Doctoral Thesis

Phosphorus recovery from municipal wastewater

The Austrian phosphorus budget and an integrated and comparative assessment of technologies to recover phosphorus from municipal wastewater

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Dissertation

Phosphorrückgewinnung aus kommunalem Abwasser

Die Österreichische Phosphorbilanz und die ganzheitliche und vergleichende Bewertung von Technologien zur Rückgewinnung von Phosphor aus dem kommunalen Abwasser

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Abstract

Phosphorus (P) is a finite and non-substitutable resource, essential to sustain high levels of agricultural productivity. However, too high concentrations of P in ecosystems can cause eutrophication, an important environmental issue. Europe does not have noteworthy raw phosphate deposits and is therefore dependent on imports, the geopolitical situation, and raw material markets.

The first aim of this Thesis was the development of a detailed and complete phosphorus budget for Austria based on the methodology of a material flow analysis (MFA). As a first result of an Austrian P budget assessment, the dependence on mineral P fertilizers application (2 kg P $cap^{-1}yr^{-1}$) was confirmed. In contrast to that, further analysis highlighted considerable, but often unexploited P-loads in municipal wastewater (~1 kg P $cap^{-1}yr^{-1}$) and animal by-products such as meat and bone meal (~0.5–0.6 kg P $cap^{-1}yr^{-1}$).

Numerous recycling technologies have been developed and partially implemented over the past years to recover P from different sources of wastewater treatment plants (secondary treated effluent, digester supernatant, and (digested) sewage sludge) as well as from sewage sludge ashes. This work describes a methodology for a comparative technical, environmental and economic assessment of P-recycling and shows its applicability for 19 selected technologies. Out of this application, useful information for a possible future implementation can be derived for decision making.

The recovery technologies have not only been assessed as stand-alone processes, but also with regard to the impact on the total process chain – from the wastewater treatment plant influent to the final disposal of all occurring liquid and solid wastes. With this methodological approach interplays on the whole process chain in respect to technological requirements, environmental impacts and costs are included into the assessment.

The results show that there is not one final indicator based on which various technologies can be sufficiently compared and the best solution identified. The overall performance of a recycling technology under specific circumstances is described by the analysis of numerous assessment criteria. This work reveals for example, that technologies to recover P from digester supernatant are ready for application from a technical point of view. Consequently, there are already many cases where these technologies have been implemented on a full-scale, and in some cases, they even pay back economically. Simultaneously a clean and very good plant-available P rich material is produced.

However, to achieve the greatest recovery of P from wastewater, sewage sludge ashes should be addressed. To enable P recovery from ashes, the co-incineration of combustibles low in P, as well as high ash and high heavy metal contents should be avoided. In this context, the necessary structures such as the mono-incineration of sewage sludge have to be increased in Austria as prerequisite of implementation of these technologies. Further advantages of a mono-incineration strategy include the potential of the combined usage of other P rich flows (e.g., meat and bone meal) and the possible temporal storage of the occurring ashes for a future P recovery (creation of an Austrian P-mine).

On a national scale, the recovery of P from sewage sludge ash (SSA) will hardly pay back economically with costs being dependent on the required removal of pollutants and/or the quality of the recovered material or product. There is a wide range of available technologies for the production of P-fertilizers: Some fertilizers have very low heavy metal contents, a good P availability at relatively high costs, and moderate recycling rates. In contrast to that, a complete recycling of P in SSA would have moderate to no extra costs, compared to a reference system with ashes being put into landfills. However, this approach would result in a low P availability and would not include heavy metal depollution. Therefore, which technologies will finally be applied to recover P from SSA will depend on (legal) requirements on product quality and economic incentives from national authorities to support their implementation.

Kurzfassung

Phosphor (P) ist eine endliche und nicht substituierbare Ressource, essentiell für die hohen Erträge in der Landwirtschaft. Emissionen von Phosphor in die Gewässer können jedoch zu einer Überversorgung und erheblichen Gewässergüteproblemen durch Eutrophierung führen. Da Europa über keine nennenswerten P Rohphosphatlagerstätten verfügt, ist es folglich auf Importe angewiesen und abhängig von geopolitischen Entwicklungen sowie den aktuellen Rohstoffmärkten. Nationale P Bilanzen bestätigen auf der einen Seite die Abhängigkeit von mineralischen Düngern auf Rohphosphatbasis (~2 kg P pro Einwohner und Jahr), verdeutlichen jedoch gleichzeitig, dass kommunales Abwasser (~1 kg P $E^{-1}a^{-1}$) sowie weitere P haltige Abfälle, wie z.B. Tiermehle (~0.5–0.6 kg P $E^{-1}a^{-1}$), eine potentielle aber gegenwärtig zumeist ungenutzte P Ressource sind.

Zahlreiche Technologien wurden in den letzten Jahren mit dem Ziel der P-Rückgewinnung aus verschiedenen Teilströmen einer Kläranlage (Ablauf, Schlammwasser, Faulschlamm) sowie von Klärschlammaschen entwickelt und teilweise bereits großtechnisch umgesetzt. Im Rahmen dieser Arbeit wurden 19 ausgewählte P-Rückgewinnungstechnologien nach einer eigens entwickelten Methodik zusammengefasst und nach technischen, ökologischen und ökonomischen Kriterien bewertet. Dabei wurden sowohl die P-Rückgewinnungstechnologien an sich, als auch in Bezug auf die gesamte Prozesskette – vom Kläranlagenzulauf bis hin zur fachgerechten Entsorger sämtlicher anfallender flüssiger und fester Reststoffe – betrachtet. Dies ermöglicht die Miteinbeziehung von Wechselwirkungen mit dem bestehenden Entsorgungssystem in Hinblick auf technische Anforderungen, Umweltauswirkungen und Kosten in der Bewertung. Die Ergebnisse liefern auch Grundlagen für Gesetzgeber und politischen Entscheidungsträger, die für Entwicklungen eines Konzeptes für ein zukünftiges optimiertes P-Management genutzt werden können.

Ein wichtiger Aspekt der Ergebnisse ist, dass kein einzelner finaler Bewertungsindikator zur Bewertung der teils sehr komplexen und verschiedenen technologischen Ansätze sinnvoll ist. Vielmehr ergeben die zahlreichen ausgewählten Bewertungskriterien ein Gesamtbild, welches eine Rückgewinnungstechnologie und deren technische Ausgereiftheit, deren Umweltauswirkungen und Kosten ganzheitlich beschreibt.

Diese Arbeit zeigt zum Beispiel, dass technisch bereits ausgereifte und vielfach bereits großtechnisch umgesetzte Methoden zur Rückgewinnung von P aus Schlammwasser, unter gewissen Voraussetzungen wirtschaftlich betrieben und gleichzeitig reine, sehr gut pflanzenverfügbare Endprodukte erzeugt werden können.

Mit dem Ziel einer größtmöglichen Nutzung des abwasserbürtigen Phosphors wäre zukünftig jedoch eine Rückgewinnung aus Klärschlammaschen anzustreben. Bei der Verbrennung des Klärschlammes sollte dabei eine Vermischung mit P-armen und schadstoffhaltigen Brennstoffen vermieden werden. Die dafür notwendigen Strukturen wie z.B. Monoverbrennungsanlagen müsste dazu allerdings noch ausgebaut werden. Weitere Vorteile einer „Monoverbrennungs-Strategie“ sind zum einen die Möglichkeiten der Nutzung weiterer P-reicher Stoffströme (z.B. Tiermehl) und zum anderen die Möglichkeit einer Zwischenlagerung der Asche mit dem Ziel einer späteren Rückgewinnung (Aufbau einer österreichischen Phosphormine).

Insgesamt ist nicht davon auszugehen, dass sich eine Implementierung von P-Recycling Technologien auf volkswirtschaftlicher Ebene finanziell rentieren würde. Die Kosten der eingesetzten Technologien werden dabei stark von den Anforderungen an Recyclingraten und Produktqualitäten bestimmt. So reicht die Spanne von Technologien, mit denen ein gut pflanzenverfügbares Produkt, mit sehr geringer Schwermetallbelastung, zu vergleichsweise höheren Kosten und mit geringer P-Rückgewinnung erzeugt werden kann, bis zu Technologien, bei denen mit im Vergleich zu einer Entsorgung der Klärschlammasche auf Deponien ohne oder mit nur geringen zusätzlichen Kosten, sehr hohen P-Recyclingraten, aber keiner Entfernung von Schwermetallen und geringer Verfügbarkeit des Phosphors zu rechnen ist. Welche Technologie(n) zur Behandlung der Klärschlammasche schlussendlich zum Einsatz kommen, wird von den (rechtlichen) Anforderungen an Recyclingraten und an die Produktqualität und von ökonomischen Anreizen durch die Verwaltung abhängen.

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Table of Contents

1. Introduction

Phosphorus (P) is an essential key element for all living organisms and cannot be substituted by any other element. In modern agriculture there is a high demand for fast available and therefore water soluble mineral fertilizers based on phosphate rock (PR). Worldwide 80–90% of mined PR is used in agriculture (Van Kauwenbergh, 2010). With a growing population and changes in diet due to rising living standards in emerging nations and developing countries, agriculture's demand for P will increase and consequently its criticality (Reijnders, 2014; Van Vuuren et al., 2010).

A closer look on national levels illustrates that only 20–25% of the P applied in agriculture actually reaches the final food consumer (Scholz et al., 2014, Dijk et al., 2016). Usually, up to 30–40% of PR is lost during mining and processing. At a global scale, a 50% loss can be detected in the food chain between farm and fork looking, and only half of all manure is recycled back into farmland (Cho, 2013).

With P being a non-renewable resource, concerns were raised about the P supplies in the long-term future. At the beginning of the 21st century, the "peak P" (comparable to "peak oil") was discussed as the PR reserves[1] showed a static lifetime[2] of about 70–90 years (Cordell et al, 2009). However, reserves are dynamic and with increasing prices for PR within the past few years, certain resources[3] have been upgraded to a reserve. As a consequence, the static lifetime of PR is now stated to be ~300 years (USGS, 2015; Edixhoven et al., 2014; Van Vuuren et al., 2010; Van Kauwenbergh, 2010).

90% of the global PR reserves are located in just five countries: Morocco, China, South Africa, the U.S. and Jordan. Morocco alone accounts for up to 85% of the currently known economically exploitable P reserves worldwide. Austria, like all European countries, is lacking P deposits and is therefore entirely dependent on imports from partly geopolitically unstable regions (e.g., North Africa, Jordan). Consequently, Austria/Europe is highly vulnerable to price fluctuations in the fertilizer sector (World Bank, 2016).

Furthermore, studies revealed that the quality of PR deposits is declining: P contents are getting lower, while the pollution with (radioactive) heavy metals such as Cd, U, and Tl is increasing (Pfundtner, 2015, Kratz et al., 2016). At the moment, European countries still try to

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[1] Reserves: Mineral reserves are valuable and legally, economically, and technically extractable

[2] Statistic lifetime in years: Economically extractable reserves divided by the current depletion rate

[3] Resources: Mineral resources that are potentially valuable, and for which reasonable prospects exist for economic extraction

import PR with low levels of heavy metal pollution due to strict cadmium (Cd) limit values in the EU (EC, 2003). However, there is a very limited availability of low-cadmium phosphate rock. With a decrease in PR quality, the energy demand to mine and process P will increase. The adaption of new limit values e.g., on Cd (current: 75 mg Cd kg $P_2O_5^{-1}$; expected top-down limit: 60 mg Cd kg $P_2O_5^{-1}$ to a decrease to–20 mg Cd kg $P_2O_5^{-1}$) which are discussed within the revised European Fertilizer Regulation (Oosterhuis et al., 2000; EC, 2003), will require the implementation of decadmiation technologies (Evans, 2014). Thus, in the near future, prices for fertilizers will increase (Cichy et al., 2014).

Finally, the processing technologies for the extraction of phosphoric acid from PR, a diverse and important raw material for the feed-, food- and fertilizer industry, also raise environmental concerns. The most important waste by-product from P mining is phosphogypsum (PG), which is mainly composed of gypsum including high levels of P, but also high levels of heavy metals and other impurities such as fluorides, sulphates, and even naturally occurring radionuclides such as radium, uranium, and thorium (Greenpeace, 2013). The composition and content of the impurities in PG are strongly influenced by the phosphate ore quality (Tayibi et al., 2009). Up to 5 kg of PG are generated for producing one kg of phosphoric acid (~110 million tons of PG yr^{-1}).

Phosphoric acid is therefore often produced in countries with lower environmental standards, where the PG is deposited in (unsecured) stockpiles, polluting groundwater and nearby rivers or coasts (Greenpeace, 2013). Rutherford et al. (1994) describe the main environmental concerns associated with PG as followed: (1) movement of impurities below PG stacks into groundwater supplies, rivers and oceans; (2) radon-222 exhalation which may pose a health risk to workers on-site or people living close to stacks; (3) acidity (pH: ~1); and (4) radon-222 exhalation from soil into residential homes when agricultural land previously treated with PG is converted into settlement areas. These external (environmental) costs need to be considered, especially in the discussion of P recycling from national sources. However, in some countries, where PG reveals low concentrations of impurities, PG can be seen as a material for soil improvement and a raw material for construction (Hilton, 2009). Due to the previously mentioned challenges, in 2014, the EU listed phosphate rock as a critical raw material (see EC Critical raw materials; EC, 2014). Within the past few years, these challenges have also been intensively discussed on scientific and political levels.

One of many measures to reduce import dependency and to re-establish broken nutrient cycles is the recovery of P from obviously available but currently often unexploited national P sources such as municipal and industrial wastewater, meat and bone meal (MBM), and other organic wastes (Scholz et al., 2014).

1.1 Aim of the Thesis and research questions

The fundament for any optimization is the knowledge about the status-quo of a system. Therefore, the first aim of this Thesis was the development of a detailed and complete phosphorus budget for Austria based on the methodology of a material flow analysis (MFA). With this MFA the following research questions were answered:

- How much P is currently imported as fertilizers, food, and feed?
- How much P is applied to agricultural soils with conventional mineral and organic fertilizers?
- How great is the P potential in certain waste flows (e.g., municipal sewage sludge, meat, and bone meal), which are discussed as possible substitutes for raw phosphate rock/mineral fertilizer imports?
- Where does P finally end up in the current system and is this P recoverable?

Based on the knowledge of the large P potential in a WWTP (effluent, digester supernatant, sewage sludge) and sewage sludge ashes, manifold promising technologies have been developed and, in some cases, already full-scale implemented to recover P from these wastewater related streams. However, so far no methodology is known, which not only accepts the challenge to holistically assess the technologies for themselves, but also in the context of existing systems. Therefore, the following research questions arise in the context of P recovery technologies:

- Which criteria are required to perform a holistic technical, environmental, and economic assessment?
- How do the technologies perform in relation to P recovery and removal of unwanted substances such as heavy metals and organic micropollutants?
- How to assess newly recovered P rich materials?
- Which methodological approaches need to be applied to perform a comparative assessment of these newly developed technologies?
- How to assess possible positive and negative impacts on a WWTP, or even on the entire system – starting from the treatment of wastewater to the final disposal of occurring liquid and solid wastes?

- Which indicator should be selected to properly assess these technologies and compare the results with current fertilizers production based on raw phosphate rock?
- How to evaluate the relevance of potential emissions to the environment?

1.2 Structure of the Thesis

This Thesis is the outcome of a cumulative and continuous research work on P recovery from wastewater based on three publications in peer-reviewed scientific journals:

1. At the beginning a detailed P balance was created in order to understand the national P flows and to identify current P losses as well as potential waste streams for substituting e.g., mineral fertilizer imports. The results of this study were published in the journal *Resources, Conservation and Recycling* (Egle et al. 2014a) and are presented in *Chapter 2* of this Thesis.
2. Manifold technologies have been developed in recent years to recover wastewater P. The basis for any technology assessment is the knowledge about the process fundamentals, the paths of nutrients and potential hazardous substances as well as the resource demand. These information, together with substance flow models for selected technologies, are presented in a Review Paper on P recovery technologies from municipal wastewater in the journal *Resources, Conservation and Recycling* (Egle et al. 2015; *see Chapter 3*).
3. Using the data from the review paper, 19 newly developed technologies and industrial processes have been selected to perform an integrated and comparative technology assessment, taking into account technical, environmental and economic criteria. The methodology and results of this assessment were accepted on July 3rd 2016 within the journal *Science of the Total Environment* (Egle et al. 2016 in press; *see Chapter 4*).
4. Chapter 5 contains supplementary results from an environmental assessment (greenhouse gas emissions, emissions with an acidification potential, cumulative energy demand) of the technologies. These results have not yet been published in a journal and can be seen as supplementary data to complete the picture of the considered recovery technologies.
5. After presenting the scientific publications, the overall conclusions are presented in a separate chapter. Finally, supplementary data and results are attached, including additional substance flow models, data on recovery technologies.

2. The Austrian phosphorus budget as a basis for resource optimization

2.1 Introduction

Phosphorus (P) is essential for supporting life on earth. It is a non-substitutable substance that serves vital functions in all living organisms. P application with mineral fertilizers, obtained from sedimentary or and magmatic deposits, is one of the factors that permits increased crop yields and livestock production, on which the world's fast-growing population depends. However, P is a non-renewable resource with a limited temporal availability. According to recalculated estimates of mineable Preserves (USGS, 2012), most recent studies and scenario analyses show that the worldwide Preserves will not be depleted in the short-term future (Scholz and Wellmer, 2013 and Van Vuuren et al., 2010). Nonetheless, additional factors such as the uneven distribution of P deposits worldwide, their concentration in geopolitically unstable regions and the rising contamination of deposits (heavy metals such as Cd and U) affect the availability of P fertilizer and therefore its price worldwide (Silva et al., 2010 and Van Kauwenbergh, 2002). More than 90% of the worldwide Pore is produced in Morocco, China, the USA, Russia and South Africa. Furthermore, poor recycling rates of P in economies and strategic decisions of P-rich countries also affect P prices. Figure 1 shows the decreasing trend of mineral P fertilizer sales and application in Austria and the strong price fluctuations in global P fertilizer markets in recent years (World Bank, 2013 and Green Report, 2012).

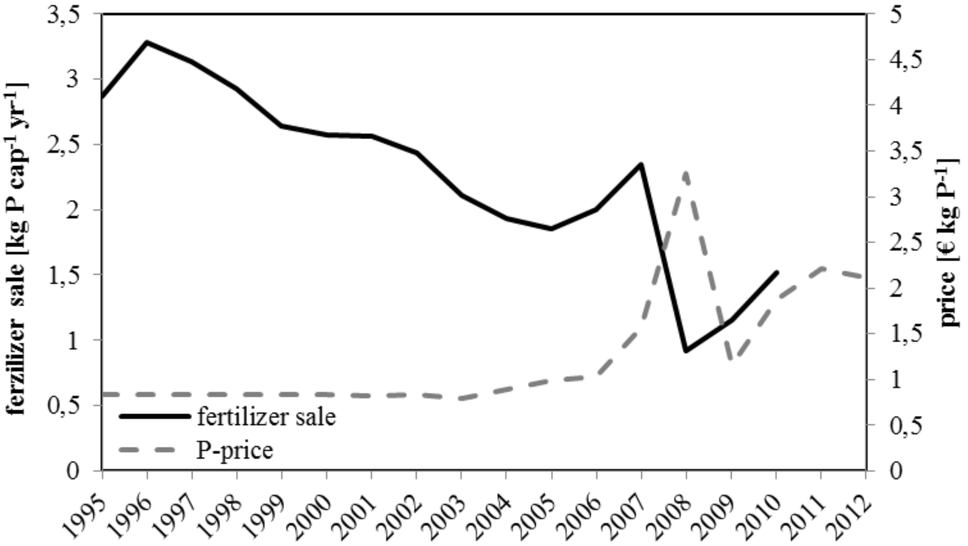


Figure 1: Trend of P fertilizer price and sale in Austria (Price: Euro per kilogram phosphorus; Fertilizer sale: kilogram phosphorus per capita and year).

