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Capturing the philosopher's stone

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Chapter 4

Phosphorus Recovery Potential from Wastewater: Creating a Viable Pathway for Realizing a Circular Phosphorus Economy

Abstract: Life on earth is heavily reliant on a consistent and readily available supply of the natural element phosphorus (P). Yet, the current way in which phosphorus is managed from mine to fork does not secure such a supply for the future. In order to reach a circular phosphorus economy, it is essential to define the specific flows and recovery potential of the greatest P losses in the cycle. This study provides for the first time such a complete, quantitative overview and highlights the potential for P recovery of these flows with a focus on the Dutch wastewater sector. Appropriate P recovery technologies are available to prevent phosphorus losses at wastewater treatment plants or via sewage sludge ash treatment, yet these are not widespread implemented. We formulate herein several P recovery scenarios to illustrate the P recovery potential for The Netherlands and its relation to the current national P demand. This study shows that The Netherlands can be self-sustainable in P based fertilizers for agriculture, if P from the prospective urban mines is recovered and recycled efficiently.

4.1 Introduction

Phosphorus (P) is, next to nitrogen (N) and potassium (K), an essential component in fertilizers that is currently mined from the non-renewable resource phosphate rock.^[1] The high economic importance of P based fertilizers for the European Union to facilitate food production, together with the dependency on supply from outside the EU, resulted in the inclusion of phosphate rock in the list of European critical raw materials.^[2] Astonishingly, next to being limited in supply, phosphorus is also wasted. The current linear use of phosphorus causes an increase in P concentration in water bodies due to the discharge of P containing fertilizers, detergents, and waste flows (e.g., excreta),^[3] which results in uncontrollable algae bloom (eutrophication), lower (drinking) water quality and fish death.^[4] In a circular economy, phosphorus should not end up as waste causing environmental concerns, but should be recovered and reused as a secondary P source. In order to realize a circular phosphorus economy, it is essential to have a clear grasp on the current anthropogenic phosphorus flows and how these P flows can be modified. Van Dijk et al. showed that the greatest area of P loss in Europe is the wastewater sector (32% of the total system losses, which resembles 390 Gg P/ year for the EU only), next to food processing (339 Gg P/ year), food waste during consumption (170 Gg P/ year), and other losses (318 Gg P/ year).^[5] Interestingly, the required technologies for P recovery in the wastewater sector are available. Yet, a complete overview on the individual P flows within the wastewater sector is lacking, which makes it difficult to assess its P recovery potential. This study provides such an overview and its implementations, and focusses on the national recovery potential of The Netherlands that we took as an example due to its frontrunner's position in P recovery. We compared the P recovery potential of The Netherlands with the

national P demand and estimated that WWTPs (wastewater treatment plants) have great potential to function as urban mines and can contribute to realizing a circular P economy. Our study focusses on the following four interrelated elements: (1) quantifying the overall trends in wastewater influent in the Netherlands, (2) quantifying the P influent and effluent flows in the Dutch wastewater sector, (3) calculating the P recovery potential using scenario analyses to estimate whether the P recovery potential is sufficient to cover the national P demand, and (4) policy recommendations for the recovery and recycling opportunities of phosphates from wastewater.

4.2 P recovery potential in the Netherlands, a case study

Phosphorus removal from wastewater is obligatory throughout the EU,^[6] and as a result more than 98% of the WWTPs in The Netherlands have implemented P removal technologies. Currently, approximately 85% of the P in the influent of WWTPs ends up in the resulting sludge via Chemical P removal and/or Biological P (Bio-P) removal, and the remaining 15% P stays in the aqueous effluent.^[7-11] While in 1995 only 10% of the WWTPs applied Bio-P removal,^[12] we found that this increased to 69% in 2017, making Bio-P removal the most used method for P removal in The Netherlands. P in the effluent as well as in the sludge can be recovered using several methods, of which two mature ones are incineration of the sewage sludge and implementation of a struvite precipitator.

