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Phosphorus Recovery from Municipal Wastewater. A Case Study : the Wastewater Treatment Plant of Linz Asten

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supervised by

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Affidavit

I, **AGNES CHANUT**, hereby declare

1. that I am the sole author of the present Master's Thesis, "PHOSPHORUS RECOVERY FROM MUNICIPAL WASTEWATER. A CASE STUDY / THE WASTEWATER TREATMENT PLANT OF LINZ ASTEN", 50 pages, bound, and that I have not used any source or tool other than those referenced or any other illicit aid or tool, and
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Abstract

Phosphorus (P) is an essential nutrient, which is vital for living organisms. Before the industrial revolution, it used to follow a natural life cycle, which is now broken: the P-cycle has become a linear process starting in the phosphate-rock mines, travelling through wastewater treatment plants (WWTP) and ending in landfill and in the aquatic environment. Phosphorus is mostly used in agriculture, entering in the composition of fertilisers. Due to the fact that it is a broken cycle as well as the fact that its resources are concentrated in three non-European countries, it is becoming a critical raw material in Europe. P is also a pollutant, which is at the origin of eutrophication problems in surface waters. In order to limit eutrophication, more and more WWTPs in Europe treat phosphorous, which ends in sludge and not in effluent. The P-flow estimation at a global scale, as well as the European and Austrian scale shows the great possibilities of recycling through wastewater treatment. A view on the European and Austrian legislation shows that the concern is traditionally on phosphorus as a fertiliser and pollutant. However, a new EU regulation on fertilisers - which is still in draft form - tends to begin to consider P as a critical raw material that must be saved and recycled.

This thesis aims to test the possibilities of installing recycling process on the WWTP of Linz (Austria), which has a capacity of 950 000 population equivalent and a phosphorus treatment process. Three main techniques exist, depending on their location: the treatment of digested supernatant, the sludge or the ashes. All techniques have advantages and disadvantages in terms of costs, establishment and implementation easiness. Moreover, the final products, struvite, calcium phosphate or ashes, usually sold as fertilisers, have different plant availability. The techniques tested in this thesis take part in the European project P-REX: Struvia, Ostara/Pearl, AirPrex, Gifhorn, Stuttgart, Leachphos, Ecophos, AshDec. As a methodology, several MFAs with the software STAN were built on the flows of phosphorus at Linz WWTP. It includes multiple scenarios: the actual state, as well as the projected state with the set-up of the different recovery techniques. A cost-benefit analysis and a multi-criteria analysis were used to rank the techniques that are the most adaptable to Linz WWTP. Almost all the techniques are economically feasible, and the most relevant ones including all the criteria are ECOPHOS (ash leaching) and STRUVIA (liquor precipitation). The implementation of Ecophos would require a change in the wastewater management practices in Austria, with a mono-incineration plant instead of the current co-incineration. The thesis concludes with the importance of the political willingness and an adapted legislation in order to close the phosphorus cycle.

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Introduction

Phosphorus is without a doubt an essential nutrient for humans and plants. It is the main component of the skeleton of living organisms, an essential structural component of RNA and DNA and has an energy storage function in the composition of ATP. Thus, it is an important resource for agriculture and enters in the composition of most of the fertilisers.

Phosphorus resources are limited. Nowadays, the majority of phosphorus production comes from mining and recycling rates are low. In addition, a synthetic substitute for phosphorus has not yet been developed. Moreover, phosphorus deposits are unequally distributed around the globe. The phosphorus market is very volatile, and the long-term economic trend has been a growth in price. Tensions in the market are growing since China, one of the main producers, has increased export taxes on phosphates (Sinaï, 2013). It is for this reason that the European Union listed phosphorus as a critical raw material in 2014 (Corazza, 2014).

Therefore, in Europe there is an increasing interest for alternative solutions for phosphorus recycling. In March 2013, the first European Conference on Sustainable Phosphorus took place, gathering industrialist experts, and European commissioners and deputies (Sinaï, 2013). The outcome document declaration of the conference affirmed the challenge and need for phosphorus recycling:

> *"[G]reater recycling and use of organic phosphorus where it is needed could stabilize the amounts of mined phosphate required and mitigate the soil contamination and water pollution issues. This will then put us on track to close the phosphorus cycle in the long term, when the physical limitations of the resource will become increasingly important"* (European Commission, 2013).

A great recycling potential is found in wastewater treatment plants (WWTPs). Indeed, the endpoint of the phosphorus cycle is faeces and urine and thus phosphorus is transported to WWTPs. Apart from being a nutrient, phosphorus is also a pollutant that can be harmful to natural water bodies, creating a phenomenon called "eutrophication", characterised by excessive algae production, anaerobic sedimentation and biodiversity depletion. In this way, WWTPs usually also treat phosphorus (to a greater or lesser extent depending on the process). At treatment plants, the phosphorus contained in the wastewater can only leave the plant either in the effluent or in the sewage sludge or its treatment by-products.

As the recycling of phosphorus from wastewater is of great social and environmental interest, many research papers, pilot projects, patents and start-up businesses deal with phosphorus-recycling techniques. However, as new technologies are yet to be proven effective and feasible, phosphorus recycling is still not a common practice.

The European P-REX project tries to fill this gap by setting up full-scale P-removal on various WWTPs. For every technique, developed by different companies, this European project analyses real operational performance, quality of the products obtained, demand in energy and chemical compounds, and costs (Kabbe, 2016). The next logical step will come when effective and feasible techniques arrive on the market. An operator or decision-maker willing to implement phosphorous recycling (or P-recycling) at a WWTP will have to select the best solution adapted to the specific local situation. To understand the challenges of such a situation, this thesis takes the WWTP of Linz, Austria, as a case study for P-recycling.

After a review of phosphorus chemistry and the European and local legislation concerning phosphorus, this thesis describes the main recycling techniques available. Following this, the WWTP of Linz is presented and the different recycling techniques are applied.

1 Phosphorus: cycle and chemistry

In 1669, a German physician, Henning Brand, first discovered phosphorus while looking for the “philosopher’s stone”. Unsurprisingly for anyone who is aware of the phosphorus cycle, he isolated phosphorus while processing urine; hence, the discovery of phosphorus shows the importance of wastewater in its cycle. After a review of the phosphorus cycle this chapter will examine the chemistry of phosphorus in wastewaters.

1.1 Phosphorus: a broken cycle

In the past, the phosphorus cycle was in equilibrium. The natural concentration of phosphorus in the soil was guaranteed by the decomposition of organisms, the excretion of animals and the natural weathering of rocks. Phosphate solubilising bacteria as Pseudomonas, Bacillus or Rhizobium also play a major role in the natural P-cycle: they increase P-uptake by plants, transforming insoluble P-rock into plant-available phosphorus by mineralisation (Rodríguez and Fraga, 1999).

Since 1950, the natural P-cycle has changed due to human activity. Nowadays, the agricultural sector is the main driver of phosphorus demand (Egle et al., 2015). As mentioned in the introduction, the majority of phosphorus comes from mining. Once mined and processed, phosphorus is added to soil in the form of mineral fertiliser as a nutrient for enhanced crop production.

After fertilisation, plants immediately take up part of the available phosphorus. The rest remains as excess phosphorus. A part of this - depending on the type of phosphorus - stays in the soil where it is available for further utilization as a phosphorus stock. In addition, part of the excess phosphorus can also be transferred to surface waters or groundwater by run-off and percolation. These processes constitute one cause of eutrophication and algal bloom.

The phosphorus taken up by plants is either returned to the soil by plant degradation or enters the food chain through consumption/ingestion by animals and/or humans. The food chain has several levels, but in every case the phosphorus ends in the faeces and urine of animals or humans. A large part of excreta from animal breeding activities is applied directly to the soil as manure. In developed countries, most of the human excreta end in municipal wastewater treatment plants. Thus, phosphorus can be found in the final products of a municipal wastewater treatment plant: effluent and sewage sludge in a proportion depending on the treatment process. It is at this stage of the anthropogenic P-cycle that recycling can take place. Phosphorus can be recycled directly through the application of sewage sludge, but the acceptance level for this process is very low in developed countries due to the potential risks for human health that result from the presence of organic pollutants and pathogens in sludge (European Commission, 2010).

For this reason, sewage sludge is often incinerated or co-incinerated, a process whereby the phosphorus present is stored in the ashes and cannot be reused (Egle et al., 2015). In Austria, 40% of sludge is incinerated (European Commission, 2010).

1.1.1 Phosphorus uses

Agricultural use represents 80% to 90% of the world demand for phosphorus. In addition to this, phosphorus is traditionally used in the production of detergents. However concerns over eutrophication have led to these detergents being increasingly phased out in developed countries (European Commission, 2010). Phosphorus is also used in industry, including in metal surface treatment, flame-retardants, special glasses or porcelain production. However, these applications represent a minor proportion of total phosphorus use (European Commission, 2013). Figure 1 illustrates the phosphorus cycle.

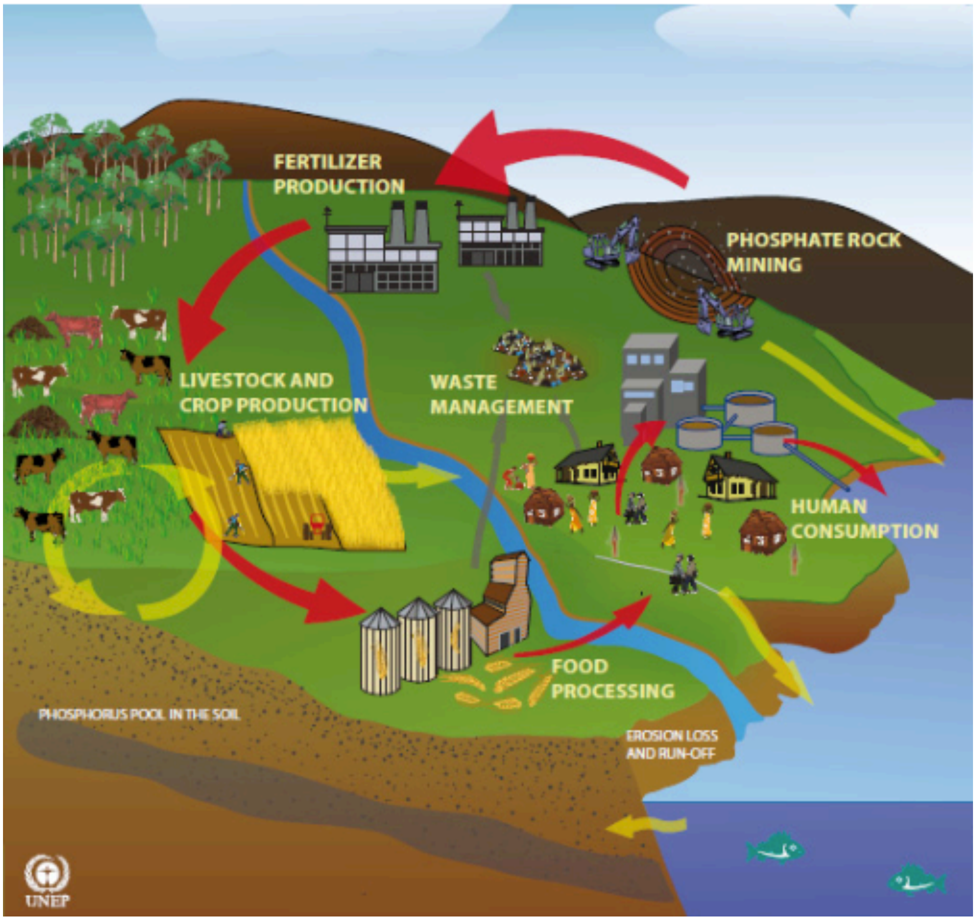

Figure 1: Phosphorus cycle (UNEP, 2012)

1.1.2 Estimation of the flows

1.1.2.1 *Global system*

Clift & Shaw (2012) analysed the yearly total flows of phosphorus in the global system (Table 1). They estimate that the total input to food production is around 27 million tons, of which more than half is mined. About 3 million tons of phosphorus ends up in food. The rest, about 24 million tons, is stored in agricultural soils or lost and discharged in oceans. Looking at these numbers, it is clear that the phosphorus cycle deserve its name as the “broken cycle”: it is more of a linear process than a cycle, where the main flow of phosphorus originates from rock mining and ends in the ocean or increased storage of phosphorus in soils (which also increases the eutrophication potential by erosion).