In the past, phosphorus and nutrient-rich wastes in general from all sectors were directly recycled in closed or nearly closed loops (Ashley et al., 2011). Urbanization significantly changed these pathways to systems in which most nutrient-rich flows are economically irrecoverable (Cordell et al., 2009 and Smil, 2000). Studies on the cycle of P through the anthropogenic metabolism have been carried out at both the global (Cordell et al., 2011, Liu et al., 2008, Villalba et al., 2008 and Smil, 2000) and European level (Ott and Rechberger, 2012), highlighting major issues pertaining to P use and availability. However, there is also a need to assess the P cycle on national and regional scales to investigate specific problems and opportunities that may differ considerably in different countries. Furthermore, it is easier to implement measures and policies on a national level. For these reasons, in the past few years, researchers started to carry out P budgets on a regional scale in, e.g., Finland (Antikainen et al., 2005, Antikainen et al., 2004 and Sokka et al., 2004), Sweden (Neset et al., 2008), China (Yuan et al., 2011 and Liu et al., 2004), Switzerland (Binder et al., 2009), Japan (Mishima et al., 2010), the Netherlands (Smit et al., 1955), the USA (Suh and Yee, 2011), Germany (Gethke-Albinus, 2012) and France (Senthilkumar et al., 2012). These regional balances differ considerably from each other because of the different approaches and methodologies used in the different studies. In particular, due to the complexity of the systems, there is a tendency to aggregate flows. This study, on the contrary, is an attempt at a comprehensive analysis of all the components of the anthropogenic system that are related to P, conducted to try to quantify every specific flow at a very detailed level, focusing in particular on the *Wastewater-* and *Waste management* sectors, due to their high potential for management improvements. This detailed approach was adopted because it offers some important advantages. Every process is dependent upon several input and output flows, the data for which are derived from different sources. Because for this type of study, direct cross-checking is not always possible because of the lack of data from different sources, the high number of input and output flows tends to minimize the alterations due to statistical reconciliation. Furthermore, this detailed and comprehensive model delivers more specific and reproducible results, which may allow improved comparisons between P balances carried out indifferent countries or regions. A very detailed description of the P balance also has considerable added value as decision support tool for resource and waste management. The assessment of P flows through several specific processes provides an essential basis for thoroughly understanding and evaluating the current situation and investigating the potential impacts and benefits of different management options in terms of P flows and stocks.

2.2 Methods and data

The investigation and quantification of all the relevant flows and stocks of elemental P in Austria for the time period 2004–2008 are carried out according to the Material Flow Analysis (MFA) methodology (Brunner and Rechberger, 2004). The freeware STAN is applied to perform the P balance. STAN makes it possible to visualize complex MFA systems and perform error propagation and data reconciliation, by taking into account data uncertainty (Cencic and Rechberger, 2008). The reconciliation, based on the method of least squares regression, alters the input data to eliminate existing contradictions in the mass balance. The initial uncertainty in the data is the weighting factor that determines the extent to which data can be altered, and as result, the final uncertainty is reduced. This study is based on various types of data, such as statistical data, values reported in the literature, measurement values and the authors' own estimates, which are characterized by various levels of uncertainty. This study applies the data uncertainty method developed for urban heavy metal data collection (Hedbrant and Sörme, 2000). It is a two-step method. The first step consists of assigning to each input data component a pre-defined uncertainty level (UL), based on the type and reliability of the data source. The second step is the calculation of an uncertainty factor (UF), based on the previously defined uncertainty level, to obtain an uncertainty interval with a 95% probability of encompassing the actual value. Table 1 shows the uncertainty levels assigned to the different types of data available for this research.

Table 1: Methodology of the uncertainty concept (Hedbrant and Sörme (2000)).

Size category [$\text{PE}_{\text{COD120}}$]	Number of WWTP	actual capacity [$\text{PE}_{\text{COD120}}$]	percentual on actual capacity [%]	sewage sludge [t DS yr^{-1}]	Phosphorus [kg P $\text{cap}^{-1}\text{yr}^{-1}$]
51-1,999	1,206	308,756	2.4	6,253	0.02
2,000-10,000	371	1,074,214	8.2	21,754	0.06
10,001-50,000	200	3,108,011	23.8	63,941	0.02
50,001-100,000	31	1,285,190	9.8	26,027	0.30
>100,000	30	7,309,374	55.9	148,025	0.35
total	1,837	13,085,544	100	265,000	0.75

2.2.1 Description of the system

The first step in the analysis of the system is the definition of its spatial and temporal boundaries. Because the aim of the study is a national balance, the geographic boundaries are identical to the Austrian national borders. From a vertical perspective, the uppermost 30 cm of the soil is considered, due to its relevance as P stock. The atmosphere is not included in the study because it plays a negligible role in the P cycle. With respect to the temporal scale, for reasons of data availability, a 1-year period is selected. Whenever possible, the average value of the considered period between 2004 and 2008 is calculated, to level out short-time fluctuations. The basic unit of the system is kg P cap^{-f} yr^{-y} (kilograms of phosphorus per capita and per year). The Austrian population during the period of study was approximately 8.2 million inhabitants. This relative unit is chosen to allow comparison with other countries or among smaller regions within Austria. Once the system is defined, the relevant processes, stocks and flows are identified. The system is composed of 9 processes and 64 flows (Figure 3). The processes, selected for their relevance with respect to the use of P, are ordered in a top-down model from the primary sector through the treatment and disposal of waste and wastewater, to trace the flow of P through the anthropogenic metabolism. Six of the processes are also assigned with a stock. For three of them, the estimation of the existing P stock is possible (*Animal husbandry, Crop farming, and Forestry and miscellaneous soils*), while for the rest, only the annual change can be calculated. What is actually imported, exported and exchanged between the processes are goods containing different concentrations of P. Thus, the flows in the system are the result of the multiplication of two types of values: the mass flows of goods and their P concentrations. The flows are numbered according to their source-processes. The explanation of each flow is discussed within the section dedicated to the respective source-process. Import flows constitute an exception to this rule because they have no source-process. These flows are therefore described in the sections pertaining to their end-processes, after which they are also named. The Appendix contains detailed information about the selected flows (Table A 1, Table A 2 and Table A 3).

2.2.1.1 Animal husbandry

The production of living animals, animal products such as meat, milk and eggs, and the associated generation of by-products are assessed within the process *Animal husbandry*. This process encompasses import (F1.1) and export (F1.2) flows of living animals from and to other countries, excluding wild animals and fish. The data are obtained from official Austrian statistics (Statistics Austria, 2010a and Statistics Austria, 2010b), and the average values for the period 2004–2008 are considered. P concentrations are extracted from previous official studies regarding nutrients in Austria (Zessner and Lampert, 2002 and Kroiss et al., 1998). The same P concentrations are applied to all other food and animal feed goods assessed in the study. F1.4 indicates the supply of slaughtered animals, milk and eggs to agri-food industrial processes (Statistics Austria, 2010b, Statistics Austria, 2010c and Statistics Austria, 2010d). Dead animals, namely, fallen stock not suitable for human consumption, are sent to waste treatment plants (F1.6) (BAWP, 2006 and BAWP, 2011). Manure, a by-product of animal production, is divided in two different flows: F1.3, if it is applied directly to agricultural fields, or F1.5, if it is treated in biogas plants. The data are based on available information related to 200 biogas plants (E-Control, 2010) and were extrapolated to the 344 existing biogas plants present in Austria. Total manure generation is calculated using official data on livestock and official data on specific P excretion per type of animal and pitch space (BMLFUW, 2006). Manure generation and corresponding P concentrations are cross-checked and verified with data from official German reports (LFL, 2010). The stock (S1) associated with *Animal husbandry* consists of the livestock present in the country. It can vary depending on import–export rates, as well as on changes in the number of slaughtered animals or the number of dead animals.

2.2.1.2 Crop Farming

Crop farming takes into account all agricultural processes, as well as the agricultural soil acting as P storage. The associated stock (S2) contains the total surface of agricultural fields, according to official reports (Green Report, 2005–2009), and it considers an average P content of 3.750 ± 1.250 kg P per hectare (ha) in the uppermost 30 cm of soil (Kroiss et al., 1998). Changes to this stock occur due to fertilization, harvest and erosion. *Crop farming* receives an input of P, represented by atmospheric deposition (F2.5) of approximately 0.2–0.4 kg P ha^{-1} yr^{-1}on agricultural fields (Kroiss et al., 1998). In contrast, flow F2.3 reflects the loss of P from agricultural soils to *Water bodies* due to erosion, groundwater and surface runoff. For Austria, these flows have been calculated with MONERIS (Modeling Nutrient Emissions in River Systems), a semi-empirical conceptual model for the quantification of nutrient emissions from point and diffuse sources in river catchments (Behrendt et al., 1999). MONERIS has been adapted to alpine conditions and applied in Austria on a spatial scale of 367 sub-catchments with an average size of approximately 200 km^2 (Zessner et al., 2011). Calculations made with MONERIS have been validated by comparison with observed river loads for 100 river gauges across Austria. The results have been aggregated to the national level. Calculations made with MONERIS show that 5–10% (the sediment delivery ratio) of particulate P mobilized by erosion finally ends up in *Water bodies*. This estimated range is consistent with values reported in the literature. The rest is retained (re-deposited) in terrestrial soils—although the type of soil into which it is redeposited is not indicated by the calculations. According to Novotny (2003), most mobilized particulate P remains in agricultural soils and is not transferred to other types of soils (e.g., forest soils). Therefore, we made the assumption that 20% of total mobilized particulate P from agricultural soil (F2.4) is transferred to *Forestry and miscellaneous soils*. The rest, which is not transferred to *Water* *bodies* or *Forestry and miscellaneous* soils, stays in agricultural soils and thus in the process *Crop farming*. High uncertainties need to be considered as these findings about the distribution of particulate P retention in different types of terrestrial soils are based on estimates from the literature only. In contrast, the estimates of inputs into rivers are much less uncertain as they are validated measured values. Flow F2.2 reflects the production of vegetal products (arable crops, wine, fruits and vegetables) sent to the agri-food industry (BAWP, 2006 and BAWP, 2011). Non-marketable animal feed, such as grassland production, straw and chaff that is not processed by the *Industry* is transferred directly to *Animal husbandry* (F2.1) (Green Report, 2005–2009). Considerable amounts of vegetal products, mainly consisting of maize, grass, rye silage and other grasses such as alfalfa and Sudan grass, are major material inputs (F2.4) to biogas plants in Austria (E-Control, 2010).

2.2.1.3 Forestry and miscellaneous soils

All non-agricultural soils, together with forest soils and soils associated with the processing, trade and commerce of wood and paper, are included in the process *Forestry and miscellaneous soils*. Like agricultural fields, forests and non-cultivated soils receive an input of P as result of atmospheric deposition (F3.9) and experience an outflow of P (F3.7) due to erosion, groundwater emissions and urban runoff. This process is further connected with the exterior of the system, through import (F3.1) and export (F3.2) of wood and paper, as well as new and waste paper (Austropaper, 2010 and Energy Agency, 2007). Part of the wood produced in Austria is recovered in domestic and industrial biomass plants, generating heat and electricity but also ash containing P, which is quantified by flow F3.5 (Energy Agency, 2007). Flow 3.8 includes raw wood materials and manufactured products for households (Energy Agency, 2007). Due to the lack of data concerning the need for paper in *Industry* (F3.3) and *Consumption* (F3.4), a 50:50 ratio is assumed. P contained in the wastewater of the paper industry and sent to wastewater treatment plants is reflected in the flow F3.6 (Kroiss et al., 1998). The stock associated with this process (S3) considers the amount of P present in the uppermost 30 cm of soil in forests and in non-cultivated fields (Kroiss et al., 1998), as well as the P contained in the total wood stock of the forests.

2.2.1.4 Chemical industry

F4.1 and F4.2 reflect the total imports and exports of chemical products such as raw materials and insecticides (Statistics Austria, 2010g). In the case of insecticides, the calculation of the P flows is based on the molar mass of every specific chemical compound. In contrast, for detergents, official data are not available. Based on available data on P consumption of detergents per capita in Germany (0.10 kg; FEA Germany, 2010) and Switzerland (0.13 kg) (Binder et al., 2009), the amount used in Austria is estimated based on the average value (F4.4). Flow F4.3 reflects the P content of the insecticides employed in agriculture (Green Report, 2005–2009). P contained in the wastewater of the *Chemical industry* and sent to wastewater treatment plants is reflected in the flow F4.5 (Kroiss et al., 1998).

2.2.1.5 Industry (food, feed, fertilizer)

Industry includes the agri-food sector, i.e., the processing, trade and commerce of food and animal feed, the production and commerce of mineral fertilizers from phosphate ore and raw materials and further processing of paper products. Due to the absence of mineable P resources in Austria, all of the phosphate ore and other phosphate raw materials (F5.1) used for mineral fertilizer production have to be imported (Green Report, 2005–2009 and FAOSTAT, 2013). As shown in Figure 1, the sale of mineral fertilizers is subject to strong fluctuations, mainly provoked by price changes. The export of mineral fertilizers, represented by the flow F5.16, is a result of the balance, i.e., it is calculated as the difference between the total import of phosphate ore and the amount of fertilizer actually used in the country, assuming that the fertilizer industry is not building any internal stock. Austria also imports and exports P in the form of food products, represented by flows F5.2 and F5.6, respectively. These flows contain food of both vegetal and animal origin, with the exception of fish and wild animals (Statistics Austria, 2010d and Statistics Austria, 2010e). Imports and exports of animal feed are represented by the flows F5.3 and F5.7, respectively. These flows include soy, peanuts, by-products of vegetable oil and starch production, meat and bone meal, fish meal, cereal bran and spent grain (Statistics Austria, 2010f). Flow F5.4 quantifies the P associated with the production of seeds. Mineral fertilizers are mostly consumed in agriculture (F5.5; 97–99%) but are also directly consumed by final consumers, e.g., in gardens (F5.12; 1–3%) (Heinzlmaier, 2010, Strasser, 2010 and Binder et al., 2009). The P content in the final consumption of food products is reflected in the flow F5.11. The wastes generated by the *Industry* process are separated into three flows. Flow F5.13 represents the wastes of animal origin (BAWP, 2006), taking into account the high discrepancies in the P concentrations of various categories of animal waste, e.g., slaughterhouse residues with and without bones (UBA, 2001). Most vegetal by-products generated by food processing are directly reused as animal feed (F5.15); while only 18% is actually treated as waste (F5.14). Furthermore, P is also contained in non-organic industrial waste (F5.10), consisting of waste paper and waste wood (ARA, 2005–2009). F5.8 reflects the P content of industrial wastewater treated in situ, while flow F5.9 represents the wastewater originating from the agri-food industry that is collected in municipal sewage systems and thus treated in wastewater treatment plants. According to Lindtner and Zessner (2003), 22% of the wastewater collected by municipal sewer systems is generated by the industrial sector, and 60% of this is generated by the agri-food sector (Obernosterer and Reiner, 2003 and Kroiss et al., 1998).

2.2.1.6 Consumption

The *Consumption* process reflects the flows of P associated with final consumers, considering not only households per se but also biomass plants, parks and cemeteries.

The input flows comprise all consumed goods containing P, i.e., food, mineral fertilizers, wood, paper and cleaning products, which have been quantified in flows as described within their own source-processes. The output flows can be separated into solid and liquid by-products generated through the consumption of such goods. Municipal wastewater (F6.1) is collected and sent to wastewater treatment plants. The solid waste produced by final consumers is divided into several categories: residual waste (F6.2; green waste, paper and cardboard, wood), separately collected organic waste (F6.3; garden waste), waste materials (F6.4; separately collected paper and wood) and green waste from markets and the trimming and pruning of municipal gardens, parks, streets and cemeteries (F6.5) (BAWP, 2006 and BAWP, 2011; ARA 2005–2009). Due to inaccurate data on the amounts of all collected heterogeneous organic wastes and their P concentrations, high uncertainty levels need to be considered. Flow F6.6 reflects the ash produced by the combustion of wood for heating in private households. A stock is assigned to the *Consumption* process (S6), to balance, e.g., composting and fertilizer application in gardens.

2.2.1.7 Wastewater management

Due to the large expected potential offered by wastewater with respect to P recovery opportunities, the *Wastewater management* sector plays an important role in the national P balance (Figure 2).

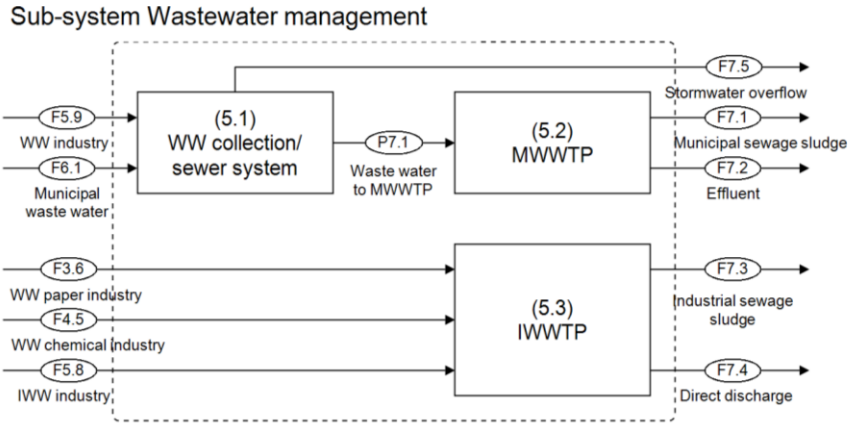


Figure 2: Conceptual model of the Austrian phosphorus budget: Sub-system Wastewater management.

The process is subdivided into municipal wastewater treatment plants (MWWTP) and purely industrial wastewater treatment plants (IWWTP) that process industrial wastewater not connected to a municipal sewer system. Wastewaters from the paper (F3.6), chemical (F4.5) and agri-food industries (F5.8) are mainly treated in situ, with generation of industrial sewage sludge (ISS) (F7.3) and direct discharge of the effluent (F7.4) to the hydrosphere. The total P load in municipal wastewater (MWW) derives from human metabolism excretions, detergents and food wastes, as well as from industries connected to municipal sewer systems. Of the total P freight in the collected MWW, 93% is sent to MWWTP, while the remaining 7%, contained in the storm water overflow, is directly conveyed to the hydrosphere (F7.5). Losses to soil and groundwater by sewer systems are not taken into account. Mechanical–biological wastewater treatment with combined simultaneous P precipitation by metal salt (Fe and/or Al) is common practice in Austria to avoid eutrophication in water streams and to fulfil strict limits on nutrient outflow (<1 mg P L^{-1}), in compliance with the requirements for sensitive areas established by the Urban Waste Water Directive (EC, 1991). Approximately 85–90% of the P contained in the influent is transferred to municipal sewage sludge (MSS) (F7.2), while the rest reaches the hydrosphere through the effluent (F7.1).

2.2.1.8 Waste Management

Due to the numerous types of wastes involved, the complex *Waste management* (WM) process has been studied in further detail, considering the state-of-the-art processes for waste treatment and recycling. The process is assigned a stock that encompasses landfills and cement kilns. Biological treatment sub-processes, such as mechanical–biological treatment (MBT), composting (C) and biogas (B), transform reactive organic wastes into stabilized products, i.e., substrates used in landscaping (F8.3), organic fertilizer applied in agriculture (F8.2) and compost used in gardens (F8.8).

In thermal treatment processes, such as municipal waste incineration (MWI) and mono-incineration (MI), P is mainly concentrated in bottom ash (83% of input P) and fly ash (98% of input P). The resulting filter cakes need to be stored in underground deposits abroad (F8.5), while bottom ash and fly ash are either deposited in landfills or utilized by the cement industry. All of the P input to the cement industry is transferred to the final concrete product. In coal-fired power plants, 96% of P is transferred to disposable ash and the remaining 4% is transferred to lime (Lederer and Rechberger, 2010). The residual waste, which is rich in organic matter, is directly treated in MWI (62%) or sent to MWI after mechanical–biological treatment (23%). Only smaller amounts are directly deposited in landfills (15%). In the rendering sub-process, animal wastes and fallen stock are transformed into meat and bone meal, 57% of which is used as secondary fuel in cement kilns, 40% is exported, 2.5% is used as fertilizer and 0.5% is treated in biogas plants (BAWP, 2006 and BAWP, 2011). Sewage sludge is distributed to a large number of sub-processes, mainly due to its varying composition and to the diversified legislation regulating its use and disposal. Industrial sewage sludge (ISS) (F7.3) is mainly used as secondary fuel in cement kilns. On the contrary, P-rich municipal sewage sludge (MSS) (F7.1) is not uniformly managed because of differences, unspecific regulations within different regions of Austria, varying degrees of social acceptance of related potential pollution (heavy metals and persistent organic pollutants) and conflicts of interest associated with agriculture (fertilizer and soil value), landscaping (soil value), energy-intensive industries (energy value and secondary fuel) and waste incineration plants (cooling value and covering overcapacity). The *Waste management* process contains a stock (S8), represented by landfills and cement kilns, from which P is assumed to be not currently recoverable.