Nowadays, 100% of the sludge in The Netherlands is mono-incinerated into sewage sludge ash (SSA), which makes P recovery from SSA a highly promising endeavor.^[13,14] The P rich SSAs are currently used as asphalt filler, in construction materials, and are disposed in landfills and salt mines. All in all, the P from SSA is not recycled, which highlights the need to bridge the gap between P recovery and P recycling.^[15] Interestingly, the quality of the SSA of the sludge obtained by Chemical P or Bio-P removal differs. Namely, SSA derived from sewage sludge from chemical dosing contains high metal concentrations, such as aluminum and iron, and 2-3% of P (dry weight), whereas Bio-P sludge contains 2-5 times more P and less metals.^[16] Thus far, the sludge in The Netherlands is incinerated in a small number of mono-incinerators by combining the two different types of sewage sludge (from chemical dosing and from Bio-P), yet ultimately Bio-P SSA would be the preferred choice, also due to its higher bioavailability that promotes P recycling.

The second mature technology for P recovery is struvite ($\text{NH}_4\text{MgPO}_4 \cdot 6\text{H}_2\text{O}$) precipitation, which is successfully implemented at several Bio-P wastewater treatment plants worldwide.^[17] A struvite precipitator reduces the maintenance costs of WWTPs, due to the prevention of clogging of pipes, and is therefore gaining in popularity.^[17–22] Struvite precipitation can be installed at WWTPs that apply Bio-P removal to recover P from the aqueous stream (the first generation struvite precipitators) or from both the effluent and the sludge (the second-generation struvite precipitators).^[23]

4.3 Materials and methods

In the Netherlands, a network of 22 governmental (regional) water bodies, called water boards, which include 340 WWTPs, facilitates the management of the wastewater sector. For this study, all water boards in The Netherlands were contacted in order to construct an up-to-date and complete overview of the P flow quantities in the wastewater industry. This resulted in a comprehensive database that includes the P (and N) volumes in the influent and effluent streams of all Dutch WWTPs, the type of P removal techniques used at each individual WWTP, and the already implemented P recovery methods, including the quantities of recovered P products.

Material flow analysis (MFA)

To visualize and structure the results of the obtained database, a material flow analysis (MFA) has been performed, which provided an overview of the current P flows in the wastewater sector in The Netherlands as well as its P recovery potential (see Figure 2). The MFA has been created with the STAN substance flow analysis software, a commonly used software program for nutrient flow analyses.^[5]

Calculating the current P recovery potential

The P recovery potential for each WWTP located in the Netherlands has been calculated individually. As the quality of the sewage sludge from the Bio-P removal and the chemical P removal technologies differs, we have calculated the two types of sludge and corresponding SSAs individually, also to be able to analyze the impact when these two types of sewage sludge will be incinerated separately. For implementing a struvite precipitator, we have included both the first-generation struvite precipitators (three different types), e.g., the Airprex technology, and the

second-generation struvite precipitators (two different types) in the scenario planning. The first generation technologies focus on the phosphorus recovery streams from either the digested sludge, the centrate or in the thermal hydrolyses upstream digester in the WWTP, while in the two second-generation technologies struvite is precipitated in the thermal hydrolyses upstream digester and the process with the highest efficiency, from the centrate, including the Wasstrip technology.^{[23-}

25]

Since these technologies have different recovery efficiency rates, we have included the five types separately to provide a complete overview of the current state of the art. The recovery percentages of these struvite precipitation techniques used in the analysis are based on literature values and data retrieved from experts, and indicate the maximum P recovery attainable under optimized conditions. As the recovery efficiency of a struvite precipitator depends on several technical aspects of the WWTP, such as the pH, P concentration, and type of Bio-P removal technology, un-optimized systems can generate reduced amounts of recovered P.

