Ca}^{2+} + 4\mathrm{OH}^- \rightarrow \mathrm{Ca}_5(\mathrm{OH})(\mathrm{PO}_4)_3 + 3\mathrm{H}_2\mathrm{O}$$

Equation 2 *– Hydroxyapatite (HAP) precipitation*
(Joko, 1985)

HAP precipitation has multiple advantages – in most cases no precipitants are needed (or in low amounts), product can be dewatered easily, and dehydrated product can be directly used as a phosphorus fertiliser. The formation of hydroxyapatite crystals is affected by phosphate concentration, calcium concentration, bicarbonate concentration and pH, the last of which has a very complex effect. Similar to MAP, HAP precipitation needs alkaline conditions, while on pH values over 9, the bicarbonate ions can partially be converted to carbonate ions (via equilibrium reaction), which can cause competing precipitation of calcium carbonate (Chen *et al.*, 2009). The ideal Ca/P value by calculations has been shown as 1.67, while calcium in excess doesn't increase phosphorus recovery efficiency (Dai *et al.*, 2018).

The process can be affected by other metal ions in the solution as well – for example high Mg^{2+} ions can change precipitation towards MAP (depending on the concentration of ammonium ions), while the overall phosphorus recovery efficiency isn't changed. Both high Cu^{2+} and Zn^{2+} concentrations can start the co-precipitation of other minerals, causing both HAP and overall phosphorus recovery efficiency to decrease slightly (around 5% reduction in P-recovery efficiency (Dai *et al.*, 2018).

With different seed materials, the generated crystallisation product can have P content over 10%, comparable to phosphate rock (Chen *et al.*, 2009). Based on different additives and Ca/P ratio, effluent phosphorus concentrations of 0.5 mg/L should be quite easy to achieve in higher pH values (Dai *et al.*, 2018), while in perfected conditions concentrations as low as 0.03 mg/L could be possible (Joko, 1985).

3.5.3 Vivianite magnetic separation

Vivianite is one of the main iron phosphate minerals ($Fe_3(PO_4)_2 \cdot 8(H_2O)$) forming during the digestion of sewage sludge when enough iron is present in the system (Wilfert *et al.*, 2018). The vivianite particles have magnetic properties, which could give a strong alternative technology to previously discussed crystallisation and precipitation technologies, especially for WWTPs using chemical phosphorus removal by dosing iron and using anaerobic digestion. Vivianite has shown to form under iron-rich, non-sulphidic conditions in close proximity to organic material (Rothe, Kleeberg and Hupfer, 2016).

The vivianite magnetic separation technologies are quite new and full-scale test applications are currently ongoing. Similar to struvite, vivianite is also responsible for mineral build-up on pipe walls, reducing the flow rates and increasing maintenance requirements in WWTP (Antoniewicz, 2018). Intentional vivianite extraction can result in extra savings due to the indirect effect on maintenance requirements. The pilot tests so far have also shown a possibility to recover iron after the magnetic separation of vivianite to reuse it as a chemical agent for chemical phosphorus removal.

In order to make the technology feasible, the amount of iron dosed in the WWTP would often need to be increased. Scientific research has shown that the increase in the share of phosphorus present as vivianite is directly proportional to the increase of Fe. While this could mean extra costs, higher iron content during anaerobic digestion also lowered the formation of H_2S and decreased the soluble P concentration in the digester effluent. No negative effects to other removal efficiencies, biogas production or sludge dewaterability has been shown as a result (Prot *et al.*, 2020), therefore the technology shows great applicability.

Vivianite however has somewhat limited bio-availability and its potential as a fertiliser is low (Schütze, Gypser and Freese, 2020). To mitigate that, vivianite re-processing into other phosphorus compounds would be necessary.

This technology is currently being developed by KEMIRA under the name Vivimag (case study in section 4.2.3).

3.5.4 Sorption processes

Alternative to mineral precipitation, phosphorus recovery from liquid state could also be possible via sorption. Different methods have been used, such as electric and chemical sorption and ion exchange process (Schaum, 2018) and may have effective uses for low phosphorus concentration wastewaters. At the same time, sorbents and ion exchange membranes can be costly and needing regeneration, therefore low-cost materials (possible waste materials) would have the best possible financial feasibility (Loganathan *et al.*, 2014).

Five different mechanisms of phosphate sorption have been described (Loganathan *et al.*, 2014):

a. Ion exchange (outer-sphere surface complex);
b. Ligand exchange (inner-sphere complex);
c. Hydrogen bonding;
d. Surface precipitation;
e. Diffusion into the sorbent.

Although different sorption processes, especially if low-cost sorbent from different waste streams can be used, could be a very potential P-recovery technique, at the current rate these processes have mostly been applied in laboratory tests. Use of sorbents needs to be tested a lot more in larger scales, in continuous column trials and using non-synthetic feeds, with emphasis on the costs for sorbent regeneration and actual phosphorus recovery technologies from the sorbent (Loganathan *et al.*, 2014). Some demonstration tests have been conducted in Japan showing potential phosphorus decrease in the effluent (from 1.0 mg/L to 0.03 mg/L) and high P-content recovered material (16%) (Schaum, 2018), while more thorough publications about these demonstrations hasn’t been published in English.

3.5.5 Wet-chemical leaching

Wet-chemical leaching is one of the most common chemical extraction processes for P-recovery from waste activated sludge and SSA (Egle *et al.*, 2016), while acidic and alkaline dissolution techniques are distinguished. Acidic dissolution is usually considered more efficient, while has a greater dissolution of heavy metals together with phosphorus, which needs to be separated later. Phosphorus is usually obtained via the use of mineral and organic acids, the least expensive of which is considered to be H_2SO_4, while many others such as HNO_3, HCl, citric and oxalic acids can be used. H_3PO_4 can also be used, while is considered the most expensive alternative. The purity of the extracted material is better with sulphuric acid due to fewer heavy metal complexes, especially compared to HCl leaching (Cieślik and Konieczka, 2017).

While the theoretical chemical demand for P-recovery is 3 mol H^+/mol P, in practice the media also contains other acid-soluble components such as calcium, magnesium, iron- potassium and aluminium oxides, calcium carbonate and calcium hydroxide. In different tests the acid demand can be from 4.4 to 14.1 mol H^+/mol P depending on the source material and used acid (Petzet, Peplinski and Cornel, 2012). Both the amount of impurities and phosphorus recovery efficiency also depend on the extraction time, as almost 100% phosphorus recovery is obtainable with 2-4 h time, while up to 100 min is considered optimal due to impurities (Cieślik and Konieczka, 2017). Other processes, such as sequential precipitation, liquid-liquid extraction, sulfidic precipitation, cationic ion exchange and nanofiltration could be used to remove cation impurities from the extracted solution, while applying any of them comes with extra financial costs (Petzet, Peplinski and Cornel, 2012).

Alkaline leaching is often considered to be less efficient and can greatly depend on the composition of minerals in the sludge. At the same time, heavy metals re-dissolution in alkaline conditions is minimal, therefore a decontamination step can be avoided and clean product delivered (Schaum, 2018). The efficiency of P-recovery depends mostly on the concentrations of Al and Ca – Al-P compounds dissolve and are later precipitated as Ca-P compounds via the addition of $CaCl_2$. When calcium content is already high, the dissolution efficiency reduces quickly. The highest efficiencies reached with Ca-poor and Al-rich SSA have been 75%, while with high Ca it can be reduced to 0-35% (Petzet, Peplinski and Cornel, 2012). The processes often use either NaOH or KOH as the leaching agents and temperature ranges of 50-90° C (Schaum, 2018).

In the marketed and full-scale applications, the wet-chemical leaching process might be more complicated – often multiple stages of leaching or even combinations of the acidic and alkaline processes can be used. As mentioned in previous sections, the applications of many P-recovery methods have to be adapted to a specific WWTP to take into account the different contents of minerals in SSA or precipitation chemicals for sewage sludge. This also means the financial feasibility of a process might depend greatly on the local conditions and can be difficult to predict without specific analysis of local streams.

An innovative new approach to wet-chemical leaching called the RAVITA process can be found as a case study in section 4.2.4.

3.5.6 Wet-oxidation and supercritical water oxidation

In order to recover phosphorus from the sludge solids, different thermochemical processes can also be used, mainly focusing on higher temperature and pressure water for the destruction of the solid particles. Both wet-oxidation and supercritical water oxidation are under this category, while the first uses water in subcritical conditions (lower energy demand) and latter in actual supercritical conditions (great increase of organic substance solubility) (Egle *et al.*, 2016). Compared to chemical leaching, these processes are considered more environmentally safe, less time and capital consuming, while with higher operational (energy) costs (Munir *et al.*, 2019).

Water enters the supercritical state after passing the critical point (375 °C and 220 bar) and attains properties between a gas and a liquid. As mentioned before, the solubility of water changes significantly, with inorganic substances becoming insoluble while the solubility of organic substances can reach almost 100%. As the diffusivity and ion mobility are higher oxygen completely mixes with water and the resulting homogenous mixture can rapidly oxidize organic compounds without usual limitations, resulting in complete destruction of molecules in less than 60 seconds. The process is exothermic and can possibly supply itself with energy and during it phosphorus is oxidized into P_2O_5 and later dissolved and precipitated (up to 90% of P recovered) (Stendahl and Jäfverström, 2003).

During the subcritical wet oxidation process, the water-sludge-oxygen mixture is heated to 150-320 °C and 20-150 bars. Phosphorus is released to the aqueous phase and organic acids are produced from sludge (Munir *et al.*, 2019). Different from supercritical oxidation, substrates are not completely oxidized (complete oxidations turns organic carbon into CO_2, nitrogen into N_2, phosphorus into P_2O_5 (Stendahl and Jäfverström, 2003)), which can potentially be beneficial to other sludge treatment processes in the integrated system. High concentrations of volatile fatty acids (VFAs) are produced (Munir *et al.*, 2019), which can potentially be used for production of biogas/other valuable materials. The P-recovery during wet oxidation (up to 95%) is achieved with subsequent struvite precipitation, the mechanisms of which are covered in section 3.5.1.

3.5.7 Thermo-chemical processes

One of the common P-recovery solutions from SSA is thermochemical treatment, which is mainly used to evaporate heavy metals from the ashes and achieve a cleaner product. The process can also greatly increase the bioavailability of phosphorus, which as previously discussed is low in untreated SSA (Cieślik and Konieczka, 2017). As organic pollutants are already destroyed during incineration, the thermochemical treatment product can have very low environmental risk-factor.

On higher temperatures (800-1000 °C) most the heavy metal oxides have low vapour pressures and therefore do not evaporate readily. With the conversion of oxides to chlorides (via the addition of HCl, KCl, $MgCl_2$ or $CaCl_2$) the evaporation temperatures are lowered considerably, allowing large fractions of heavy metals to be removed that way (Adam *et al.*, 2009). The Cd and Pb chlorides as easily volatile, Cu and Zn semi-volatile while Cr and Ni are not very volatile. The effect greatly depends on temperatures - at higher temperatures the efficiency increases significantly (Havukainen *et al.*, 2016). At the same time, the phosphorus bioavailability decreases significantly at the melting point of ashes, therefore up to a 1000 °C are commonly used.

The bioavailability of phosphorus is commonly tested in the 2% citric acid solubility test. The conventional super phosphate fertiliser has a solubility of 87% while the raw ashes usually fall between 25-40%. With secondary thermochemical treatment, the bioavailability of phosphorus can increase significantly and depending on the temperature high bioavailability indicators can be achieved (86%, 93% and 97% at 800, 900 and 1000 °C respectively) (Adam *et al.*, 2009). Therefore, the residue of SSA secondary thermochemical treatment could be use directly as a fertiliser and might have a better bioavailability than the conventional fertilisers. The process however has significant energy and fuel consumptions, especially considering the full process scheme of drying-incineration-thermochemical treatment and the price of the recovered product is currently in most cases not market-competitive (Havukainen *et al.*, 2016).

An example about thermo-chemical P-recycling from SSA, the case study of Ash Dec® by Outodec can be found in section 4.3.2.

3.5.8 Thermo-electrical processes

Thermo-electrical method often refers to the electrical resistance heating of the furnace. Electric heating is easy to control and high temperatures can be attained, while the method (taking into account the electricity generation processes), is very energy inefficient. While being phased out in municipal use, the electric heating still has benefits for industrial use as the easy control of high temperature furnaces gives enables efficient optimisation of processes.

Thermo-electrical phosphorus recovery uses very high temperatures generated in an electrically heated furnace (1500 °C), during which phosphate is reduced to P_4 (aka white phosphorus). This gaseous stream leaves the furnace together with CO and dust and can later be electrostatically precipitated. This process has also been used for phosphate extraction from phosphate rock and pre-existing factories might be used with SSA as phosphate rock substitute (Schipper *et al.*, 2004).

When SSA is used for the process, the impurities of the sludge are vital to the efficiency – iron is one of the main negative influences, being reduced to a mixture of FeP and FeP_2 and reducing the yield of white phosphorus (from 90% of recovery to as low as 50%). As iron is often used in the WWTP process as a precipitation agent, the resulting SSA might be unsuitable for the process (Fe/P ratio over 0.2 is deemed unsuitable) (Schipper and Korving, 2009). Other parameters with a negative effect to the process include volatile metals (Zn, Pb, Cd, Sn), which can also be partly evaporated; chloride causing corrosion to the system; and copper affecting the quality of the final product (Schipper *et al.*, 2004). Using The Netherlands as an example, around 17% of the produced municipal SSA was suitable for the process, while with the change to Al-based precipitation salts this number could reach around 45% (Schipper and Korving, 2009).