2.2.1.9 Water bodies

Austria receives a considerable import of P from Germany via the Danube River. Inputs to *Water bodies* from diffuse sources such as erosion and point sources such as wastewater treatment plants are assessed in flows described within their own source-processes. The Danube River itself and all other rivers in Austria that flow out of Austria represent a significant export of P from the national balance (F9.2) (Kroiss et al., 2005). P freights in the Danube River entering and leaving Austria, as well as in other major rivers leaving Austrian territory, are calculated by multiplying water mass flows as monthly averages by corresponding P concentrations determined from one or two measurements per year. Finally, freights in months in the period 2004–2008 are added and expressed as a yearly average. Nevertheless, not all P that flows into rivers is exported through *Water bodies*, due to retention in sediments and in the riparian zone. Therefore, an assumed stock increase with high associated uncertainties (S9) is assigned to this process.

2.3 Results

Figure 3 presents the resulting Austrian P balance. The results are expressed as reconciled values affected by an error, which is the reconciled uncertainty. The balance shows a total import of 8.4 ± 5% kg P cap^{-1} yr^{-1}and a total export of 6.3 ± 6% kg P cap^{-1} yr^{-1}. This implies that the national P stock is increasing at an annual rate of 2.1 ± 5% kg P cap^{-1} yr^{-1}. The import of P mineral fertilizers and raw materials is the predominant import flow, with a value of 5.8 ± 6% kg P cap^{-1} yr^{-1}. However, a large fraction of this imported P (3.8 ± 9% kg P cap^{-1} yr^{-}1) is in turn exported in the form of mineral fertilizers. Other relevant import flows are represented by the imports of animal feed (0.57 ± 11% kg P cap^{-1} yr^{-1}), food (0.38 ± 12% kg P cap^{-1} yr^{-1}) and chemical products (0.37 ± 39% kg P cap^{-1} yr^{-1}). In turn, the most important export flows, after P mineral fertilizers, consist of food (0.31 ± 10% kg P cap^{-1} yr^{-1}), meat and bone meal (0.28 ± 12% kg P cap^{-1} yr^{-1}), animal feed (0.25 ± 18% kg P cap^{-1} yr^{-1}) and chemicals (0.23 ± 63% kg P cap^{-1} yr^{-1}). Half of the national stock increase (1.1 ± 5% kg P cap^{-1} yr^{-1}) occurs in landfills and cement kilns, 23% occurs in agricultural soils, and the remaining 27% is partly due to accumulation in forestry soils, miscellaneous soils and gardens and partly due to retention in *Water bodies*.

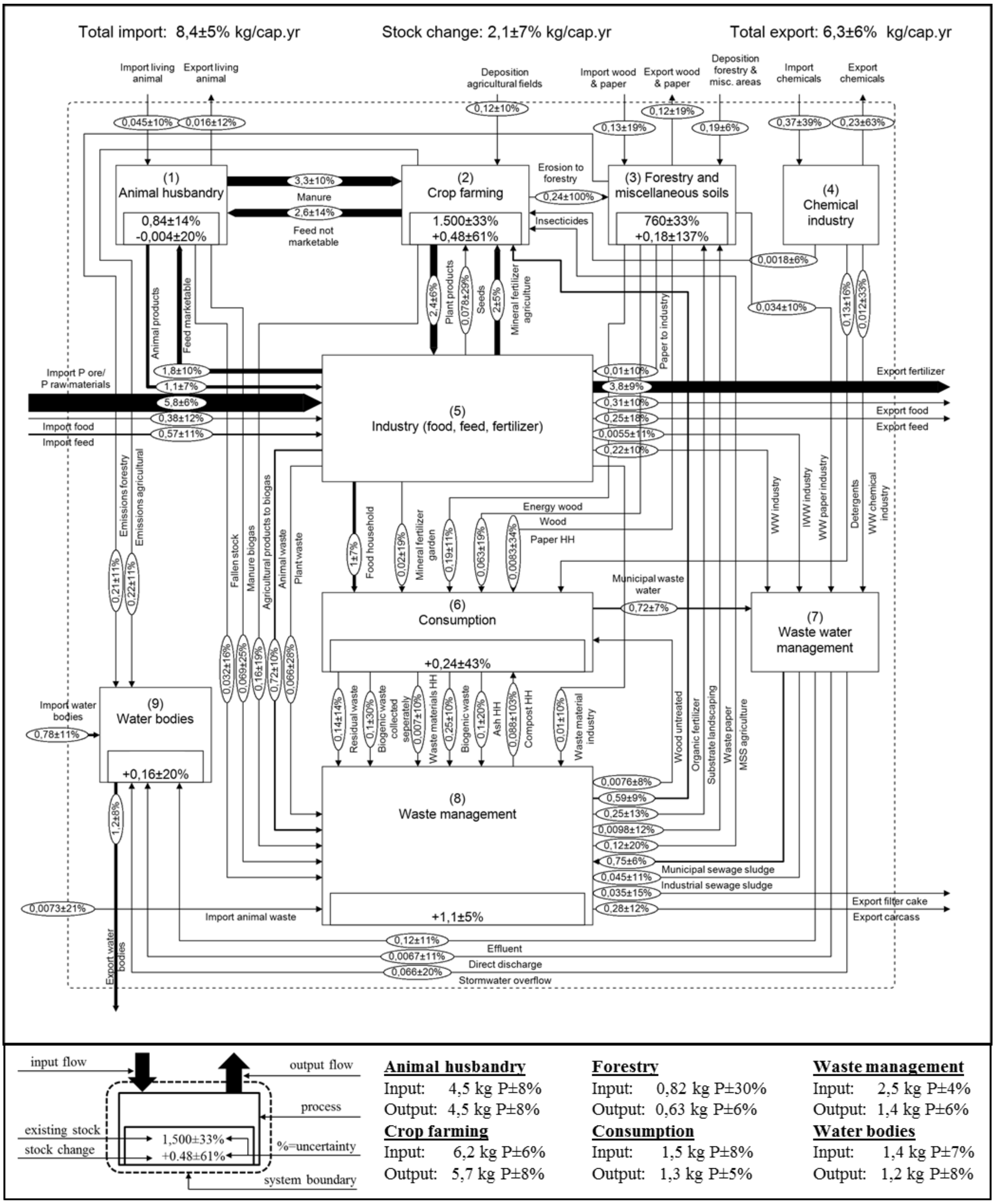


Figure 3: Results of the Austrian national phosphorus budget with reconciled data in kg P cap^{-1} yr^{-1}.

The predominant P flows in the agricultural sectors of *Animal husbandry* and *Crop farming* are manure (3.3 ± 14% kg P cap^{-1} yr^{-1}) and non-marketable feed (2.6 ± 14% kg P cap^{-1} yr^{-1}). The total food production is 3.5 kg P cap^{-1} yr^{-1}. Of this total, 1.1 ± 7% kg P cap^{-1} yr^{-1} originates from animal sources, whereas 2.4 ± 6% kg P cap^{-1} yr^{-1}originates from vegetal sources. Marketable feed accounts for 1.8 ± 10% kg P cap^{-1} yr^{-1}. In *Animal husbandry* and

Crop farming, total P loads of 4.5 and 6.2 kg P cap^{-1} yr^{-1}, respectively, are used for food production.

The balance reveals that food consumption in Austrian household's accounts for approximately 1 ± 7% kg P cap^{-1} yr^{-1}. The second-largest input flow for the *Consumption* process is represented by wood consumed for energy in biomass plants (0.19 ± 11% kg P cap^{-1} yr^{-1}), and the third-largest input flow is represented by detergents consumed (0.13 ± 16% kg P cap^{-1} yr^{-1}). With respect to the outputs of *Consumption*, the most important flow is municipal wastewater (0.72 ± 7% kg P cap^{-1} yr^{-1}), followed by green waste (0.25 ± 10% kg P cap^{-1} yr^{-1}) and residual waste (0.14 ± 14% kg P cap^{-1} yr^{-1}).

Municipal wastewaters arising from households (75%) and from industry (25%), which together carry a P freight of 0.94 kg P cap^{-1} yr^{-1}, are treated in MWWTP. Thanks to high P removal rates, approximately 0.75 ± 6% kg P cap^{-1} yr^{-1} is transferred to MSS. The P freight of ISS is much lower, amounting to only 0.045 ± 11% kg P cap^{-1} yr^{-1}.

Through their flow across the Austrian territory, the P loads of surface waters increase substantially, from 0.78 ± 11% kg P cap^{-1} yr^{-1}in the Danube River at the German border to 1.2 ± 8% kg P cap^{-1} yr^{-1}in the Danube at the Slovakian border and other rivers that flow out of Austria to the east and south-east. Point sources, represented by flows of effluent, direct discharge, and stormwater overflow, are responsible for a total P load of 0.3 kg P cap^{-1} yr^{-1}, which accounts for approximately 30% of total input to *Water bodies*. The contribution from diffuse sources, such as agriculture and forestry, is approximately 0.43 kg P cap^{-1} yr^{-1}. In addition, P is retained in sediments and in the riparian zone, resulting in a stock increase of 0.16 ± 20% kg P cap^{-1} yr^{-1}. However, this stock is subject to strong fluctuations due to discharge fluctuations such as floods.

The major P inputs to the *Waste management* process are MSS (0.75 ± 6% kg P cap^{-1} yr^{-1}) and animal waste (0.72 ± 10% kg P cap^{-1} yr^{-1}), followed by the separately collected biogenic waste from households and industry (which together account for 0.42 kg P cap^{-1} yr^{-1}) and by residual wastes (0.14 ± 14% kg P cap^{-1} yr^{-1}). Sewage sludge is involved in nearly every treatment process within this sub-system. Sixteen percent of MSS is directly reused in agriculture as fertilizer, 40% is incinerated, and 26% is treated at the fluidized bed reactor (mono-incineration) in Vienna (BAWP, 2006 and BAWP, 2011). Currently, the outputs (fly ash and filter cake slag) of the incineration process are either landfilled or exported. Another 36% of MSS is treated by biological processes such as composting, MBT and biogas, whose

P rich by-products are mainly reused in agriculture and landscaping (Figure 4a). The fraction of MSS recycled to *Crop Farming* via compost or biogas slurry is 0.10 kg P cap^{-1} yr^{-1}. During the time period 2004–2008, 9% of MSS was still directly landfilled. Compared to MSS, the P flow of ISS is very small, amounting to 0.045 ± 11% kg P cap^{-1} yr^{-1}. Almost all of this flow is currently used as secondary fuel in several co-incineration plants. Therefore, this flow of P is lost in cement kilns and landfills.

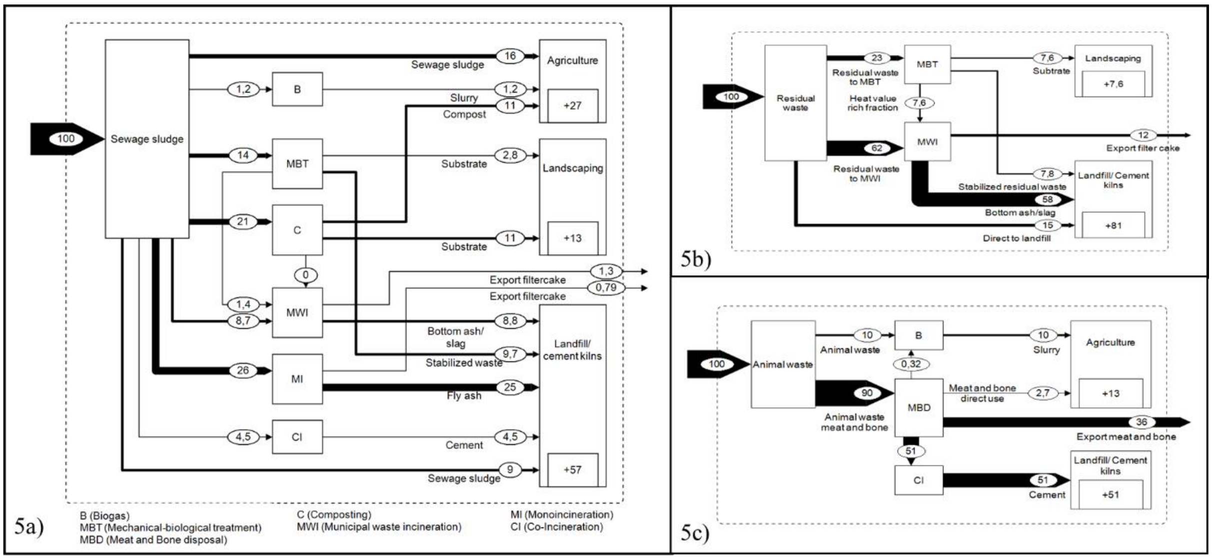


Figure 4: Specific P flow of three types of waste (a) sewage sludge, (b) animal waste, (c) residual waste.

For most of the animal waste from *Industry* (F5.13), fallen stock and imported animal waste, amounting to a combined P load of 0.65 kg P cap^{-1} yr^{-1}, rendering is the common treatment process. Fifty-seven percent of the generated meat and bone meal is used as secondary fuel in cement kilns, 40% is exported, and 3% is used as fertilizer or as substrate in biogas plants (Figure 4b).

Separately collected municipal and industrial organic wastes (F6.3, F6.5 and F5.14) amount to 0.42 kg P cap^{-1} yr^{-1}. The currently preferred treatment for these wastes is composting. Together with the input of sewage sludge, 0.62 kg P cap^{-1} $yr^{-1}$of compost is generated annually. Forty-five percent of this compost is currently applied to arable land, 35% is used as substrate for landscaping, and the remaining 20% is employed as fertilizer in gardens. However, high levels of uncertainty are associated with these flows. Compared to the above-mentioned flows, the P content in residual waste (F6.2) is quite low (0.14 ± 14% kg P cap^{-1} yr^{-1}). Additionally, P-rich fractions in residual waste are mingled with other waste fractions. Through MBA pretreatment located upstream of MSI, the high-calorie fraction is separated and the main parts of P-rich fractions are separated, ending up in landfills as stabilized waste (0.10 ± 21% kg P cap^{-1} yr^{-1}; Figure 4c).

To summarize, the final destinations of P containing waste materials within the Austrian P budget are the following: landfills, 44%; agriculture, 29%; export, 13%; landscaping, 10%; and households (gardening), 4%.

2.4 Discussion and Conclusion

Austria, like all other European countries except Finland, has no mineable phosphate deposits. Therefore, as might be expected, the import of P mineral fertilizers and raw materials represents the major flow of the entire national P balance, with a value of 5.8 ± 6% kg P cap^{-1} yr^{-1}. However, only approximately 34% of this amount is actually applied as mineral fertilizer to agricultural fields and gardens. The remaining P freight is exported and has no effect on the Austrian P budget. Furthermore, the P import of the Danube River (0.78 ± 11% kg P cap^{-1} yr^{-1}) is a passive flow that does not affect the rest of the budget. Therefore, the ratios and percentages discussed in this section are based on an adjusted total P import of 3.8 kg P cap^{-1} yr^{-1}. The adjusted import of mineral P fertilizers (actually applied in agriculture) accounts for almost 53% of the total import of P into the system. Imports of food and animal feed account for 25%, chemicals for 10%, deposition for 8%, and minor sources, e.g., wood and paper, living animals and animal wastes, approximately 5%. The major export flows in the adjusted balance are food and animal feed (34%), export via *Water Bodies* due to erosion, emissions via groundwater, point sources and other emissions from urban areas (26%), meat and bone meal (17%), chemicals (14%), wood and paper (7%) and fly ash for underground disposal (2%).

2.4.1 Food production – efficiency of food supply chain

As expected, the abundance of large flows between the processes *Animal husbandry*, *Crop farming* and *Industry* reflects the major role of food production within the national P balance. *Crop farming* and *Animal husbandry* receive a total net input (excluding circulating flows such as manure and non-marketable feed) of approximately 4.8 kg P cap^{-1} yr^{-1} for food production. This P freight is calculated by taking into account the total imports of P to *Animal husbandry* (4.5 kg P cap^{-1} yr^{-1}) and *Crop farming* (6.2 kg P cap^{-1} yr^{-1}), minus the circulating flows of manure (3.3 kg P cap^{-1} yr^{-1}) and non-marketable feed (2.6 kg P cap^{-1} yr^{-1}). With imported food (0.4 kg P cap^{-1} yr^{-1}) taken into account, the total amount of P used for food production is 5.2 kg P cap^{-1} yr^{-1}. Nonetheless, the P content of the food consumed within Austria is only 1.0 ± 7% kg P cap^{-1} yr^{-1}, while 0.3 kg P cap^{-1} yr^{-1} is exported via food. This means that approximately 24% of the P actually used for food production (marketable feed, mineral fertilizer, organic fertilizers, municipal sewage sludge, seeds and deposition) and of the P imported via food actually reaches consumers, which indicates a considerable lack of efficiency in the food supply chain. Similar findings have been reported at the global scale, with the food chain efficiency estimated at 20–25% (Cordell et al., 2009).

2.4.2 Agricultural soils – fertilization – erosion

Due to eutrophication of surface waters and good P supply in agricultural soils, overfertilization has been reduced in recent years (Figure 1). Nonetheless, agricultural soils still exhibit a relevant stock increase rate of 0.48 ± 61% kg P cap^{-1} yr^{-1}or 1.25 kg P ha^{-1} yr^{-1}, which in turn can represent an increased potential for P loss to *Water bodies*. However, the high uncertainty associated with the path of erosion from agricultural soils makes it very difficult to accurately quantify the real surplus changes in soil P contents, especially on a broad national scale. As outlined by Schröder et al. (2011), there are a number of measures that could be implemented to improve P use in agriculture beyond simply reducing overfertilization, such as land use optimization, precision farming and adjustments to livestock diets. Furthermore, consumers' dietary choices (currently, per capita meat consumption in Austria is approximately 65 kg per year, which is double the meat consumption level recommended on the basis of nutritional science) have a considerable effect on the total P needed for agriculture because meat-based food production requires almost three times the amount of phosphate fertilizer required by vegetable-based food production (Thaler et al., 2011 and Cordell et al., 2009).

2.4.3 Chemical Industry – detergents

The *Chemical industry* plays a minor role in the national P balance (Figure 3). Approximately 60% of the P imported through chemicals is in turn exported. Furthermore, the P consumption through detergents in Austria is lower than the European average of 0.15 kg P cap^{-1} yr^{-1}because of the implementation of EU regulations and to different consumption patterns of the populations (Ott and Rechberger, 2012).

2.4.4 P recovery potential from municipal wastewater

Municipal wastewater treatment plants in Austria generally have high P removal rates because P removal of more than 80% is required for all plants designed for a wastewater load of more than 1000 (population equivalent or PE). As a consequence, the balance shows that municipal sources (wastewater effluent, combined sewer overflow, etc.) are responsible for 30% of the total P load into *Water bodies*. These results are consistent with the results of the daNUbs project, which indicate that 23% of the total P load comes from point sources, while most of the input is caused by diffuse sources such as erosion, groundwater and surface runoff from the land (Kroiss et al., 2005). The findings on diffuse and point sources correspond well to the results of surveys of Central European countries such as Switzerland (Binder et al., 2009) and Germany (Gethke-Albinus, 2012). However, further improving P removal rates from 85% to 95% could reduce the total P emissions by up to 12%, simultaneously increasing the P recovery potential of municipal sewage sludge (MSS). The balance shows that the P content of MSS corresponds to almost 13% of the total input of P to agriculture (6.2 kg P cap^{-1} yr^{-1}) or approximately 40% of the annually applied mineral P fertilizers. The results of this study thus confirm the great potential of municipal sludge with respect to the recovery of P, in accordance with the efforts of researchers worldwide to develop new recycling technologies. In this regard, it is interesting to analyze the characteristics of the WWTP in Austria with respect to their size, design capacity and distribution. As Table 2 shows, more than 55% of municipal wastewater is treated in 30 plants with design capacities ≥100,000 PE. Consequently, more than 55% of the total sewage sludge annually generated in Austria (265,000 t dry matter) is produced by just 30 of the existing 1837 WWTP (BMLFUW, 2012). This reveals an intense concentration of wastewater that could help implementing recovery technologies.

Table 2: Size distribution, design capacity and amount of occurring sewage sludge in Austria's *Wastewater management* sector.