Calculating the P recovery potential based on scenario analyses

Scenario analysis was used to assess and predict several possible future perspectives of P recovery in the Netherlands. The scenario planning is based on an extrapolation of the current trends of (1) an increase of implemented Bio-P removal technologies at WWTPs, which also provides opportunities for P recovery, and (2) the widespread implementation of sewage sludge incineration (SSA production) and struvite precipitation as key P recovery methods. Note that a struvite precipitator can only be implemented in combination with Bio-P removal. Moreover, experts indicate that, as a rule of thumb, the size of the WWTP should be above 50.000 population equivalent (p.e.)^[26] for the implementation of a struvite precipitator to be

economically feasible. This notion correlates well with the German legislation, where a new law was adopted in 2017 that requires that all WWTPs bigger than 50.000 p.e. should implement P recovery technologies.

Therefore, in our scenario analyses we focus on 1) P removal technologies (current state-of-the-art *versus* all WWTPs implement Bio-P) and 2) P recovery methods (sewage sludge ash and struvite precipitation), which result in four scenarios (see Table 1). Scenarios 1 & 2 focus on the P recovery via SSA, while scenarios 3 & 4 target WWTPs (>50.000 p.e.) that can implement struvite precipitation.

Table 1. On overview of the four scenarios incorporated in this study.

P removal technologies \ P recovery method	Sewage sludge ash	Struvite precipitation (only for WWTPs with a size above 50.000 p.e.)
P removal technologies: current state-of-the-art	<i>Scenario 1</i> Infrastructure stays the same	<i>Scenario 3</i> P removal infrastructure stays the same, but all current Bio-P WWTPs >50.000 p.e. implement a struvite precipitator
All WWTPs implement Bio-P	<i>Scenario 2</i> All SSA becomes valuable Bio-P ash	<i>Scenario 4</i> All >50.000 p.e. WWTPs implement Bio-P removal and a struvite precipitator

4.4 Results and Discussion

4.4.1 Trends in P content in the Dutch wastewater influent

To calculate the P recovery potential, it is essential to know the accumulative national influent. Not unexpectedly, the concentration of P in the influent of Dutch WWTPs changed over the years, as shown in figure 1.^[12] The increase of P in the influent until 1986 can be ascribed to the increasing use of washing detergents.^[27] After 1986, the amount of P in the influent decreased (from 55.0 ton P/ day to 36.4 ton P (dry matter)/ day) due to the adoption of low-/ non-phosphate detergents for washing machines.^[27] From the early 2000s, there is a slight increase again due to the introduction of phosphate-containing detergents for dishwashers in households. Since 2007, the P content is slowly declining due to the introduction of low-phosphate detergents for dishwashers. About 20% of the P in the influent originates from detergents,^[27] the other 80% originates mainly from excreta and in smaller amounts from agriculture, waste processors, the chemical industry, and the food and beverage industry.^[28] The observed trends show that regulations have a direct and significant effect on the P content in the influent. When all dishwasher detergents become phosphate free, it is to be expected that the P content in the influent reduces to about 29.5 ton P/ day, which is still a sizeable amount that is available for P recovery and recycling. Interestingly, despite the reduction of P in the Dutch influent from 1981, the amount of P in the sludge increased considerably (42% in 1981 to 85.4% in 2015) due to stricter rules of the amount of P allowed in the discarding effluent (a removal of at least 75% of the P in the influent is mandatory).^[27]