3.5.9 Metallurgical processes

Metallurgical processes usually mean working with shaping or extracting metals, although some of the processes can as a co-product produce extracted phosphorus as well (Egle *et al.*, 2016). The smelting gasification technology, also called metallurgic melt-gassing can for example produce a raw off-gas for power generation, iron metallic phase and liquid phosphorus slag with high bioavailability and low heavy metal content (Adam *et al.*, 2015). These types of processes can be used for both sewage sludge and SSA and the process temperatures are from 1500 – 2000 °C (higher than thermochemical and thermoelectrical processes). Depending on the specific process, heavy metals can be captured in the resulting iron alloy or evaporated via the addition of chlorides (discussed in section 3.5.7) and phosphorus can either be separated via the gas stream or as a liquid slag. The recovery potentials are reported up to 90% and both phosphoric acid and white phosphorus can be produced (Schönberg, Raupenstrauch and Ponak, 2018).

The metallurgic processes need feedstock with low water content (at least 80% DM), while for subsequent high-calorific off-gas production low temperature drying processes are recommended. The energy potential is one of the main benefits of this type of process, which can potentially off-set the high energy demand of the technology. The metallic iron slag can be also recovered and reused, especially with technologies using heavy metal evaporation and collection from the dust and flue gases (Adam *et al.*, 2015).

3.6 N-recovery

Although the main emphasis in nutrient recycling is currently on phosphorus, nitrogen is also a necessary nutrient and a key part of fertilisers. While the natural cycle of phosphorus is limited (endpoint to marine sedimentation) and may one day bring forth easily accessible phosphorus scarcity, nitrogen cycle is much more complex. Inert nitrogen is also a major part of our atmosphere and nitrogen can be produced via N_2 fixation into ammonia, while requiring considerable energy input. Therefore, looking at wastewater treatment plants, where ammonia is directly converted back to N_2 and lost to the atmosphere, nitrogen recycling could be viable in the future. First commercially applicable technologies are being developed already now (EasyMining Services Sweden AB, 2020), while the financial feasibility of the practice is still under question.

Nitrogen is also a key component of multiple environmental problems: a key factor in eutrophication (Elser *et al.*, 2007) and a potential contributor to greenhouse effect as N_2O is also emitted during biological treatment. While the amount of N_2O emitted during aerobic phase of wastewater treatment contributes to about 3% of total anthropogenic N_2O emissions, the stream makes up 26% of total greenhouse gas emissions of the water sector (Kampschreur *et al.*, 2009). As both energy and climate neutrality are key factors in the near future of the sector, these emissions need to be reduced – giving way to N-recovery as a potential problem solver.

Some promising technologies for the nitrogen recovery are reject water air stripping (24% N-recovery) and vacuum or hydrophobic membrane filtration (both around 75% recovery). Other processes, such as struvite precipitation or thermal sludge drying air treatment, can recover small amounts of nitrogen as well (1.1% and 2.1% respectively in in-situ tests), while the processes are not used for N-recovery. Completely separate approach would be the separation of urine and faecal matter, which would allow to reduce N-load to WWTPs up to 60%, while requiring a completely new infrastructure (van der Hoek, Duijff and Reinstra, 2018).

Nitrogen in the sludge matrix is also recovered with direct use of sludge, while as previously discussed these approaches may not be viable in the future due to possible contamination with micropollutants. Incineration however completely evaporates nitrogen; therefore, direct use of ash or ash products only recycle phosphorus. Another way to recovery nitrogen would be the production of materials from the wastewater or sludge. Although not used in larger scales, pilot tests have shown recovery potential of cellulose from wastewater influent solids (Ruiken *et al.*, 2013) and protein production from sludge (Matassa *et al.*, 2016), which both recover significant loads of nitrogen. As WWTPs potentially move towards material recycling plants in the future, these approaches may be considered, while at the present the economic viability is questionable.

An overview of different technologies and approaches connected to N-recovery are presented on Figure 4. Most of these technologies however need significant changes in the wastewater treatment systems, as only a limited recovery is possible with integrated solutions to the current traditional layouts. Separate urine collection and treatment is one of the most significant options, while requiring completely new infrastructure and sewer systems (van der Hoek, Duijff and Reinstra, 2018). Therefore, nitrogen recovery is not deemed financially profitable at the present, as the main driver of reducing N_2O emissions is not attractive and profitable enough to carry large investments.

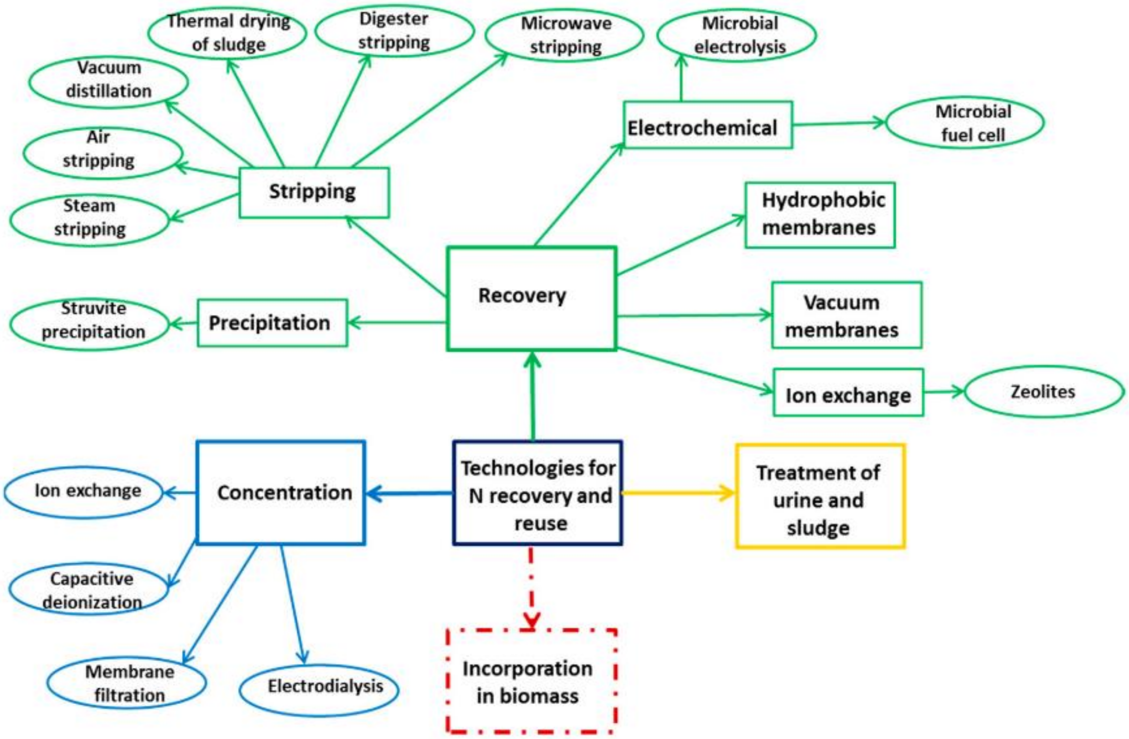


Figure 4 *– Overview of strategies with related technologies for nitrogen recovery and reuse*
(van der Hoek, Duijff and Reinstra, 2018)

Looking at the impact of N-recovery from wastewater, the main benefit would be to potential reduction of greenhouse gases produced in the treatment process. With the current tests, the recovery does not show signs of negative influences to biogas production or any other processes and could result in significant savings in aeration energy in the biological treatment (van der Hoek, Duijff and Reinstra, 2018). More conclusive data would be needed for integrated solutions to the current process scheme, such as different filtering systems. In some cases, especially for industrial wastewater treatment plants, the solutions could already be financially worthwhile due to reduction of WWTP operating costs.

Nitrogen recovery from wastewater and sludge could become more important in the future with more strict quotas on greenhouse gas emissions and good legal groundwork for recycled product market. When building completely new urban sewer networks and treatment plants, separate collection of urine should be considered already today. At a larger scale however, the development of P-recovery systems has shown the difficulties with recycled products, especially problems with legislations, marketability and profitability. Taking the background into account, large scale nitrogen recovery from the wastewater treatment plants is much less viable than phosphorus recovery, at least until significant changes are made to the global approach to wastewater and sludge treatment.

4. Cases of nutrient reuse and recovery

As discussed before, phosphorus recovery technologies have been developed rapidly for the last decades, while the full-scale implementation of them is still only beginning due to many legal, financial and technical barriers. The main products from phosphorus recycling are struvite (MAP), calcium phosphate (HAP) and phosphoric acid, while struvite precipitation is by far the most used technology to date. The distribution of different technologies and products, showing the large variety of different technological processes in development can be seen from Figure 5.

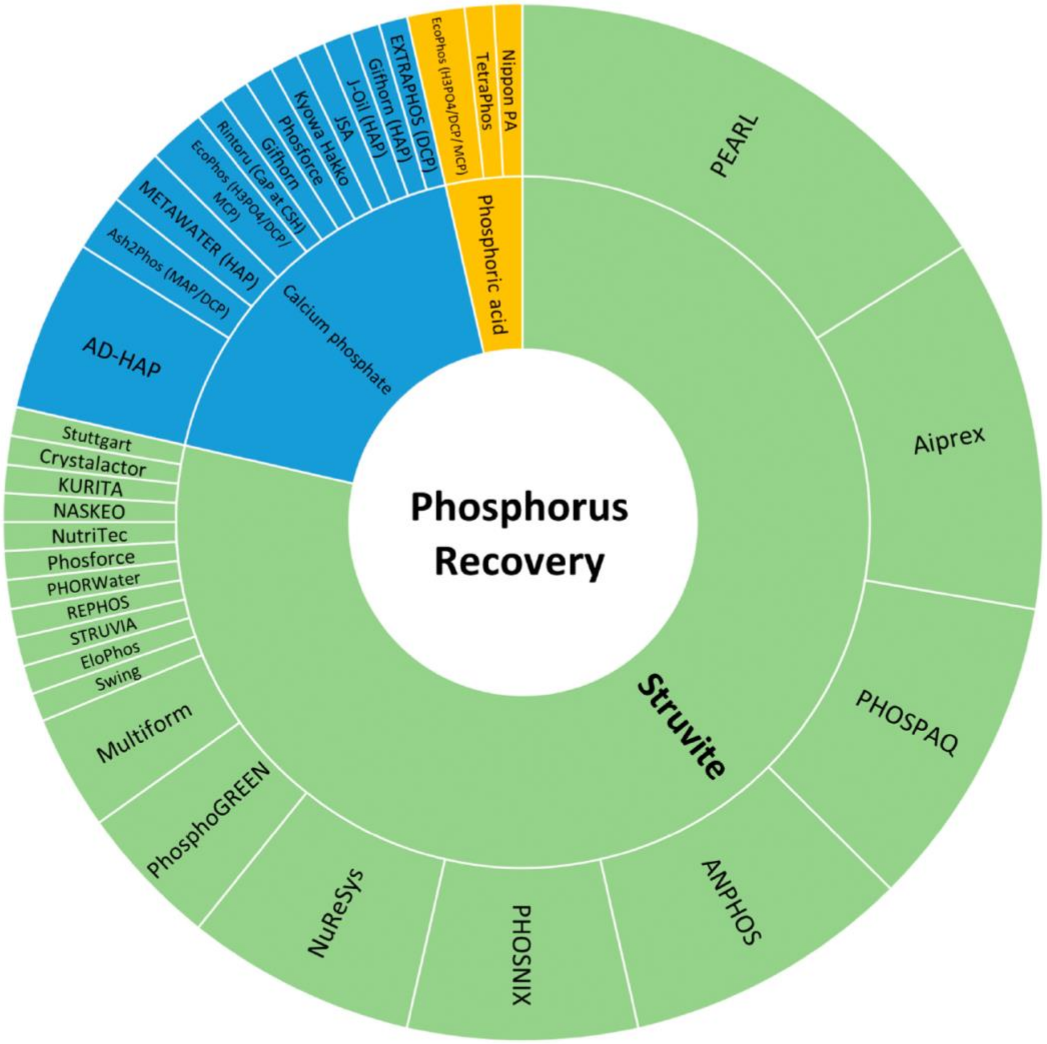


Figure 5 *– Main products and related technologies of phosphorous recovery (Reproduced)*
(Shaddel *et al.*, 2019)

As the current interest in phosphorus recovery is very high many different companies are aiming at new technical solutions and new products, with mostly laboratory and other small-scale tests. This rapid development and competitive nature of the industry also means the amount of information published, especially cost-efficiency and other financial data, is often severely lacking. Therefore, this chapter is focused on the technologies already in practice and high-promise developments with good, published information. Each principal solution is described by one sample case and a comprehensive overview of all technologies and trademarks will not be presented in this document.

The widely used technologies of direct use are also described in this chapter, as many of the more complex technologies are only suitable for medium and large WWTPs. Direct use could still be in consideration for smaller WWTPs, therefore cases for the main technologies currently in use, such as composting and humification are presented.