Table 1: global phosphorus flows (Clift & Shaw analysed, 2012)

PHOSPHORUS INPUTS	
From mineral fertilisers	15 million tons
Vegetation fed to livestock	12 million tons
TOTAL INPUT	**27 million tons**
PHOSPHORUS OUTPUTS	
Food	3 million tons
TOTAL OUTPUT	**3 million tons**
PHOSPHORUS LOSSES	
Distribution losses	1 million tons
Erosion, leaching losses	8 million tons
Post harvest and supply chain losses	5 million tons
Losses as human excreta	3 million tons
Losses as animal excreta	7 million tons
TOTAL INPUT	**24 million tons**

This analysis is interesting to see the global cycle of phosphorus but it neglects the storage of phosphorus in soils, which is very important for the P-cycle.

1.1.2.2 *Europe and Austria*

The European Commission has proposed an estimation of the P-cycle in Europe. (European Commission, 2013). Apart from limited mineral resources in Finland, Europe is largely dependent on phosphorus imports, which amount to about 2,7 million tons. By contrast, exports (mainly of agricultural products) are around 0,6 million tons. The net import of 2,1 million tons of phosphorus is roughly divided into 70% mineral fertilisers, 20% animal feed, and 10% food and non-food materials. The P-losses are

1,25 million tons. On average, about 40% of phosphorus in sewage sludge is recycled through direct application within the EU-28.

In Austria, as yearly average for the period 2004-2008, P-imports are 70 000 t, whereas exports are 52 000 t P/y. Thus, the stock is annually growing by 18 000 t. The quantity of phosphorus that enters the Austria WWTPs is 7 800 t and 6 600 t is transferred to sewage sludge by treatment (Egle et al., 2014). In 2005, only 18 % of sludge was reused in Austria (European Commission, 2010). With a potential recovery of 50 %, around 3 300 tons of P could be recycled every year in Austria, if P-recovery was applied to WWTPs. This represents around 5% of the current imports.

Austria, like almost all of the other EU-countries, is entirely dependent on imports of phosphorus. To understand one of the main incentives of a phosphorus-recycling economy, it is necessary to look a bit closer at the status of the phosphorus reserves worldwide.

1.1.2.3 Phosphorus rock reserves

Phosphate rock exists as three forms of calcium phosphate: hydroxyapatite, fluor apatite, and chloroapatite. In addition, one can distinguish between two major types of phosphate rock: sedimentary (carbon apatite) and igneous rock.

The largest reserves of phosphate rocks are located in Morocco including occidental Sahara, followed by United States and China (European Commission, 2013). These three countries that hold 85% of the world reserves are also currently the main world producers. Phosphorus resources in Europe are very limited, with Western Europe having no production at all and with Eastern Europe producing just enough for their own consumption (Nieminen, 2010). In addition to this constraint, the most common process for phosphorus extraction requires the use of sulphuric acid. Countries in the Northern Hemisphere are the main producers of this, as sulphuric acid is a by-product of the petroleum refining industry (Nieminen, 2010).

In addition to this imbalanced resource distribution, geopolitical problems are arising, which could cause supply disruptions. For example, the nature of Morocco's ownership of the Western Sahara is contested by a large proportion of the international community and in disagreement with UN resolutions. An other example is the weight of China. In 2008, as a protection measure, the government applied a series of seasonal tariffs on phosphate exports. The aim was to prevent domestic prices from global short-term fluctuations and to ensure the local availability of the product. As a result, the global price of phosphate raised dramatically. In 2015, China announced an easing on export tariff for fertilisers (Persona, 2016).

The risk of price volatility and supply disruptions is high due to the concentration of phosphate rock mining in a limited number of countries, as well as potential geopolitical instability (de Ridder et al, 2012).

On the other side of the chain, the demand is growing continuously. The United Nations estimate that the global population will reach over 9 billion people by 2050. As a result, the world food production will need to increase by 70% in the coming decades, implying an increasing phosphate fertiliser demand. This will put further pressure on the demand of phosphate rock, making it a scarce resource (de Ridder et al, 2012). Consequently, the estimates of phosphorus resources sufficiency vary between 60 and 240 years (Cornel and Schaum, 2009).

In 2008, the phosphate rock market underwent a crisis (Figure 2). Due to a high agricultural demand combined with a tightened supply of phosphate rock, the price of phosphate skyrocketed.

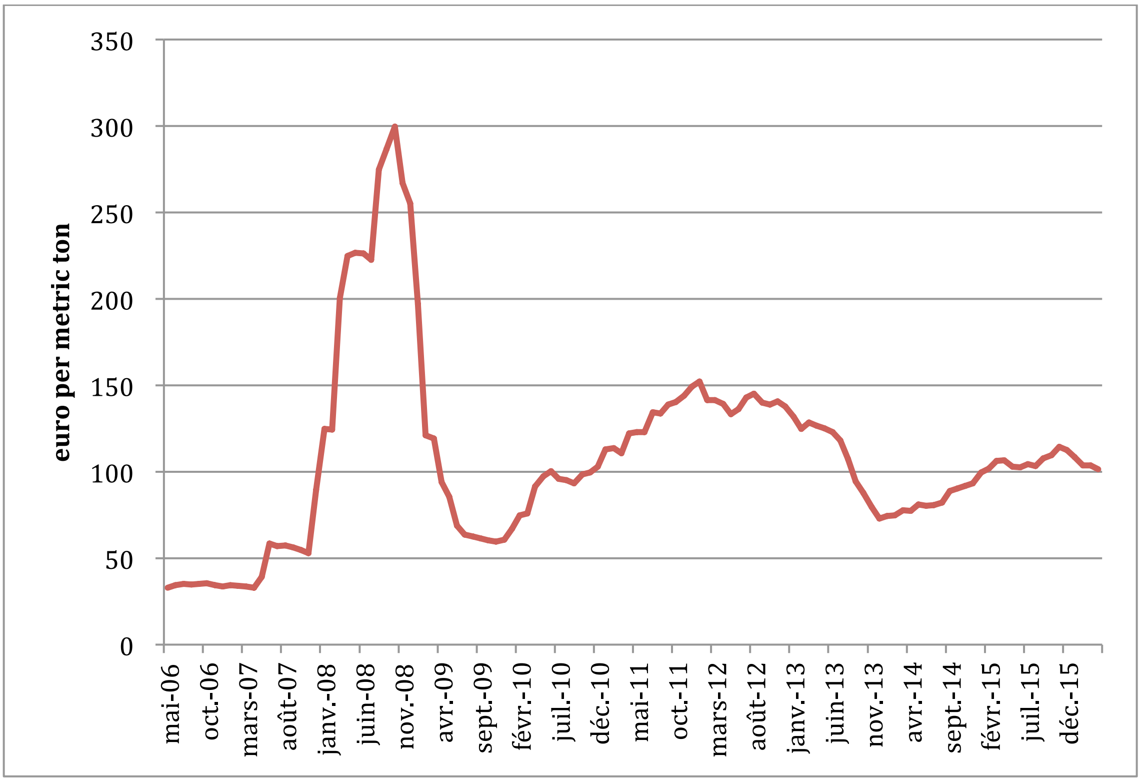

Figure 2: Price of phosphate rock (Morocco), 70% BPL (source: indexmundi, 2016), corrected with inflation

As with any other mineral resources, different quality levels of phosphate rocks exist. The quality level is judged on the basis of phosphate concentration, as well as the presence of contaminants. Typically, phosphate rock contains around 15 % P (Schipper et al. 2001). Regarding contaminants, uranium and cadmium are of particular concern because they are radioactive and toxic, respectively, and are not completely removed during the refining process. The best quality rocks require less cleaning and processing to produce mineral fertiliser and are more economically feasible. As a result, they are prioritised in the mining process. This has led to a decline in the availability of good quality phosphate rock, as well as an increasing price trend.

To summarize, phosphorus reserves are limited, the market is volatile, and a trend of increasing prices and decreasing quality is currently being observed. Thus, there is a significant interest in improving the recycling of this resource within the EU.

1.2 Phosphorus removal in WWTP

1.2.1 Phosphorus chemistry

To efficiently remove phosphorus from wastewater, sludge, or effluent, it is important to have an understanding of the behaviour of phosphorus in wastewaters. Therefore, this chapter presents the basic chemistry of the element, with a special focus on phosphorus in wastewaters.

Phosphorus has the atomic number 15 in the Mendeleïev table, has an atomic mass of around 31 g/mol, and only have one naturally occurring isotope: 31P. Phosphorus oxidation states are -III, +III, and +V. Phosphorus has three allotropes: white (or yellow), red and black phosphorus. The most common allotrope is white phosphorus, or tetra-phosphorus (P4), a very reactive and highly poisonous element, which has been used since World War I in bombs and weapons due to its their smoke-generating and burning properties. On the other hand, red phosphorus is not very reactive and is used in matches. White phosphorus transforms to red phosphorus at ambient temperature. In the natural environment, phosphorus occurs in its fully oxidised form orthophosphate PO_4^{3-}.

1.2.2 Phosphorus behaviour in wastewater treatment plants

Phosphorus is present in wastewater either in a particulate phase or in a dissolved phase. Particulate matter includes precipitates of phosphorus and phosphorus absorbed in particles. The dissolved state of phosphorus includes inorganic and organic phosphorus compounds.

Phosphorus exists in natural waters in the form of inorganic phosphates (orthophosphates and polyphosphates) or organic phosphates. Organic or organically-bound phosphates are phosphates that are bound to plant or animal tissue and faeces, they are found in organic matter and cell protoplasm, and are formed primarily by biological processes. Inorganic phosphates are phosphates that are not associated with organic material. These include orthophosphates, which are directly available for biological metabolism and sometimes referred to as “reactive phosphorus”. This is the form used by plants. Inorganic phosphates also include polyphosphates (also known as metaphosphates or condensed phosphates), which are more complex molecules. Usually, polyphosphates are unstable and undergo hydrolysis to convert to the orthophosphate form through a relatively slow process.

The typical Phosphorus concentration at the inlet of a municipal WWTP is around 10 mg/l (+/- 5 mg/l), mainly depending on the dilution (drinking water supply and infiltration of groundwater to the sewer system). In general, approximately half of this is found in the soluble inorganic orthophosphate form, 35% comprises polyphosphates (complex inorganic phosphates), and 15% comprises organic phosphates (Nieminen, 2010).

Phosphorus bound to solid particles is to a large extent removed during primary sedimentation. This counts for about 11% of total inlet phosphorus load. In the secondary treatment, the biological phase (e.g. activated sludge process) needs phosphorus as the substrate for micro-organism growth and can remove 25% to 40% of phosphorus without specific phosphorus removal processes. Most of the European WWTPs are equipped with specific phosphorus removal processes, due to the requirements for sensitive areas imposed by the EU law (Wastewater Directive). With these techniques, such as chemical precipitation and/or enhanced biological P-removal, the rate of phosphorus removal can reach 95% (Nieminen, 2010).

1.2.3 Removal techniques

As a prerequisite for the set-up of phosphorus recycling in a WWTP, the plant must first remove phosphorus from the wastewater. Understanding the processes of phosphorus removal is important to understand and adapt the techniques of phosphorus recycling. Phosphorus removal can be divided into two main approaches: chemical and biological processes.

1.2.3.1 Chemical phosphorus removal

Chemical precipitation of dissolved phosphorus from wastewater is a simple and effective method to remove phosphorus from the wastewater. Aqueous solutions containing two or three valent metal salts are added to the wastewater. These react with the dissolved phosphorus, forming solids that can be easily removed with the sludge. The most commonly used phosphorus removal chemicals are iron salts containing ferric and ferrous ions. Aluminium and calcium are also used as precipitants. The chemical precipitation reaction can happen at several locations in the wastewater treatment plant, but the most common chemical removal procedure takes place during secondary treatment, through the so-called simultaneous precipitation procedure (Metcalf and Eddy 2003).

Although chemical precipitation is efficient, it also has some negative effects. For example, it causes an increase in solid load (Lenntech, 2016), and is sometimes unfavourable for phosphorus recovery processes. In addition, the operating costs of the chemical process are depending on the prices of the precipitants, normally estimated at less 1 €/p.e/y.

1.2.3.2 Biological phosphorus removal

Phosphorus Accumulating Organisms (PAOs), are the major actors in biological phosphorus removal. They are bacteria, the most common being Acinetobacter sp. PAOs can work under both aerobic and anaerobic conditions. Under aerobic conditions, they store energy rich polyphosphates in their cells, and in the anaerobic stage, they are able to accumulate energy rich organic material by using the energy contained in the polyphosphate by releasing the accumulated phosphorus in form of phosphate.

Biological phosphorus removal has several advantages compared to chemical phosphorus removal. First of all, the sludge produced has a higher content of plant-available phosphorus and thus better value in agricultural use. Moreover, the existence of biological phosphorus removal improves the possibility for several phosphorus recovery processes. The additional sludge production is lower than for precipitation. However, the enhanced biological P-removal also has disadvantages. In particular, it is reported that the thickening properties and dewaterability of excess sludge from bio P removal plants deteriorate. In addition, the excess sludge must not be thickened together with primary sludge, because this causes anaerobic conditions in the thickened sludge resulting in the release of the stored P to the water phase again.