Size category [PE_{COD120}]	Number of WWTP	actual capacity [PE_{COD120}]	percentual on actual capacity [%]	sewage sludge [t DS yr^{-1}]	Phosphorus [kg P $cap^{-1}yr^{-1}$]
51-1,999	1,206	308,756	2.4	6,253	0.02
2,000-10,000	371	1,074,214	8.2	21,754	0.06
10,001-50,000	200	3,108,011	23.8	63,941	0.02
50,001-100,000	31	1,285,190	9.8	26,027	0.30
>100,000	30	7,309,374	55.9	148,025	0.35
total	1,837	13,085,544	100	265,000	0.75

PE, Population equivalent; COD120, Chemical oxygen demand in mg/l (Daily calculation load per PE: 120 g); WWTP, Wastewater Treatment Plant; t DS yr^{-1}, Tons of dry substance per year.

Currently, only 27% of MSS is directly or indirectly (e.g., as compost or biogas slurry) applied to agricultural fields. Roughly the same amount of MSS is mono-incinerated. The arising fly ash shows high P-contents in the range of raw phosphate ore (8–10%), which is suitable for the application of new phosphorus recycling technologies. These results, particularly concerning the indirect application and mono-incineration of MSS, are only obtainable by having a detailed balance and deep insight into current treatment options.

The main reason that such a small fraction of MSS is recycled is the generally low acceptance of direct MSS application, due to the potentially hazardous contents such as heavy metals and persistent organic pollutants (POPs). In addition, the heterogeneity of regulations concerning direct MSS application among the different regions of Austria contribute to this situation, reflecting a lack of consistent and standardized regulation on both the national and international scales. For instance, in Switzerland and the Netherlands, MSS application in agriculture is prohibited, while in Spain, all MSS is directly applied in agriculture (Eurostat, 2012).

2.4.5 P recovery potential from animal waste

Another flow that shows great potential for P recovery is meat and bone meal obtained through rendering of animal waste. For this material, however, the current recycling rate is less than 3%, which is even lower than that for MSS. Mono-incineration of meat and bone meal is an option for avoiding dispersal or loss of P and offers the possibility of future recovery from separately landfilled fly ash. MSS and the total meat and bone meal output from rendering (0.75 and 0.65 kg P cap^{-1} yr^{-1}, respectively) together have a substitution potential of approximately 70% of the annually applied mineral P fertilizers (2.0 kg P cap^{-1} yr^{-1}). Taking into account the fertilizer sales in 2010 (1.5 kg P cap^{-1} yr^{-1}), almost all of the current mineral fertilizer used could be substituted.

2.4.6 P recovery potential from miscellaneous organic wastes

Separately collected municipal and industrial organic wastes (F6.3, F6.5 and F5.14) have moderate recycling potential of 0.42 kg P cap^{-1} yr^{-1}, corresponding to half of the P load of MSS. Due to biological treatment such as composting and biogas production, most of the P potential is already used as compost in agriculture and as substrate in landscaping. Residual waste, in contrast, carries a low P load (but high concentrations of, e.g., heavy metals) and should not be targeted as a priority source for future recycling of P.

2.4.7 P stocks and sinks in the Austrian balance

The outcomes of the study show that Austria is building up a considerable P stock, increasing at an annual rate of 2.1 ± 5% kg P cap^{-1} yr^{-1}. Similar results have been reported for Switzerland (1.6 kg P cap^{-1} yr^{-1}), while the annual rates of P stock increase in France and the Netherlands are more than twice as high, at 4.6 and 4.8 kg P cap^{-1} yr^{-1}, respectively, according to estimates based on analyses of these countries' P budgets (Senthilkumar et al., 2012, Smit et al., 1955 and Binder et al., 2009). The *Waste management* process experiences a total input of 2.5 ± 4% kg P cap^{-1} $yr^{-1}$and an annual stock growth of 1.1 ± 5% kg P cap^{-1} yr^{-1}, which reflects the importance of this sector in the Austrian P budget. The main reason for the high annual rate of stock growth in the *Waste management* process is the incineration of P-rich flows, such as sewage sludge and meat and bone meal, with final disposal of the ashes generated in landfills and cement kilns. Similar estimates have been obtained from the analysis of the Swiss P budget: the *Waste management* sector experiences an input of 1.7 kg P cap^{-1} $yr^{-1}$and a stock increase of 1.1 kg P cap^{-1} yr^{-1} (per capita calculation from results published by Binder et al., 2009). In contrast to the expected outcome of P accumulation in agricultural soils, a quite surprising result is the considerable stock increase rate in the *Consumption* process (0.24 ± 43% kg P cap^{-1} yr^{-1}). A possible interpretation is that the large diffusion of home composting and application of biomass ash from wood heating is provoking over fertilization in gardening, creating a soil P imbalance. If this were the case, then it would most likely be more appropriate to collect green wastes and compost in centralized facilities and redistribute fertilizers for garden use in only the needed amounts. However, there is a high level of uncertainty associated with this estimate, requiring more accurate and specific studies.

One of the goals of sustainable economies is to close regional material cycles fairly. A good example is the agricultural sector, which exhibits internally circulating flows, given the exchanges of animal feed and manure between *Crop farming* and *Animal husbandry*. Nonetheless, P management in the agricultural sector can still be greatly improved to reduce losses caused by erosion, through implementation of better practices of cultivation and fertilization. In contrast, the current systems of food and animal feed processing and consumption and the final treatment of liquid and solid wastes are not designed for conservative handling of P resources, and this strengthens the evidence that natural nutrient cycles are apparently interrupted. This balance also illustrates the great potential for P recovery and possible starting points for significant improvements in P recovery and reduction of P losses. The basis for this assessment is the detailed evaluation of generated wastes and their treatment in the *Wastewater*- and *Waste management* sectors. The results reflect the very inefficient way in which P is currently managed in the *Waste Management* sector: nearly half of it is landfilled or lost in cement kilns. Currently, the focus is on safe disposal of wastes. The balance reflects almost all of the pathways followed by all types of wastes containing P and confirms and quantifies the high potential for P recovery offered by sewage sludge, meat and bone meal and other green wastes.

The results of this study will be the basis for the development of concepts and scenarios for future improvement options for P management, with a particular focus on recycling P from wastewater. The national P balance detailed in this study can be used to demonstrate the potential results of implementing various P recycling technologies. Furthermore, this study will serve as a preliminary basis for the development of a quasi-dynamic model for the P budget of Austria, to investigate changes overtime and the dynamics and drivers of the system.

3. Overview and description of technologies for recovering phosphorus from municipal wastewater

Wastewater

Sewage sludge

Sewage sludge ash

Recovered P product

3.1 Introduction

Given the important role of phosphorus (P) containing mineral fertilizers in the total global supply of P (~80%; Prud'Homme, 2010), future demand will clearly be driven by developments in the agricultural sector. Agriculture's demand for P will primarily be affected by population growth and changes in diet in part due to rising living standards in emerging and developing countries (Metson et al., 2012 and Heffer and Prud'Homme, 2011). Countries lacking P deposits are entirely dependent on imports and are vulnerable to market fluctuations in fertilizer prices to ensure agricultural production and food security. The availability of the resource P is dynamic and dependent on price and technology (Scholz and Wellmer, 2013). National P balances demonstrate that European countries with enhanced wastewater collection and treatment (biological carbon removal and P removal, see Section 3.2.2) possess a large but often exploited and inefficiently used potential source of P in waste streams, especially in municipal wastewater of ∼1 kg P cap^{-1} yr^{-1} (Cordell et al., 2011, Egle et al., 2014a, Gethke-Albinus, 2012 and Binder et al., 2009). However, globally human extractions are a very small part of the global anthropogenic P flows. Considering P losses and efficiency, proper manure management is certainly at least as important as sewage management (Scholz et al., 2014). In some countries, the imported P with feedstuff even submerges P in sewage. Direct agricultural application of wastewater (still practiced in many parts of the world) and sewage sludge is the simplest method of P recycling, although the plant availability of sewage sludge P is debated (Kahiluoto et al., 2015 and Krogstad et al., 2005). Due to potential environmental and health risks primarily from heavy metals (HM), persistent organic pollutants (POPs), and pathogens, acceptance of direct sludge applications is low or decreasing in many European countries (Ott and Rechberger, 2012). Consequently, alternative disposal methods focus on co-incineration (cement kilns, power plants or municipal solid waste incinerators) where P is the irretrievably lost.

Potential methods of P recovery from wastewater consist of direct the separate collection of urine, secondary treated effluent from wastewater treatment plants (WWTP), digester supernatant, sewage sludge (SS) and sewage sludge ash (SSA) (Montag, 2008). These flows differ widely in terms of volume, P concentration, the form of P (dissolved as orthophosphate or biologically/chemically bound), the characteristic of the source (liquid, liquid/solid, solid), pollutant content (HM, POPs, pathogens) and the theoretical recovery potential (Table 3). An ideal approach would feature a high P recovery rate, economic efficiency, and a useful product with low environmental risks. Currently, well-developed and large-scale approaches differ appreciably in terms of these criteria. This article focuses exclusively on approaches for recovering P from municipal wastewater streams. The P recovery approaches address WWTP with strict European standards in P removal for landlocked countries (EC, 1998) and thermal sludge treatment options, namely fluidized bed reactors, which are state of the art in Europe. The general procedures of P recovery approaches have been published frequently, but the important details are frequently lacking (Montag et al., 2011).

Table 3: Characteristics of the potential flows for P recovery (Europe countries, Montag, 2008; modified).

Source	Mass flow	P concentration/ PO_4-P concentration	P specifics	P recovery potential
Untreated wastewater	200 L $cap^{-1}d^{-1}$	~10 mg P L^{-1}	bound/dissolved	100%
(1) Urine	~1.5–2 L $cap^{-1}d^{-1}$	~150–250 mg PO_4-P L^{-1}	dissolved	30–50%
(2) Secondary treated effluent	200 L $cap^{-1}d^{-1}$	~5–10 mg PO_4-P L^{-1}	dissolved	50–70%
(3.1) Digested sewage sludge (SS) (~3.5% TS*)	1.6 kg $cap^{-1}d^{-1}$	dissolved part: 20–400 PO4-P mg L-1	partly dissolved (10–30%)	10–30%
(3.2) Digester supernatant	1–10 L $cap^{-1}d^{-1}$	20–400 PO_4-P mg L^{-1}	dissolved	10–30%
(4.1) Digested sewage sludge (~3.5% TS*)	1.6 kg $cap^{-1}d^{-1}$	1.4 g P kg^{-1} sludge	bound (bio/chem); partly dissolved	90%
(4.2) SS thickened (10% TS*)	0.6 kg $cap^{-1}d^{-1}$	4 g P kg^{-1} sludge	bound (bio/chem)	90%
(4.3) SS dewatered (30% TS*)	0.2 kg $cap^{-1}d^{-1}$	12 g P kg^{-1} sludge	bound (bio/chem)	90%
(5) Sewage sludge ash	0.03 kg $cap^{-1}d^{-1}$	50–130 g P kg^{-1} TS	bound (chem)	~90%

*TS (Total Solids)

Some approaches have received more attention than others have in the past and as such, we have varying degrees of knowledge about them. P recovery by precipitation from sources of dissolved P (orthophosphate) has been investigated in detail (Muster et al., 2013, Rahman et al., 2014 and Doyle and Parsons, 2002). Therefore, optimum process parameters, resource demands, effects on WWTPs and characteristics of the products are well known. To recover P from sewage sludge, various sludge treatment options such as anaerobic treatment, thermal hydrolysis, (wet-) oxidation or wet-chemical leaching are necessary as a first step to dissolve P. The behavior of P and process inhibiting ions (Fe, Al, heavy metals) has been well studied and extensively described (Section 3.4). This knowledge is fundamental for taking further steps in pollutants removal and final P recovery. The same applies to procedural challenges, practicability, waste flows, and possible effects on the functioning of WWTPs. For metallurgic approaches, there is a lack of reliable data regarding mass balances and the fate of heavy metals within the process, and only the results of a few trials are available (Ingitech, 2009). Surprisingly, the current literature on P recovery from ash primarily describes approaches with little realistic potential for prospective practical application (Petzet et al., 2012, Donatello et al., 2010, Franz, 2008 and Levlin, 2001). In contrast, SSA is already used to create recycled products using industrial processes (e.g., ICL Fertilizers®: fertilizer industry; Thermphos®: production of P_4; EcoPhos®: production of phosphoric acid or animal feed). Numerous approaches have been developed in universities and private companies currently operating at pilot scale or full-scale, but details are not yet widely published. In general, many approaches for recovering P from ash are similar to those for treating raw phosphate ore (Nielsson, 1989). This article provides an overview on known approaches looking in detail at those with potential for full-scale implementation or which are already implemented. These approaches are characterized in the context of P removal from wastewater and at other access points of recovery (Figure 5). Thus, a first criterion is the characterization of the optional P flows, thereby outlining the challenges for recovery technologies. Based on this knowledge, our review serves as a database for further integrated, comprehensive and comparative technical, environmental and economic assessments. The following items were investigated:

- Fundamentals of process engineering (e.g., process steps).
- Possible technical challenges.
- Quantification of resource demands (e.g., chemicals, energy).
- Effects on WWTP and resulting waste products.
- Illustration of the fate of potentially harmful substances.
- Characteristics of the final product (e.g., chemical compound, plant availability, heavy metals (Table A 7)).
- P recovery potential of a technology (1) related to the input flow (e.g., sewage sludge) and (2) recovery potential in relation to the WWTP influent.

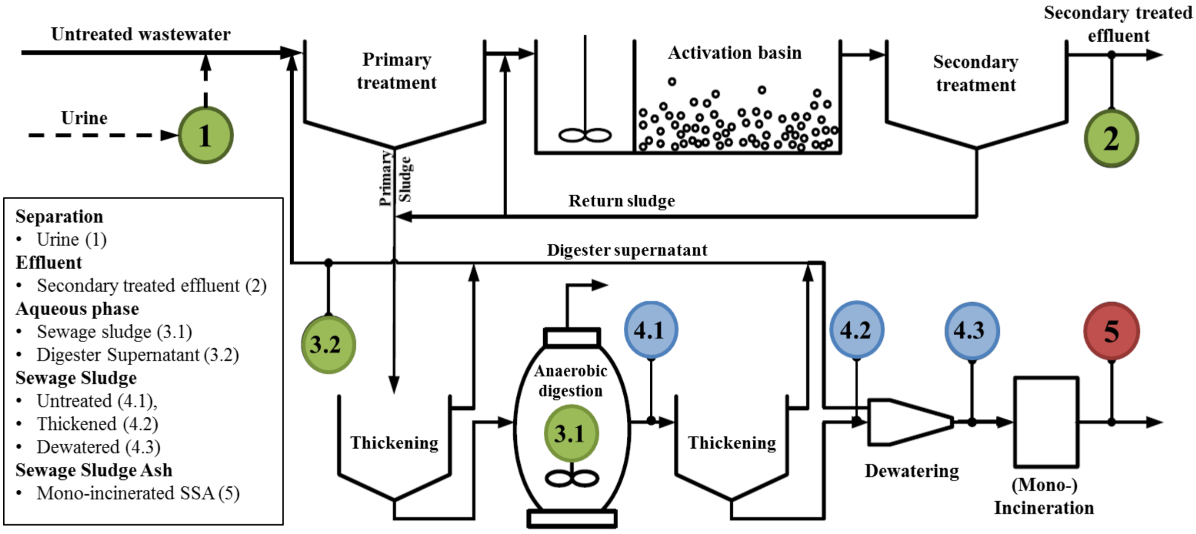


Figure 5: Various possible access points for P recovery approaches during wastewater and sewage sludge treatment or before/after incineration (Montag, 2008; sketch modified).

In addition to the literature review, the database contains information from interviews of plant operators, researchers, commercial companies, and by visiting existing plants. For selected technologies detailed material flow models are provided, generated with the Software STAN (Cencic and Rechberger, 2008). These models are the basis for material flow analysis (MFA; Brunner and Rechberger, 2004). Utilizing MFA input- and output flows will be balanced for a technology. Furthermore, MFA is an appropriate methodology to track the path of P and pollutants for assessing P recovery and depollution potential.

The recovery technologies are summarized in the Appendix and present their status of implementation, the technological approach, the final product and the efficiency with regard to the WWTP influent (Table A 4, Table A 5, Table A 6 and Table A 7). A comparison of costs is not part of this work, as a fair comparison requires a standardized initial situation (reference system) and technologies in a similar development stage. An attempt for economic assessment however, should be an essential part of future work.

3.2 P in wastewater and its removal and recovery

3.2.1 P in wastewater

P enters the wastewater stream primarily in the form of excreted human metabolic products (urine, feces), food residues, and industrial source emissions. Additional to these P sources, detergents are a source of P in European countries. Therefore, a typical daily P load in municipal influenced wastewater in Europe is 1.5–2 g cap^{-1} (Henze et al., 2002, Zessner and Lindtner, 2005 and Richards et al., 2015). P is present in many suspended and dissolved, inorganic, and organic compounds (Baumann, 2003).

3.2.2 P removal from wastewater

P compounds can be removed from wastewater by biological and/or chemical processes to avoid eutrophication of water bodies (Jarvie et al., 2006). Through biomass growth, approximately 30–40% of P is already eliminated by conventional biological uptake (Levin and Shapiro, 1965). P accumulating organisms (PAO) are capable to store ≥5% (other bacteria 1–2%) of their dry weight in P under specific process configurations (enhanced biological P removal; EBPR or Bio-P). Under specific sludge treatment conditions, biologically bound P is dissolvable and thus is easily recoverable (Section 3.3.3).

To guarantee strict effluent limit values in landlocked countries and sensitive areas year-round, chemical precipitation of P by adding Fe or Al is a common method in Europe. The advantage of precipitation is that P removal is not dependent on microorganisms, which can be sensitive to inhibiting substances in wastewater or temperature. With biological and/or chemical P removal up to 95% of the influent P is transferred to sewage sludge. However, chemical precipitation is associated with a lower recovery potential from the aqueous phase and occasionally from sewage sludge, due to the low dissolution of metal-phosphate compounds. Also for recovery from SSA, metal-phosphate compounds seem to be problematic (Section 3.5.2).

To date, the primary purpose of a wastewater treatment is the reliable removal of organics and nutrients to ensure human and environmental health. Thus, any P (or other resource) recovery technologies must not have negative effects on treatment processes that ensure such objectives are met.

3.2.3 Access points for P recovery in the wastewater treatment process

P recovery is possible directly from separated waste flows such as urine, secondary treated effluent and digester supernatant. Sewage sludge and sewage sludge ash are also relevant mass flows for P recovery. Figure 5 shows the different access points for recovery technologies. Subsequently, the characteristics of these potential flows for P recovery are presented in Table 4 and show the challenges for technologies depending on the source. Additionally, an overview on the theoretical P recovery potential is presented.

3.2.4 Principles of P recovery by formation of low soluble salts

Precipitation of dissolved P in the form of different salts with low water solubility is a common step in numerous recovery technologies, thus the principles are presented in this section to avoid repetition within this work. By adding appropriate precipitants, typically magnesium (Mg^{2+}) or calcium (Ca^{2+}) dissolved P and also available NH_4^+ can be recovered. Magnesium phosphates are precipitated primarily as magnesium-ammonium-phosphate (Struvite), whereas for calcium phosphates (CaP) the range of possible compounds is wider. Typical forms are hydroxyapatite (HAP), hydroxydicalcium phosphate (HDP) but also amorphous calcium phosphate (ACP), brushite or octacalcium phosphate (Angel, 1999 and Maurer et al., 1999) (Eq 1, 2 and 3).

$$\text{(Eq 1) Struvite: } Mg^{2+} + NH_4^+ + HPO_4^{2-} + 6H_2O \rightarrow MgNH_4PO_4 + 6H_2O + H^+$$

$$\text{(Eq 2) HAP: } 5Ca^{2+} + 3HPO_4^{2-} + 4OH^- \rightarrow Ca_5(PO_4)_3(OH) + 3H_2O$$

$$\text{(Eq 3) HDP: } 2Ca^{2+} + HPO_4^{2-} + 2OH^- \rightarrow Ca_2HPO_4(OH)_2$$

Common precipitants are alkaline substances (MgO, $MgOH_2$, CaO, $Ca(OH)_2$) and various salts ($MgCl_2$, $CaCl_2$) (Hug and Udert, 2013). Key factors for P precipitation are in order of importance the (1) molar ratio of P and precipitation agent, (2) concentration of ions, (3) pH and (4) temperature (Muster et al., 2013). In addition, the presence of suspended solids, COD content, and recirculation flow rate affects precipitation. The required molar Mg:N:P ratio for Struvite precipitation is 1:1:1 and the required Ca:P ratio for HAP precipitation is 1.7:1. To initiate precipitation, higher Mg or Ca ratios are required. A slightly alkaline pH of 7.5–9 is essential and preferably above pH 8.5 (Rahman et al., 2014). Caustic soda (NaOH) is commonly applied for pH adjustment, but stripping of CO_2 by simple aeration of wastewater has also proven to be feasible (Fattah et al., 2010). With higher PO_4-P and/or precipitant concentrations, precipitation takes place at a lower pH resulting in lower process costs (Wilsenach et al., 2007). Generally, the precipitants are added hyperstochiometrically at an excess of 5–50% (Doyle and Parsons, 2002). The formation of desired salts occurs when the concentrations of the precipitants and P ions exceed their solubility product at a certain pH. Under optimal conditions, P recovery using this process is as much as 98–99.9% with reaction times of less than 1 h (Rahman et al., 2014). With longer retention time under appropriate conditions and the use of fluidized bed reactors, larger particles or even small pellets can be formed (crystallization process; Le Corre et al., 2007 and Mullin, 1993). The struvite that precipitates in other reactors is a wet, sometimes brownish or grayish slurry, that needs further processing, such as dewatering, pelletizing, and drying, to create a commercial product that meets market specifications. Struvite and different forms of CaP are accepted slow-release fertilizers that exhibit plant uptake within one growing period (Römer, 2013).