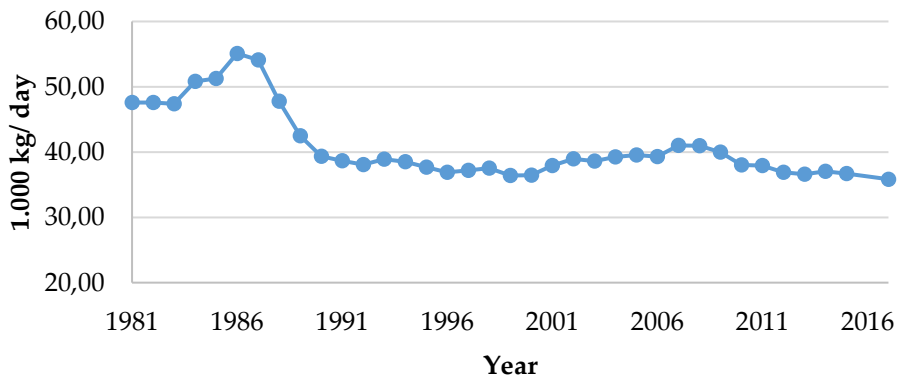


Fig. 1. The P content in the influent of WWTPs in the Netherlands from 1981 until 2017 based on data from CBS (1981-2015) and this study (2017).

4.4.2 Quantifying the P flows in The Netherlands

Next, to quantify the P recovery potential of The Netherlands, we studied the fate of P after it enters the WWTP. Dutch WWTPs use Bio-P removal, chemical P removal, or a combination of these two techniques to remove the majority of P in wastewater and capture it in the sludge. Based on our overview of P volumes in the influent and effluent streams of all Dutch WWTPs in the year 2017, we created a material flow analysis visualizing all P flows (Figure 2), where the difference between the P in the influent and the effluent corresponds to the amount of P that is obtained in the sludge.

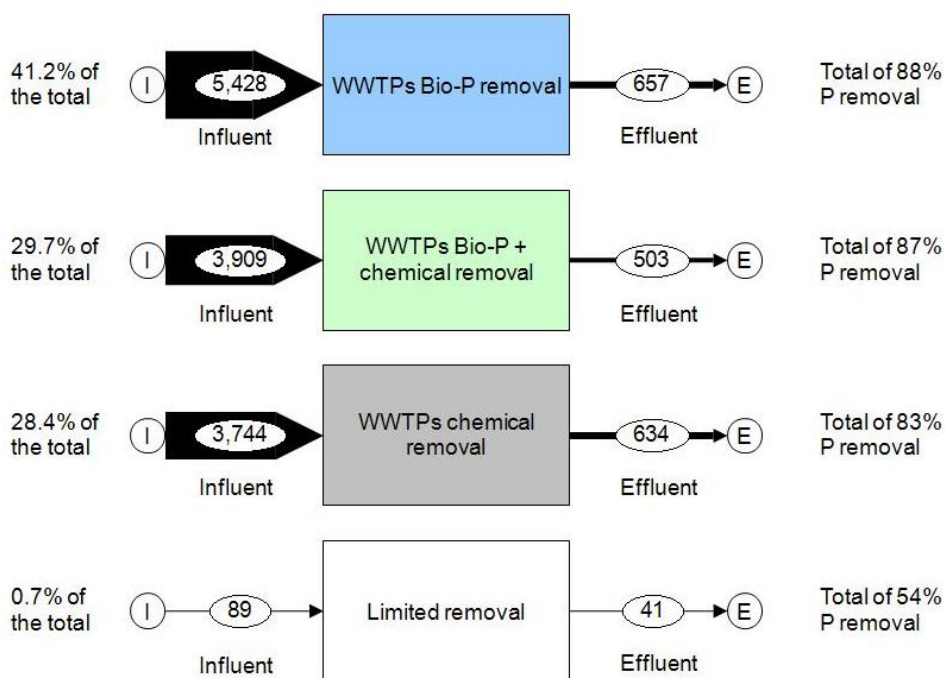


Fig. 2. Material Flow Analysis of the Phosphorus flows in the Dutch wastewater sector (amounts in ton per year).