4.1 Direct reuse of nutrients from sewage sludge

Direct use of treated sewage sludge is still a viable option for most of the Baltic Sea region, especially for smaller WWTPs. During the recent decade, there has been a tendency towards ceasing direct use and seeking other alternatives, mainly due to a wide variety of pollutants in the wastewater and sludge that can potentially cause environmental harm. As these pollutants include pathogens, heavy metals, organic micropollutants and microplastics, the analysis of the safety of the sludge is almost impossible due to the sheer number of different analytes. Germany for example has directly banned the direct use of sludge for large WWTP-s. The current WWTPs have a 12- or 15-years transition period (for plants over 100 000 PE or 50 000 PE respectively) to start recycling phosphorous from sewage sludge with other methods.

Current conventional sludge treatment technologies stabilize sludge and can achieve hygienisation by concomitant processes during stabilisation or by special technological processes. Testing the content of heavy metals in the sludge has become affordable and today most municipal WWTP-s have reduced heavy metal content in the wastewater. Recent studies (including IWAMA project sludge benchmarking and audit) have demonstrated, that the WWTP-s of the BSR have relatively low heavy metal content in the sludge and could conform to the limit values defined for usage of biowaste (Saveyn and Eder, 2014).

Contrary to pathogens and heavy metals, organic micropollutants are often not degraded in the conventional processes and therefore can be unpredictable. Measuring the wide variety of organic contaminants is difficult, expensive and due to the lack of background data often inconclusive. This problem with both known and unknown pollutants is one of the main reasons the perspective on the direct use of sludge has decreased.

One of the main issues however is that most of the phosphorus recycling technologies attain potential financial feasibility only on large scales. Very few tested methods are suited for WWTPs below 50 000 or even 100 000 PE, which greatly reduces the possible options. Centralisation of all sludge treatment in one big facility could be a possible option for densely populated areas, but large parts of the BSR are scarcely populated. Therefore, until a cheap and very well scalable technology for P-recovery is developed, the only recovery option left for small WWTPs is the direct use of treated sludge.

4.1.1 Composting of sewage sludge – Helsinki HSY

Composting of the sewage sludge is a widely used technology for the stabilization and hygienisation of sewage sludge. Although composting in the ambient conditions is often associated with smell problems, the main advantage of the technology is the hygienisation achieved in the process due to elevated process temperature (above 60°C). Furthermore, composting has been also shown to reduce the concentration of some POP-s. During sludge or manure composting, the antibiotics removal range is between 17–100% (Ezzariai *et al.*, 2018).

The composting result is usually a stable soil-like product, which can be used as a fertilizer (soil improvement product) in agriculture, as well in greenery or recultivation as lawn soil or garden soil. In the BSR, sewage sludge composting is widely used in Finland, Estonia and Sweden. These countries have also established their own assurance systems to control the process and product quality. In Sweden the Swedish Water and Wastewater Association in cooperation with other organisations has created a voluntary REVAQ sludge certification system. In Estonia a certification system has been established on the legal basis for End of Waste for sewage sludge. These quality assurance systems are tools to facilitate sewage sludge compost of good quality to be directly recycled for agriculture and greenery.

Although, composting is mostly used in medium size WWTP-s (2 000-100 000 PE), is can used also in large sludge treatment facilities, such as the Metsäpirtti composting site in Helsinki.

Helsinki Region Environmental Services Authority (HSY) is the environmental body operating water and wastewater services for more than one million residents of the Helsinki metropolitan area. Since 1994 HSY operates also sludge composting site at Metsäpirtti, where about 85 000 t of sludge (TS content of 30%) is treated yearly (Väänanen, 2018). The composting site spans on 18 hectares (Figure 6) and is covered with watertight surface. The runoff water is collected and treated in Viikinmäki WWTP (Helsinki).

The quality assurance control system at the composting site is well established and complete with the tracking of each product batch, including the composition, achieved temperatures, stabilisation and hygienisation parameters. The quality control of the incoming sewage sludge is part of Viikinmäki WWTP continuous control system. The reporting of the process is done annually to the Finnish Food Safety Authority (EVIRA).

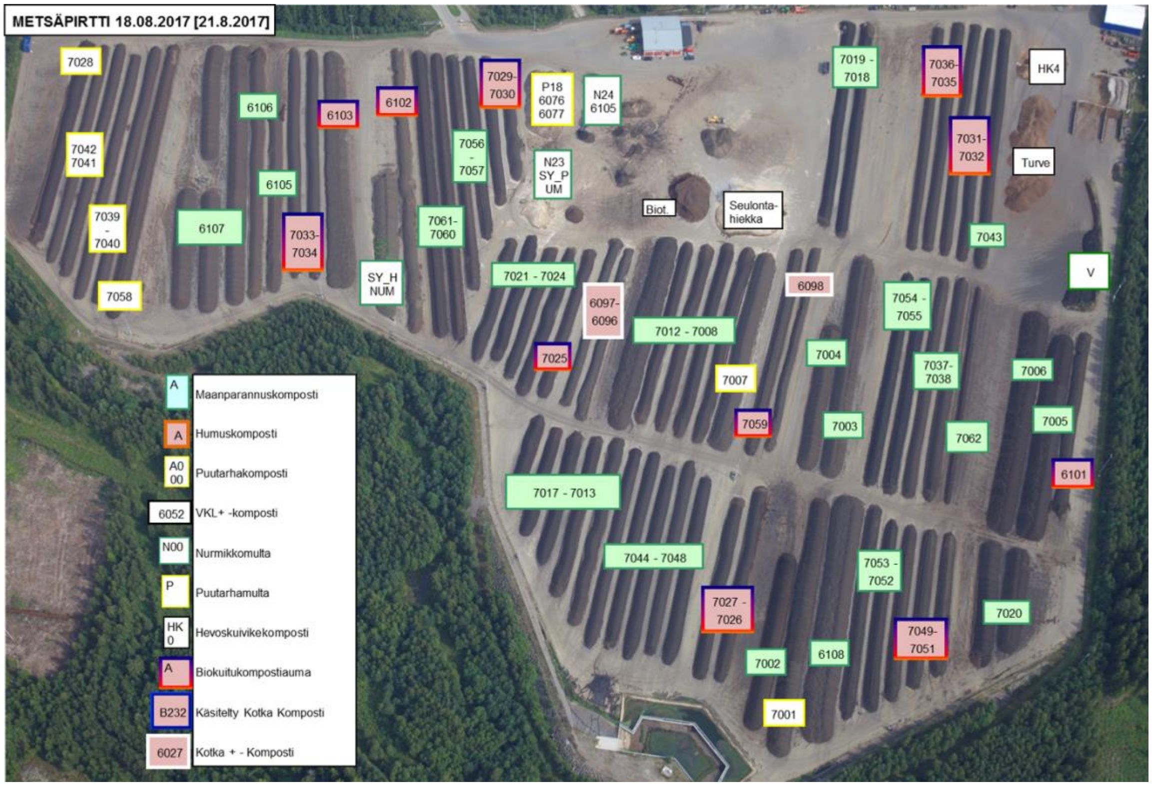


***Figure 6** – Identification numbers of windrows at Metsäpirtti composting site*
(Väänanen, 2018)

HSY sells three different compost products (data from 2016): lawn soil (48%), garden soil (18%) and soil improvement compost (34%). In 2016 the total sales of compost from the Metsäpirtti composting plant were 120 500 m^3. As the heavy metal content of wastewater has been decreasing in Finland during the last years, the accumulation in sludge is low (Table 3).

Annual operation cost of the composting site is around 4,7 M€, out of which personnel costs 6%, material costs 34% and outsourced services 60%.

- Unit cost of the sludge is about 38 €/t of sludge.
- Income from compost sales is 1,6 M€.
- Composted soil price including VAT and transport is about 20-33€/ m^3.
- Compost prices range between 2-5 €/ m^3 depending on the load size and delivery location.

***Table 3** – The highest permitted heavy metal concentrations of farming sludge and sludge mixture comparing to the concentrations of Metsäpirtti lawn soil* (Väänanen, 2018)

	Limit values mg/kg of solid matter	Average values of analytical data 2010-2013	Lawn soil concentration's percentage of the limit value
Arsenic	25	3.8	15%
Cadmium	1.5	0.24	16%
Chrome	300	17	5.7%
Copper	600	91	15%
Mercury	1.0	0.13	13%
Nickel	100	8.5	8.5%
Lead	100	7.8	7.8%

4.1.2 Thermal conversion and pyrolysis of sewage sludge – PYREG®

Pyrolysis is thermal technology during which partial degradation of organic material takes place in the absence of oxygen. The temperature of the pyrolysis stage is usually between 500°C and 700°C. The specific products of pyrolysis vary depending on the process temperature and include gaseous (syngas), liquid (biofuels) and solid fractions (biochar). Pyrolysis is considered to be a technology with very high potential for phosphorus recycling, as different organic micropollutants also carbonize at higher temperatures. This can result in greatly reduced POP concentrations in the products and lower associated environmental risks when using the products for fertilization.

Although pyrolysis is a common process for biofuels production from wood and biowaste (around 326 active biochar companies based on 2015 Internation Biochar Initiative report (International Biochar Initiative)), pyrolysis of sewage sludge is not a very common practice. While pyrolysis has been lately considered as one of the technologies for P-recovery from sewage sludge, only a few companies (PYREG® and AVA Cleanphos®) have been currently developing and testing such applications.

PYREG GmbH is a company providing various systems and technologies related to carbonisation of biomass and sewage sludge. The continuous process can reach energy autonomous state as the syngas produced during the pyrolysis can be used as a fuel in the process itself (PYREG GmbH, 2020).

The PYREG® process uses the principle of dried carbonisation, which means dewatered or pre-dried sludge (>50 % DS) is fed into a reactor heated to 500-700 °C. Sewage sludge goes through both the degassing and carbonisation processes, greatly reducing the total quantity. The resulting product is called carbonate-ash, in which micropollutant concentrations are lowered and pathogens are destroyed due to thermal hygienisation effect of the process. The concentration of P in the final product is up to 15% P_2O_5 and the plant-availability is around 80% (Eliquo Technologies, 2020).

Syngas forming as a result of the degassing and carbonation reactions in this slow pyrolysis process is subsequently burned in a chamber with a FLOX®-burner at 1,250°C. Low-NO_x producing burner technology has been developed, which avoids secondary gas cleaning. As an example, PYREG® compact module P500 has an annual throughput of 1100 t DS, producing around 610 t of the carbonate-ash (P content 10-20%). The power consumption is around 18 kW electrical energy and up to 150 kW of thermal energy can be produced.

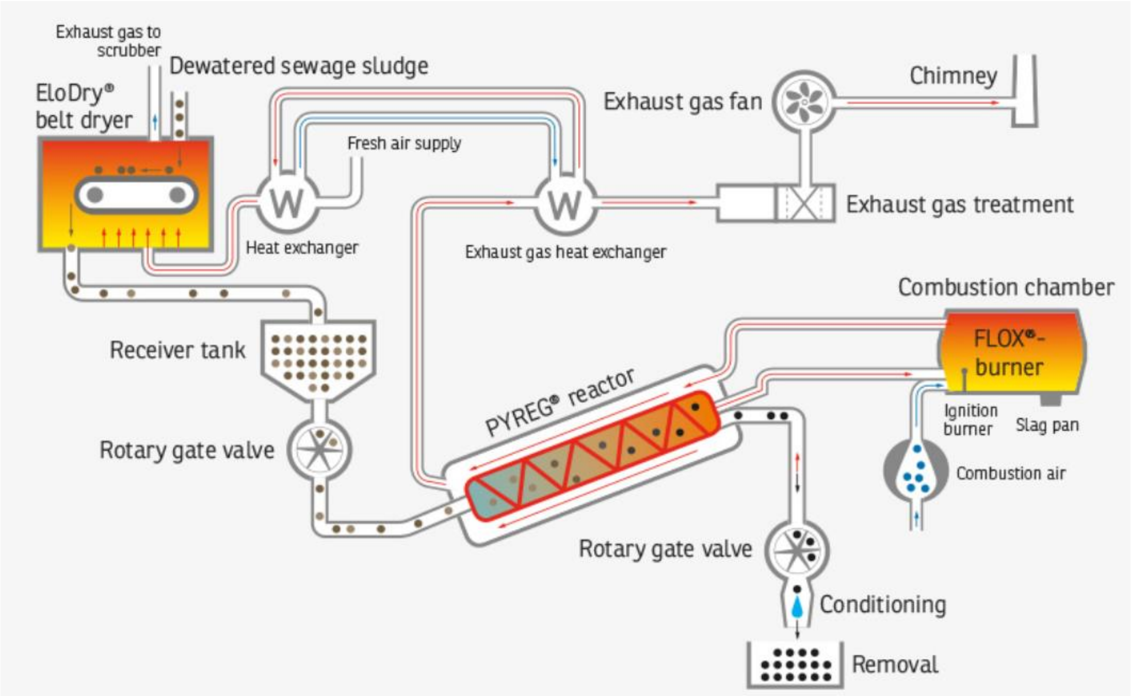


Figure 7 *– Process scheme of PYREG® pyrolysis technology*
(Eliquo Technologies, 2020).

The PYREG® pyrolysis technology is currently used in more than 30 full scale applications, including 4 sewage sludge pyrolysis plants: Unkel, Germany (1200 t DS/y, since 2015); Homburg, Germany (1200 t DS/y, since 2016); Redwood, California (1200 t DS/y, since 2016); Hammenhög, Sweden (1200 t DS/y, since 2016) (European Sustainable Phosphorus Platform, 2020). The PYREG® sewage sludge biochar is also registered as a fertiliser in Sweden (PYREGphos). In 2015 PYREG launched a petition for a biochar fertiliser amendment at the German Ministry of Agriculture and got accepted as phosphorus recovery method in 2017.