3.3 Detailed characterization of P recovery approaches

3.3.1 Urine separation

For nutrient recovery from urine, so-called NoMix installations or urine separation toilets are installed to avoid dilution and fecal contamination (Vinnerås and Jönsson, 2002). Urine contains approximately 50% of the total P and 80% of the total N of household wastewater, whereas its mass flow is less than 1% (valid for European combined sewer systems: 1.5–2 L cap^{-1} d^{-1}) (Larsen and Gujer, 1996). Urine recovery in NoMix toilets is usually in the range of 30–50% (Zessner and Lindtner, 2005). This percentage limits the P recovery potential. The dissolved P concentration is about 200 ± 50 mg PO_4-P L^{-1} (Grau et al., 2012 and Etter et al., 2011).

Heavy metals and pathogens play a minor role, whereas organic pollutants such as pharmaceuticals and hormones are present in urine (Escher et al., 2006). The recovery of P and N is technologically simple and is achieved via Struvite/CaP precipitation (Section 3.2.4). With respect to possible pollutants in urine, Ronteltap et al. (2007) reported that common pharmaceuticals (e.g., Propranolol, Ibuprofen, Diclofenac and Carbamazepine) transfer into the precipitated products in only very small quantities i.e., at rates ranging from 0.01% (Diclofenac) to 2.6% (Propranolol) in the product vs. their amounts in urine. Heavy metals behave differently as 20% (As) to 63% (Pb) of their initial amounts are present in the final product (Ronteltap et al., 2007), but in general the heavy metal content already in urine is very low (Ronteltap, 2009 and Jönsson et al., 1997). No pathogens can be detected in the final crystalline products. Pilot or even full-scale plants are implemented e.g., at airport Schipol (De Graaff et al., 2014) and by SaNiPhos® (GMB, 2010).

3.3.2 Secondary treated effluent

To recover P from secondary treated effluent to a great extent, pre-condition is the abstinence of P elimination during wastewater treatment. As such, the recovery process simultaneously acts as a final P barrier. The hydraulic flows and accordingly the P concentrations vary greatly depending on the time of day and weather. Heavy metals, pathogens, and POPs are also present in the effluent. Because the P concentration is low (~5–10 mg PO_4-P L^{-1}) and the volumetric flow is high (~200 L cap^{-1} d^{-1}) preceding enrichment is essential. By installing selective ion exchangers, dissolved nutrients such as HPO_4^{2-}, NH_4^+ and K^+ are concentrated in the ion exchanger regeneration brine (Liberti et al., 2001). The **REM-NUT®** technology (Liberti et al., 1986a, Liberti et al., 1986b) works using two ion exchange units (cationic and anionic) housing a natural zeolite or a strong alkaline resin (Figure A 1). As much as 90% of P and N is removed from wastewater. Liberti et al. (2001) demonstrated that in addition to nutrients, due to the poor selectivity of the PO_4^{3-} anion, other components of the wastewater are adsorbed, including suspended solids, HM, POPs (>65%) and microorganisms (~90%). These components partly adsorb irreversibly, leading to a decrease in P recovery and the need to replace the ion exchange material. Alternative selective adsorbents e.g., a hybrid anion exchanger (HAIX; Blaney et al., 2007) or polymeric ion exchangers (Petruzzelli et al., 2003, Sengupta, 2001 and Zhao and Sengupta, 1998) have been developed to reduce these shortcomings.

P is finally recovered from the brine by precipitation in a nearly sterile struvite with low pollutant content (Liberti et al., 2001). Currently, no large-scale implementation is known.

Taking into account a reduced recovery rate of ion-exchangers over time, recovery potential related to WWTP influent is ~50 to 70%.

3.3.3 Aqueous phase (anaerobic sludge and digester supernatant)

For only biologically removed P (Bio-P) the P release during anaerobic digestion is 15–23% of the total P of sludge (Ewert, 2009), leading to PO_4-P concentrations in anaerobic sludge of 200–400 mg L^{-1}. With additional preceding sludge treatment and disintegration methods as acidification (Section 3.4.1), thermal treatment (Section 3.4.2), and oxidation (Section 3.4.3) of even higher concentrations above 1.000 mg L^{-1} are possible, thereby affecting the recovery potential and economic feasibility (Adnan, 2002). The recovery potential depends strongly on the type of P removal (biological vs. chemical) from the wastewater. However, high levels of dissolved P can cause undesirable incrustation, and cause a significant reduction of the dewatering efficiency due to strongly hydrophilic colloid systems and an increase of nutrient backflow.

3.3.3.1 Dissolved fraction from anaerobic sludge

With the patented **AirPrex**® approach, anaerobic sludge is aerated in a separate tank, thereby leading to an increase in pH to 8 due to stripping of CO_2 (Heinzmann and Engel, 2006). Adding precipitants such as $MgCl_2$ (Mg:P = 1.5:1), 80–90% of the former dissolved P is crystallized in the sludge, primarily as struvite, and settles as a result of its higher density than the sludge (struvite: 1.7 g cm^{-3}) on the bottom of the tank where it can be harvested (Figure A 2). After a cleaning step, the struvite is sold as a trademarked fertilizer (*Berliner Pflanze*®; Heinzmann, 2009). The heavy metal content of the final product is low and no pathogens are present (Lengemann, 2012). The recovery potential depends on the PO_4-P concentration and harvesting efficiency, but is only 7–22% with respect to the WWTP influent. In combination with sludge disintegration methods as e.g., CAMBI® significant higher P recovery is possible (Kabbe, 2015). The **AirPrex**® approach is primarily a technology to improve operation and dewatering properties, and P recovery is a side benefit. Several large-scale implementations in Germany and the Netherlands are known (e.g., Berlin Waßmannsdorf, Mönchengladbach, and Amsterdam). Petzet and Cornel (2012) demonstrated that CSH (calcium-silicate-hydrate), known from the **P-RoC®** approach (Section 3.3.3.2), is also appropriate for fixing dissolved P in anaerobic sludge (**FIX-Phos**).

3.3.3.2 Digester supernatant

Another possibility is the recovery of P from the digester supernatant, which is the liquid and nutrient rich flow after thickening and/or dewatering of sewage sludge. An advantage of P recovery from this stream is a constant flow rate (1–10 L cap^{-1} d^{-1}), which simplifies the design of the recovery components. Little procedural effort is needed for recovery as the P is already present as PO_4-P. Heavy metals and POPs are not a major concern. During normal operation, high concentrations of dissolved P in the digester supernatant are undesirable due to incrustations (e.g., formation of struvite) in pipes, pumps and dewatering units (Doyle and Parsons, 2002 and Jaffer et al., 2002). Consequences of such incrustations include high maintenance costs or even the breakdown of plant components (Heinzmann and Engel, 2003). Therefore, the primary motivation driving controlled P precipitation from digester supernatant is to avoid these incrustations and to minimize the nutrient content of the reject water. The recovery of P is a side benefit. Crystallization in cylindrical reactors, some with the diameter increasing with height for forming pelletized products, are common approaches producing, in certain cases, a certified commercial fertilizer: **Nishihara Reactor** (Nawamura et al., 2001); **NuReSys**® (NuReSys, 2015); **Phospaq**® (Abma et al., 2010); **Ostara Pearl Reactor**® with *Crystal Green*® (Britton, 2009 and Britton et al., 2007); **Phosnix Unitaka** (Ueno and Fujii, 2003); **Phostrip** (Kaschka and Weyrer, 1999); **REPHOS**® (Lebek and Lohmar, 2013); **STRUVIA™** (Mêlé et al., 2014). Optimal conditions for struvite or CaP formation need to be provided by pH adjustment (commonly by NaOH or CO_2-stripping) and dosing of Mg or Ca. Some process parameters for precipitation (powdery characteristic) and crystallization for gaining pelletized products are listed in Table 4.

Table 4: Variations in process parameters in precipitation and crystallization (Rahman et al., 2014 and Britton et al., 2007).

Approach	pH	molar ratio (Mg:P)	Retention time of precipitated material	Final product	Recovery (%)
Precipitiation	8.5-9.0	1.5:1	1 h	powdery/amorph	90-99
Crystallization	7.5-8.5	1.1:1	several days	pellets	80-95

Commonly, the reactors are partially filled with suitable seeds such as sand (Pietkema, 2004) or already precipitated struvite crystals (Adnan et al., 2003) to facilitate the initial crystallization. Depending on the process parameters and retention time very pure and sterile pellets of various grain sizes (diameters of 1–4 mm), low HM concentrations and very low POP contamination are produced (Britton et al., 2007). After drying (atmospheric or thermal) and classification, a commercial fertilizer is obtained. No waste occurs with these processes.

A method using a product of the cement industry called calcium-silicate-hydrate (CSH) as the crystallization seed is referred to **P-RoC®** (Ehbrecht et al., 2011 and Berg et al., 2006, Figure 6). This Ca-rich material is added to the digester supernatant at an optimal solid/solution ratio of approximately 5 wt.%. Hydroxyl ions are released, thereby increasing the pH (8.5–9.5) and creating ideal condition for crystallization of PO_4^{3-} with Ca^{2+} ions at the CSH surface (Okano et al., 2013). The residence time of the digester supernatant is approximately 1–2 h and the final product is a P rich CSH (Berg et al., 2007 and Berg and Schaum, 2005). The recovery potential from digester supernatant with respect to the WWTP influent is 10–25%.

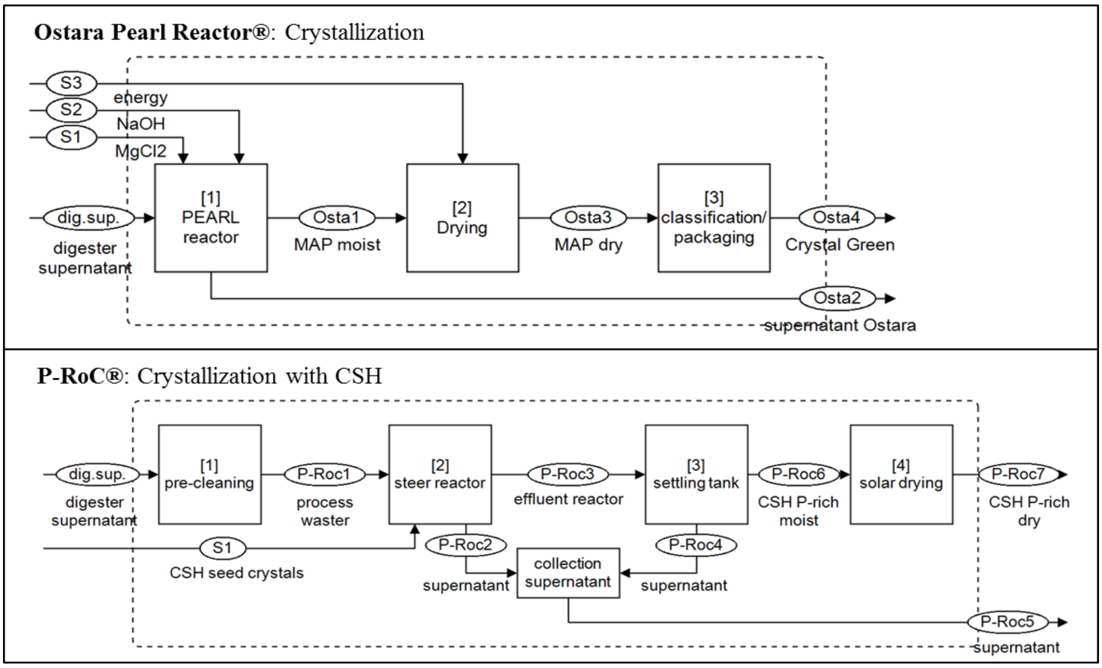


Figure 6: Material flow models for technologies to recover P from the digester supernatant (above: Ostara Pearl Reactor®, below: P-RoC®).

3.4 Sewage sludge (SS)

Sewage sludge is the residue resulting from the wastewater treatment process. This material is a heterogeneous, semi-solid material containing essential plant nutrients (P, N, Ca, Mg, and K) among other constituents. However, sewage sludge also contains heavy metals (Kupper, 2000 and Zessner, 1999), POPs as e.g., pharmaceuticals, and endocrine disrupting compounds (Roig et al., 2012, Kupper et al., 2004, Peysson and Vulliet, 2013, McClellan and Halden, 2010 and Yu and Wu, 2012) and pathogens (Sahlström et al., 2004). Thus, its acceptance as an agricultural fertilizer is often low and its application to fields is partially restricted or even prohibited in certain countries (e.g., Switzerland, the Netherlands). P concentration and the mass flow are dependent on P removal efficiency, its thickening and level of dewatering (Table 3). Depending on the removal steps, P is biologically and/or chemically bound. As such complex procedural and resource-intense P recovery approaches have been developed (Figure 7) for the recovery of P from wastewater sludges.

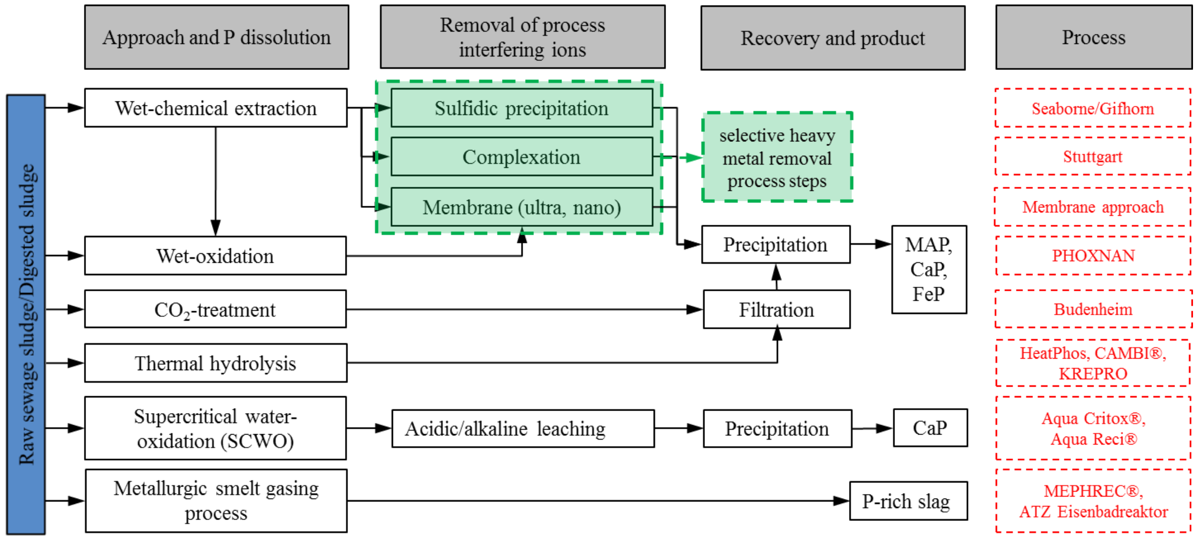


Figure 7: Overview of the approaches to recover P from raw and digested sewage sludge.

3.4.1 Wet-chemical approach

Complex technological approaches are required to recover P from sludge and simultaneously achieve a significant reduction in contaminants. Sludge from anaerobic digestion is preferred over raw sludge in wet-chemical treatment as complex organic compounds are already reduced during digestion. Wet-chemical approaches decrease the pH by adding a strong acid, e.g., sulfuric acid (**Seaborne/Gifhorn process**: Esemen, 2013; Günther et al., 2007 (Figure A 3); **Stuttgart process**: Antakyali et al., 2012; Meyer et al., 2012) (Figure 9). At low pH (<2), more than 80% of the initially bound P is dissolved. However, process interfering ions dissolve simultaneously to various degrees (Figure 8).

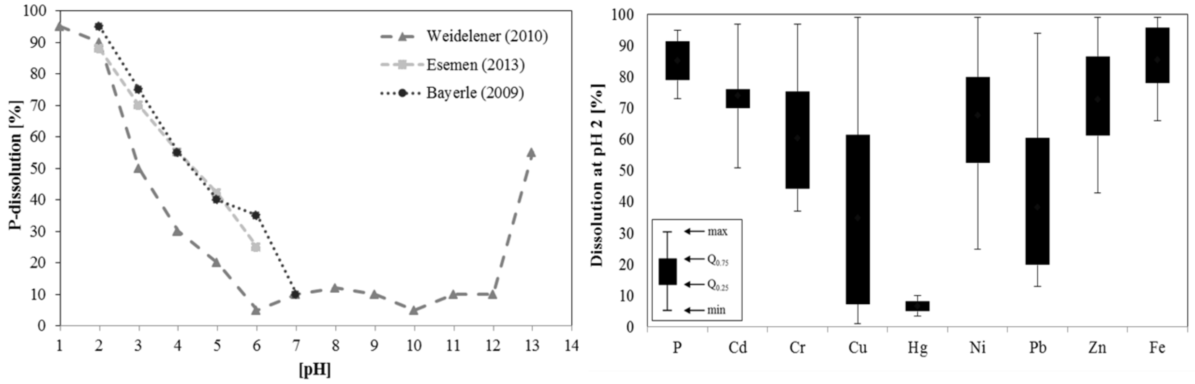


Figure 8: Dissolution [%] of P from digested sewage sludge at various pH values (left) and variability in dissolution of heavy metals at pH 2 (right; Bayerle, 2009; Günther, 2011; Weidelener, 2010; Bouda et al., 2009; Naoum et al., 2001) (first ($Q_{0.25}$) and third quartile ($Q_{0.75}$) and minimum (min)/maximum (max)).

The targeted pH should be 3–4 to reduce the amount of interfering ions. This pH leads, however, to decreased P dissolution (30–75%) and thus lower recovery potential. The acid demand is proportional to the dry matter content of the sludge. Table 3 presents the amount of acid (kg H_2SO_4 [98%] per kg TS) required to treat various sludges (2 to 5% TS) at different pH levels.

Table 5: Comparison of pH, H_2SO_4 (98%) demand [kg kg^{-1} TS] and P dissolution [%] in wet-chemical sewage sludge treatment (Esemen, 2013; Günther, 2011; Weidelener, 2010; Bouda et al., 2009).

pH	mean	min	max	P dissolution [%]
1.5	0.55	0.45	0.65	>90
2	0.50	0.36	0.59	75-90
3	0.35	0.23	0.39	50-75
4	0.27	0.20	0.34	30-55

Following the dissolution step, the **Seaborne approach** continues with the precipitation of dissolved interfering ions, primarily iron, which is achieved by pH increase to ~5.7 using NaOH and Na_2S dosing (~3 L 15% Na_2S per m³ sludge), in advance of the step of solid/liquid separation. This way, potential interfering ions are transferred to the solid sludge phase which is then discarded (Esemen, 2013). Alternatively, the **Stuttgart approach** aims to avoid the simultaneous precipitation of dissolved interfering ions by complexation with citric acid (10–12 L 50% citric acid solution per m³ filtrate) in the digester supernatant (after dewatering) (Weidelener, 2010; Meyer et al., 2012). In both approaches MgO or $Mg(OH)_2$ is added and NaOH is used to increase the pH level for the precipitation of struvite. The supernatant from the **Stuttgart approach**, which is rich in complexed ions, is returned to the WWTP influent, leading to an accumulation of heavy metals in the sewage sludge and/or higher load of heavy metals in the effluent (Egle et al., 2014[c]). Güney et al. (2008) tested a cation-exchange membrane for metal ion removal (Al, Ca, Fe) of the supernatant to minimize contamination in struvite production, although with unsatisfactory results. Consequently, the product quality may not fit legal thresholds.