In order to fulfil the standards for phosphorus removal at any time -especially during winter time- in WWTP, it is reasonable to combine both processes. It may especially be the case for sensitive areas subject to the Urban Wastewater Directive.

1.2.4 Phosphorus flow at sludge treatment and disposal

The wastewater treatment essentially transfers phosphorus from the liquid to the sludge phase, therefore, the sludge treatment processes selected have a great impact on phosphorus management of a WWTP and on the quantity and availability in the different streams: primary sludge, secondary sludge, and digested supernatant.

1.2.4.1 Sludge treatment by anaerobic digestion

Anaerobic digestion is a very common method for sludge stabilisation in WWTPs. It is a process that allows the sludge to be stabilised (for odour abatement) and reduced in solids load without the use of oxygen. Under anaerobic conditions, micro-organisms convert organic compounds into gaseous products, mainly CH_4 and CO_2. As long as the digestion process is operated at constant low fatty acid concentrations (with a sludge detention time > 20 days), P-release to the liquid phase is quite limited by the availability of fatty acids. As a result, side streams from digested sludge (supernatant from thickening and dewatering) remains low in P. Accumulation of fatty acids in the digester due to bad operation results in a nearly complete release of the stored polyphosphates to the supernatant.

1.2.4.2 Mono-incineration

After its treatment, the sludge can be disposed of as it is in a landfill or used on fields as P-fertiliser, or incinerated. Incineration has several advantages: it considerably reduces the volume and the solids load, produces heat that can be used as energy and produces a chemically inert material. Moreover, incineration eliminates organic contaminants and pathogens: organic matter is combusted to CO_2 and pathogens are removed with water vapour. Thus, incineration avoids odour problems, hygienic risks by pathogens and also destroys organic micropollutants.

Phosphorus is not volatile and concentrates in the ashes. As a result, sewage sludge ashes are potentially good fertilisers. In the European Union most of the incinerated sewage sludge is co-incinerated with municipal waste, bark or coal products. This prevalence of co-incineration over mono-incineration is due to economic reasons. As a result, the ashes from co-incineration plants are much lower in P-content (dilution effect) and can contain additional contaminants as heavy metals. The ashes from co-incineration plants are usually sent to landfill.

Mono-incineration is more costly but is favourable for the recycling of the phosphorus in the ashes. To be used as a fertiliser, sludge can be incinerated with other products rich in phosphorus as ground animal bones, food waste or manure.

2 Legislative context

An understanding of the different levels of legislation is crucial to be able to propose solutions for feasible phosphorus removal. Two levels are drivers in Austria: European and national legislations.

2.1 European legislation

2.1.1 Phosphorus as a pollutant

The four main directives concerning phosphorus in the European Union are considering phosphorus as a pollutant: the Water Framework Directive, the Urban Waste Water Directive, the Sewage Sludge Directive and the Waste Framework Directive. These directives are legally binding for each member state. National authorities must adapt their laws and legislations to meet these goals, but they decide how they want to achieve them. In contrast to directives, regulations are directly binding in every member state. Member states may also take measures regarding phosphorus in line with the Common Agricultural Policy.

In addition to directives and regulations, conventions contribute to national or regional legislation.

The purpose of the Water Framework Directive (2000/60/EC, WFD) is to protect EU surface waters and groundwater. Many daughter directives derive from it, for example the Nitrates Directive or the Groundwater Directive. The main goal of the WFD is to achieve and to maintain a “good status” for all surface waters and ground waters by 2015 (Amery and Schoumans 2014). The concentrations of total phosphorus, as well as PO_4^{3-}, take part in the status definition of the water body, the limits are locally fixed state by state.

The Urban Waste Water Directive 91/271/EEC is a part of the Water Framework Directive. It defines the discharge limits for total phosphorus concentrations in wastewater treatment plant effluents. In “sensitive” areas subject to eutrophication, the concentrations of total phosphorus should be less than 2 mg/l when the size of the WWTP is between 10 000 to 100 000 population equivalent (p.e) and less than 1 mg/l when the size is larger than 100 000 p.e.. Moreover, the minimum reduction rate of phosphorus is set at 80% of the load of the influent. Depending on the local conditions, only one or both of the limit values should be applied (Nieminen, 2010).

The Sewage Sludge Directive 86/278/EEC has encouraged and regulated the recycling of sewage sludge in agriculture since 1986. The aim is to prevent harmful effects on humans, animals, soil and vegetation. It prohibits the use of untreated sludge on agricultural land, unless it is incorporated or injected into the soil. The Directive also requires that the use of sludge takes into account that the quality of the soil and of the waters is not impaired, as well as that the nutrient requirements of plants is fulfilled It gives maximum permissible concentrations of potentially toxic elements in sludge-treated soils regarding Cd, Cr, Cu, Hg, Ni, Pb and Zn.

The Waste Framework Directive 2008/98/EC sets the definitions as well as the basic concepts of waste management. It also defines “end-of-waste criteria”, which specify when waste is a not a waste anymore and becomes a secondary raw material or a product. From now, only some products as iron, steel aluminium scrap, glass cullet and copper scrap have their criteria. Sewage sludge is considered as a waste.

In addition to these major directives, the Common Agricultural Policy (CAP) has defined the agricultural policy of the European Union since 1962. It defines, among other things, a system of agricultural subsidies. In accordance to CAP, farmers have to keep their land in a good agricultural and environmental condition. Moreover, Agri-Environmental Programmes are compulsory for every Member State, and phosphorus application restrictions can be included in these (Amery and Schoumans 2014).

2.1.2 Phosphorus as a nutrient and an essential raw material: a new regulation

The Fertilisers Regulation in force in the EU since 2003 ensures free movement for traditional fertilisers, but it does not include a clearing procedure for organic fertilisers, which are made from recycled products. Thus, a new Fertiliser Regulation, available since March 2016 as a draft, is now replacing the old regulation. After adoption by the European Parliament and the Council, it will be immediately applicable. This proposal defines new rules on organic and waste-based fertilisers in the EU and affirms that phosphorus is an essential raw material for European agriculture. The text emphasizes the possibilities offered by domestic waste and in particular sewage sludge, which contains large quantities of phosphorus. Recycling this phosphorus within a circular economy model could cover about 25 % (+/- 5%) of European Union's demand for phosphate fertilisers. Moreover, implementing P-recycling at a large scale would decrease Europe’s dependence and increase its resource efficiency. To achieve this goal, the proposal contains several rules such as free movement for all CE marked fertilised or updates for CE marking on organic fertilisers. New regulation will be set-up for bio-waste (composts and digestates). If marked as “CE marked fertilisers”, bio-waste will no longer considered to be waste within the meaning of the Waste Framework Directive. With this new directive, it is clear that P-recycling will be encouraged in the next few years within the EU.

2.2 National legislation

In Austria, the total P concentration requirement at the effluent of a WWTP is 1 mg/l for treatment plants with a size above 5 000 p.e and 2 mg/l for treatment plants that are between 500 p.e. and 5000 p.e.. Concentration requirements are not set for treatment plants under 500 p.e (Bundesrecht konsolidiert, 2016).

In relation with the WFD, the Austrian "Water Act" set up the limits for PO_4-P in running water bodies depending on the bioregion they belong to and their initial trophic state. The reference condition for the trophic index only depends on the elevation of the water body. For example, the Danube near Linz belongs geographically to the bioregion AV (Alpine Molasse Bayerisch- Österreichische Alpenvorland). The elevation being below 500 m above sea level, the trophic index is mesotrophic (mt). The Water Act sets-up two concentrations limits for PO_4-P depending on the status: the PO_4-P concentration must be lower than 0,020 mg/l to achieve a very good trophic status and lower than 0,050 mg/l to achieve a good status. The concentration calculations are based monthly data for 2 years and a 90-percentile compliance (Bundesrecht konsolidiert, 2016).

Regarding the use of sludge in agriculture, legal requirements differ from federal state to federal state in Austria. Some states have banned sewage sludge application in agriculture. The best practices are described in the Austrian Guideline 17 issued by the Austrian Water and Waste Association (ÖWAV). Sludge must not be applied during winter or late autumn. A minimum storage capacity of 6 months is necessary to fulfil this requirement (Miller, 2016).

Austria does not currently have any P restrictions or maximum phosphorus application rates (Amery and Schoumans, 2014). Moreover, since the main risk related to phosphorus fertilisation is eutrophication, Austria, as most other European countries, has defined buffer zones along waterways as additional legislation, where no fertilisation is allowed. The width of this buffer zone is 0,5 m, corresponding to the minimum value that is allowed according to European legislation. Grazing is allowed in the zone in order to limit erosion (Amery and Schoumans, 2014).

According to Austrian legislation, the minimum content of a phosphate market fertiliser must be 4,3 % P. The legislation also limits heavy metals as follows:

- Lead: 100 mg/kg TM
- Cadmium: 75 mg/kg P_2O_5
- Chrom VI: 2 mg/kg TM
- Nickel: 100 mg/kg TM
- Mercury: 1 mg/kg TM

- Vanadium: 1500 mg/kg TM
- Arsenic: 40 mg/kg TM

Finally, legislation also limits hygienic parameters: Escherichia coli Salmonella sp. Campylobacter sp and Listeria monocytogenes must be absent in a given 50 g sample. Additionally, the legislation limits organic pollutants, radioactivity and residues.

3 Phosphorus recycling techniques

Numerous methods exist for phosphorus recovery from wastewater. The procedures differ according to the location of the process within the WWTP, the form of phosphorus, the recovery rate potential and the costs. The ideal method would combine a high phosphorus recovery rate with a low production cost, and final products low in hazardous compounds, which can easily be applied in agriculture or industry and can be marketed.

Phosphorus can be removed at different locations in the treatment process: from urine when separation toilets are used, from wastewater or from sludge treatment. Ashes from sewage sludge incineration are also very promising for P recovery.

Phosphorus removal from liquids can be achieved through different processes: chemical treatment, biological processes, or a combination of both. These processes are often the first step in P recovery from wastewater.

3.1 Location of the techniques

A wastewater treatment plant offers several locations for phosphorus recovery, presented in the figure below. Depending on the location, different raw materials can be used for phosphorus recovery: urine, wastewater, sludge or ash (fig.4).

Several techniques are only effective in regard to phosphorus in its soluble form of orthophosphate; it therefore is important to know the concentration of orthophosphate in the liquids to be treated for P removal.

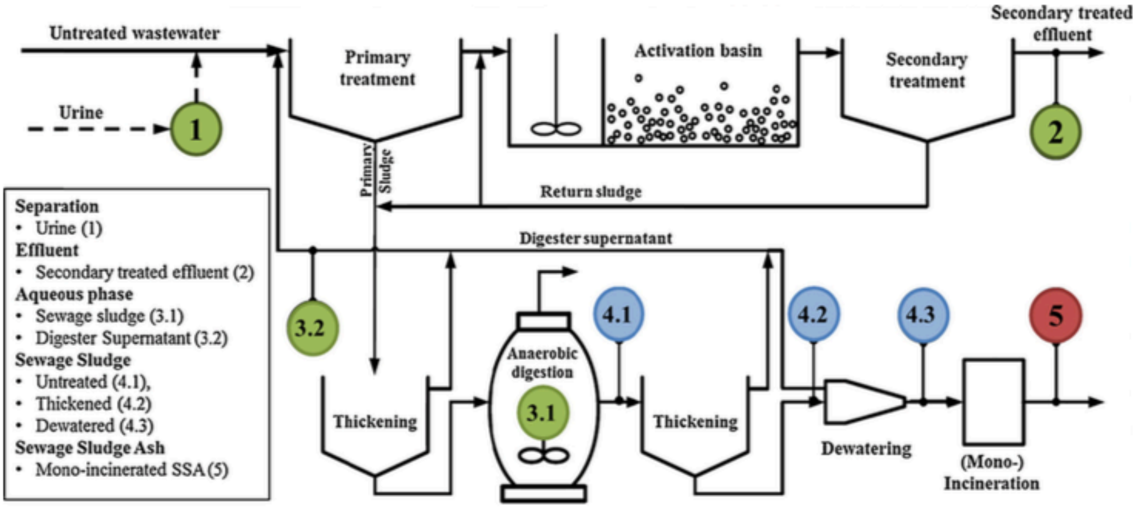

Figure 3: Possible access points for P-recycling approaches in a WWTP plant (Egle et al., 2015).