However, sewage sludge biochar is not included in the current EU Fertilising Products Regulation (Huygens *et al.*, 2019). The companies' home page (Eliquo Technologies, 2020) states, however, that the obtained phosphorus substrate is legally permissible throughout Europe as a fertilizer.

4.1.3 Direct use of incinerated ash – Ulm WWTP

Incineration is another thermal treatment process, taking place at higher temperatures (800 – 1200 °C) compared to pyrolysis discussed in the previous sub-chapter. During incineration organic substances are fully degraded, therefore all organic pollutants and pathogens are destroyed, while even the content of heavy metals can be lowered to some extent. Although direct use of sewage sludge incineration ash is not very common, the ash can for example be used in Germany if it complies with the requirements of the German fertilizer ordinance (Plank, 2018).

With mono-incineration of sewage sludge and very careful operation, it is possible to reach the compliance without large extra investments or complete change of technology. For example, the Ulm WWTP (220 000 PE) in Germany uses a 2-step strategy: first the transfer of P into sewage sludge needs to be as high as possible (effluent value kept under 0.1 mg/L), second the sewage sludge is kept as clean as possible. This means very strict control of incoming streams of wastewater and external sludge in order to reduce the amount of heavy metals transported to the sludge (Schaum, 2018).

Phosphorus is chemically precipitated in the Ulm WWTP using aluminium salts and the total output of ash is around 600 t P/a. One of the main contaminants in both mineral and recycled P-fertilisers is cadmium, and the resulting ash from Ulm has a cadmium concentration of 7-8 mg Cd/kg P_2O_5, which is significantly lower compared to other fertiliser products (33 mg Cd in super phosphate, 67 mg Cd in triple-super phosphate, 26 mg Cd in MAP) (Plank, 2018). As the German fertilizer ordinance sets two different contaminant concentration limits (maximum allowed contaminant value and declaration to end-user value), the sewage sludge incineration ash is below all the maximum values, while only Ni exceeds the declaration value (max value 80 mg/kg DM, declaration value 40 mg/kg DM and Ulm WWTP value 50 mg/kg DM) (Schaum, 2018). This allows the ash to marketed as a phosphate fertiliser and is marketed by sePura GmbH (as soliPur® 160P) (Plank, 2018).

The main benefit of this practice is the relative cost of the production – no additional costs to regular WWTP treatment are required. The only related costs are due to the transportation of the ash to the distributor, while in exchange the landfilling costs are significantly lower. The main problem however is the low plant-availability of the ash-based P-fertilisers, especially the short-term availability (48% solubility of P in citric acid tests) (Schaum, 2018). This means not all phosphorus in the fertiliser can be quickly taken up by the plants, reducing the overall efficiency of fertilisation. Due to this low use efficiency the ash from Ulm will potentially be used in the new AshDec® (the case study for the technology can be found in section 4.3.2) facility being built to the Altenstadt incineration plant (RePhoR, 2021).

4.2 Integrated P-recovery at WWTP

The most straightforward approach to recover phosphorous from the wastewater is to perform it within the process of wastewater treatment. Those processes are classified as integrated P-recovery technologies. While the cost-benefit of P-recovery is still very low due to low sale price of recovered phosphorus, many of these technologies have been implemented in full-scale due to relevant operational benefits to the WWTPs (Schaum, 2018). Therefore, the integrated P-recovery solutions might have beneficial side-effects to the treatment process, such as increased dewaterability and reduced unwanted struvite precipitation in pipes.

Based on the location of the technological intervention the integrated phosphorous recovery methods are divided as:

- crystallisation in the sludge matrix (AirPrex®);
- crystallisation in the sludge water after dewatering (Pearl®, Struvia®, Phosphogreen®, AD-HAP®).

The essential part of most integrated technologies is the availability of soluble phosphate at the point of crystallisation. Since chemical phosphorous removal from the wastewater by precipitation with Fe^{3+} or Al^{3+} forms an insoluble precipitate, the integrated recovery is preferably combined with biological phosphorous removal technology. Some newer research using magnetic separation (Vivianite) could also be used together with chemical phosphorus removal, while these approaches are still under development (Prot *et al.*, 2019).

Ostara (Pearl® and WASSTRIP®), Veolia (STRUVIA™), Suez (Phosphogreen) and Phosnix are the main full-size technologies used to recycle phosphorous in the form of struvite. Hydroxyapatite and mixes of phosphate salts have been used by Metawater alkaline ash leaching in full scale (METAWATER, 2019) or at the Karlsruhe Institute of Technology (Karlsruhe Institute of Technology, 2015).

In many cases a technological solution for enhanced redissolution of phosphorous is established prior crystallisation technology to increase the crystallisation yield. There are several P-redissolutions technologies on the market:

- Cambi® process (Cambi Group AS) uses thermal hydrolysis (TH) to extract phosphorous from sewage sludge. The TH reactors can be installed in the influent, bypass or effluent of an anaerobic digester. This results in enhanced soluble P and improved dewaterability.
- Lysotherm® (Eliquio Stulz GmbH) is thermal pressure hydrolysis (similar to Cambi®), which is based on a tube-in-tube heat exchanger.
- Pondus™ process (Pondus Verfahrenstechnik GmbH) is a thermo-chemical hydrolysis at 160-180°C to increase P-solubility before the sludge is fed into an anaerobic digester.
- Wasstrip® process uses the anaerobic conditions after EPBR to release phosphorus, magnesium and calcium from the sludge before the thickening process. P can be recovered from the thickener reject water with elevated P concentrations.
- Other similar technologies - HCHS process (Harsleev company), Bio Thelys™/Exelys™ (Veolia Water Technologies), TurboTec® method (Sustec), ZERO SLUDGE (NewLisi S.p.A.) and MultiWAS™ (Multiform Harvest Inc.).

The P redissolution technologies are relatively mature technologies and, in many cases, have been installed at the WWTP-s independent of the P recovery process to increase biogas yield during anaerobic digestion or to increase dewaterability. As they are independent of P-recovery methods and can be integrated on their own, these technologies are not studied in detail in this document.

4.2.1 Struvite recovery from digested sludge matrix – AirPrex®

Struvite aka magnesium-ammonium-phosphate (MAP) can be recovered from the digested sludge matrix prior to the sludge dewatering process. Enhanced biological phosphorus removal in the biological wastewater treatment is a prerequisite of process. P-redissolution technologies discussed in the previous section (4.2) are also often used together with these processes.

AirPrex® technology by CNP – Technology Water and Biosolids (patent by Berliner Wasser Betriebe) is one of the most mature examples of struvite recovery from digested sludge matrix (Centrisys CNP, 2020). The process takes place between anaerobic digestion and dewatering and consists of an additional process reactor and a separation unit (Figure 8).

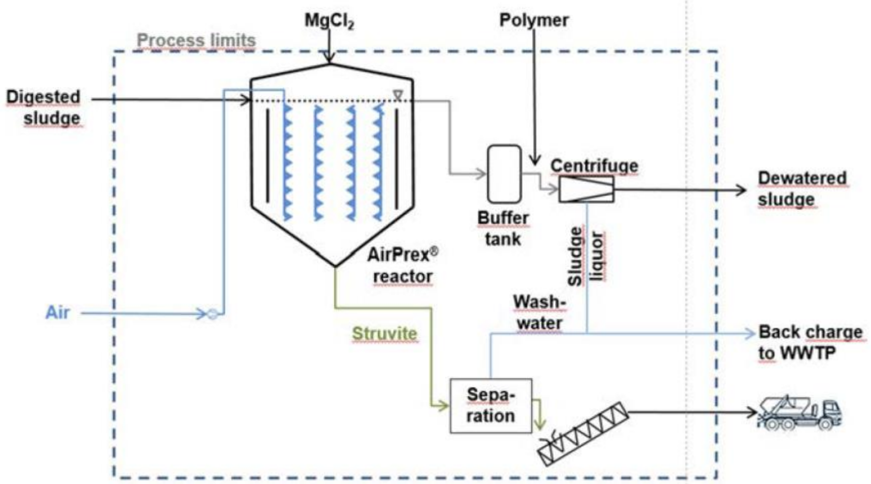


Figure 8 – Process scheme of AirPrex® struvite crystallisation technology (Ortwein, 2018).

The digested sludge is fed to the air stripping reactor, where the concentration of CO_2 is decreased and pH is raised to about 7.8 to 8.2. $MgCl_2$ is simultaneously added to the reactor, which facilitates the formation of struvite crystals. The specific configuration of the reactor is a patented solution made to speed up the growth and separation of bigger crystals.

The increase in the dewaterability of the sludge is often seen as a major side-advantage of the process – the decrease of phosphate ions and the proportional increase of bivalent metal ions lead to reduced water-binding capacity in the sludge. The results of full-scale references have shown that the increase of dry solids concentration of dewatered sludge is about 2–4% and a reduction of polymeric flocculants consumption is up to 35% (Ortwein, 2018).

The AirPrex® process is promised to be relatively easily integrated into the main process of WWTP-s with anaerobic sludge stabilisation. From 2009 to 2020 13 full-scale implementations of the technology have been finished and the process is readily available in the market (Centrisys CNP, 2020). The largest AirPrex® plant has been built in Amsterdam, with around 160 000 t of sludge treated per year. The plant is equipped with bio-P and an advanced AirPrex® reactor configuration (3 tanks in a series with 10 h HRT), further increasing the recovery rate of phosphorus with about 2-3 tons of crystalline struvite discharged daily. The investment cost of the process was 3 million euros, with annual savings on maintenance and disposal alone around 400 000 €. These circumstances suggest the present payback time of the technological investment to be about 7-8 years, not taking account possible extra revenue from the sale of produced struvite (Ortwein, 2018).

4.2.2 Phosphorous recovery from sludge liquor matrix – Pearl®

P-recovery from the sludge liquor is the most widely used concept until to date with many different companies offering full-scale technologies. The more in-depth case study will focus on Ostara Nutrient Recovery Technologies Inc with a combination of two technologies on the market (Pearl® and WASSTRIP®). Though also viable for separate use, the combination results in a very high efficiency P-recovery from sludge liquor (Figure 9).

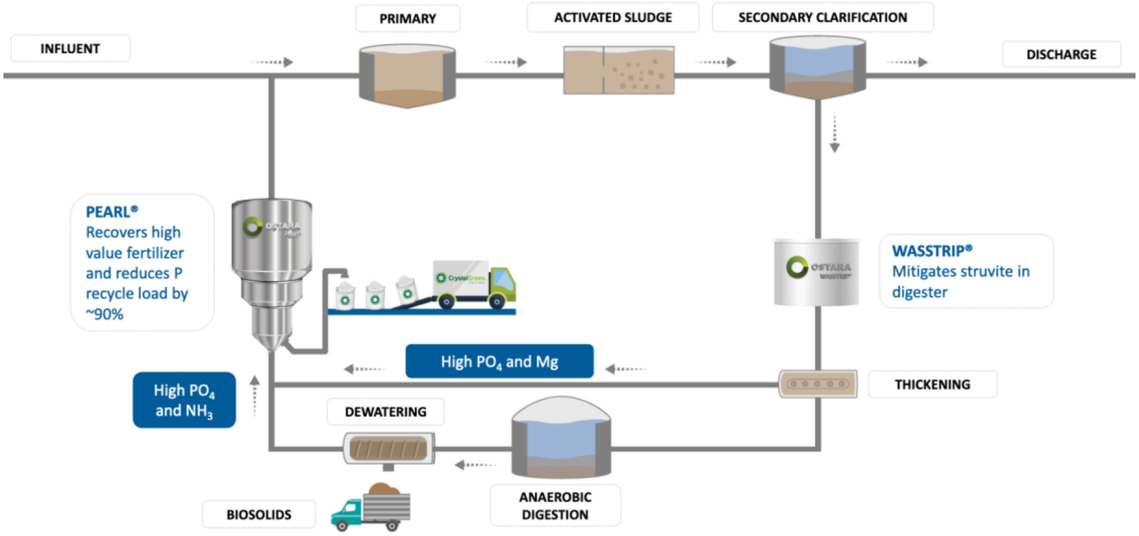


Figure 9 – Process of Pearl® and WASSTRIP® struvite crystallisation technology
(Lee, 2018)

The Pearl® process recovers phosphorus from wastewater liquors through the controlled precipitation of struvite via the addition of soluble magnesium salts. The process takes place in an expanding up-flow fluidized-bed reactor with specific reactor geometry to support efficient nutrient removal and recovery of high quality, commercial fertilizer. The main influent to the Pearl® process is the reject water from the dewatering process after anaerobic digester.

If the WWTP uses biological P-removal, a separate anaerobic reactor for excess sludge (WASSTRIP® process) is used before thickening and digesting, resulting in the re-release of soluble phosphates from PAOs. This approach decreases the uncontrolled struvite precipitation in the digester, improves dewaterability (up to 4%) and reduces polymer consumption (Ostara Nutrient Recovery Technologies Inc, 2017), providing an additional advantage to P-recovery and decreasing payback time of the technology.

The combined technology by Ostara has an overall P recovery rate of about 40-45%. To date Ostara has 18 operational Pearl systems worldwide with capacity from 65 to 1260 kg PO_4-P per day. The recovered struvite granules are branded under name CrystalGreen® (Gysin, Lycke and Wirtel, 2018), which is registered as a commercial fertiliser (Figure 10).