About two fifths (41.2%) of the Dutch influent in 2017 entered a WWTP that solely used the Bio-P removal technology,^[9] of which 88% of the P is removed into the sludge affording 4.770 ton of P material per year (5.428 [influent] – 657 [effluent], see Figure 2). Note that Bio-P removal processes also have the ability to remove nitrogen simultaneously,^[10,11] with Enhanced Biological Phosphorus Removal (EBPR) being the most widely applied Bio-P process in The Netherlands. The remaining 12% P that ends up in the effluent is disposed of in water bodies and enters the environment. The second largest fraction of the influent (29.7%) is treated at WWTPs that use both Bio-P and chemical removal, traditionally to achieve better results, with a combined removal efficiency of 87% P into the sludge affording 3.406 ton/ yr

(3.909 – 503, see Figure 2), while the remaining 13% P stays in the effluent. Note that chemical P removal involves the dosing of a metal salt, such as alum ($\text{KAl}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$), or the frequently used iron salts (iron(III) chloride (FeCl_3) and ferrous sulfate ($\text{FeSO}_4 \cdot x\text{H}_2\text{O}$)), thereby causing precipitation of an insoluble metal phosphate, which enables the separation of P from wastewater in the formation of sludge.^[7,8,29] Thus, the third fraction (28.4%) of the influent flow enters WWTPs where only chemical dosing is being applied, resulting in the capture of 83% of the P in the sludge (representing 3.110 ton/ yr), and 17% of P still ending up in the effluent. The remaining six WWTPs (2% of the total), typically the smaller WWTPs, accounting for 0.7% of the total P influent, have limited or no controlled P removal at all, as the implementation of a removal technology is relatively expensive for these small volumes. Still at these WWTPs 54% of the P (48 ton/ yr) is removed from the influent mainly by limited chemical dosing, while 46% of the P stays in the effluent.

Currently, seven out of the total 340 municipal WWTPs in The Netherlands have struvite recovery installations installed, affording 3.400 ton/ yr of struvite (note that this is the mass of struvite, not solely P). The implemented first-generation technologies recover P from the aqueous phase of the digested sludge (which ends up as effluent) are Airprex (2x, 902 ton/ yr), NuReSys (2x, 1050 ton/ yr) and Phospaq (2x, 550 ton/ yr). So far, only one second-generation struvite precipitator is implemented (Amersfoort; Pearl, Ostara), which precipitates struvite from the centrate with Wasstrip and yields 900 ton/ yr of struvite.

4.4.3 Dutch P recovery potential using scenario analyses compared to its national demand

Based on the current state-of-the-art that quantifies the amount of P in the Netherlands that enters the sludge and is subsequently mono-incinerated to obtain SSA, the amount of P that is recovered as struvite, as well as the P content that remains in the effluent, we can now calculate the total P recovery potential of the Netherlands using WWTPs as urban mines and compare the outcomes with the national P demand. In 2016, according to Statistics Netherlands (CBS, Centraal Bureau voor de Statistiek), the total demand of P in the form of phosphates was 81.000 ton, with 75.000 ton used as animal feed/ feed concentrates, 4.000 ton as fertilizer in agriculture and about 3.000 ton for other applications.

Scenario 1: Current state-of-the-art with no changes in the current infrastructure.

For the calculation of the recovery potential of the currently produced sewage sludge ash in the Netherlands a conversion factor of 98% of sewage sludge into sewage sludge ash was used, which is the theoretical yield for converting sewage sludge into sewage sludge ash reported by STOWA (Dutch foundation of applied water research).^[25] We found that 145 out of the 340 WWTPs in The Netherlands have implemented solely Bio-P removal, 100 WWTPs implemented solely chemical removal, and 60 WWTPs have installed Bio-P in combination with chemical removal, while the remaining 6 WWTPs use limited or no P removal at all. This results in the production of 4.533 ton/ yr of SSA from Bio-P (98% of 4.770), 3038 ton/ yr of SSA from chemical dosing (98% of 3100), a mixed fraction of 3338 ton/ yr of SSA from Bio-P and chemical removal (98% of 3406), and an un-optimized P recovery potential of 46 ton /yr of SSA (98% of 47; Table 2).