3.2 Final products for phosphorus recovery

3.2.1 Struvite

Struvite, or "Magnesium ammonium phosphate" also called MAP, has the formula $MgNH_4PO_4$-$6H_2O$. Struvite is the optimal phosphate mineral for recovery; since it also contains nutrients such as nitrogen and magnesium, it is very useful in the fertiliser industry. Moreover, it consists of easily accessible phosphorus and has slow-release properties both are in favour of agricultural application.

The optimal conditions for struvite precipitation occur under alkaline conditions (pH > 8,5). As the pH of wastewater is usually around 7, the formation of struvite requires an adjustment of pH. This adjustment is done by dosing alkalinity or, in digested sludge, by CO_2 stripping. The precipitation process can be limited by P, NH_4 and Mg. Without adding chemicals, P removal would be limited by a lack of magnesium, which is present in wastewater only at low concentrations. Normally P-precipitation can only be effective by addition of Mg. In some liquids, ammonia can also limit P-precipitation, even enough Mg is added.

The two possible magnesium sources are $MgCl_2$ and $Mg(OH)_2$. The first one, $MgCl_2$, is more common, because it is easier to carry and stock and it disassociates faster, requiring a shorter retention time during the reaction. The second one $Mg(OH)_2$, also has the advantage of aiding the adjustment of the pH level.

Struvite precipitates according to the simplified reaction:

$$Mg^{2+} + NH_4^+ + PO_4^{3-} + 6H_20 \rightarrow MgNH_4PO_4.6(H_20)$$

From this reaction, the stoichiometric presence of Mg and NH_4 can be derived for highly efficient P-removal.

Struvite formation can be separated into two stages: nucleation and growth. The first stage, nucleation, takes place when the constituent ions combine to form a crystal embryo. Following this, the second stage, crystal growth, takes place until an equilibrium is reached in the solution. In systems such as WWTPs where the struvite constituents are continuously replenished, crystal growth may continue indefinitely. Several parameters control struvite precipitation: temperature, pH, degree of supersaturation and the concentration of other ions (Doyle and Parsons, 2002).

Uncontrolled struvite-scaling in pipes and equipment is a common problem in the wastewater treatment industry. It occurs even in the anaerobic digestion tank and post digestion processes where or when the pH increases towards 8.5. Therefore digested sludge and digested sludge supernatant pipes are more frequently subjected to clogging.

Struvite deposition in pipes leads to an increase in pumping costs due to higher friction losses or even clogging of pipes. Struvite-scaling in pipes can result in reduced plant capacity or even to complete stand still in the case of clogging. Cleaning and replacing pipework are expensive and disruptive procedures. In addition to pipes also pumps, centrifuges and aerators can also be affected by the uncontrolled MAP precipitation (Doyle and Parsons, 2002).

3.2.2 Calcium phosphate (apatite)

As calcium phosphate is similar to phosphate rock, it is a very attractive form of recovered Phosphorus. For this reason, its utilisation is possible in both industry and agriculture.

The most thermodynamically stable form of calcium phosphate is hydroxyapatite (HAP), which has the chemical formula: $Ca_5H(PO_4)_3O$. However, calcium phosphates can also have other chemical compositions.

Calcium phosphate crystals do not form spontaneously in solutions with low concentrations. They only precipitate in P-rich liquids when calcium is added and the pH is above 8,2.

In wastewater, the formation of calcium phosphate crystals requires a very high oversaturation of component ions, as competition exists between all possible reactions. For this reason, the precipitation of calcium phosphate requires seed material, for example sand or calcium silicate crystals.

3.2.3 Ash

Ash is the final product of sludge incineration. It consists of two components: coarse particles and aerosol particles (also called flue ash). The incineration process has some important advantages for phosphorus recovery, such as the fact that it concentrates the phosphorus content as compared to dewatered and dried sludge. Moreover, incineration destroys organic matter, including organic micro-pollutants. Sewage sludge ashes are usually landfilled. There are applications in road construction or cement production but in both cases the phosphorus is “lost” for future generations.

Incineration destroys the organic matter and the nitrogen compounds contained in sewage sludge. Both compounds are relevant in agriculture but the potential contribution of sewage sludge is comparatively low. Moreover, incineration does not remove heavy metals from the ash; on the contrary, the process concentrates them in the ash, particularly in the aerosol fraction.

Typically, sewage sludge ash in WWTPs with P-removal contains approximately 8% P. The other main chemical constituents are SiO_2, CaO, Al_2O_3 and Fe_2O_3, depending on the chemical phosphorus precipitant.

3.3 Market opportunities for phosphorus recycling

Sewage sludge has a potential commercial relevance in agriculture as a fertiliser, due to the fact that it contains phosphorus, other nutrients and organic materials (Kroiss, 2016).

At first sight, the direct spreading of sludge on agricultural fields is the easiest and the most economical process for the recycling of phosphorus and other nutrients. However, the sludge contains micro-pollutants, pathogens and heavy metals that are perceived as harmless to soils, animals and humans. The harmless is not certified by research as long as the related standards are met. Thus, public opinion on this practice is strongly different in different regions depending on historic development and legislation. In Austria, the direct agricultural use of sewage sludge is only around 18% (European Commission, 2010). The application of sludge in agriculture has to be included into the fertiliser management (including market fertiliser and manure) of farmers in order to avoid negative consequences (e.g. ground water pollution). It also is linked to specific monitoring and reporting requirements.

3.4 Recycling methods using digested supernatant

Recycling methods using supernatant from digested sludge dewatering are usually easy to implement. Most of the phosphorus is present as orthophosphate and can easily be precipitated by chemical addition but supernatant only contains a small portion of the total flow of phosphorus within the WWTP. Two main techniques are presented here: Struvia and Ostara.

3.4.1 Struvia

Struvia is a crystallisation process producing struvite or calcium phosphate from sludge liquor.

3.4.1.1 Principle

According to Veolia Water Technologie, the owner and developer of Struvia, the process has three phases: precipitation, liquid/solid separation and struvite harvesting.

The precipitation phase takes place in a tank reactor. The tank is continuously fed with digested sludge and enhanced mixing takes place with the help of a special mixing system called Turbomix^{TM}. An increase of pH and the addition of magnesium salt boost the reaction of struvite precipitation.

Then, in a second phase, an integrated lamella settler ensures the separation of the produced struvite prills and the treated effluent. In municipal applications, the effluent goes back to the inlet of the WWTP.

Finally, the struvite products that have been formed are pumped and directed to a draining and storage facility, before being sent to the packaging unit.

3.4.1.2 Final product

The struvite or the calcium phosphate produced have both a high plant-availability. STRUVIA^{TM} is a certified commercial fertiliser and can be directly used on fields (Egle et al., 2015).

3.4.1.3 Existing plants

In 2013-2014, Véolia set-up a prototype plant in Brussels-North WWTP, in the framework of the P-REX project. The treatment of the centrate generated from digested sludge showed an efficiency of 85% for dissolved phosphorus removal.

In addition to Brussels, Véolia is currently running 3 WWTPs using Struvia technology: in the Urabandai plant, Hakusyu distillery and Kyoto distillery.

3.4.2 Ostara/Pearl

The Pearl® process is a crystallisation process that takes place in a fluidised bed reactor, with sewage sludge liquor as an input product. The end-product is struvite in the form of crystalline pellets or “prills”.

3.4.2.1 Principle

The crystallisation reactor is directly localised after the dewatering unit of the WWTP to treat the sewage sludge liquor. In the reactor, the precipitation of struvite occurs through the addition of magnesium chloride ($MgCl_2$), while the dosing of sodium hydroxide (NaOH) increases the pH to its optimal level. A process of internal recirculation in the crystallisation reactor insures the proper mixing.

As soon as struvite “seeds” begin to form, they grow in diameter as a pearl until they reach the desired size (0,9 mm to 3,0 mm). They are harvested after they sink to the bottom of the reactor. After a phase of drying in a fluidised bed dryer, the final product is ready.

The economic feasibility of the process lies in the range of 75 – 95% PO_4- P removal.

3.4.2.2 Final product

The struvite produced is a certified commercial fertiliser and is sold under the name of Crystal Green®. It has a high plant availability and low heavy metal content compared to industrial fertilisers, except for Cu which is 3 to 4 time the concentration usually found in single super phosphate (Egle, 2015). The product is still in conformity with Austrian law, which do not regulate Cu content in fertilisers.

3.4.2.3 Existing plants

The Canadian firm OSTARA Nutrient Recovery Technologies Inc. developed and commercialises the Pearl Process. Pilot plants have been tested since 2001 and the first industrial scale reactor opened in Edmonton (Canada) in 2007. Pearl® is now being operated at several treatment plants in the United Kingdom and North America.

3.5 Methods using sludge

The methods treating sludge are usually more difficult to implement on WWTP. As they treat the main flow of P, their recovery rate is better but still quite restricted.

3.5.1 AirPrex

The AirPrex process removes phosphorus from the digested sludge liquid through the formation and removal of struvite. The struvite can be directly used as a phosphate fertiliser. At first, P-recovery was only a side effect of the process, which was designed to improve the dewatering of the sludge. Then the recovering properties were discovered (Kabbe, 2016).

3.5.1.1 Principle

The process takes place in a reactor tank placed after the digestion tank. The operator injects magnesium chloride into the tank to form a precipitate called struvite. At the same time, the digested sludge is aerated in order to strip CO_2 resulting in a raise of the pH and to enhance the mixing of sludge and the reactant (magnesium). To collect the formed struvite, the reactor bottom is intermittently tapped. Smaller crystals settle in a second tank.

The process was originally developed by BWB (Berliner Wasserbetriebe) in collaboration with the TU Berlin (Technische Universität Berlin). The aim was to prevent struvite clogging in pipes and other equipment after digestion at some WWTPs using enhanced biological phosphorus removal.

3.5.1.2 Final product

The final product conforms to fertiliser regulations in Germany and is marketed under the brand “Berliner Pflanze”. It has a high plant-availability combined with a low heavy-metal content and a slow release of nutrients, which helps to avoid eutrophication problems (Kabbe, 2016).

3.5.1.3 Existing operating plants

Several WWTPs in Germany and the Netherlands are currently using the AirPrex process, such as Berlin-Wassmannsdorf (Germany), Mönchengladbach (Germany), and Echten WWTP (Netherlands).

3.5.2 Gifhorn process

The Gifhorn process is a sludge leaching process, which includes acidic dissolution and precipitation. The final product is a mix of struvite and hydroxylapatite, with a P-concentration corresponding to 12% P.

The P-recovery performance is 49 % of phosphorus from the sludge input (Kabbe, 2016).

3.5.2.1 Principle

The process consists of three main steps: acid leaching, removal of heavy metals and formation of struvite. In addition to these steps, the process includes stripping for ammonium recovery.

The first phase is a dissolution phase. The extraction of phosphorus from the digested sewage sludge takes place with the addition of sulphuric acid. The process takes place at a low pH, around 4,5.

The second step is the precipitation of dissolved heavy metals, such as sulphides, with the proper dosing of sodium sulphide Na_2S. The addition of NaOH allows the optimal pH of 5,6 to be reached.

The last step is a solid/liquid separation. The pH is again adjusted with NaOH to reach the optimal value of 9. In the decanter, the addition of $Mg(OH)_2$ initiates the precipitation of phosphorus in the form of a mix of struvite and calcium phosphate. The addition of magnesium has to be done below the stoichiometric ratio to provoke the complete precipitation of calcium. This stoichiometric ratio is useful in preventing scaling in the reactor but produces a major fraction of hydroxylapatite in addition to struvite.

3.5.2.2 Final product

The final product in that way a mix between struvite and hydroxylapatite. If struvite has a high-plant availability, hydroxylapatite has the disadvantage of low availability for plants.

The heavy-metal content is low as compared to market fertilisers as single super phosphate or triple super phosphate (Egle et al., 2015).

3.5.2.3 Existing plants

The German Seaborne Environmental Laboratory initially developed the process in 2000 to extract phosphorus from liquid manure at the pilot plant of Owschlag. The plant has a capacity of 10 000 p.e, producing 50 kg struvite per day.

Following the initial development, the laboratory was able to optimise the process through a revision of the heat requirement calculation, as well as by taking economic factors into consideration. Between 2005 and 2006, the process was implemented on a full-scale in the wastewater treatment plant of Gifhorn, Germany.

The Gifhorn WWTP has a capacity of 50 000 p.e. and is able to produce 270 kg struvite/d. However, due to economic and technical reasons, the plant is currently operating under altered conditions and performance is limited (Müller et al., 2005).

3.5.3 Stuttgart process

The Stuttgart process is an acidic leaching reaction from dewatered sludge to deliver struvite (Kabbe, 2016).