Figure 10 *– Crystal Green® product*
(Ostara Nutrient Recovery Technologies Inc, 2019)

On average Pearl® facilities have been promised to achieve a payback on capital investment in 3–7 years and operate with over 95% uptime (Figure 11). The operating costs depend on the size of the WWTP and decrease with higher populations served.

Following operation costs represent a WWTP of about 250 000 PE (Gysin, Lycke and Wirtel, 2018):

- Electricity demand of 1.6 kWh/kg $P_{recovered}$
- Heat demand of 3.0 kWh/kg $P_{recovered}$
- Chemical demand of 2.4 kg $MgCl_2$/kg $P_{recovered}$ and 0-2 kg NaOH/kg $P_{recovered}$

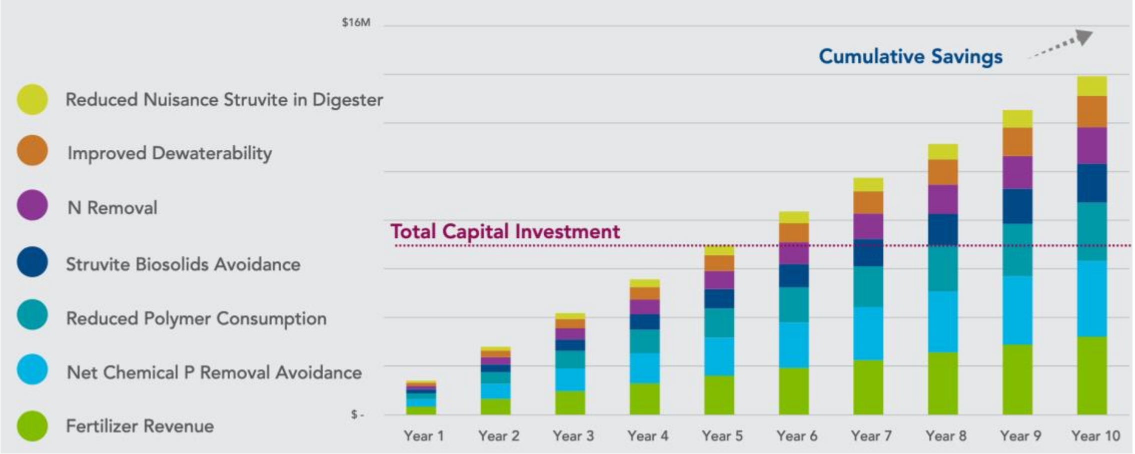


Figure 11 *– Investment recouped in 5 years (average based on standard installations of two systems)* (Ostara Nutrient Recovery Technologies Inc, 2017).

4.2.3 Phosphorus recovery via vivianite magnetic separation – Vivimag (KEMIRA)

As a new and innovative alternative to MAP or HAP precipitation, vivianite precipitation and subsequent magnetic separation is currently under a lot of interest. No reports about a long-term full-scale application of the technology, similar to previous case studies, have currently been published. Nevertheless, the potential technology currently developed by KEMIRA has been identified as one of the most promising new innovations for P-recovery in the region.

As discussed previously in section 3.5.3, the magnetic separation of vivianite is potential method for WWTPs relying on chemical phosphorus removal (Wilfert *et al.*, 2018). Pilot-scale tests have also been done with a subsequent recovery module after vivianite production, allowing for fertiliser production and iron recovery from the mineral. The main benefits of this combined process have been described as the reduction of sludge disposal costs, re-use of iron as a raw material, production of a high-quality fertiliser and decreased vivianite scaling issues at different pumps and valves (Wetsus, 2018).

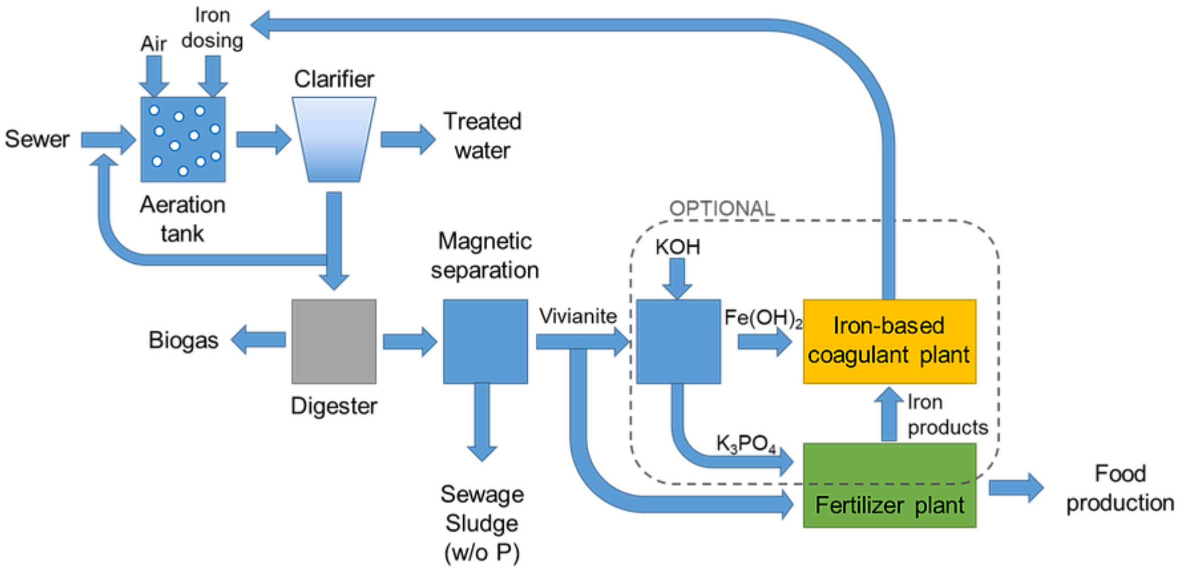


Figure 12 *– Vivianite precipitation, magnetic separation and subsequent dosing agent recovery* (Wetsus, 2018)

4.2.4 Phosphoric acid recovery from chemically precipitated sludge – RAVITA process

RAVITA process is one of the very promising innovative phosphorus recovery technologies, which is currently developed by the Helsinki Region Environmental Service Authority. RAVITA process is based on phosphoric acid recovery from wastewater and is currently studied in 1000 PE technical scale pilot in Viikinmäki WWTP in Helsinki (Helsinki Region Environmental Services HSY, 2020). This new approach has garnered a lot of international interest and was also one of the three winners of the BONUS Return competition (Rosmarin and Ek, 2019).

Phosphorus in the RAVITA process is extracted from the chemical precipitation sludge, meaning bio-P is not a prerequisite of the technology. The chemically precipitated sludge is dissolved into a metal phosphate solution, from which phosphoric acid is produced. Most the chemicals during the whole process are regenerated and can be used circularly.

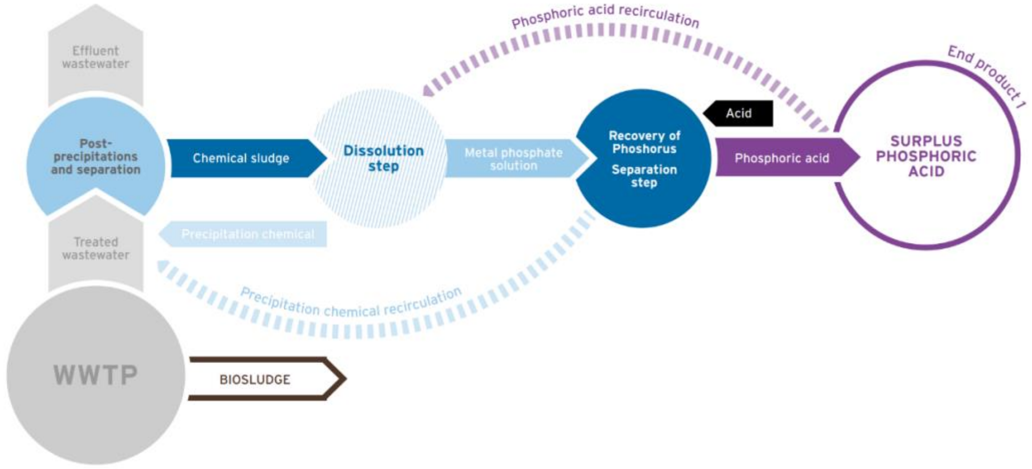


***Figure 13** – The RAVITA process technological scheme*
(Helsinki Region Environmental Services HSY, 2020)

The development of the technology has been steadily going – the first small pilot scale tests were run in 2018 and in the beginning of 2020 the technical scale pilot in Viikinmäki WWTP started running in order to quantify important data, such as the consummation of energy and chemicals, mass balances of the process and costs and quality of the products. The first pilot scale tests show more than 80% of the phosphorus is recoverable from the chemically precipitated sludge and around 55-60% of influent phosphorus could be recovered in total (Baltic Smart Water Hub, 2020).

4.3 Downstream P-recovery at WWTP

Compared to the integrated solutions, the technological advancement of downstream P-recovery is mostly still in the research phase and has not yet reached the phase of implementation and full-scale testing. For most technologies good data can only be found in bench and pilot scale applications, with a first few technologies currently on the move to full-scale application (Schaum, 2018; Remondis Group, 2019). As such large scale investments pose a huge financial risk, the implementation of the downstream P-recovery technologies have been plagued by bankruptcies (Thermphos® in 2012, EcoPhos® in 2020 (Prayon, 2020)) and technologies have changed hands between companies (Italmatch Chemicals Group, 2020), delaying many planned implementations.

So far main large-scale investments have been done for chemical extraction of phosphorus from the SSA, some of the most promising technologies for the Baltic Sea region include (European Sustainable Phosphorus Platform, 2020):

- TetraPhos® (Remondis) – SSA dissolution in phosphoric acid and subsequent fractionation, production of phosphoric acid and side products such as gypsum and precipitation salts;
- Ash2Phos® (EasyMining) – SSA dissolution in hydrochloric acid, production of calcium phosphate and side products such as precipitation and coagulation agents;
- EcoPhos® - SSA dissolution in phosphoric acid, production of phosphoric acid and precipitation salts (declared bankruptcy in 2020, patents changed ownership to Prayon (Prayon, 2020);
- PHOS4Green (Glatt) – SSA treatment with phosphoric acid and subsequent granulation to pellets;
- AshDec® (Outotec) – thermo-chemical extraction of phosphorus from SSA;
- RecoPhos® thermal (Italmatch) – thermo-electrical production of phosphoric acid from SSA.

Sewage sludge ash can also be added to the conventional phosphate rock based fertilizer production, which is done by ICL (ICL, 2019) and was done previously at Thermphos® (Schipper and Korving, 2009).

4.3.1 Acidic wet-chemical process for phosphoric acid recovery from SSA – TetraPhos®

TetraPhos® is a P-recovery process from SSA by REMONDIS, with multiple pilot-scale plants working for years and a full-scale production planned to be commissioned in 2020. The process uses phosphoric acid to dissolve the SSA and the enriched phosphoric acid is then treated with sulphuric acid to produce high-quality phosphoric acid, with gypsum and metal salts as by-products (Rak and Lebek, 2016).

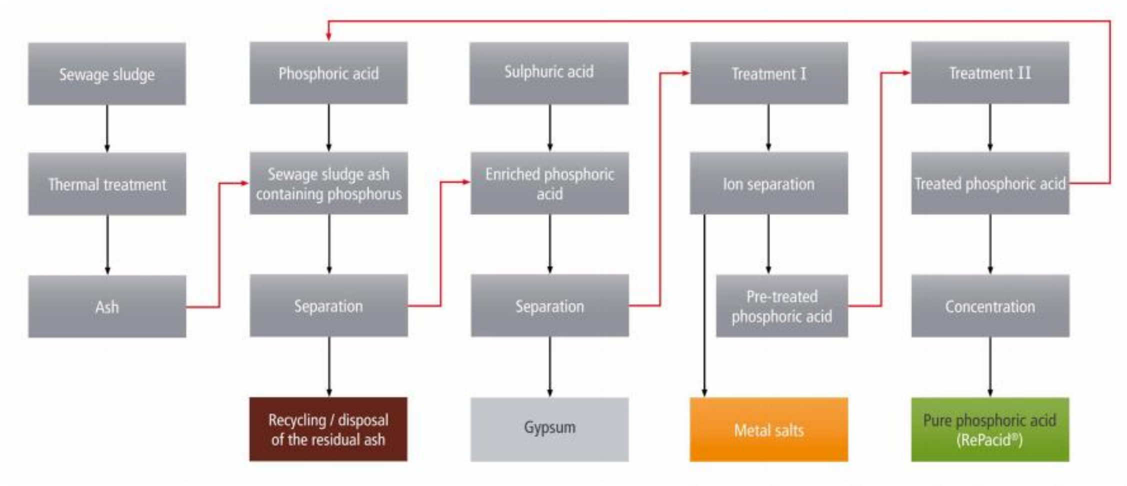


***Figure 14** – Overview of the TertaPhos® process scheme*
(Rak and Lebek, 2016)

The process can achieve a recovery rate around 80% of P in the SSA by first treating the ash with phosphoric acid, then purifying the leachate with sulphuric acid and finally using ion-exchange and selective nano-filtration to produce industrial quality phosphoric acid (Remondis, 2018). The schematic of the whole process can be seen on Figure 14. The product has been branded as RePacid®.