The **Budenheim process** provides an alternative to the acid leaching processes, due to far lower chemicals consumption at comparable recovery rate (Stössel, 2013) (Figure 9). With CO_2, the sludge is treated at a pH of 4–5. This is an economical decision following the trade-off between recovery rate and effort. Operating at pH 5 provides the advantage of low re-dissolution of interfering ions and thus no specific HM removal.

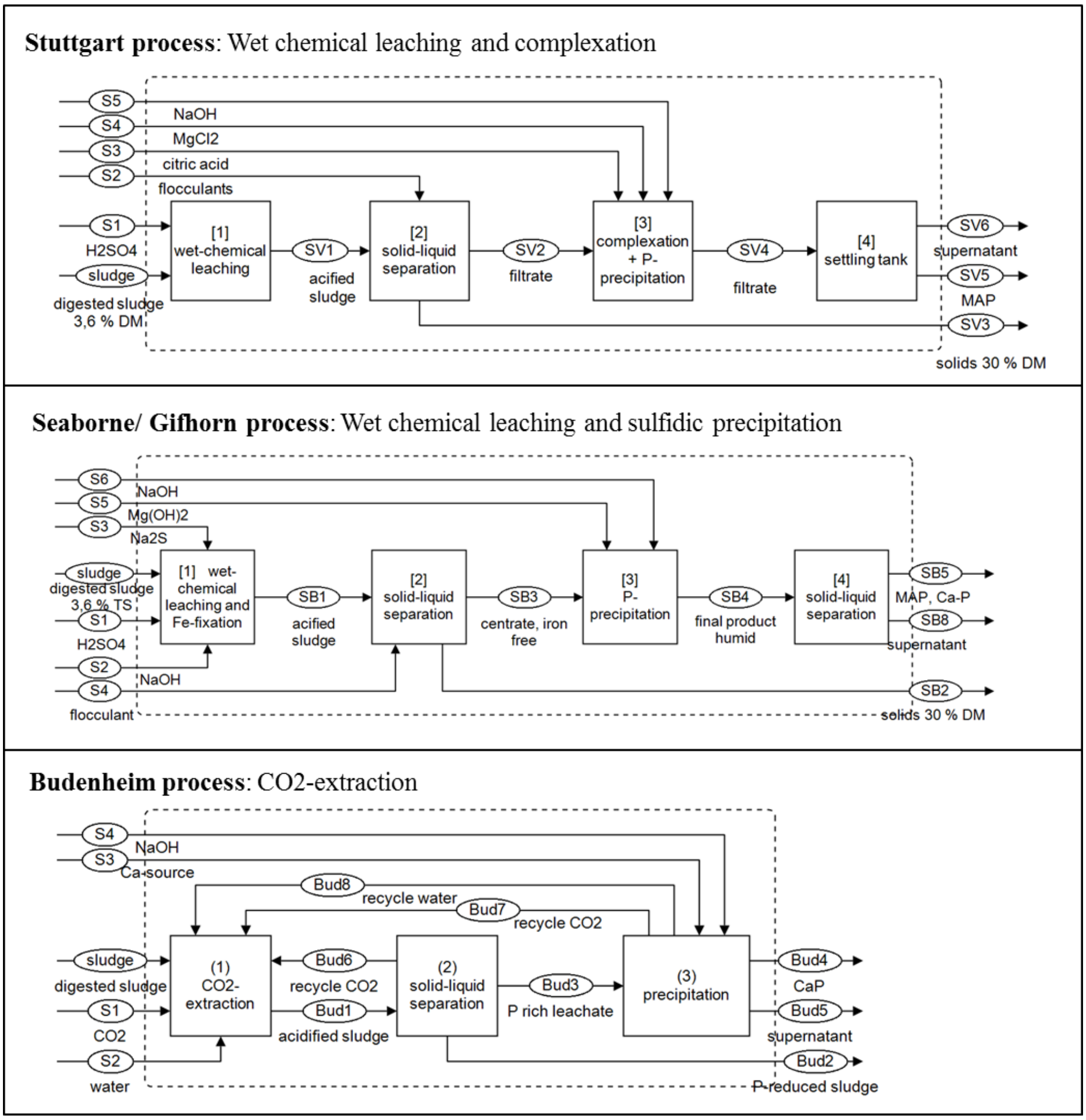


Figure 9: Material flow models for technologies to recover P from the digested sludge by wet-chemical leaching with mineral acids (above: Seaborn/Gifhorn, middle: Stuttgart) or CO_2 (below: Budenheim).

Generally, the final products of these approaches contain low concentrations of heavy metals (Table A 1). POPs in low concentrations cannot be excluded. Pathogens are not detected. The demand for chemical agents varies greatly depending on the sludge characteristics (e.g., water content), and the pH during acid treatment and thus the dissolution rate of P and other ions. The by-product of these technologies, the acidic sludge, and the different procedural steps create a number of challenges (Esemen, 2013; Meyer et al., 2012):

- Foaming during acidification, degasing of CO_2 and even toxic H_2S creates the need for headspace and air filters.
- At pH 2 or 3 common flocculants for dewatering may fail, creating a need for alternative flocculants or a higher pH (=lower P recovery potential).

- An acidic sludge waste product with a high sulfur content (corrosive), which leads to transportation issues and further sludge treatment (e.g., incineration).
- Possible heavy metals enrichment of the activated sludge and WWTP effluent via the recovery of heavy-metal-rich supernatants.
- Iron recovery due to the recirculation of supernatants.

Theoretically, 99.9% of P could be recovered with the Seaborne and Stuttgart approach, but the recovery potential is limited by the cost. A realistic recovery potential is 40–50% with respect to the sewage sludge input. With respect to the WWTP influent, 35–45% can be recovered (Esemen, 2013). Whereas the **Seaborne process** has been modified and implemented full-scale under the name **Gifhorn process**, the **Stuttgart process** has been realized at pilot scale, which is expected for the **Budenheim process.**

3.4.2 Thermal treatment and thermal or chemical hydrolysis

Takiguchi et al. (2003) demonstrated the release of significant amounts of polyphosphate (PolyP) from EBPR sludge by simple heating, i.e. thermal treatment, due to the disintegration of internal cell storage structures and cell wall membranes (**HeatPhos**; Hirota et al., 2010). Nearly the entire PolyP is released at temperatures of 70–90°C within <60 minutes (Kuroda et al., 2001; Takiguchi et al., 2003). The primary advantage of this approach is the recovery of CaP without pH adjustment by adding $CaCl_2$. Thermal hydrolysis focuses on disintegration of dewatered raw sludge (14–18% DS) at higher temperatures (165°C) and higher pressure (6 bar) (Carrère et al., 2010). Under these conditions, water is more reactive and is able to break high to low molecular compounds and P is released after a retention time of 20 minutes (Hii et al., 2014). For example, the **Cambi®** process is performed in a three-step system in which steam ruptures the cells (Bormann and Sievers, 2011; Bormann et al., 2009, Sievers et al., 2005). The biologically bound P is dissolved and thus P recovery in combination with known technologies (e.g., AirPrex®; full-scale implemented at Lingen, Uelzen, Salzgitter, Wolfsburg) (Kabbe, 2015) becomes possible. This process and also the chemical hydrolysis approach (PONDUS) is primarily a disintegration technology that is beneficial for enhanced sludge treatment at higher digester loads and is associated with higher biogas yields and improved dewatering properties (Hii et al., 2014). The recovery potential depends on the former biologically bound P (EBPR).

3.4.3 Wet-air oxidation (WAO) and supercritical water oxidation (SCWO)

At temperatures of 160–325°C, pressures of 50–175 bar and a residence time of ~1 h and with the addition of pure oxygen, WAO ensures oxidation of the organic matter in sludge (Debellefontaine and Foussard, 2000) (Figure 10). The carbon reduction is as high as 95% and biologically bound P is released. The recovery potential from an iron-containing sludge is low because the oxidized iron (Fe^{3+}) reacts immediately with released orthophosphate and forms $FePO_4$ (Niewersch, 2013). Due to these conditions, the **PHOXNAN** approach, which is based on a wet-chemical treatment (pH: 2–1.5; H_2SO) as a first step, was developed. Low pH prevents the immediate formation of undesired $FePO_4$. The acidified sludge is fed to a low-pressure wet oxidation tank (1–4 h, 160–200°C, ~20 bar. (LOPROX®). Oxygen is fed at rates depending on the COD load (1 kg O_2 kg COD^{-1}). This process is exothermal, and the surplus heat can be used (Debellefontaine and Foussard, 2000). The residuals (organic content <5%) exhibit good settling properties, whereas the P and other ions are dissolved in the liquid phase. POPs are almost completely destroyed. To recover the P from the acidic liquid phase by precipitation, two filter units, and ultra-filtration followed by nanofiltration are used to separate Fe/Al and heavy metals from P (Blöcher et al., 2012). The concentrates, which are rich in (heavy) metals, require proper treatment. The selectivity of the filtration steps is low and thus the recovery potential with respect to the input sewage sludge is only 55% (Blöcher et al., 2011).

Supercritical water oxidation (SCWO) is based on the use of water in its supercritical state to destroy organic matter (Stenmark, 2003). At temperatures above 375°C and a pressure of 220 bar, water enters the supercritical phase (Stendahl and Jäfverström, 2004). By adding technical oxygen (1 kg O_2 kg COD^{-1}), 99.99% of the organic matter is destroyed within one minute. SCWO, like WAO, is an exothermic process that generates surplus energy that can be recovered. POPs are completely destroyed (Veriansyah and Kim, 2007). The outputs are a liquid phase and an inorganic ash, which contains the majority of the P and heavy metals that are oxidized to their highest valence (Zuh et al., 2011; Stendahl and Jäfverström, 2003). Two approaches, **Aqua Reci®** (Stenmark, 2003) and **Aqua Critox®** (O’Callaghan and O’Regan, 2009), are known to recover P from reactive SCWO ash (Figure 10). To dissolve P from the reactive ash an extraction step is needed in which acid (HCl) or caustic salt (NaOH) is added (Levlin et al., 2004). Up to 90% of the P redissolves at an alkaline leaching, which can be precipitation to gain plant available hydroxyapatite (HAP) (Levlin, 2007). The recovery potential is 50% (**PHOXNAN**) to 70% (**Aqua Reci®**, **Aqua Critox®**) with respect to the WWTP influent. The oxidation technologies have been implemented either on full-scale (WAO, **LOPROX®**) or at pilot scale (SCWO). P recovery associated with these processes has been only tested on the pilot scale so far, however no further development is noticeable.

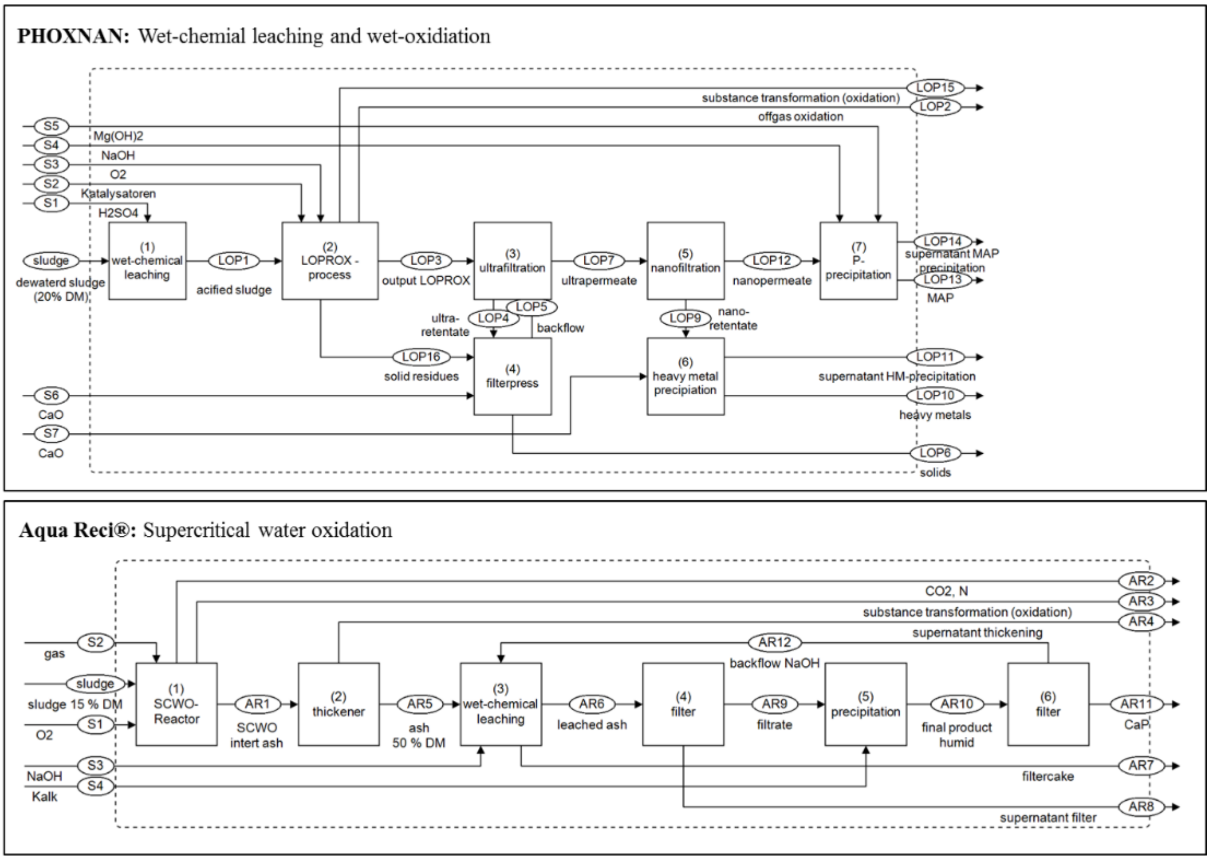


Figure 10: Material flow models for technologies to recover P from sewage sludge by wet-oxidation and supercritical water oxidation (above: PHOXNAN, below: AquaReci®).

3.4.4 Metallurgical smelt-gasification process

Organic-rich matter such as sewage sludge can be considered as a source for renewable biogas production and the generation of a P containing slag by applying the smelt-gasification process. A molten iron bath is produced in a cupola furnace at temperatures above the ash melting point (>1,450°C) (**MEPHREC®**; Scheidig et al., 2013; Ingitech, 2009; Scheidig et al., 2009) (Figure 11). The **ATZ iron bath** method follows a similar approach, but no implementation has been published (Mocker et al., 2010). Metallurgical coke is used as an energy supply and reducing agent. Furthermore, technical oxygen or hot oxygen-rich air is required. The sewage sludge, which has been pressed into cement briquettes is added to the cupola furnace. Under reducing atmospheric conditions all of the organic matter initially degasses. At lower levels in the furnace, mineral components melt and together with other additives (limestone) and coke ash form the P containing slag. The liquid slag is then collected within the lower compartments of the coke bed (porous zone). Due to its higher density, the metal slag accumulates below the P containing slag. This liquid slag is separately tapped and forms fine grained porous granules when released into a water bath. As a pilot plant is under construction in Nürnberg (KRN-Mephrec, 2015), currently no reliable data on the fate of heavy metals are available. However, the product quality will be mainly dependent on the sludge quality. The P containing slag exhibits good plant availability when used on alkaline soils but surprisingly low availability on acidic soils (Römer, 2013). The recovery potential is 60–80% with respect to the input and approximately 50–70% with respect to the WWTP influent.

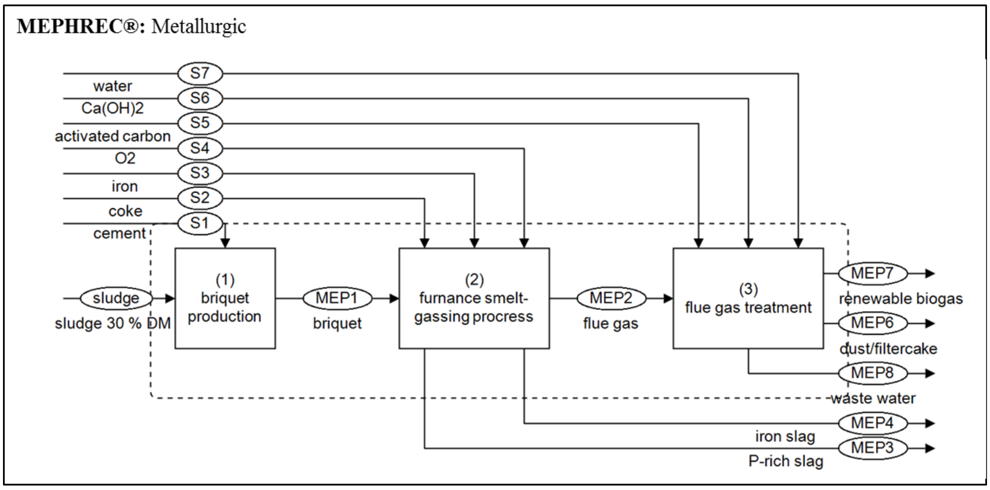


Figure 11: Material flow model for the metallurgic MEPHREC® technology to recover P from sewage sludge.

3.5 Sewage sludge ash (SSA)

Organic matter, including organic pollutants, is nearly entirely oxidized during thermal sludge treatment (800–900°C) (Deng et al., 2009; Park et al., 2008; Werther and Ogada, 1999). 97–99.9% of the P in sewage sludge accumulates in the SSA. Heavy metals, except mercury (which evaporates due to its low boiling point), behave similarly (Lederer and Rechberger, 2010; Van de Velden et al., 2008). The predominant P containing mineral phases in the SSA are low-solubility minerals such as whitlockite (Maier and Scheffknecht, 2007). Occasionally, other minerals such as brushite, stanfieldite, estanite or even natural apatite are present (Gajic, 2011). Furthermore, iron/aluminum phosphates are usually present due to their chemical precipitation during wastewater treatment. Consequently, the P in SSA is not soluble in water and thus has low immediate plant availability (Römer, 2013; Maier and Scheffknecht, 2007). These properties are similar to those of raw phosphate rock, which requires processing. The P concentration of SSA range from 50 to 100 g kg^{-1} TS. In comparison, the typical P concentration of raw phosphates from North Africa and Asia is 130±40 g P kg ore^{-1} (IFDC, 2010; Van Kauwenbergh, 2010).

Dilution of sewage sludge with other wastes low in P or high ash content before incineration should be avoided because low P concentrations increase the cost of recovery (Egle et al., 2014[b]). The preferred incineration technology is the fluidized bed reactor, which produces a powdery ash, thereby simplifying further process operations (Donatello et al., 2010). Because the P in SSA is not water-soluble and the SSA contains heavy metals in high concentrations, several procedures have been developed (Figure 12).

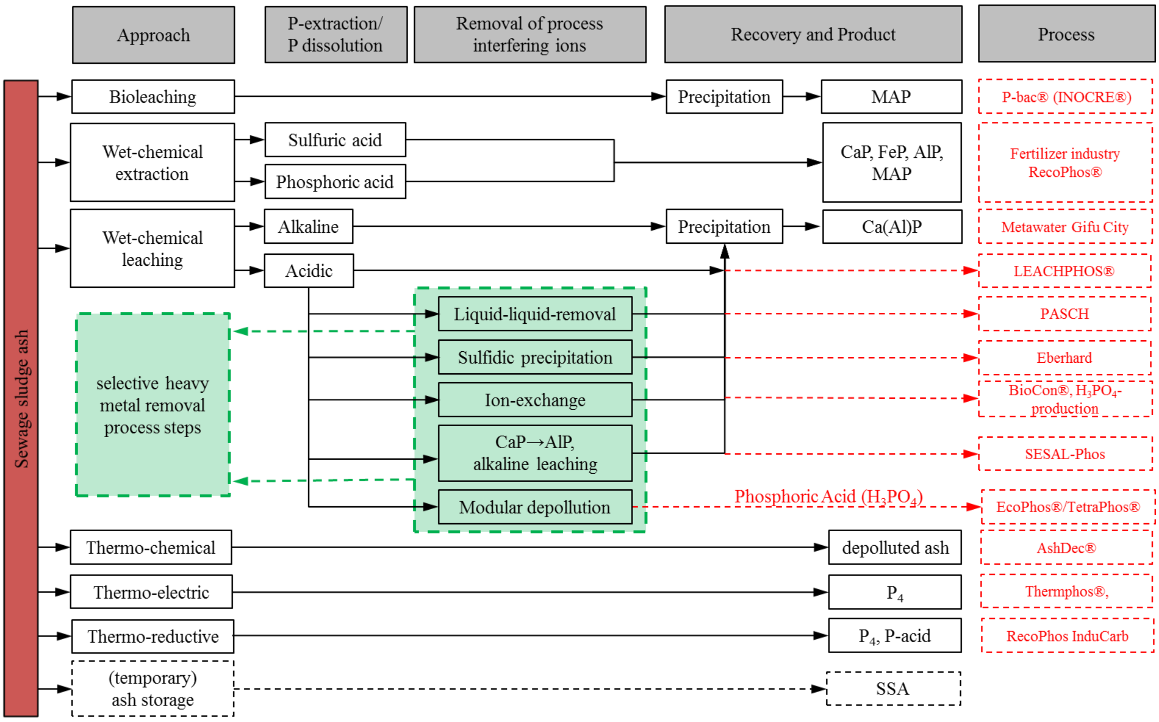


Figure 12: Overview of the approaches to recover P from sewage sludge ash.