3.5.3.1 Principle

The process is based on acidic extraction of P from digested sludge at pH 4 with the addition of H_2SO_4. After solid/liquid separation, dissolved Fe and heavy metals in liquor are masked by citric acid to prevent their transfer into the P product. Struvite precipitation is initiated by the dosing of MgO and the raising the pH to 8. Adjustments to the pH are made with NaOH. Finally, struvite is harvested as a powder by solid/liquid separation and dewatering/drying.

3.5.3.2 Final product

The struvite produced has a high plant availability. Moreover, according to Egle et al. (2015), it has a very high phosphorus concentration, more than 27% P, which is higher than conventional fertilisers.

3.5.3.3 Existing plants

The Stuttgart process for P recovery from digested sludge of chemical P removal WWTPs was developed by the Institute for Sanitary Engineering (ISWA) at the University of Stuttgart. A pilot plant in Offenburg has been in operation since 2011. With a size of 8 000 p.e, it delivers 50 kg struvite per day.

3.6 Methods using ashes

3.6.1 Acid leaching

The principle is to first dissolve the phosphorus contained in the ashes by adding strong acids. After this, a solid/liquid separation recovers the phosphorus. The dephosphorised ash requires proper treatment because of its acidity. In addition, the leach liquor needs to be decontaminated as interfering ions (Fe, Al) and heavy metals dissolve simultaneously with phosphorus (Egle et al., 2015).

3.6.1.1 Ecophos

Ecophos is a company based in Belgium, which has developed several technologies for the phosphorus industry. These technologies include the valorisation of low-grade rocks but also phosphorus recovery from fly ash.

The process is under brevet and as a result the details not fully known, but the principle is to add HCl to the ashes in a digestion reactor (ECOPHOS, 2016). The resultant products are a purified phosphorus acid (H_3PO_4) with 62% concentration, as well as impurities, Al/Fe solution, Mg/CA solution and silicate residue.

The final product has a particularly low concentration of heavy metals (Kabbe, 2016).

A first pilot-plant in Bulgaria was built to treat fly ashes and low-grade rocks. Now, a full-scale plan in Dunkerque is currently in project. After its completion in 2017, it will have a production capacity of 220 000 tons/year of dicalcium phosphate dihydrate, used for animal feed.

In addition, Ecophos has also several references in low-grade rock treatment plant in Syria, Peru and Namibia. The process is comparable to ash treatment.

3.6.1.2 Leachphos

The process consists of a wet acidic chemical leaching process (Kabbe, 2016).

Through the addition of diluted sulphuric acid, the extraction of phosphorus from sewage sludge ash takes place in a reactor tank. The addition of sodium hydroxide or lime adjusts the pH.

During the process, interfering ions (Fe, Al) and heavy metals (Cd, Pb, Zn) are partially dissolved in the leachate. A filtration step separates phosphorus in the form of a mixture of aluminium-, ferric- and calcium phosphate. With the addition of a precipitating agent and at a pH above 9, the precipitation of the remaining heavy metals in the filtrate takes place. The filter cake is then separately disposed. The final product of the Leachphos process is CAP or wet struvite with 15% P of dry matter (Kabbe, 2016). With this technique, the removal rate is about 70%. The process was developed by BSH Umweltservice GmbH. A pilot unit was built in 2012/2013 with a capacity of 2 t ash/h.

The final product has a high phosphorus concentration, comparable to phosphorus rock (Kabbe, 2016). The experiments conducted within the P-REX project also showed very high plant availability in the product if applied on acidic soil and good plant availability on neutral soil. On the other side, regarding heavy metals, the concentration in Cu and Zn are relatively high, respectively 500 and 1 500 g/kg TS (Egle et al., 2015). Even if it is still in compliance with Austrian legislation that does not regulate these metals in fertilisers, this element has to be taken in consideration for further analysis.

3.6.2 Thermal process

3.6.2.1 AshDec (Ash Decontamination)

The AshDec technology is a thermal decontamination process comparable to calcination. The thermal treatment allows for the removal of heavy metals from the final product.

3.6.2.1.1 Principle

To begin with, the ash is pre-heated if necessary, than it is mixed with a reactant chloride donor such as NaCl. Following this, the sludge ash enters a rotary kiln where the phosphate is transformed into $NaCaPO_4$ at a temperature of around 900 – 1 000°C. The temperature is above the boiling point of the heavy metal chlorides, thus separating them. The retention time is at least 20 minutes. Then, the flue gas cleaning process takes place; it requires reactant as $Ca(OH)_2$, $MgCO_3$ and $NaHCO_3$.

3.6.2.1.2 Final product

The final product from the furnace is round pellets or granules that have a size of 2 to 5 mm with a porosity of 20 to 40%. $NaCaPO_4$ is a product that is directly available for plants. The process is supposed to have high removal of heavy metals: 99% of Cd, Hg, and Pb; over 90% of Cu and Zn; and about 50% of Sn and Mb. The final product has still high concentration in As, Cu and Zn (Egle et al., 2015).

As an alternative reactant, $MgCl_2$ can be used. In this process, phosphorus is transformed into calcium-magnesium phosphate. Its availability for plants is limited to acidic soils, but the heavy metal removal process is more efficient.

3.6.2.1.3 Plants in operation

The process has been jointly developed by the Finnish firm Outotec and the German institute BAM (Bundesanstalt für Materialforschung und –prüfung).

A pilot plant in Leoben (Austria) was built in 2008; it is designed to treat about 7 to 10 t/d ash. A full-scale plant is in operation in Zurich, Switzerland (Outotec, 2016). The sludge thermal treatment plant of Werdhölzli treats all of the sewage sludge produced in the Zürich region, which amount to 100 000 tons per year. It generates electrical power and heat. The project is to add an AshDec process.

3.6.2.2 Mephrec® (Metallurgisches Phosphor Recycling)

The Mephrec process is a reducing shaft melting gasification that uses sewage sludge and/or ash as an input material (Kabbe, 2016). The main output is slag enriched with P and depleted in heavy metals. The by-products of the process are iron alloy with P-content and, with sludge as an input, raw gas with a high calorific value.
The German company Ingitec is the developer of the process.

3.6.2.2.1 Principle

To begin with, the dewatered sewage sludge (>25 % dry matter) is dried to 80% and is pressed into briquettes. If ash is used, it is also directly pressed into briquettes. Following this process, the briquettes are thermally treated through gasification at temperatures above 1 450 °C in a shaft furnace. Under these high temperatures, a reduction of the heavy metals occurs. The most volatile ones (Cd, Pb, Zn, Hg) evaporate and the non-volatile ones (Fe, Cu, Cr, Ni) go into a liquid phase. Moreover, the phosphorus present turns into silico-phosphate ground basic slag (Kabbe, 2016). The raw gas produced can be directly injected into a municipal waste incineration plant or, after refinement, into a combined heat and power (CHP) plant.

3.6.2.2.2 Final product

With sewage sludge, the final product contains from 2 to 5% P with over 90% citric acid solubility. The possibility to produce electricity and heat with the gas that is formed is an additional benefit. The final product contains high levels of Ca and Cu (Egle, 2015). With sewage sludge ash, the P content can reach 10%, but energy recovery is not possible.

3.6.2.2.3 Existing plants

A pilot plant in Nuremberg has run with a capacity of 8 t/h briquettes since 2015.

3.7 Conclusion

The techniques using sludge liquor are usually simple and economical, giving a very good final product in terms of quality and plant availability. On the other side, as sludge liquor does not represent the main flow of phosphorus is a WWTP, these techniques have a low recycling potential (Egle et al., 2015).

The processes using sewage sludge as a raw material are usually more complicated, more costly, but they have a better recycling rate (Egle et al., 2015).

The process using ashes have the advantage of permitting a high removal rate of heavy metals. Moreover, they are usually economic. For some techniques (AshDec, MEPHREC), a decision has to be made between a high P recovery or a low heavy metal concentration (Egle et al., 2015), but some processes as ECOPHOS can do both. On the other hand, all the processes using ashes require a mono-incineration, which is actually not the most commonly used technique in the EU.

4 Linz wastewater treatment plant

The Linz wastewater treatment plant treats the wastewater from the whole Linz urban area, as well as from 39 surrounding municipalities. This corresponds to a catchment area of about 900 km^2. The plant also receives a large portion of industrial wastewaters, coming from heavy industry, such as the metallurgical complex, Voestalpine, or the DSM chemical plant. These industrial wastewaters represent 40% of the COD charge (Linz AG Abwasser 2016).

4.1 General description of the process

The design capacity of the treatment plant is 950 000 p.e (population equivalent). This treats about 30% of industrial wastewaters in volume. The design peak flow is about 8,8 m^3/s, and the maximum dry weather flow is 2,7 m^3/s. The treatment plant is a conventional mechanical-biological plant designed for carbon and nutrient removal. The primary and secondary sludge is thickened and digested by an anaerobic sludge digestion (Linz AG Abwasser 2016).

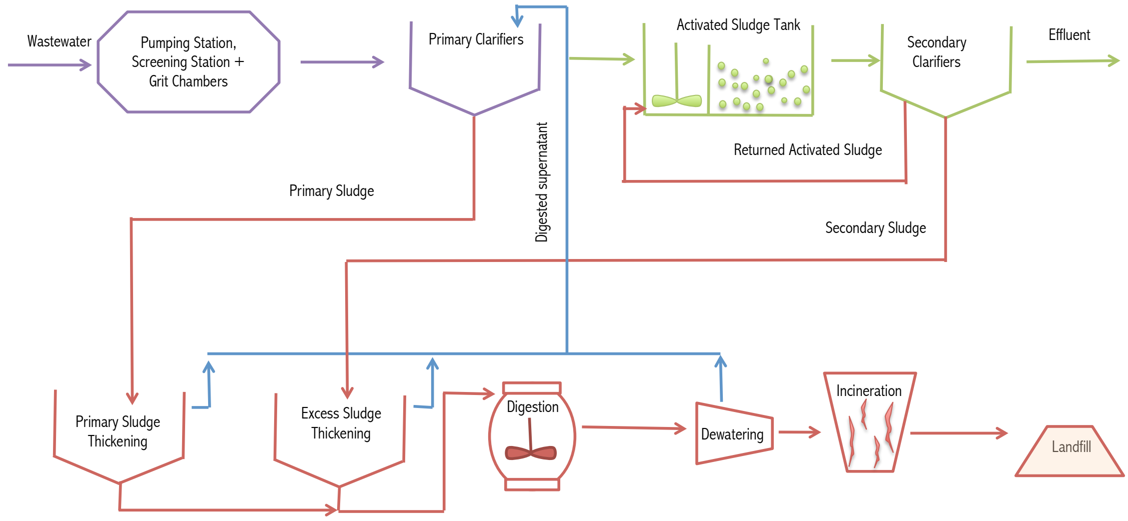

Figure 4: WWTP scheme

The first process is the mechanical treatment. After the inlet pumping station, the wastewater enters the pre-treatment area, which consists of a screening station and grit chambers. After that, the pre-treated sewage flows into two primary settling tanks, with a capacity of 6 900 m^3 each, where primary treatment takes place.

The water then enters the biological treatment stage, which is an activated sludge process. The biological treatment of the Linz-WWTP consists of four preliminary activated sludge tanks, which have a capacity 11 000 m^3 each, and four aeration tanks, with capacities of 12 000 m^3 each. The eight secondary clarifiers have a capacity of 8 750 m^3 each. Due to bacteria growth, there is a daily excess sludge production of about 4 500 m^3/d. After this treatment, treated water is discharged to the Danube.

The sludge enters the sludge treatment stages. This process begins with thickening. The primary sludge is subjected to thickening by two pre-thickeners, with volumes of 1 500 m^3 each. The secondary sludge is thickened in 4-belt thickeners. After thickening, the anaerobic digestion of the mixed sludge takes place in three digesters, each of which have a capacity of 10 400 m^3 and retention time of about 30 days. The biogas formed is used in the biogas plant to produce electrical energy and heat.

Following this, the digested sludge, with a dry solids content of about 3,6 %, enters the mechanical dewatering process using centrifuges. With the addition of polymers the solid content of the waste sludge is increased to 23 %. The sludge is transported to the Linz waste-to-energy plant, where it is co-incinerated with solid waste.

4.2 Data analysis

This thesis uses the daily operation data of the WWTP for a complete year, from October 2013 to September 2014. Mean values with an uncertainty of a 95% confidence were used. The volume balance analysis is complex as the model contains wastewater as well as sludge with differing water contents. Thus, this thesis concentrates on the phosphorus balance. The phosphorus content in digested supernatant is given in terms of concentration. As a result, it is possible to compute the data using the flow. Some data was not available, namely, the phosphorus concentration in the supernatant of the belt thickener. It was assumed to be negligible. This assumption is considered realistic since most of the phosphorus is bound at this stage of the process and there is almost no phosphorus recycled to the primary settling tank.