The demonstration pilot-scale application for the processes is located at a sewage sludge incineration plant WFA Elverlingsen GmbH, Werdohl Germany, with the SSA input mass of approximately 50 kg DM/h, producing around 18 kg of H_3PO_4 hourly (Remondis, 2018). A full-scale plant in Hamburg with an expected throughput of SSA being around 20 000 t/a (European Sustainable Phosphorus Platform, 2020).

Specific details about the financial feasibility haven't been shared while the profitability of the technology has been stated in an official press release (BioEnergy International GmbH, 2018). Besides phosphoric acid, gypsum and different metal (iron or aluminium) salts are also produced and can increase the sustainability and feasibility of the technology.

4.3.2 Thermo-chemical enrichment of phosphorus in SSA – Ash Dec®

Ash Dec® is a thermochemical treatment process currently registered as a brand by Outotec. The process uses higher temperatures of 800-1000 °C in a rotary kiln, where most heavy metals are evaporated due to the addition of chlorine ($MgCl_2$ and $CaCl_2$) or sodium (Na_2SO_4) donors (Desmidt *et al.*, 2015). This process also increases the bio-availability of phosphorus, as depending on the donors the phosphates are transformed at higher temperatures (Adam *et al.*, 2015). Examples of the minerals formed include farringtonite ($Mg_3(PO_4)_2$), stanfieldite

($Ca_4Mg_5(PO_4)_6$) and rhenanite ($CaNaPO_4$) (Desmidt *et al.*, 2015), which in pot- and field experiments have shown to increase the fertilisation performance of the extracted phosphorus (Adam *et al.*, 2009). These treated ashes are later mixed with other nutrients, pelletized and are currently marketed under the PhosKraft brand (NPK fertilizer with N-P_2O_5-K_2O rate as 20-5-8). The product also contains significantly less cadmium and uranium than equal phosphate rock based fertilizers (Desmidt *et al.*, 2015).

Similar to other downstream P-recovery processes, Ash Dec® has so far only been operated in large-scale pilot phase (ash input capacity of around 7 t/day) (Desmidt *et al.*, 2015). While the plant could stand alone separate from other facilities, significant savings considering heat energy requirements can be reach when combined with a mono-incineration facility. Further savings from equipment, joint off-gas treatment and other systems make combined facilities more economically viable (Adam *et al.*, 2015). A full-scale AshDec® facility is currently planned to be built at Altenstadt incineration plant in Bavaria, Germany and should be ready and operational by 2023 (RePhoR, 2021).

Besides SSA and ion-donor, the process also requires a reducing agent, to facilitate the reaction between ion-donor material and phosphorus phases in the ash. In Ash Dec® process, granular dried sewage sludge (over 80% dry matter) is used as the reducing agent. The flow chart of the process with all the necessary input streams is shown on Figure 15.

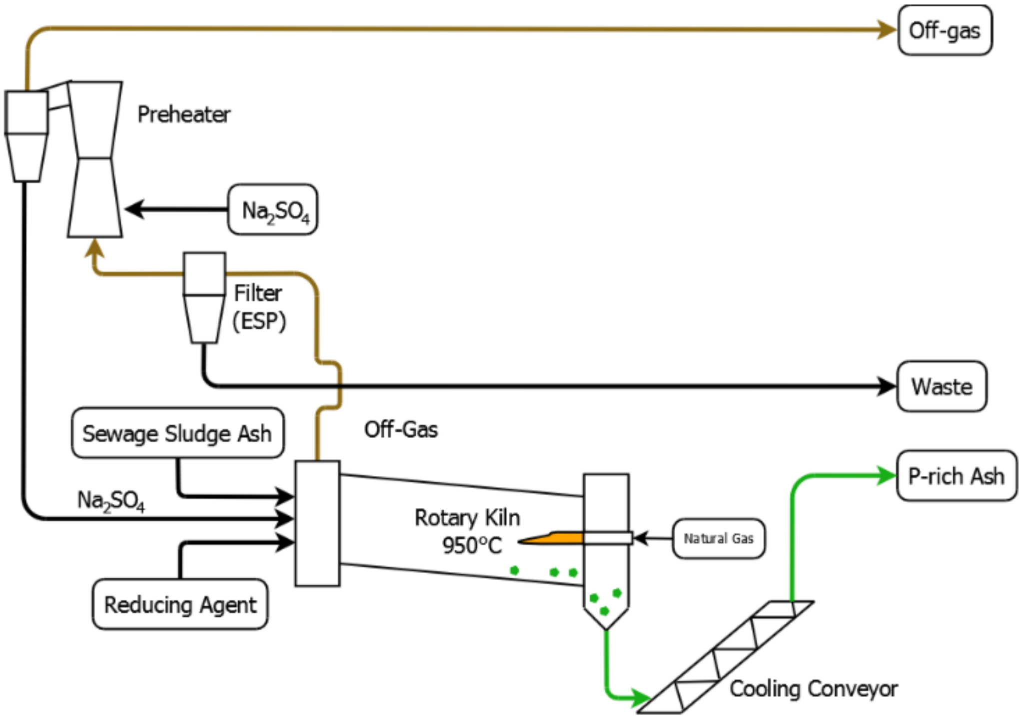


Figure 15 *– Flow chart of the Ash Dec® process using Na_2SO_4 as sodium donor*
(Adam *et al.*, 2015)

Pilot-scale operation of the Ash Dec® process has achieved a continuous P-recovery rate of over 90%, with final P content in the product 5-10% (Adam *et al.*, 2015). Increased bio-availability and deceased concentrations of Cd, Cu, Pb and Zn have been shown, while specific decontamination effect of heavy metals depends on the ion-donor (higher decontamination with chlorine than sodium donors) (Schaum, 2018). In combination with a sludge mono-incineration plant, positive energy balance can be possible if BAT for energy recovery is used (Adam *et al.*, 2015).

No specific cost-analysis data has been published for the Ash Dec® process, while the estimated extra costs of the technology compared to mono-incineration has been shown as 5-20% (lower costs for using sodium and higher costs for using chlorine). The annual cost of the Ash Dec® technologies is estimated to be approximately 2 € per kg recovered phosphorus (Egle *et al.*, 2016).

4.3.3 Full-scale thermo-electrical processes – Thermphos®

Thermphos process was the first full-scale P-recovery technology in Europe extracting white phosphorus (P_4) from the SSA. The company Thermphos International is large producer of different phosphorus derivatives from the phosphate rock and in 2006 started to test replacing some of the non-renewable phosphate sources with sewage sludge ashes from the Noord-Brabant incineration plant (Schipper and Korving, 2009). The full-scale process ran from 2007 until 2012 using SSA as the source of phosphorus on some P_4 production lines. In the end of 2012 the company declared bankruptcy and all production was halted (DutchNews, 2012). As the first and only full-scale downstream recovery process in Europe, the technology, economical view and problems will be evaluated in-depth.

Thermphos adapted the same process used for phosphate rock (Figure **16**) to P-recovery from SSA, the ashes were reduced with coke and gravel (SiO_2 for slag formation) in a submerged arc furnace at high temperatures (heated to 1500 °C via electric resistance heating). In these temperatures phosphate leaves the furnace as a gas (P_4) together with CO and dust and can be condensed. The residues left in the furnace form a slag which could be reused in construction work (Schipper *et al.*, 2004).

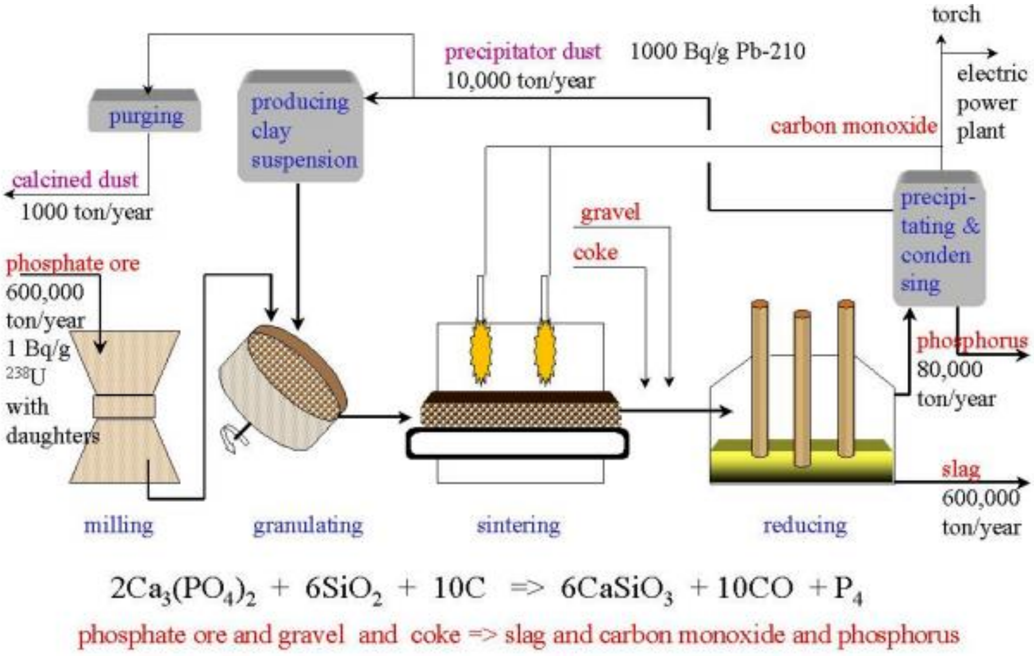


Figure 16 *– Thermphos process flow diagram*
(Erkens, 2018)

As iron is also reduced in the process, high iron concentrations in the source material result to a mixture of FeP and Fe_2P instead of P_4. This decreases the yield of the process and increases energy and raw material consumption (Schipper and Korving, 2009). While in phosphate rock the Fe/P ratio is usually less than 0.05, the ration varies in the sewage sludge ashes from 0.1 to 1.5. The maximum suitable molar ratio was deemed to be 0.2-0.3 mol Fe/mol P (Desmidt *et al.*, 2015). Therefore, the Thermphos process could only use ashes from WWTPs using EBPR or aluminum dosing for phosphorus removal, as iron dosing for precipitation increased the Fe/P ratio to unfeasible values. As 64% of WWTPs used iron as a precipitation agent (25% used EBPR and 11% were small enough that no additional P-removal was necessary) in the Netherlands, the amount of feasible SSA for the process was a potential problem from the start (Schipper and Korving, 2009).

One of the main benefits of the Thermphos process was the use of existing infrastructure, which greatly reduced the starting costs of the process. Via cooperation with local WWTPs it was possible to increase the amount of low-iron sludge produced and a separate line in the incineration plant was designated for low-iron ashes (Schipper and Korving, 2009). The recovery potential of the Thermphos process was very high (85-95%) and P_4 is a universal source of phosphorus, which can be used in fertilizers, feed, chemical industry etc. As the process was adapted from phosphate rock processing, the additional costs of phosphorus recycling are seen as quite low and are mainly due to lower initial phosphorus concentration in the substrate (Egle *et al.*, 2016).

Thermphos declared bankruptcy in 2012 as European market was flooded for years with foreign P_4 product, which made both phosphate rock and SSA based production financially unfeasible. No references to the bankruptcy being tied to SSA recycling has been reported and impossible market prices set by a foreign company have been shown as the main reason (DutchNews, 2012).

5. Economic feasibility of nutrient-recovery

One of the main barriers of phosphorus recycling from wastewater, sewage sludge and ashes is the economic feasibility of the processes. Although the phosphate rock reserves have been often reported to be at end, causing mild panic worldwide, the long-term prices are have been decreasing since 2008 (Figure 17). As many different reports have shown the reserves are still plentiful for hundreds of years (Ulrich and Frossard, 2014), there might not be a significant financial initiative to start recycling phosphorus for many years to come. Although the fertilizer prices are projected to increase 2-3 times during this century (Van Vuuren, Bouwman and Beusen, 2010) the phosphorus value is still low. Economic feasibility of many solutions might instead come from other operational benefits (common with integrated solutions), not from the sale of the recycled phosphorus products.

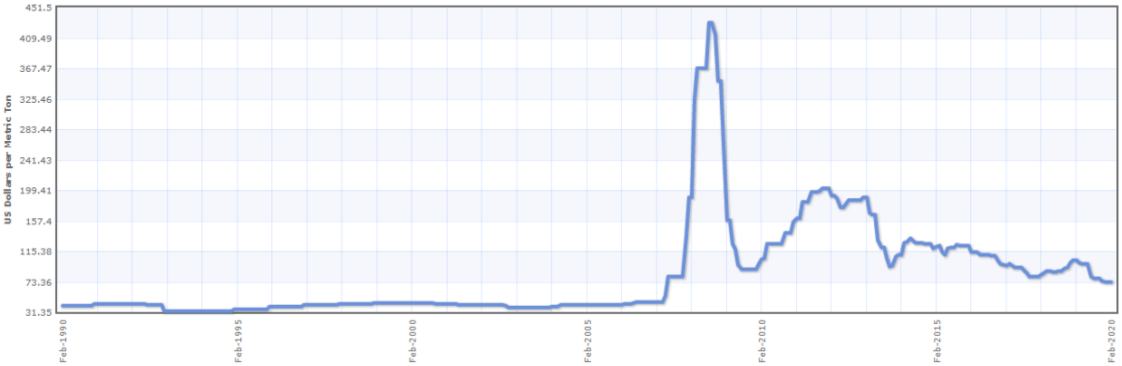


Figure 17 *– Phosphate rock monthly prices (Morocco) from 1990 to 2020*
(IndexMundi, 2020)

Significant portion of the potential savings could also come from reduced handling, transport and disposal and lower energy consumption (Schaum, 2018). Cost comparisons between production of struvite against common fertilizers indicates that cost-effective P-recovery can be achieved if high recovery rates and regional demand are combined (Gaterell *et al.*, 2000). One of the largest financial uncertainties however is the recycled product itself and marketability – all studies use the common market value of elements in estimations, as there currently does not exist a market for recycled products. That approach also doesn't take into account the bioavailability of materials, which could increase the market price of some recovered products (Egle *et al.*, 2016).