Table 2. The recovery potential of scenario 1 and 2.

	Sewage sludge ash (ton/ yr)			
	Bio-P ash	Chemical ash	Chemical + Bio-P ash	Limited removal ash
Scenario 1	4.533	3.038	3.338	46
Scenario 2	10.955			

Scenario 2: All WWTPs implement Bio-P removal and all SSAs will become Bio-P SSA

As currently Bio-P removal is the most promising technology for P recovery, scenario 2 targets the amount of SSA that can be produced when all WWTPs switch to Bio-P removal. This means that next to the 145 WWTPs that implemented Bio-P already (affording 4.533 ton of Bio-P SSA/ yr) an additional amount of 6.553 ton of Bio-P sludge can be generated, that amounts to 10.955 ton of Bio-P SSA/ yr in total (Table 2). Interestingly, the quantity of this potentially recovered P is more (~275%) than the amount of fertilizer currently used in the Dutch agriculture that is produced from mined phosphate rock. This indicates that when competing recycling techniques become available to convert Bio-P SSA into fertilizers, The Netherlands can become self-sufficient for its own fertilizer production from secondary (renewable) phosphates.

Scenario 3: All current Bio-P WWTPs with a capacity of more than 50.000 p.e. implement a struvite precipitator

In The Netherlands, currently 165 WWTPs are large enough to allow an economically viable installment of a struvite precipitator (> 50.000 p.e.), of which 97 WWTPs have installed Bio-P or a combination of Bio-P and Chemical P already that makes this combination technically feasible. By implementing the struvite precipitator technology in these existing 97 Bio-P WWTPs with from the least effective (first gen. from digested sludge; recovery rate of ~10%^[30]) to the most effective method (second gen. from the centrate with Wasstrip; recovery rate of ~40%^[25]) 539–3.187 ton of P/ yr can be recovered in the form of struvite (Table 3), as 4.401–26.027 ton of struvite/ yr. This corresponds with 13.5-56.7% of the Dutch fertilizer demand.

Table 3. The recovery potential of scenario 3 and 4.

	Struvite precipitation (ton/ yr)				
	First generation			Second generation	
	From digested sludge	From centrate	Thermal hydrolyses upstream digester	Thermal hydrolyses upstream digester	From centrate with Wasstrip
Scenario 3	539	1.992	1.195	2.390	3.187
Scenario 4	1.144	2.862	1.717	3.434	4.579

Scenario 4: All WWTPs with a capacity higher than 50,000 p.e. will implement Bio-P removal and a struvite precipitator

Scenario 4 targets the implementation of Bio-P removal at the 165 large WWTPs in The Netherlands to allow the installment of a struvite precipitator at all of these 165 WWTPs as the key P recovery technique. Installment of the five different struvite precipitators in these WWTPs will afford 1.144–4.579 ton of P per year, which corresponds to 9.342–37.395 ton struvite/ yr. In terms of P, this covers 28.6–114.5% of the Dutch fertilizer demand, in the case that struvite could directly substitute the currently used fertilizers.

Combining the P recovery technologies struvite precipitation and recovery from SSAs?

So far, we investigated P recovery from either SSA or by struvite precipitation. Ultimately, a combination of struvite precipitation and sewage sludge ash production should lead to the highest recovery of P from the influent, yet the technical feasibility of this concept is currently not well developed. Namely, struvite precipitation at WWTPs lowers the P content in the sludge and SSA derived thereof, which makes P recycling from SSA more challenging.^[25] Typically, sewage sludge ashes of a WWTP without struvite precipitator contain 127 kg P/ day (per 100.000 p.e.). This reduces to 93 kg P/day (per 100.000 p.e.) when the Airprex (first-gen.) technology is installed, and to a mere 56 kg P/day (per 100.000 p.e) when the most efficient struvite precipitator (2nd gen, from centrate with Wasstrip) has been implemented.^[25]