3.5.1 Bioleaching

Inocre® invented a two-step P recovery process by biological leaching (**P-bac®**) (Inocre, 2013). Based on the knowledge of the ability of specific microorganism to produce acid (Ehrlich, 2001; Zimmermann, 2010; Sarlin et al., 2013), P and HM are leached from the ash by the microbial production of sulfuric acid (Chi et al., 2006). After a solid-liquid separation step, P is assimilated by specific microorganisms and thus separated from HM. The P enriched biomass is then separated from the liquid phase which contains the HM. The HM remain dissolved and are subsequently chemically precipitated and concentrated for disposal. No further information on the chemical demand and final product is available. According to **Inocre®**, as much as 90% of the P may be recovered.

3.5.2 Wet-chemical extraction

Wet-chemical extraction forces the transformation of P that is not immediately plant available to P that is water-soluble and thus plant available. This is the process commonly used in the fertilizer industry to produce a commercial fertilizer from raw phosphate ore (apatite) (Figure 13). By adding sulfuric acid (70% H_2SO_4) to ground raw phosphate, single superphosphate (SSP) is produced (Eq 4). The H_2SO_4 demand is 0.38–0.4 kg kg ore^{-1} (Silva and Kulay, 2005; Patyk and Reinhardt, 1997).

$$\text{(Eq 4) SSP: } Ca_3(PO_4)_2 + 2\ H_2SO_4 \rightarrow Ca(H_2PO_4)_2 + 2\ CaSO_4$$

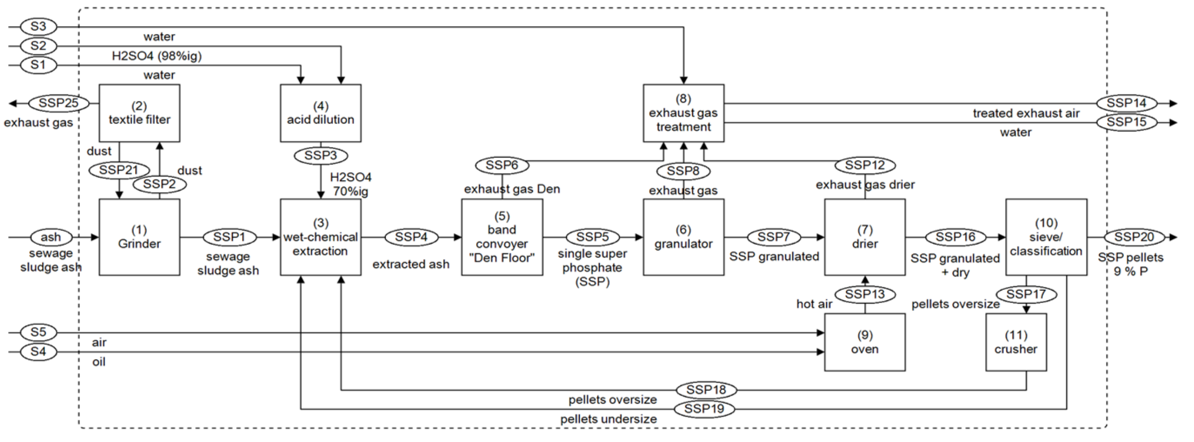
P availability from SSA can be raised through the same process (Donatello and Cheeseman, 2013). This approach has already been realized by **ICL Fertilizers®** (Ten Wolde, 2013). However, further investigation is necessary to understand the behavior and chemical bonding of P, heavy metals and Fe/Al during extraction (Herr et al., 2013). Petzet and Cornel (2011a) found, that ash containing aluminum from the precipitation of P at a WWTP exhibited significantly better water-solubility after H_2SO_4 extraction than did Fe-containing ash. Furthermore, treated Fe-rich ash exhibits a sticky characteristic, which impedes the further treatment steps as granulation or drying (Petzet and Cornel, 2011b).

In order to achieve a higher P content, industrial grade phosphoric acid (~52% H_3PO_4) is added to SSA in a rotary kiln to produce a product similar to triple superphosphate (**RecoPhos®**, Weigand et al., 2011; Bohndick, 2012) (Figure 13). The following extraction reaction of stanfieldite in SSA takes place to generate a water-soluble calcium or magnesium phosphate (Eq 5):

$$\text{(Eq 5) } Ca_4Mg_5(PO_4)_6 + 12\ H_3PO_4 + 2\ H_2O \rightarrow 4\ Ca(H_2PO_4)_2 + 5\ Mg(H_2PO_4)_2 + 12\ H_2O$$

H_3PO_4 demand is dependent on the P content of the SSA. For an SSA with a P content of 8.5%, ~0.65 kg H_3PO_4 kg ash^{-1} has to be added. Nevertheless, the study of Krüger and Adam (2014) reveals that the P content of SSA from a mono-incinerated sludge varies strongly (3–13% P) and thus the H_3PO_4 demand. The two purposes of the **RecoPhos®** approach are greater P solubility and P enrichment. After pelletizing and classification the certified *RecoPhos® P38* product has a P content of 16.6% with good water-solubility. Its rate of plant uptake is comparable to that of commercial fertilizers (RecoPhos, 2011; Von Tucher et al., 2011). The grain size is in the range of 0.8–5 mm and the grain strength fulfills the requirements for application with modern spreaders (RecoPhos, 2011). However, due to the lack of a decontamination step, the entire heavy metal load is transferred to the product with the wet-chemical extraction processes. Therefore, this process is only applicable for fertilizer grade ash (Weigand et al., 2013). For both approaches, i.e., **Fertilizer Industry** and **RecoPhos®**, the recovery potential is ~100% with respect to the ash input and thus ~85% with respect to the WWTP influent.

Fertilizer industry: Wet-chemical extraction with sulfuric acid



RecoPhos®: Wet-chemical extraction with phosphoric acid

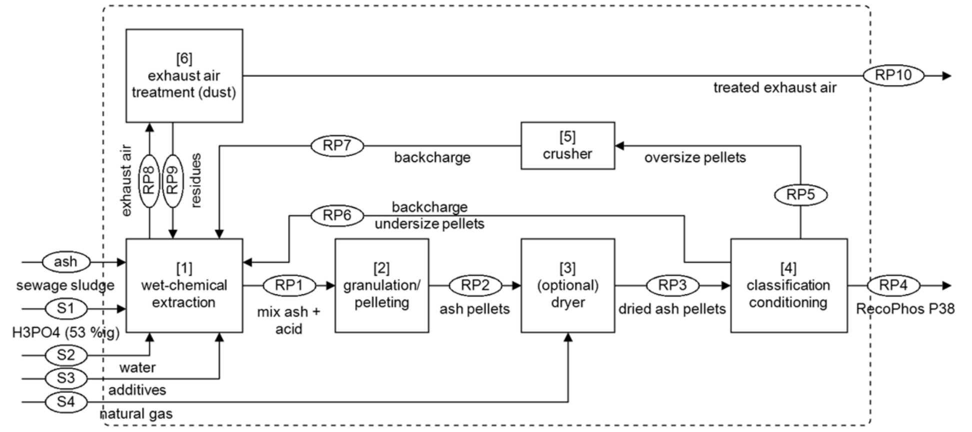


Figure 13: Material flow models for technologies to recover P from SSA by wet-chemical extraction (above: fertilizer industry, below: RecoPhos®).

3.5.3 Wet-chemical leaching

In wet-chemical leaching P is converted from a solid-bound to a dissolved form in a liquid (leach liquor). Changing the pH and thus the solubility by adding strong acids such as H_2SO_4 or HCl or bases as NaOH are well-known methods used to achieve 90% P dissolution (Schaum et al., 2004; Schaum, 2007). In contrast to extraction, a separation of solids from the P rich leach liquor is necessary.

3.5.3.1 Alkaline leaching

One alkaline leaching approach (**LOTUS-Project**; Takaoka et al., 2010) has been implemented full-scale in Gifu City, Japan. P is leached from SSA by adding a strong base (NaOH or KOH) at temperatures of 50–90°C (Figure A 4). The leaching is performed in two-steps to increase the P yield from the ash. The extraction rate is about 60–70%. The NaOH demand is ~0.12 kg kg ash^{-1}. As heavy metals barely redissolve under alkaline conditions, no heavy metal removal is required. Calcium hydroxide ($Ca(OH)_2$) is added to the P rich leachate in a stirring reactor at a molar Ca:P ratio of 1.5:1. As much as 100% of the dissolved P is precipitated as CaP. The alkaline supernatant is recycled back to the initial leaching step. The solid fraction (dephosphorized ash; 0.76 kg kg ash^{-1}) can be used as a raw material for construction materials after the addition of sulfuric acid for pH neutralization (0.04 kg kg ash^{-1}). Thus, no wastes arise from this recovery approach (LOTUS, 2007).

3.5.3.2 Acidic leaching

A pH below 2 and a residence time of 0.5–2.0 h are necessary to dissolve up to 90% of the P from SSA (Montag, 2009). H_2SO_4 and HCl can be used and their demand amounts to approximately 0.3–0.5 kg pure acid kg ash^{-1}. For wet leaching, the acids are diluted with water (~8–9 m^3 kg ash^{-1}) to generate a liquid phase. Realistically, 80–90% of the initial P is present in the leach liquor after the solid-liquid separation. The dephosphorized ash with a mass flow of 0.65–1 kg kg ash^{-1} requires proper treatment because of its acidic characteristic. Interfering ions (Fe, Al, heavy metals) dissolve simultaneously to P, although to varying degrees (Figure 14). Particularly at pH level below 2, there is considerable dissolution (Herr et al., 2013; Ottosen et al., 2013).

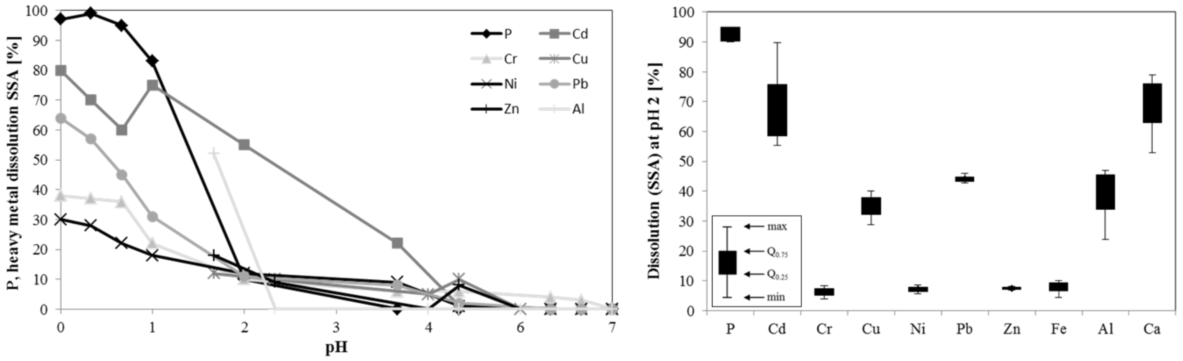
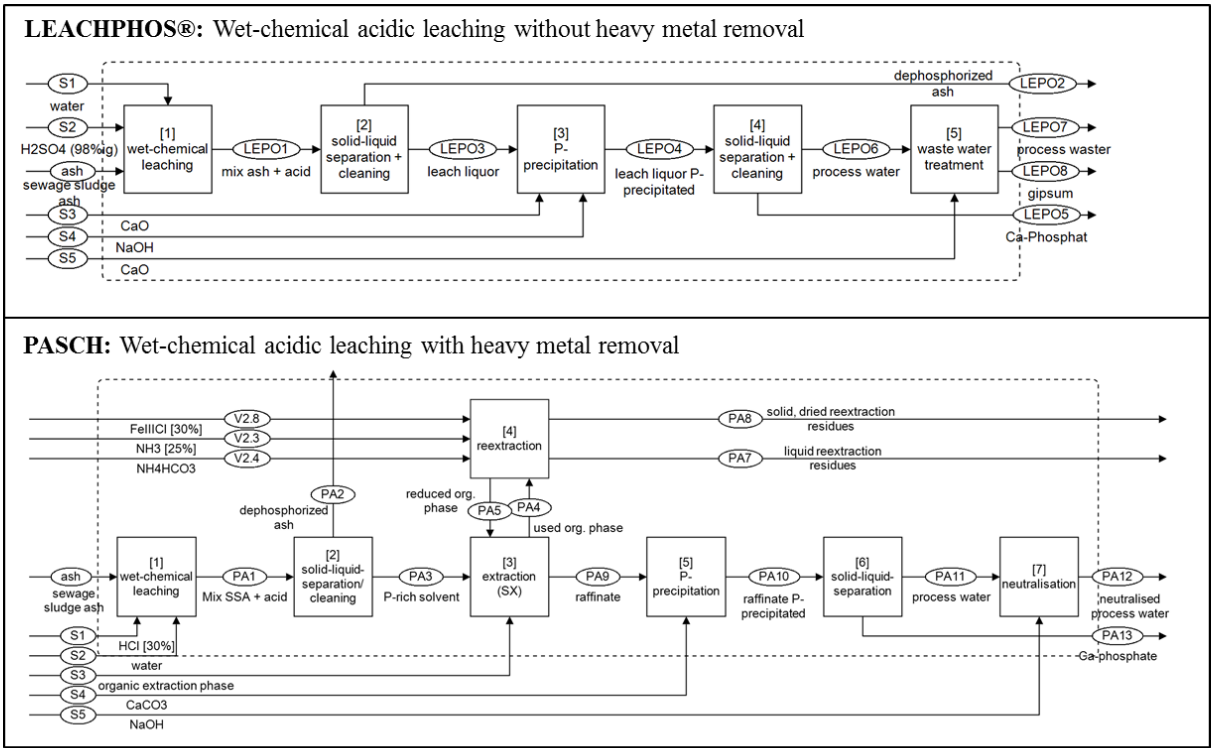


Figure 14: Dissolution [%] of selected elements from SSA at various pH level (left, Herr et al., 2013 and dissolution [%] of P and HM at pH 2 (right, PASCH; Montag et al., 2011) (first ($Q_{0.25}$) and third quartile ($Q_{0.75}$) and minimum (min)/maximum (max)).

Due to presence of interfering ions in the leach liquor, several approaches for decontamination have been investigated (multi modular decontamination: **EcoPhos®** (de Ruiter, 2014) and TetraPhos® (Remondis, 2015); solvent extraction: **PASCH** (Montag et al., 2011); sulfuric precipitation: **Eberhard** approach (Franz, 2008); ion exchange: **BioCon®** (Levlin, 2001) and approach from Donatello et al. (2010) for **H_3PO_4-production**; P rearrangement and leaching: **SESAL-Phos**: (Petzet et al., 2012). In contrast, the developers of **LEACHPHOS®** (Morf, 2012; Morf, 2013) even preclude specific decontamination (Figure 15).


LEACHPHOS®: Wet-chemical acidic leaching without heavy metal removal

PASCH: Wet-chemical acidic leaching with heavy metal removal

Figure 15: Material flow models for technologies to recover P from SSA by wet-chemical acidic leaching to produce calcium phosphate without and with specific heavy metal removal (above: LEACHPHOS®; below: PASCH)

The **EcoPhos®** process is a patented, already full-scale implemented multi-step approach to valorize low grade phosphate rock and also P rich ashes to high quality market products as phosphoric acid or animal feed (EcoPhos, 2015) (Figure 16). First step is the leaching of the ash with HCl. Undergoing different modules, which are kept confidential, a purified phosphoric acid for fertilizer or food and feed industry is produced. At the same time most of the occuring by-products are sellable products as $CaCl_2$, gypsum, silicate as well as iron- and aluminum chlorides. With the **TetraPhos®** process, Remondis developed and implemented a similar approach in pilot scale (Hamburg) using phosphoric acid instead of HCl to the leach the ash (Remondis, 2015) (Figure 16). Due to the lack of data, no conclusions can be drawn on resource demand and fate of pollutants. P recovery potential related to the input SSA is 80–90%.

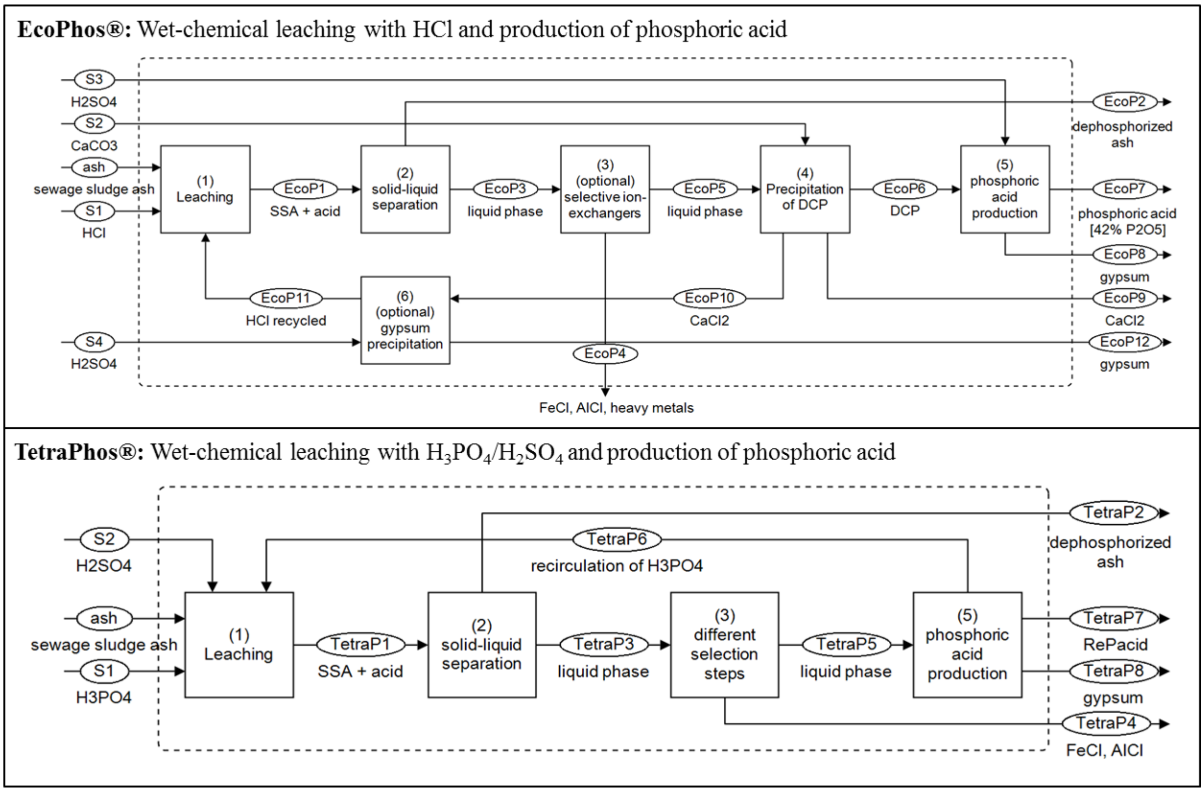


Figure 16: Material flow models for technologies to recover P from SSA by wet-chemical leaching technologies to produce phosphoric acid (above: EcoPhos®; below: TetraPhos®)

The **PASCH** approach uses solvent extraction (liquid-liquid extraction) for targeted heavy metal removal (Dittrich et al., 2009; Montag, 2009). The organic solvent phase (2 kg t ash^{-1}) consisting of various resolvents and eluants that are thoroughly mixed with the leach liquor from which, after a short residence time of 15 minutes, interfering ions settle out. Cd, Cu, Pb, Zn and Fe are removed at a rate of 80–99%, whereas Al, Cr and Ni are predominant in the treated leach liquor. Ninety-eight percent of the P pass this solvent extraction step and is then precipitated as CaP by adding NaOH and lime (Montag et al., 2011). With around 5% the aluminum content is high. The heavy-metal-rich solvent phase is decontamination by a reextraction step and can be reused for heavy metal removal. Heavy metal rich solid waste (~20 kg t ash^{-1}) and wastewater (0.25 m^3 t ash^{-1}) are generated from the reextraction. The supernatant from the precipitation process needs to be neutralized. The recovery potential with regard to the input ash is approximately 80% and thus 70% with regard to the WWTP influent.