The main data about phosphorous are given in the table 2 below:

Table 2: P loads and concentration at WWTP-Linz

	Total P	PO_4-P
Influent WWTP	941 kg/d	342 kg/d
After primary settling tanks	793 kg/d	368 kg/d
Effluent WWTP	65 kg/d	20 kg/d
Sludge treatment liquid	78 mg/l	/
Digested supernatant	231 mg/l	146 mg/l
Digested sludge		165 mg/l

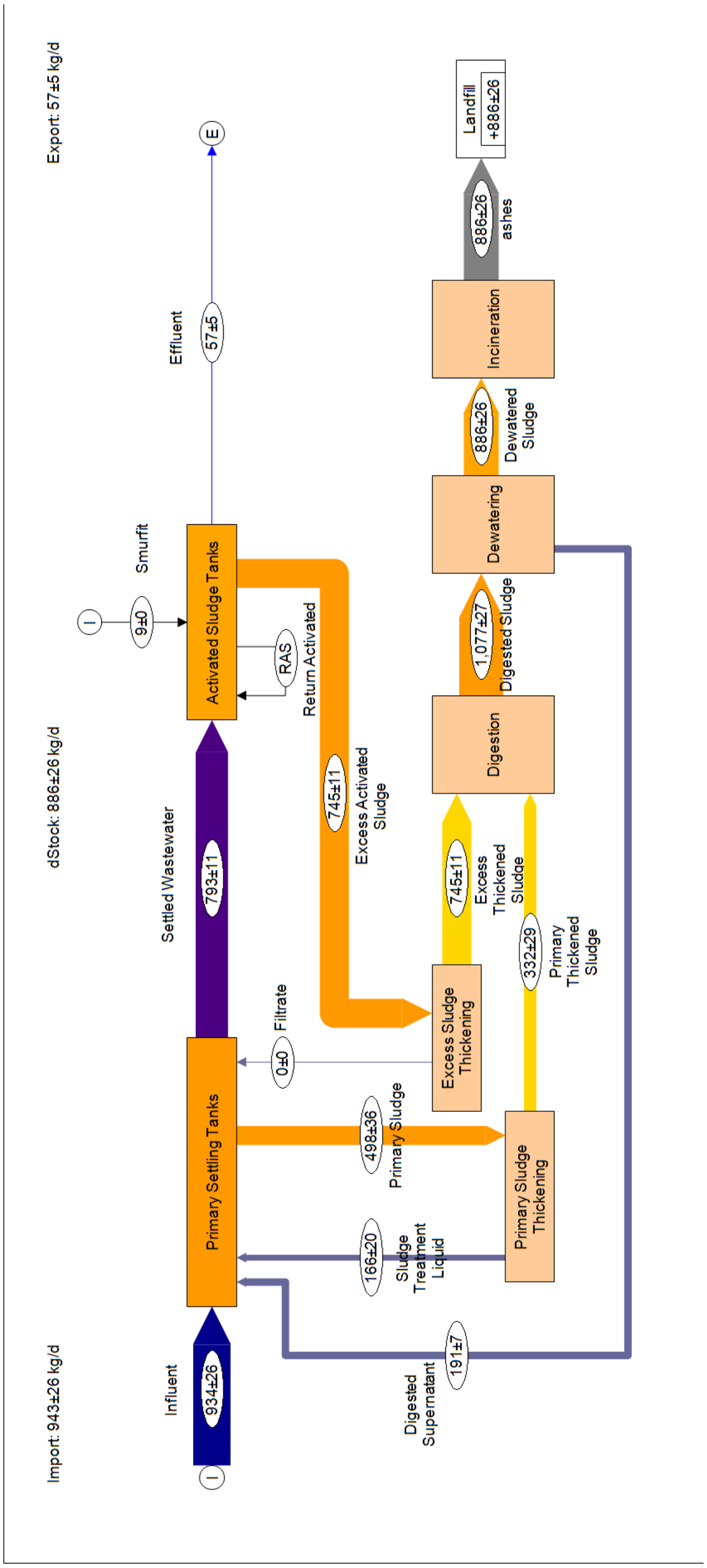

Figure 5: STAN-model (actuel)

The STAN model built on this basis (Figure 7 above) shows that 941 kg/d of phosphorus enter the wastewater treatment plant, and 934 kg/d go through the treatment process, whilst 7 kg/d go directly to the outlet with the rain overflow. 74 kg/d are found in the effluent and 886 kg/d are contained in the ashes.

The analysis of the actual operation of the Linz-WWTP shows two important facts. First of all, the P-content in the influent and the sludge is quite low compared to other WWTPs, for example the Vienna WWTP, which has an actual loading of about 3 million p.e and a daily total P-load of 5,7 t. This is due to the fact that the Linz-WWTP receives a large proportion of industrial waters. These waters come from industries that manufacture paper or steel, which do not use phosphorus and produce less nutrients compared to municipal wastewater or wastewater that comes from the food industry. Secondly, the dissolved phosphorus content after digestion (165 mg/l) is relatively high due to the anaerobic digestion process.

4.3 Application of P-recycling techniques

This chapter makes a cost benefit analysis of all the methods, when adapted to the Linz-WWTP.

Assumptions were made about the different costs, and these results may change if there is a change in the prices. The main costs are taken from the report “Endbericht Phosphorrückgewinnung aus dem Abwasser” (Egle, 2014). All the quantities of reactants and the energy needed are from the P-REX fact sheets, as well as the recovery efficiency. We set up the price of struvite at 400 €/t, which may seem a bit high but is realistic as the trend is for a growth in price. The gain in sludge disposal is 52 €/t (PCS, 2014).

Among the techniques presented in the previous chapter, Stuttgart and Leachphos were not studied because the investment costs were not available. As the quality of the final product is disputable, Leachphos is anyways not the best solution a priori. Three main types of techniques are tested here: P-recovery on supernatant, on sludge, and on ashes.

4.3.1 Methods using digested supernatant

4.3.1.1 Struvia

In Linz WWTP, the digested supernatant contains around 63% of dissolved phosphorus. According to Veolia, the rate of soluble-P removal at pH =7,4 is 80 % (Veolia, 2014). As a consequence, the phase of pH adjustment with NaOH can be avoided, in order to save costs. The harvesting rate is around 90 % (Véolia, 2014). The Stan model built on this basis gives an amount of recovered phosphorus of 87 kg/d or 31 tons/year and a struvite production of 245 t/y. The reactor STRUVIA 3200 is the most adapted to the project, with an investment price of around 700 000 € (Véolia, 2016).

Table 3: Cost/Benefit analysis of the implementation of the Struvia process to WWTP-Linz

STRUVIA			
Investment Costs	**Quantity**	**United Price**	**Total price**
Senior loan: 70 %, 4 % interest rate, 15 years			*700 000 M€*
Annual Costs			
Capital Costs	0,7 x 0,0899 x $0,7.10^6$		44 051 €/y
Maintenance + repair costs	2,5% of investment costs		17 500 €/y
Personal costs	0,15 employee	50 000 €/empl.	7 500 €/y
MgCl	3,2 x 31 tons/year	240 €/t	23 808 €/y
Electricity demand	1,3 kWh/kg x 31 t/y	10 c / kWh	4 030 €/y
		Total annual costs	**96 889 €/y**
Revenues			
Struvite Selling	245 t/y	400 €/t	98 000 €/y
Clogging avoided			22 000 €/y
		Total benefits	**120 000 €/y**
Balance			**+ 23 111 €/y**

Even if the digested supernatant is not the location where the greatest quantity of phosphorus is available, the Struvia process is economically feasible because of its low investment cost.

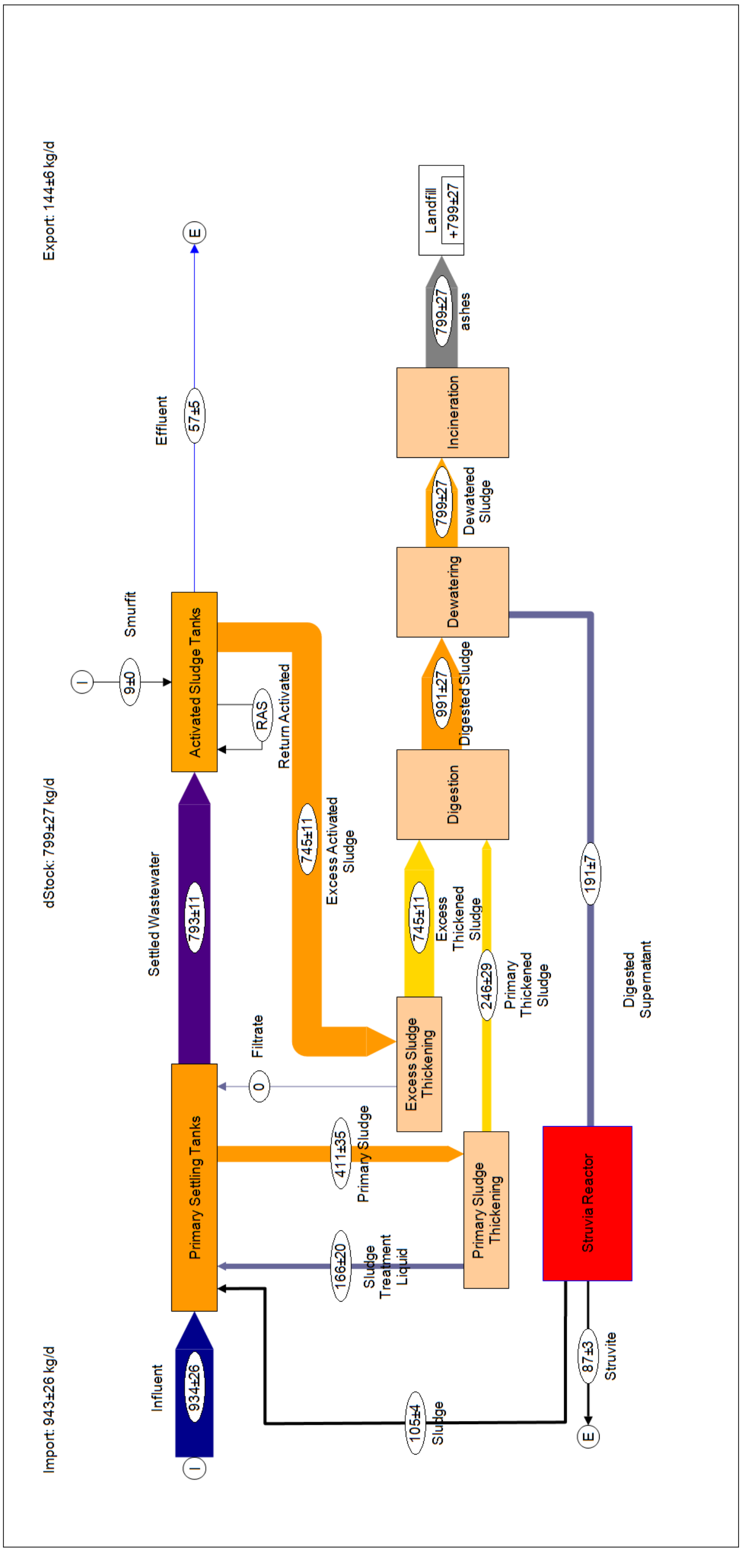

Figure 6: STAN-model for Struvia process

4.3.1.2 Ostara/Pearl

The optimal P-concentration is above 60 mg/l PO_4-P. On this basis, the process typically removes 85 % of dissolved phosphorus (Ostara 2010). The initial STAN model was modified to account for this. It gives a quantity of 102 kg/d of P recovered, which represents 37 tons of phosphorus annually.

Electricity is currently used for drying the pellets; however, heat from the nearby waste-to-energy plant could be used instead. The capital costs are estimated to be around 3 M€ (Nieminen 2010).

Table 4: Cost/Benefit analysis of the implementation of the Ostara/Pearl process to WWTP-Linz

OSTARA			
Investment Costs	**Quantity**	**United Price**	**Total price**
Senior loan: 70 %, 4 % interest rate, 15 years			3 000 000 €
Annual Costs			
Capital Costs	0,7 x 0,0899 x 3.10^6		188 790 €/y
Maintenance + repair	2,5 % of investment costs		75 000 €/y
Personal costs	0,15 employee	50 000 €/empl.	7 500 €/y
Reactant: MgCl	3,2 x 37 tons/year	240 €/t	28 416 €/y
Electricity demand	1,3 kWh/kg x 37 t/y	10 c / kWh	4 810 €/y
		Total annual costs	**304 516 €/y**
Annual Revenues			
Struvite Selling	290 t/y	400 €/t	116 000 €/y
Clogging avoided			22 000 €/y
		Total annual revenues	**138 000 €/y**
Balance			**- 166 516 €/y**

The process is not economically feasible, largely because of a high investment cost.