Bridging the gap between P recovery and P recycling

We highlighted in our scenario analyses that the amount of P-based fertilizer used in The Netherlands and the amount of P that can be recovered from the wastewater

via the existing WWTPs is in balance. Unfortunately, the recovered material has not the same properties as the currently applied fertilizers. For example, struvite does contain the macronutrients P and N, yet is sparsely soluble at neutral pH, but more soluble at low pH or above a pH of 9.^[31] Therefore, the acidity of the soil is crucial for its bioavailability to crops and thus its effectiveness as fertilizer on the fields.^[32] Struvite contains a high percentage of the micronutrient magnesium, which when in surplus can have detrimental effects that reduces the availability of other nutrients in the soil (antagonistic effect), such as calcium and potassium.^[33] Furthermore, due to the different struvite precipitation techniques, also the quality of the struvite and its P content varies, between 11–26%, see chapter 2.^[34]

Also the reuse of sewage sludge has its challenges. The direct use of (treated) sewage sludge as a biosolid to enrich agricultural soils is prohibited in The Netherlands due to the high quantities of heavy metals and organic pollutants.^[35–37] After incineration, the sewage sludge ashes (SSA) are free of organic pollutants, pathogens and pharmaceuticals,^[38] but require additional treatment to increase the bioavailability of P and to stay within the threshold limit values of several heavy metals that are still present.^[38] A promising avenue for recycling of the recovery P is to convert SSA into the highly soluble phosphoric acid, which is a known fertilizer. Currently, up to 97% of the P in SSA can be converted into phosphoric acid via leaching,^[30] yet this technology has not been implemented yet on large scale.

4.5 Conclusions and Recommendations

The largest phosphorus losses occur in the wastewater treatment sector. A particular solution that has shown relevant opportunities in Europe is via phosphorus removal at wastewater treatment plants by generation of various P recovery products, such as struvite and sewage sludge ash. Various types of policies can stimulate the recovery of P from communal wastewater. This study provides policy makers the needed quantitative and qualitative insights to evaluate existing policies and legislation and to define potential policy measures on P recovery and recycling.

In The Netherlands, 100% of the sewage sludge is mono-incinerated. With the amount of P in Dutch SSA, the Netherlands could easily meet the supply demand for fertilizers (275%). Currently, 57% of the dry weight of our SSA is derived from sewage sludge from chemical dosing which contains high metal concentrations. An increase of the amount of Bio-P SSA, now 43%, would increase the quality of the SSA and promote recycling, due to its higher bioavailability and higher P concentration.

The second P recovery technology from communal wastewater is struvite precipitation. If all Bio-P WWTP bigger than 50.000 p.e. (the size to make the implementation of a struvite precipitator economically feasible) would implement the most efficient and industrial applied struvite precipitator, 79,7% of the total P demand for fertilizers in the Netherlands could be covered. In case all WWTPs bigger than 50.000 would implement a struvite precipitator, this will even be 114,5% of the Dutch demand for fertilizers. This could even increase if all smaller WWTP would merge into bigger (>50.000 p.e.). Therefore, introducing legislation, such as the new German legislation that obliges all WWTPs (> 50.000 p.e.) to recover P, could

stimulate P recovery. The next step after recovery is recycling. Therefore, it is vital for the creation of a circular phosphorus economy that the end-products have a market demand.

In this study, we focused on the P recovery potential from WWTPs as urban mines. There is still much room for improvement, since only 7 out of 340 WWTPs have implemented P recovery so far. With the European fertilizer regulation currently under revision, the economic feasibility of the implementation of P recovery technologies and the untapped and the local potential of the urban mines, this is the time to develop these opportunities. In addition to communal wastewater, there are other P rich waste stream as in the food processing (28% of total losses of the system) or food waste (15%) sector that show great potential for P recycling too.^[5]

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