As discussed previously, many technologies and approaches are still in laboratory or pilot-scale, therefore it is difficult to correctly estimate all the relevant costs and savings. One of the few comparisons possible, is the recovery potential of phosphorus from different streams and with various technologies. As the overall cost of the technology is usually viewed as € per tonne of P or P_2O_5, the recovery rate is one of the most important values to consider – high recovery yields often relate to much higher economic viability while the actual cost-efficiency also depends on the cost of investment and operation. A comparison of the recovery rates of different technologies is represented on Figure 18.

Technologies extracting phosphorus from aqueous phase (influent, digester supernatant) are generally with the lowest recovery rates (15-25%), with some exceptions (REM-NUT® and DHV Crystalactor® shown to achieve up to 40-50% recovery) (Schaum, 2018). Most of these technologies require EBPR to be used for phosphorus removal from wastewater, therefore are not applicable for precipitating WWTPs. This can also be a major influencer for P-recovery from sewage sludge, as different extraction or disintegration is required whether phosphorus is organically bound or minerally bound (Quist-Jensen *et al.*, 2019). Although around 90% of influent phosphorus is bound in sewage sludge, the recovery rates of different technologies are around 40-70%, as the phosphorus concentration in sludge is not high enough for very efficient recovery. Higher recovery rates could be possible with extensive use of disintegration and chemicals, while achieving extra recovery yields can increase operational costs significantly, destroying the financial feasibility potential.

The highest recovery potential is in sewage sludge ashes (70-85%), as organic matter is already removed and phosphorus concentrations are much higher than in other phases. Although full-scale P-recovery from SSA is still in development, the larger scale, centralisation of incineration units and high recovery yields show this strategy could be with greatest potential among different approaches (Egle *et al.*, 2016).

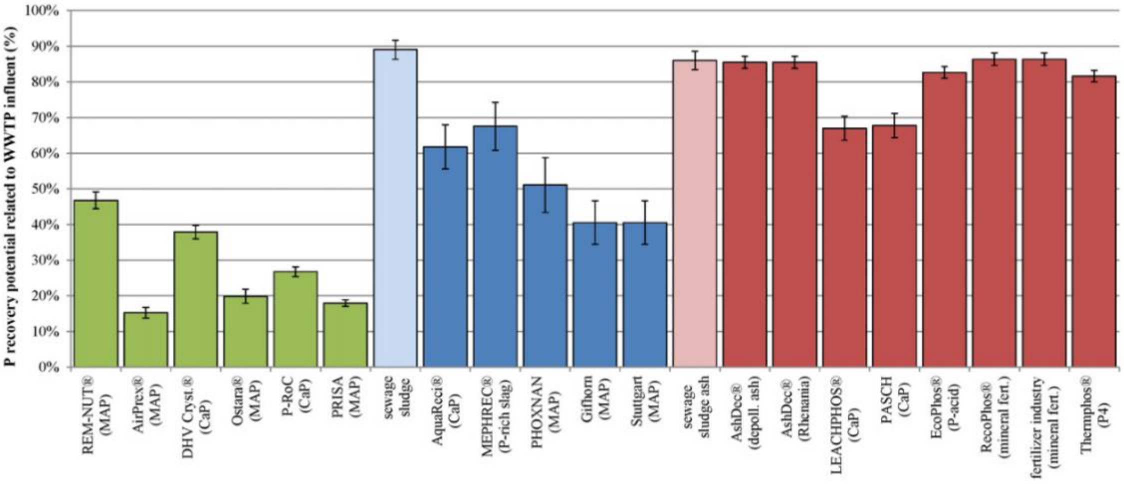


***Figure 18** – Phosphorus recovery potential of different technologies (compared with WWTP influent), green represents technologies extracting P from aqueous phase, blue from sludge and red from incineration ashes*
(Egle *et al.*, 2016)

Taking these yields into account, estimations for the costs of P-recovery with different technologies has been done in previous literature (Figure 19, (Egle *et al.*, 2016)). As revenues from product sales are uncertain and savings from operational costs, energy demand and transport depend on specific cases, they have been excluded from the calculation on the figure. The very high costs of some technologies are due to extensive need for resins and chemicals, while on average the main costs are due to investment costs for equipment (crystallisation reactors). Different processes from sludge (wet-chemical, wet-oxidation) are generally more expensive and extensive chemical use doesn't get cheaper in larger scales. Inversely, P-recovery from SSA is expected to scale well with larger recovered loads, especially thermal technologies (Ash Dec®) not using extensive chemical treatment. The chemical extraction from SSA is more expensive per PE, while very low product specific costs can be achieved (RecoPhos®) (Egle *et al.*, 2016).

As the comparative baseline in the industry is the price of phosphate rock-based fertilisers, some technologies have the potential to reach similar costs with very good feedstock and operation. When taking into account the factors not included into that calculation (revenues and savings), cost-efficient solutions are possible in many cases (Schaum, 2018). Still the risks of P-recycling investments are very high, as the production prices are barely competitive with market prices. Looking at the costs alone, phosphorus recovery at the WWTPs is currently not profitable and cost-efficiency depends too much on the local parameters. This also reduces the marketability and transferability of P-recovery technologies, as in many cases overall financial efficiency is only made apparent after thorough research or pilot-scale testing.

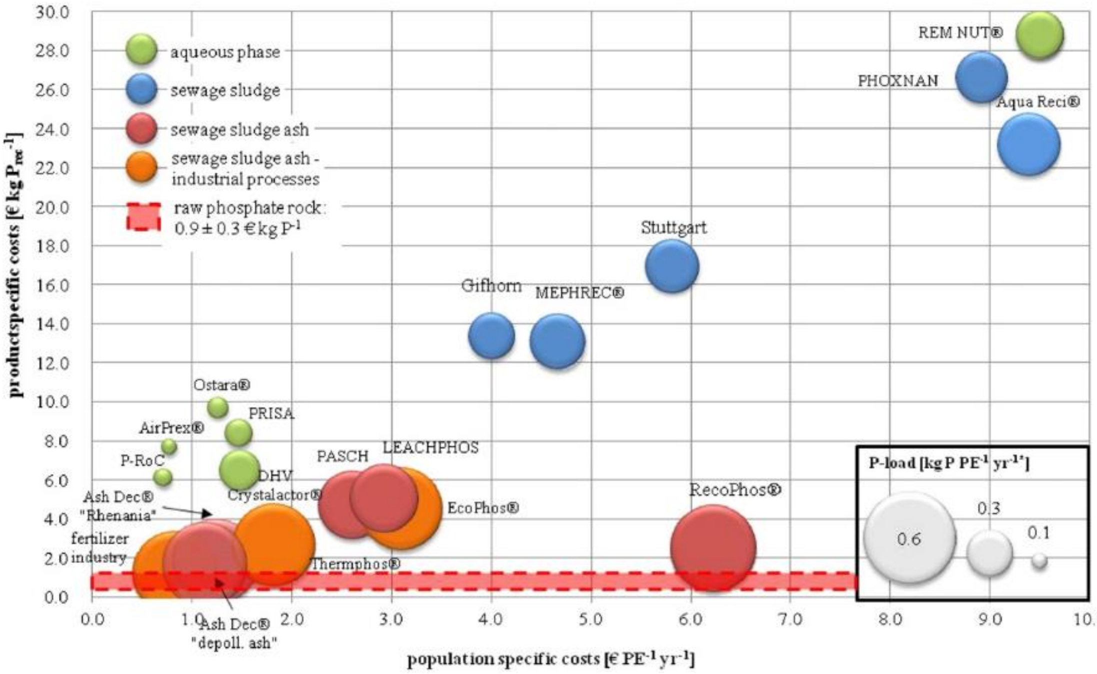


Figure 19 – Annual costs of different P-recovery technologies, both compared to PE and produced material. Savings from operational costs and revenue from the sale of recovered products are not taken into account
(Egle *et al.*, 2016).

Widespread application of different P-recovery technologies in the WWTPs around the Baltic Sea region might therefore be difficult at present. At the same time, the technologies mature with every passing year with considerable improvements made at the profitability and transferability. Together with this development, the actual business potentials and costs are slowly being revealed. First business models of P-recycling are already on the market, with Ostara being one of the frontrunners. For example, the PEARL® process is sold as capital purchase or as treatment fee model (no installation costs, monthly treatment fees), while both forms come with purchase agreement for recovered P-material. This makes adapting P-recovery easier – WWTP can decide between capital investment or long-term monthly fees, while guaranteed a buyer for the generated product without entering the market (Otoo and Drechsel, 2018). As more companies reach marketable P-recovery solutions, the variety of different business models is expected to increase, greatly influencing the actual costs and risks for the WWTPs.

6. Use of the recycled nutrients

At the moment, the main emphasis on the use of recycled nutrients is on the production of different fertiliser products, the first of which are already entering the market. Previously, recycled phosphorus was also used to produce P_4 for potential use in chemical industry and manufacturing, while this stopped with the bankruptcy of Thermphos®. With chemical dissolution, such as previously mentioned TetraPhos® and EcoPhos®, phosphoric acid can also be produced for different industrial applications, but the technologies have not been implemented at full scale (European Sustainable Phosphorus Platform, 2020).

As due to legal frameworks there is currently no wide support to help recycled phosphorus products, surviving in the market competition is a main barrier most products need to face (Rosmarin and Ek, 2019).

In the fertiliser market two important factors are important – the bioavailability of nutrients in the product and the environmental impact. As the main competition is against phosphate rock-based fertilisers, common fertilisers on the market currently are used as the basis of the comparison. Following sections will showcase studies on both areas.

6.1 Bioavailability of recycled phosphorus

When looking at fertiliser products, one of the most important characteristics is the bioavailability of nutrients. In commercial fertilisers, the plant uptake efficiency is very high, which at the same time doesn't mean all of the nutrients are actually taken up by the plants. Bioavailability shows the possible uptake, while fertilisation practices, rainfall and erosion can significantly lower the efficiency. The plant uptake can also depend on the type of soil, its pH value and many other factors.

Most often the bioavailability is measured by a variety of laboratory methods, such as extraction tests with water, citric acid, ammonium citrate or mineral acids, while the results of different methods vary greatly (Weinfurtner, 2011). In-situ tests, such as pot and field tests are more trustworthy, although there is often not enough data available in the literature.

Figure 20 summarises data from the literature, showing the nutrient concentrations and differences between total phosphorus and bioavailable phosphorus. MAP products are most often compared to regular fertilizers, as they have nearly 100% plant available P content, especially in acidic soils. The P concentrations of MAP are around 10-12% and it's not soluble in water (Egle *et al.*, 2016). Calcium phosphate products (HAP, 13-17% P) are less consistent in plant availability, better with acidic soils, but poor in neutral and alkaline soils. In sewage sludge ashes the bioavailability is very low, but different recovered materials show better results. For example, AshDec® products have up to 90% of plant uptake on acidic and neutral soils, while alkaline soils are again problematic, overall better bioavailability can be reached using sodium than chlorine sources in the thermal treatment. Similar results can also be seen with other technologies (Egle *et al.*, 2016).

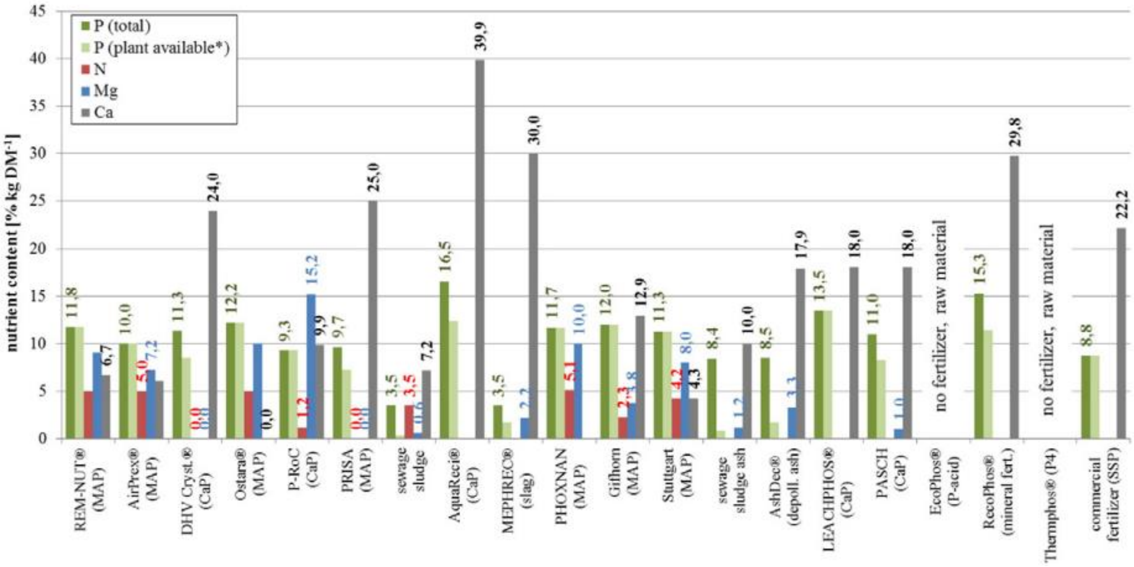


***Figure 20** – Bioavailable phosphorus and other nutrient contents in recycled products*
(Egle *et al.*, 2016)

6.2 Environmental risk-assessment of recycled phosphorus

As the main limitations for the direct use of sewage sludge are different hazardous components, the recycled phosphorus products also need to be assessed for the environmental safety. As most recycling technologies use heavy oxidation and acid leaching, persistent organic pollutants are commonly degraded, while heavy metals can still be included in the products (Bloem *et al.*, 2017). As in market competition, the recycled phosphorus products are mainly trying to replace commonly used fertilisers such as superphosphate and different mineral fertilisers, the environmental risk is assessed via comparisons of possible danger.