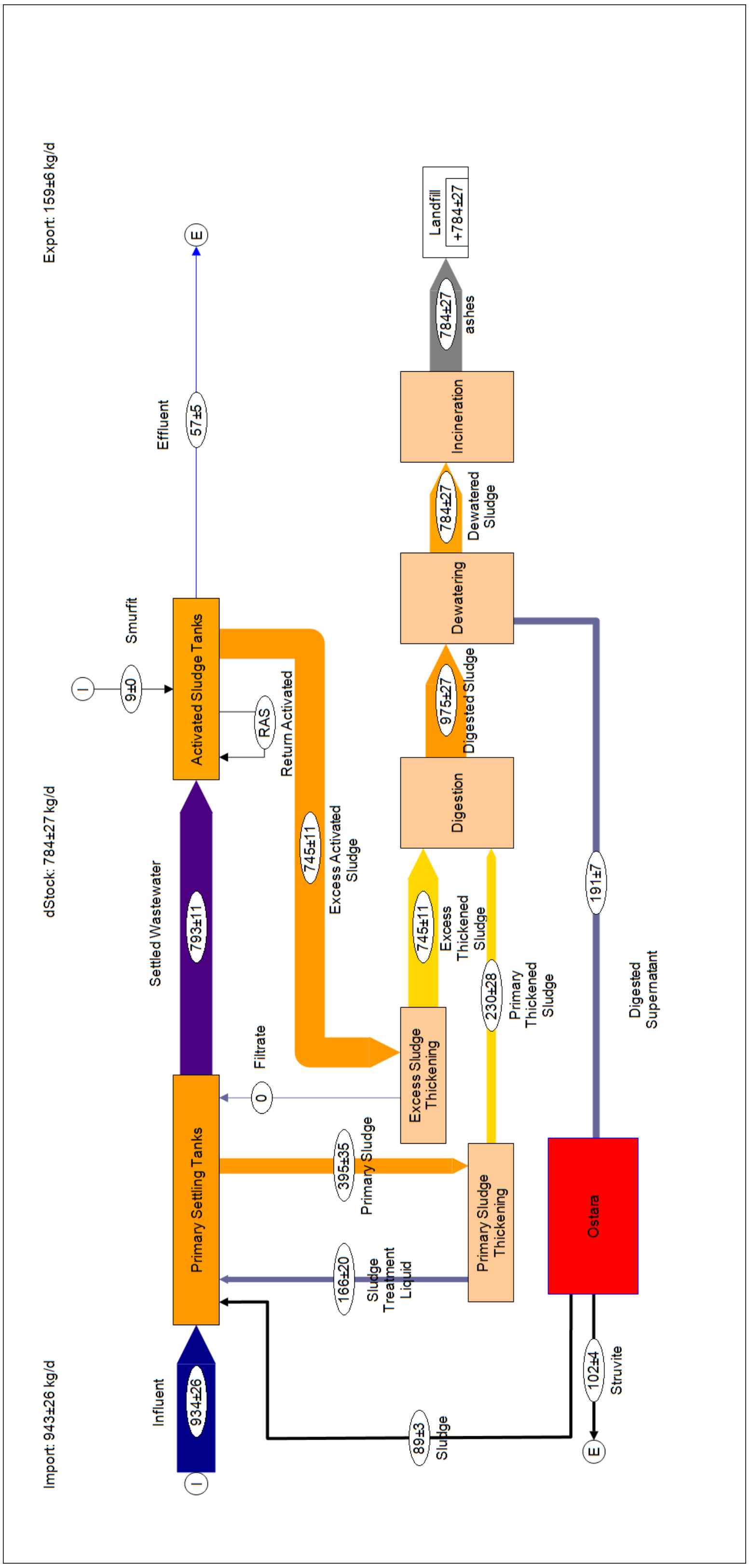

Figure 7: STAN-model for Ostara process

4.3.2 P-recovery on digested sludge

4.3.2.1 *AirPrex*

We modified the STAN model by adding an AirPrex reactor after the sludge digester. An efficiency of 85% of dissolved phosphorus removal was assumed. As a result, the amount of phosphorus recovered in struvite is 135 P kd/d, (= 49 t P/y).

The investment costs are 2 millions euros for a 1 million p.e process (TU Wien, Lebensministerium, 2015). The following calculations are based on 49 t/y P production, which corresponds to 205 t of struvite.

Table 5: Cost/Benefit analysis of the implementation of the AirPrex process to WWTP-Linz

AIRPREX PROCESS			
Investment Costs	**Quantity**	**United Price**	**Total price**
Senior loan: 70 %, 4 % interest rate, 15 years			*2 000 000 M€*
Annual Costs			
Capital Costs	0,7 x 0,0899 x 2.10^6		125 860 €/y
Maintenance + repair	2,5 % of investment costs		50 000 €/y
Personal costs	0,15 employee	50 000 €/empl.	7 500 €/y
$MgCl_2$	14,5 x 49 t/y	240 €/t	170 520 €/y
Electricity demand	10 kWh/kg x 49 t/y	10 c / kWh	49 000 €/y
		Total annual costs	**402 880 €/y**
Benefits			
Struvite selling	387 t/y	400 €/t	154 800 €/y
Clogging avoided			22 000 €/y
Polymer saved	25 % x 115,30 t/y	2 480 €/t	71 500 €/y
Gain in sludge disposal	1 600 t/y	52 €/t	83 200 €/y
		Total benefits	**331 500 €/y**
Balance			**- 71 380 €/y**

The process is not economically feasible, even if in complement to struvite selling, it has side advantages as avoiding the clogging, saving polymers and drying the sludge.

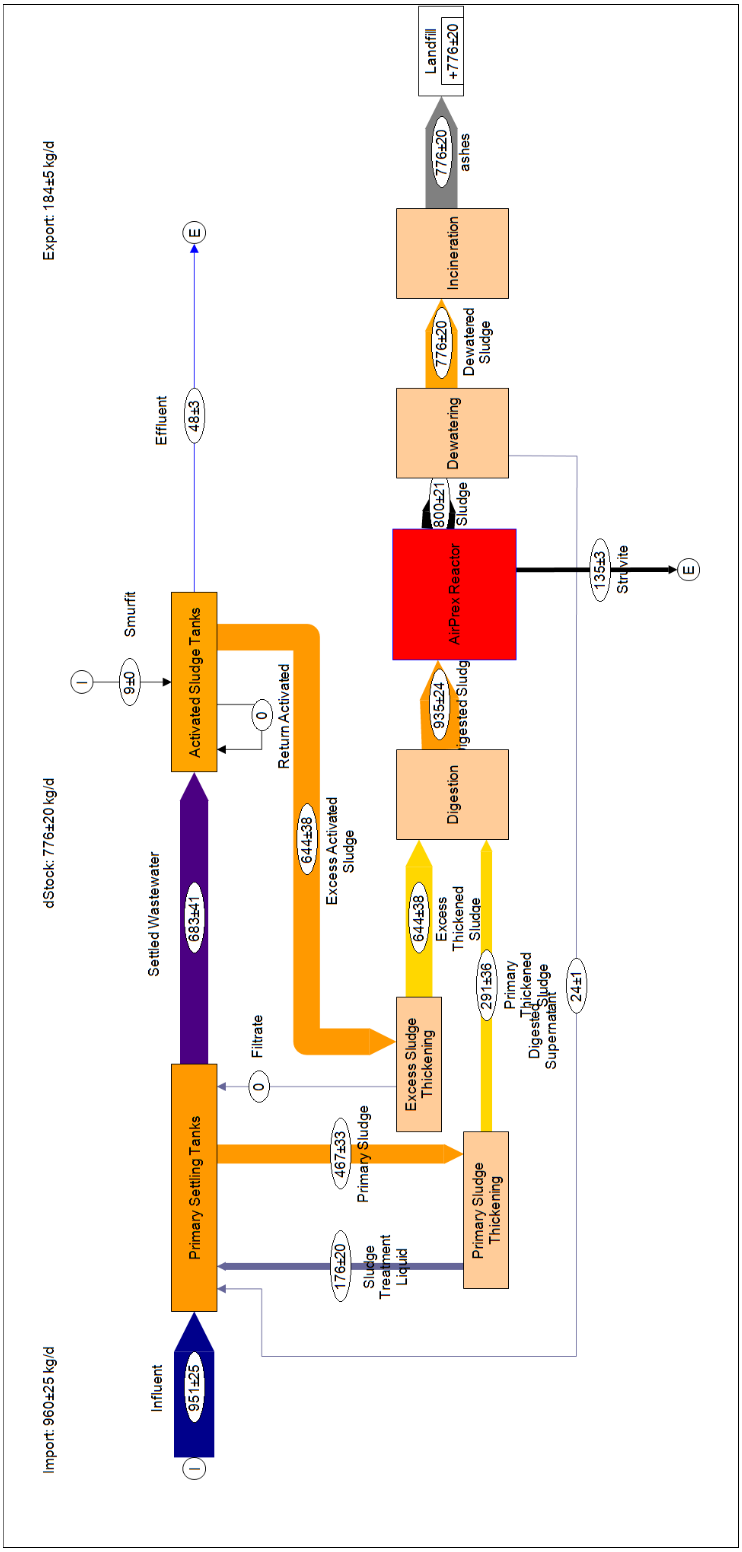

Figure 8: STAN-model for Air-Prex process

4.3.2.2 Seaborne/Gifhorn

As with the other P-recovery techniques using sludge, Seaborne only recovers dissolved phosphorus. According to our WWTP data, at pH=7,4, the digested sludge contains 17 % dissolved phosphorus. At pH=3, the ratio of dissolved phosphorus raises to about 60 %. With the Seaborne process, the rate of recovery is 95%.

The modified STAN model gives a production of phosphorus of 527 kg/d (190 t/y), which corresponds to 1 520 tons of struvite per year.

The investments costs for the process on a 50 000 p.e WWTP is around 430 k€ and the price for a 1 M p.e will be multiplied by 5 (TU Wien, Lebensministerium, 2015).

Table 6: Cost/Benefit analysis of the implementation of the Seaborne process to WWTP-Linz

SEABORNE PROCESS			
Investment Costs	**Quantity**	**United Price**	**Total price**
Senior loan: 70 %, 4 % interest rate, 15 years			2 150 000 M€
Annual Costs			
Capital Costs	0,7 x 0,0899 x 2,15.10^6		135 300 €/y
Maintenance + repair	2,5 % of investment costs		53 750 €/y
Personal costs	0,15 employee	50 000 €/empl.	7 500 €/y
H_2SO_4	8,2 x 190 t/y	150 €/t	230 700 €/y
NaOH	2,9 x 190 t/y	330 €/t	181 830 €/y
$Mg(OH)_2$	0,2 x 190 t/y	230 €/t	8 740 €/y
Na_2S	0,8 x 190 t/y	680 €/t	103 360 €/y
Electricity demand	6,9kWh/kg x 190.10^3 kg/y	10 c / kWh	131 100 €/y
		Total annual costs	**852 280 €/y**
Benefits			
Struvite selling	1 520 t/y	400 €/t	608 000 €/y
Clogging avoided			22 000 €/y
Polymer saved	30 % x 115,30 t/y	2 480 €/t	85 800 €/y
Gain in sludge disposal	1 600 t/y	52 €/t	83 200 €/y
		Total annual benefits	**799 000 €/y**
Balance			**- 53 280 €/y**

The process is economically negative.

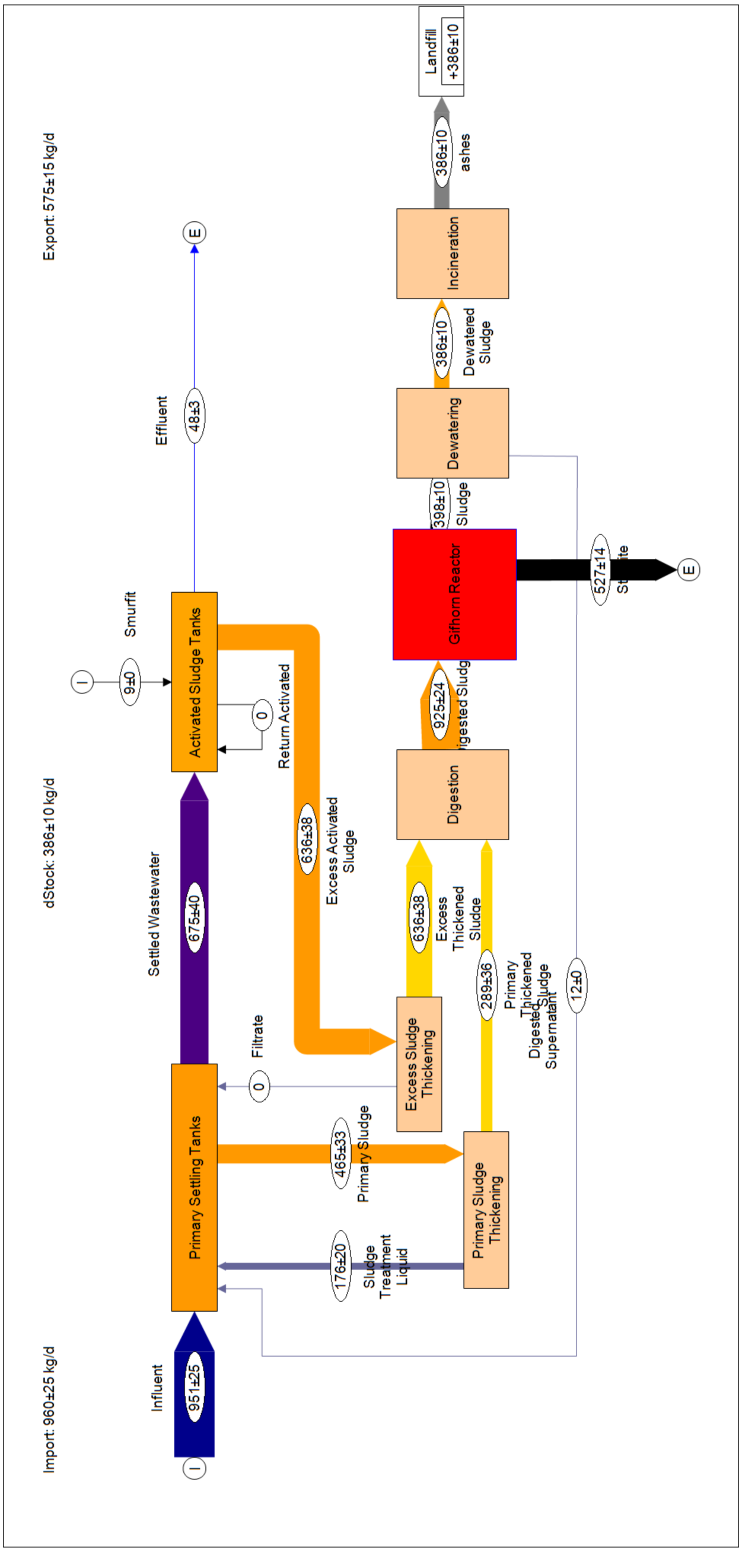

Figure 9: STAN-model for Gifhorn process

4.3.3 P-recovery on ashes

Today, the sewage sludge of the WWTP goes into the Linz waste-to-energy plant where it is co-incinerated with municipal waste. Therefore, it will be necessary to build a new incineration plant in order to be able to treat sewage ashes separately. The cost of a mono-incineration plant is not included in the cost analysis.

The Linz WWTP produces 981 m^3/d of digested sludge with a total solid content of 3,6 % and an ignition loss of 59,6%. After a mono-incineration, it would give a production of 5 178 t/ashes a year. These ashes would contain 886 t of phosphorus, from which a part can be recovered, depending on the techniques used.

It was not necessary to build a STAN model for P-recovery in ashes.

4.3.3.1 Ecophos

The Ecophos process as a P-recovery rate of 97 % and can recover 860 t of phosphorus per year after a sludge mono-incineration. According to the company, the approximate investment cost for a system treating 5 000 t of ashes/year would be between 5 and 7 million euros.

The analysis of the annual costs and benefits for Ecophos shows that the process is able to make a profit:

Table 7: Cost/Benefit analysis of the implementation of the Ecophos process to WWTP-Linz

ECOPHOS			
Investment Costs	**Quantity**	**United Price**	**Total price**
Senior loan: 70 %, 4 % interest rate, 15 years			6 000 000 €
Annual Costs			
Capital Costs	0,7 x 0,0899 x 6.10^6		378 000 €/y
Maintenance + repair	0,5 % of investment costs		30 000 €/y
Personal costs	1 employee	50 000 €/empl.	50 000 €/y
Reactant H_2SO_4	0,5 x 3 800 t/year	150 €/t	285 000 €/y
Electricity demand	6 kWh/t x 3 800 t/y	10 c / kWh	2 280 €/y
		Total annual costs	**745 280 €/y**
Annual Revenues			
Selling DCP	3 800 t/y x 62%	850 €/t	2 002 600 €/y
		Total annual benefits	**2 002 600 €/y**
Balance			**+ 1 257 320 €/y**

4.3.3.2 Mephrec

The Mephrec process can recover 81 % of the phosphorus in ashes, which represents 718 t/year of recovered phosphorus for Linz WWTP. The investment costs on a

Mephrec plant treating 5 000 t/y is around 6 M€ (TU Wien, Lebensministerium 2015). The annual costs and benefits are presented below:

Table 8: Cost/Benefit analysis of the implementation of the Mephrec process to WWTP-Linz

MEPHREC				
Investment Costs		**Quantity**	**United Price**	**Total price**
Senior loan: 70 %, 4 % interest rate, 15 years				6 000 000 M€
Annual Costs				
Capital Costs		0,7 x 0,0899 x 6.10^6		377 580 €/y
Maintenance + repair		0,5 % of investment costs		30 000 €/y
Personal costs		1 employee	50 000 €/empl.	50 000 €/y
Reactant	coke	2,5 x 718 t / y	360 €/t	646 200 €/y
	O_2	0,75 x 718 t / y	80 €/t	43 080 €/y
	dolomite	1,3 x 718 t / y	100 €/t	93 340 €/y
	$Ca(OH)_2$	0,1 x 718 t / y	140 €/t	10 050 €/y
Electricity demand		1,2 kWh/kg x 718.10^3 kg/y	10 c / kWh	86 160 €/y
			Total annual costs	**1 366 410 €/y**
Annual Benefits				
Slag selling with 10 % P		20 000 t/y	100 €/t	**2 000 000 €/y**
Balance				**+ 633 590 €/y**

4.3.3.3 AshDec

The AshDec process can recover 98 % of the phosphorus in ashes, which represents 870 t/year of recovered phosphorus per year for Linz WWTP.

The investment costs on a full-scale AshDec plant, treating 30 000 t/y, is estimated to be 20 M€ (TU Wien, Lebensministerium 2015). With its capacity, the AshDec treatment in Linz will have an investment price around 5 M€. The annual costs and benefits are listed here:

Table 9: Cost/Benefit analysis of the implementation of the AshDec process to WWTP-Linz

ASHDEC				
Investment Costs		**Quantity**	**United Price**	**Total price**
Senior loan: 70 %, 4 % interest rate, 15 years				5 000 000 M€
Annual Costs				
Capital Costs		$0,7 \times 0,0899 \times 5.10^6$		314 650 €/y
Maintenance + repair		0,5 % of investment costs		25 000 €/y
Personal costs		1 employee	50 000 €/empl.	50 000 €/y
Reactant	Na_2SO_4	3,3 x 870 t / y	90 €/t	258 390 €/y
	$Ca(OH)_2$	0,1 x 870 t / y	140 €/t	12 180 €/y
	NaOH	0,1 x 870 t / y	330 €/t	28 710 €/y
Electricity demand		0,85 kWh/kg x $870\ 10^3$ kg/y	10 c / kWh	73 950 €/y
Natural gas demand		5,2 kWh x $870\ 10^3$ kg/y	4 c / kWh	180 960 €/y
			Total annual costs	**943 840 €/y**
Annual Benefits				
$NaCaPO_4$ selling		4 400 t/y	270 €/t	**1 188 000 €/y**
Balance				**+ 244 160 €/y**

4.3.4 Intermediate conclusion

Among the methods using digested supernatant, Struvia is the most interesting from an economic point of view, even if the phosphorus recovery potential is low (10%).

All three methods using ashes gave very interesting economic results, too. The phosphorus recovery is between ~80 and ~98% on total P in ashes.

For the methods using sludge, AirPrex gives similar economic results as Seaborne, but Seaborne as a better P-recovery (57%) than AirPrex (15%) on phosphorus in sludge input.

In addition to recovery potential and economic factors, other important criteria have to be considered. First, the quality of the final product: the product has to be low in heavy metals and should have high plant availability. Moreover, the process has to be proven at an industrial scale or, at least, at an online pilot scale. The size (in terms of capacity) of the plant is of great importance as the professional qualifications of the operator team are increasing with the size of the WWTP. Finally, the process for implementation has to adapt to the human resources. This is easy for the processes using supernatant but has to be proved for the three ashes processes as they are linked to applying mono-incineration In the following analysis, the economic criteria are weighted with 2, while all other criteria with 1.

Table 10: Multicriteria analysis of the implementation of various P-recovery process to WWTP-Linz

Process	P-recovery in sludge input	Economy		Final product	Proven process	Imple-mentation	TOTAL
Ostara	15 %	-	-	+	++	++	+++
Struvia	10 %	+	+	+	+	++	+++++
AirPrex	15 %	-	-	+	+	+	++
Seaborne	57 %	0	0	-	-	+	-
Mephrec	81 %	++	++	0	-	-	++
Ecophos	97 %	+++	+++	+	0	0	+++++++
AshDec	98 %	++	++	0	+	-	++++

With the hypotheses used, the ash leaching method Ecophos is the most interesting method in the case of the Linz WWTP. It is economically feasible and gives a high quality final product. It requires sludge mono-incineration, which could be designed for sewage sludge alone or as co-incineration with animal and bone meal, having a high phosphorus content and a high calorific value.

Struvia process, with P precipitation from sludge liquor is easier to implement and operate, produces also a high quality fertiliser and could result in a net benefit of around 23 k€ per year (which represents ~5 ct/pe/a or ~ 0,5% of the total yearly operating costs of the WWTP Linz). It represents a good alternative if the option of a mono-incineration plant is not to be considered. Struvia can also be used on digested sludge. This option merits to be further investigated, as P-recycling from digested sludge could result in relevant savings in sludge treatment and disposal.

These results cannot be easily transferred to other case studies and are markedly dependent on a number of assumptions. The analysis shows that P-recycling is already economically feasible with a high struvite price of 400 €/t (3,3 €/kg P). This price is high compared to the actual price (2016) but reflects the trend of the P-market.

5 Conclusion

The recycling of phosphorous, this essential resource for all living organisms has become more and more a concern in Europe. The continent is wholly dependent on imports, and as the trend for the market price is upwards, the economic incentives are growing. In addition, the legal incentives are also increasing. European water legislation used to consider phosphorus primarily as pollutant causing eutrophication and favoured the removal of phosphorus from the wastewater. European waste legislation is already in favour of a transition from a linear to a circular economy. The recently started European project P-REX aims to harmonise technical solutions for P-recycling in WWTPs and to prove their feasibility at full-scale. Recycling of phosphorus in Europe and especially in Austria however is not yet a common practice.

This thesis tested several solutions for P-recovery from wastewater at the wastewater treatment plant of Linz, Austria. The majority of the solutions are or might become economically and technically feasible. The solutions were ranked through a multi-criteria analysis. The ash leaching solution Ecophos seems to be the most promising for Linz WWTP, with a high P-recovery of 98 %, followed by the liquor precipitation process Struvia with a much lower P-recovery of 10%, but which could be improved in implementing it on the sludge.

These conclusions are based on sound data from the WWTP Linz-Asten and results from scientific investigations published in literature. The assessment of the different processes uses a number of hypotheses that influence the results. If the prices of P-market fertilisers continue to rise and the costs for the engineering solutions decrease, it can be concluded that the implementation of P-recycling techniques not only in Austria will become increasingly interesting even from the economical point of view.

The most promising technique for the Linz, WWTP (1 Mio p.e), would require a change in sewage sludge treatment and disposal techniques. The actual co-incineration plant for solid waste and dewatered sewage sludge would have to be adapted by the construction of a new mono-incineration plant for the sewage sludge. At the Vienna main treatment plant (4 Mio p.e), mono-incineration of sludge is already in place and would favour P-recovery. Even more favourable would be co-incineration of sewage sludge with animal meal or other P-rich organic substances with low levels of contaminants. This solution would markedly enhance the national P-recycling potential and would profit from the economy of scale.

The implementation of P-recycling in Austria is not only linked to the solution of the technical and economic problems. Also the organisational aspects and all responsibilities will have to be elaborated. The wastewater service sector is dominated by public utilities. Some plants are operated on the basis of a public private partnership. In both cases, the main aim is the public interest in water protection by wastewater treatment. Waste legislation could serve as the starting point for a P-recycling policy with an adequate legal framework as this legislation has already a strong market economy and public private partnership background. Political commitment and an adapted legal framework are the keys to success.

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