One of the common comparisons is to the cadmium content in organic, mineral and recycled fertiliser products, which can show recycled products very favourably (Figure 21). Cadmium is one of the heavy metals often evaporated during incineration, which leads to very low cadmium concentrations in the recycled products from SSA, while rock phosphate has very high cadmium content which is transferred to all the products (0.8 mg Cd/kg P_2O_5 in recycled products from SSA versus 25-105 mg Cd/kg P_2O_5 in other products) (Kratz, Schick and Schnug, 2016). Not all heavy metals are evaporated during SSA thermal treatment (Cu, Fe, Ni etc), therefore for better comparison of the potential danger of different fertilisation products, heavy metals should be summarised and looked as one package.

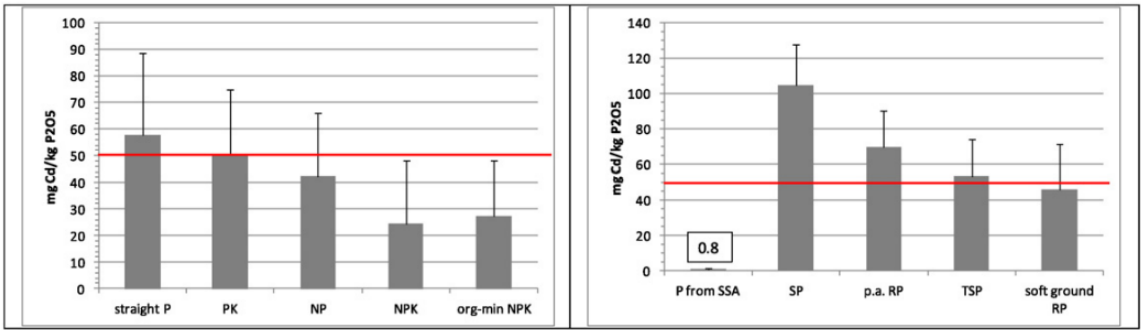


Figure 21 *- Mean concentrations of cadmium in different fertilisers (PK, NP, NPK - macro-element fertilisers (phosphorus, nitrogen, potassium, SSA - sewage sludge ashes, SP - superphosphate, p.a. RP - partly acidulated rock phosphate, TSP - triple superphosphate)*
(Kratz, Schick and Schnug, 2016)

One of the methods to show total potential harm of a product as a coefficient is represented on Figure 22. Damage units are calculated comparing each heavy metal concentration to reference value (in this case from organic compost), which are summed and related to P content of the product. In this summarised way, it becomes clearer that both products extracted from liquid phase and sludge phase are very low in heavy metal content and potential danger (Ueno and Fujii, 2001), while most of the SSA based products have higher danger unit values (Egle *et al.*, 2016). All of the recycled products however are lower in danger units than the commercial superphosphate, while using data from the literature, superphosphate danger units could be up to 0.4 due to very high average cadmium concentrations (Kratz, Schick and Schnug, 2016).

As mentioned previously, heavy metals are the main concern looking at environmental risk assessment of recycled phosphorus products. Traces of other pollutants, such as organic micropollutants and pathogens, have been detected in low concentrations from wet-chemical processes of sludge and in struvite, while in far lower concentrations (Antakyal *et al.*, 2012). When looking at SSA however the incineration process destroys all pathogens and organic micropollutants, therefore derived products could be higher in danger units for heavy metals while safer for persistent organic pollutant content.

Overall, all of the recycled phosphorus products have significantly lower environmental risk from use than direct use of sewage sludge, sewage sludge ashes or even commercial rock phosphate-based fertilisers. Therefore, the contamination of the recycled products should not be viewed as a barrier, as the approaches increase the safety of fertilisation. As previously discussed, the main barriers on the market remain financial, as the processes of phosphorus recovery may have difficulties competing with traditional fertiliser production costs.

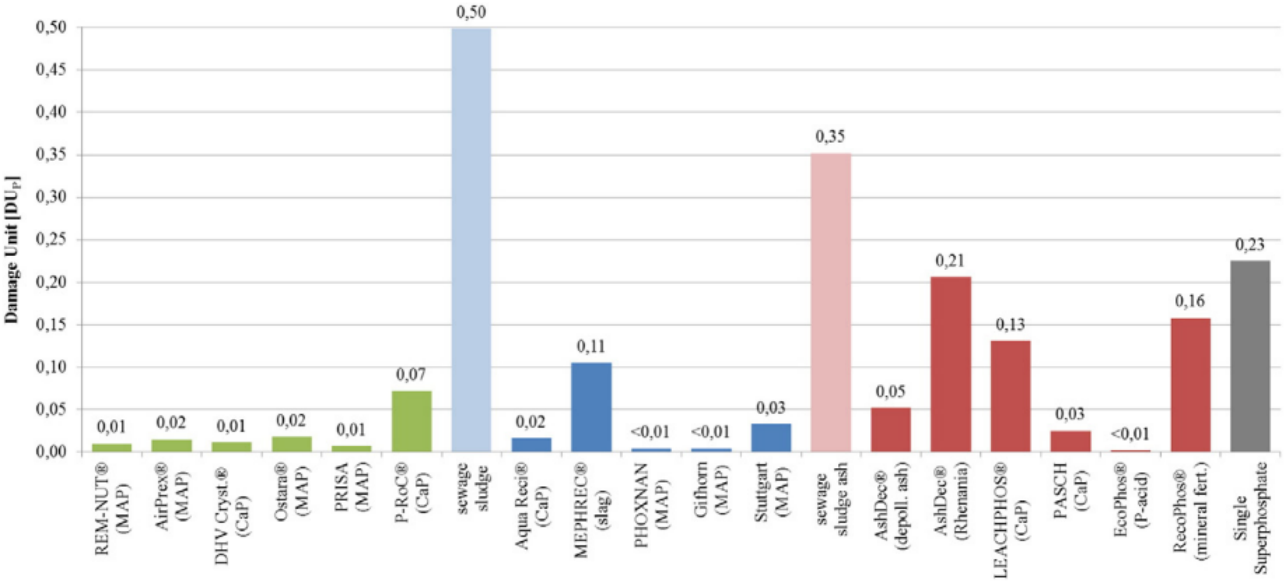


Figure 22 *– Damage units of different recovered materials compared to single superphosphate. Damage units show in reference to organic compost, how much larger is overall heavy metal concentration (As, Cd, Cr, Cu, Hg, Ni, Pb, Zn) per gram of phosphorus*
(Egle *et al.*, 2016).

7. Policy summary

One of the major barriers with transitioning to a nutrient recycling society is the state of current legislations, which are often not intended for a circular economy point of view. The first circular economy package was put out by the European Commission in the end of 2015 and the transition towards it is a lengthy process, as it encompasses all the member states of EU. Nutrient recycling was outlined as one of the key points in the first circular economy package (European Commission, 2015), while the differing quality and environmental standards of fertilisers across the member states was outlined as the main barrier. The Commission proposed revising the EU regulation on fertilisers to support the recognition of organic and waste-based fertilisers and help them in the market situation.

The new fertiliser regulation (European Commission, 2019) was adopted in June 2019, with the goal of harmonising the conditions of making organic and recycled fertilisers and promoting their increased use to further develop the circular economy. The CE marking for fertiliser products was established with strict requirements and bearing it will benefit from the free circulation in the EU's market. At the same time, fertilisers not bearing the CE marking were allowed in the national market's based on a member states own legislation. The new regulation covers all types of fertilisers (mineral, organic, soil improvers, growing matters, etc), while the full regulation will come into force in three years from the publishing date (16th of July 2022). The new fertiliser regulation is one of the first ones to confer the "End-of-Waste" status to the new products (European Sustainable Phosphorus Platform, 2019a).

When looking at the new regulation from sewage sludge perspective, the new rules are very strict and the CE marking cannot be achieved using sludge, sludge compost or sludge digestate. No specific category for recycled sludge products is included in the 11 Component Material Categories (CMCs), while CMC10 and CMC11 are quite vaguely defined and might include those. At the same time, STRUBIAS working group, a sub-class of the Commission Working Group on fertilisers has proposed 3 new CMCs directly connected to recycled products from sludge (CMC 12: Precipitated phosphate salts or derivatives, CMC 13: Thermal oxidation materials or derivatives, CMC 14: Pyrolysis or gasification materials) to be added to the new fertiliser regulation (European Sustainable Phosphorus Platform, 2019b).

Connected to the circular economy package, the waste legislative framework was also revised and entered into effect in July 2018 (European Union, 2018), while an official evaluation of the Water Framework Directive was concluded, resulting in the decision against revising it (Directorate-General for Environment European Commission, 2019).

7.1 National regulations and policy action

Even though the new European Union fertiliser regulation establishes a common ground for fertiliser products in all the member states, using and manufacturing fertiliser products not adhering to the new established rules is still allowed on the national markets. Many countries have already established their own End-of-Waste criteria for fertiliser products, including sludge compost. The REVAQ certification system in Sweden has been used as a success story in many different occasions, as at the highpoint more than 50% of the treated sewage sludge in Sweden was REVAQ certified (European Sustainable Phosphorus Platform, 2019a). Similar certification systems are in use around the Baltic Sea region, while some have found more success and others have been mostly unused due to public opinion or strict requirements.

In the last few years, the danger of different organic micropollutants and pharmaceutical residues has become more and more apparent, which has also changed the public and scientific opinion of using treated sewage sludge for agricultural applications. Even the well-working REVAQ system in Sweden is currently under scrutiny, as different environmental organisations are putting pressure on the government to stop the land use of sewage sludge. An inquiry was launched by the Swedish government and while the proposal to completely ban sludge reuse was pushed back, new and stricter quality requirements and five-yearly reviews were proposed instead. Still many businesses, such as Swedish flour mills, do not accept grain fertilised with sewage sludge and crops grown that way cannot be certified as organic (Stockholm Environment Institute, 2020).

While in Sweden the discussion is still mostly ongoing, Germany has already decided for more strict regulations of sludge treatment and reuse. Within the next 12-15 years, recovery of phosphorus will become mandatory for WWTP over 50 000 PE, if the phosphorus content in sludge is over 2% DM. The operators of the WWTP have to submit their phosphorus recycling concept to the authorities by the end of 2023 (Pollution Control Service GmbH, 2017). Smaller WWTPs might still have the option for direct sludge use, but that is severely limited by the new German fertilising regulation (Nitrates directive). The main emphasis in the legislation is toward mono-incineration of sewage sludge and subsequent P extraction from the sewage sludge ashes. Mandatory P-recycling is also under discussion in Austria and Switzerland (European Sustainable Phosphorus Platform, 2017b, 2017a).

7.2 Main Barriers

Although phosphorus recycling in WWTPs is taking major steps forward, there are still many problems that need to be solved. As the recycled phosphorus products are entering the fertiliser market, they have to compete against the mineral fertiliser industry, with very high production volumes, long-term investments and decades of experience and optimisations (Rosmarin and Ek, 2019). In order to enter the fertiliser market, very large investments into P-recycling are needed. National and even EU-wide incentives will be one of the key deciders, on how wide-spread P-recycling in WWTPs will be.

One of the main problems with economically feasible nutrient recycling from wastewater and sludge is that for most technologies the production costs are likely to exceed the market value of the produced fertiliser product. While indirect savings can be achieved with these technologies (lower maintenance costs for anaerobic digestion with struvite precipitation, etc), phosphate rock global market price is still very low, reducing the economic efficiency of the technologies (Carolus *et al.*, 2018).

Another issue potentially lowering the economic feasibility of nutrient recycling at WWTPs, are the transport costs of the products. As these costs depend greatly on the specific situations and business models, they are an important topic to be considered (Carolus *et al.*, 2018). While most products derived from sewage sludge (struvite, ash) are significantly lighter in weight than sewage sludge (greatly reduced water and organic matter content), transporting sewage sludge to a centralised incinerator, the ashes to a recovery facility and the recover product to distribution centres and markets can add up. Localised business models can have significantly lower transportation costs, but the larger centralised systems needed for a countrywide P-recycling might be very inefficient, especially in areas with low population density.

Additional active regulatory support, such as recycling obligation or subsidies from EU will be vital to overcome these barriers (Rosmarin and Ek, 2019). Changes to the new fertilise regulation to include the recycled phosphorus products from wastewater and sludge as proposed by the STRUBIAS work group will be necessary to overcome barriers on the trade of the products and harmonise quality criteria (European Sustainable Phosphorus Platform, 2019b). New business models and new operations need active support to come to the market dominated by the large mineral rock based fertiliser production companies.

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