Franz (2008) demonstrated with the **Eberhard** approach, the effective removal of heavy metals from leach liquor, by sulfide precipitation (Na_2S). The Na_2S solution is added to the leachate at concentrations between 10 and 80 g L^{-1}. Most of the heavy metals immediately form low soluble sulfide compounds even in a strong acid solution. The black precipitates are separated and the dissolved P is precipitated by adding limewater. This approach has been tested only at laboratory scale, and no further data are available.

The multistep **SESAL-Phos** approach starts with an acidic pre-treatment of SSA (~pH 3–4.5, residence time of 60–90 min) to convert CaP to aluminum phosphate (Al-P) (Petzet et al., 2012; Petzet et al., 2011; Figure A 5). A necessary precondition therefore is an aluminum-rich ash. After separation, NaOH is added to the ash to redissolve the Al-P at pH 13. As much as 75% of the P dissolves and compared to pure acidic leaching the heavy metals either do not (Cu, Ni, Pb, Zn) or barely redissolve (Cd, Cr). The dephosphorized ash is separated and 99% of the P is precipitated primarily as hydroxyapatite ($Ca_5(PO_4)_3OH$) by adding $CaCl_2$. The aluminum-rich supernatant, can be recycled back to the WWTP for P precipitation. The acid demand is shown in Table 4. Approximately 0.32 kg of NaOH, 0.3–0.36 kg of $CaCl_2$ and 12 L of water are required per kg of ash. The recovery potential related with regard to the input SSA is 70–75% (Petzet and Cornel, 2011).

Table 6: Comparison of applied acid, pure acid demand [kg kg ash^{-1}], pH and P dissolution [%] (Petzet et al., 2012; Montag et al., 2011; Franz, 2008; Schaum et al., 2007; Takahashi et al., 2001)

Acid	mean	min	max	pH	P dissolution [%]
H_2SO_4 (98%)	0.39	0.30	0.50	≤ 2	80-90
HCl (33%)	0.45	0.31	0.46	≤ 2	90-95
HCl (33%)	0.18	0.15	0.22	3	-[1]

[1] SESAL-Phos: no leaching, converting P-compounds

Research into the removal of interfering ions from leach liquor by ion exchange yielded shows promising results (Mendes and Martins, 2004) but also revealed difficulties regarding selectivity due to the low pH of the leach liquor (Franz, 2008). Sequential recovery of iron chloride, calcium hydrogen carbonate and finally phosphoric acid from the leach liquor was the basis of the **BioCon®** approach. Donatello et al. (2010) demonstrated the possibility of producing phosphoric acid from ion-exchanger brine.

3.5.4 Thermo-chemical

Thermo-chemical approaches (**AshDec®,** part of OUTOTEC®) are aimed at removing heavy metals from SSA at high temperatures of up to 1,050°C with the addition of chloride additives, e.g., $MgCl_2$, $CaCl_2$, KCl_2 or $NaCl_2$ (Hermann, 2008) (Figure 18). Following initial warming to 600°C, the SSA is mixed with the chlorides and treated in a rotary kiln at temperatures of 750–1,050°C for 40–120 minutes (Adam et al., 2009[a]; Adam et al., 2009[b];

Mattenberger et al., 2008). Due to chlorine donation volatile heavy metal compounds with low evaporation temperatures are formed. Shown below is an example of this process of heavy metal decontamination in which Zn is removed by addition of $CaCl_2$ as a chlorine source. Depending on the humidity of the reaction atmosphere, either HCl or Cl_2 is formed. These compounds diffuse through the ash matrix resulting in the following reactions (Eq 6, 7):

$$\text{(Eq 6) } 2\,HCl + ZnO \rightarrow H_2O + ZnCl_2$$

$$\text{(Eq 7) } Cl_2 + ZnO \rightarrow \tfrac{1}{2}\,O_2 + ZnCl_2$$

$ZnCl_2$ has a significantly lower evaporation temperature than ZnO and escapes to the flue gas. However, heavy metal chlorides may also react with the ash matrix by adsorption, thereby forming stable silicates or aluminates. Unfortunately, these reactions exhibit reduced levels of decontamination. Depending on the temperature, the type and quantity of the chloride and the residence time in the rotary kiln, the heavy metal removal rate is variable (Figure 17). Although very good decontamination of >90% have been reported for Cd, Cu, Pb and Zn, other heavy metals such as As, Cr and Ni remain primarily in the ash (Nowak et al., 2011[a], Nowak et al., 2011[b]).

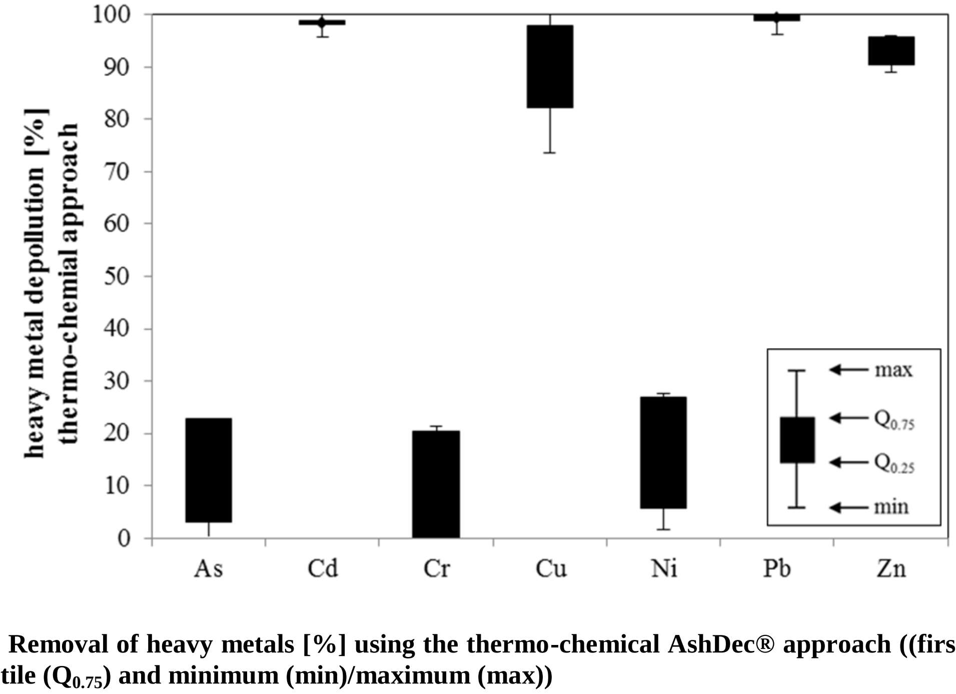


Figure 17: Removal of heavy metals [%] using the thermo-chemical AshDec® approach ((first ($Q_{0.25}$) and third quartile ($Q_{0.75}$) and minimum (min)/maximum (max))

The HM containing flue gas is treated in a three-step flue-gas treatment, equipped with filters, chloride absorption by $MgCO_3$ and SO_2 removal by $NaHCO_3$. Table 7 shows the chemical demand for three scenarios: (1) the **SUSAN-Project** (2) an approach based on a direct connection to an incineration plant (using the hot ash) and (3) an approach based on a centralized plant (cold ash) (Hermann, 2013). In addition to the benefits of the decontamination, the P remains almost entirely in the ash and the formation of a new mineral phase can be observed, possibly implying higher bioavailability of P (Mattenberger et al., 2010). By mixing the treated ash with additional nutrients (P, N and K) a certified commercial fertilizer known as *PhosKraft*® was produced and marketed (Nanzer et al., 2014). Due to the low bioavailability of the output ash, an alternative approach was explored. By addition of sodium sulfate ($Na_2(SO_4)$) compared to former Cl-donation a calcium-sodium-phosphate, similar to the known calcined phosphate approach (“Rhenania”) is produced (Scharrer, 1950). Indeed, the removal of heavy metals is significantly lower (e.g., Cd: 80%, Pb: 50-70%, As and Zn: partial, Cu: none). However, the resulting P exhibits much higher bioavailability (Hermann, 2013). The P recovery potential of both approaches is ~98% with regard to the ash input and thus ~85% with regard to the WWTP influent.

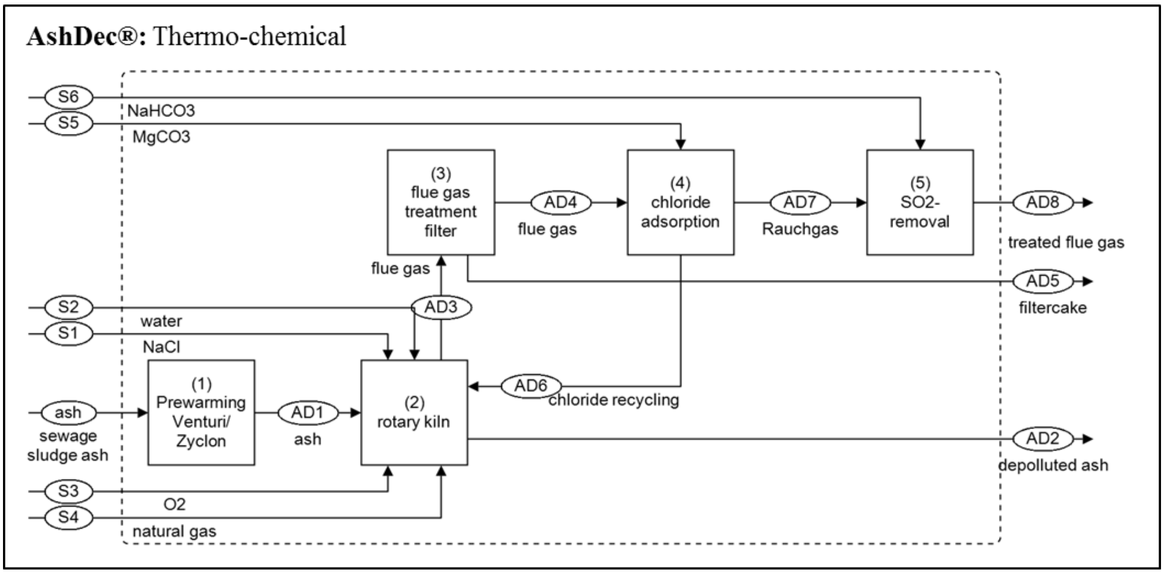


Figure 18: Material flow model for the AshDec® technology to depollute SSA with a thermo-chemical approach

Table 7: Resource demands and occurrence of filter cake in flue gas treatment [kg kg ash^{-1} or kWh kg ash^{-1}*] in various scenarios of thermo-chemical ash decontamination (Hermann, 2013)

kg kg ash^{-1} or kWh kg ash^{-1}	**Rotary kiln**			**Flue gas treatment**			**Energy***		**Filter-cake**
	$MgCl_2$	NaCl	O_2	$Ca(OH)_2$	$MgCO_3$	$NaHCO_3$	gas	electr.	
SUSAN	0.20	-	-	0.17	-	-	0.93	-	0.060
Cold ash	-	0.042	0.048	-	0.076	0.012	0.58	0.107	0.064
Hot ash	-	0.038	0.085	-	0.081	0.008	0.31	0.08	0.064

3.5.5 Thermo-electric

The thermo-electric process is typically used to produce pure white phosphorus (P_4) from raw phosphate ore (**Thermphos**®, Schipper et al., 2001 and Schipper et al., 2004), however SSA can be incorporated. SSA is mixed with raw phosphate and clay, is sintered (CO-burner) and is subsequently fed to an electric arc-furnace at temperatures above the ash melting point (1500 °C) together with coke (reducing agent) and pebbles (SiO_2, for slag formation) (Figure A 6). Under these conditions the P is reduced to P_4 and leaves the furnace as a gas together with CO and dust. After the flue gas treatment (electrostatic precipitator), the P_4 condenses and is stored in a water bath with a purity of 99.99%. The waste product remaining in the furnace is a liquid calcium-silica slag that can be used in road construction. Lower P concentrations in the SSA than in the raw phosphate ore cause more slag, which adversely affect the energy efficiency of the process, however. Due to the presence of iron among other metals in the SSA, a separate iron phosphate is formed in addition to other metal compounds. Because the iron phosphate is an undesired by-product (lowering the P_4 yield), SSA with a low iron content is required and the iron to P ratio should be below 0.25 (Lefferts, 2012). Besides iron, other heavy metals as Cd, P and Zn are undesired in the process and enrich within the system (Schipper et al., 2004), thereby creating a need for costly purges and disposal. Resource demand is shown in Table 8 (Schipper, 2012a, Schipper, 2012b, Hirschberg, 1999 and Breil, 1970). In 2008–2011, **Thermphos**® processed already 1000–3000 tons of SSA per year, but they became insolvent in 2012 due to Kazakhstan price dumping (van der Weijden et al., 2013). The recovery potential is approximately 95% with regard to the ash input and thus ~83% with regard to the WWTP influent.

Table 8: Resource demand [kg kg ash^{-1}; kWh kg ash^{-1}] in the thermo-electrical approach (Schipper, 2012; Hirschberg, 1999; Breil, 1970)

Process	Sintering	Furnace	P_4-storage	Unit
Natural gas	7.8-97[1]			kWh kg ash^{-1}
Coke		72		kg kg ash^{-1}
Clay		100		kg kg ash^{-1}
Quartz		220		kg kg ash^{-1}
Water			140	kg kg ash^{-1}
Electricity	14[1]	930[1]		kWh kg ash^{-1}

[1] variance due to different data source

3.5.6 Thermo-reductive

Using the **InduCarb** reactor (RecoPhos InduCarb process, Leoben, Austria), P and other compounds derived from SSA or sewage sludge are reduced at temperatures between 1,300 and 1,600°C. All kinds of phosphate compounds (CaP, $FePO_4$ and $AlPO_4$) are reduced or vaporized to pure P (Schönberg et al., 2014; Rapf et al., 2010). P and CO leave the reactor in the gaseous phase. Due to the presence of vaporized heavy metals, flue-gas treatment is required. CO can be used for energy production in an afterburning chamber. The solids produced in the reactor consist of slag (solidified), molten metal, and an iron-P alloy. The heavy metals with higher vaporization temperatures are transferred to the various slags. These processes are currently the topic of intense research, but data regarding resource demands and the transfer of P and heavy metals are insufficient (Schönberg et al., 2014).

3.6 Discussion

This overview of P recovery technologies from municipal wastewater highlights that although P recovery is intensively discussed on the international level, there is no “magic-bullet” solution and appropriate technology choice should be based on local conditions that determine several aspects of wastewater quantity, composition, treatment, and reuse. For many technologies, their technical feasibility has already been demonstrated and replicable data for detailed material- and energy flow analysis are frequently available. Additionally, costs for implementation of different technologies are reported. Still, the question remains for municipalities and regions around the world: what is the most appropriate technology for investment and eventual implementation considering technological, ecological and economic realities and goals?

P recycling and possible solutions are bound to vary from region to region (Metson et al., 2015). For example, conventional wastewater treatment technologies currently used in Europe may affect the selection of recovery technologies. As such solely using information on ecological impacts or related costs currently reported in the literature could be very misleading as accounting methods and the applied system boundaries used in different studies differ significantly from each other. Important limitations related to such comparison include that the technologies address different P sources with varying characteristics and that the approaches and their aims differ greatly (e.g., avoid struvite incrustation, depollution of heavy metals, pure P product, use energy content of sewage sludge as well). Furthermore, the occurring P products show different characteristics (e.g., P and pollutant content, plant availability), technologies have often been investigated without taking into account connected up- or downstream processes, positive or negative effects on the WWTP are often neglected, occurring wastes or by-products and their disposal are not considered and possible environmental effects are not systematically included in comparisons and assessments. For an economic assessment, costs calculations and their transparency are missing and vary strongly.

In addition to difficulties with comparing the appropriateness of technologies for a particular city, and comparing technologies to one-another, it also remains difficult to compare (again with respect to environmental and cost criteria) P fertilizer production using phosphate ore based on results from available reports and articles. Therefore, a critical area of research building on this article is the development of an appropriate methodology to allow an objective, integrated and comparative assessment between the recovery of P sources to create fertilizers. Such a method of assessment should consider technical, environmental and economic aspects. A modular reference system needs to be developed (including a defined reference WWTP and different reference sludge treatment options meeting European wastewater and sludge treatment standards) where the different technologies can be applied and the effects on connected up-or downstream processes can be considered. Material, energy and cost accounting should be based on reported data and recalculated using a uniform methodology, guaranteeing a better comparability of different approaches for application in different P rich wastewater related flows. The data compiled in this article is a fundamental building block for such an assessment. We will present such a model in an upcoming article. As such, we restrict our conclusions in this article to more general aspects of the different recovery technologies and avoid very distinct statements on the superiority of one or the other approach.

3.7 Conclusions

P in wastewater represents a high recycling potential. With an efficient use of sewage sludge, theoretically up to 50% of annually applied mineral P fertilizer in agriculture could be substituted in Europe (Egle et al., 2014a and Schoumans et al., 2014). Taking into account other important P imports as feedstuff and food, human excrements play a minor role in the anthropogenic P cycle. Currently, a wide range of approaches exists to recover significant amounts of P from wastewater. From a technical point of view, P recovery from wastewater does not pose a great challenge, and numerous approaches are ready for full-scale implementation. The options include:

Recovery from secondary treated effluent

P can be directly recovered by precipitation or after a step of P enrichment by e.g., ion exchanger. However, ion exchangers have problems with the complex composition of wastewater (low selectivity and unwanted adsorptions). A reliable technology is important as this P removal step is the last barrier before receiving waters. Recovery potential is around 50–70% with regard to WWTP, but currently no demonstration of full-scale plant is known.

Recovery from the aqueous phase

For the recovery of already dissolved P by precipitation/crystallization, numerous technologies are known and already implemented on a full-scale. Their implementation and benefits primarily involve the improvement of plant operations, i.e., avoiding maintenance costs due to removal of incrustations, improvement of dewatering properties, and reduction of nutrient back flow with digester supernatant. As a benefit, these technologies can be used to affordably deliver a high-purity, plant available and market ready products (struvite, CaP). The recovery potential is low, a maximum of 30% with regard to the WWTP influent. In addition, the use of these technologies in advanced WWTPs equipped for chemical P removal reduces the recovery potential. Higher P yield is possible for these technologies when combined with disintegration or enforced P re-dissolution technologies beforehand (e.g., extraction or leaching).

Recovery from sewage sludge

Due to the complex composition of sewage sludge theses recovery approaches are technically complex and resource intense, particularly when applied to sludge resulting from chemical precipitation with Fe or Al. Negative effects on the WWTP are possible and such processes can create new waste streams that require further treatment (e.g., HM rich supernatants, acidified sludges, and HM-containing sludges). The recovery potential using wet-chemical approaches is approximately 40% with regard to the WWTP influent. This recovery rate is a trade-off with the chemical consumption and thus the costs. Using CO_2 instead of mineral acids is a promising option as the same recovery potential can be observed but with significantly lower consumption of chemicals and no need of a depollution step for heavy metals. Technologies that simultaneously recover energy from the sewage sludge and thus yield an inert product (wet-oxidative, metallurgic) appear to be promising. Their recovery potential is approximately 50–70% with regard to the WWTP influent. However, no further development is noticeable (wet-oxidative) or further research is required (metallurgic).

Recovery from sewage sludge ash

Technologies to recover P from SSA exhibit ideal characteristics for recovering P in large amounts (70–85% of WWTP influent). It is possible to destroy all pathogens and POPs as organic matter is incinerated. The mixing of sewage sludge with combustibles low in P or high in heavy metals should be avoided. Ash quality with regard to P and heavy metals can vary strongly due to sewage sludge quality. Thus, certain recovery approaches can only be applied on a small fraction of the total SSA (e.g., **RecoPhos®**). An advantage with those strategies is independence from the location of a WWTP and making the installation of a large central unit possible, yielding economies of scale. Possible drawbacks are costs associated with incineration infrastructure. Three general types of approaches for future SSA P recovery have been identified with the following possible positive and negative effects:

1. No heavy metal removal, high recovery potential (85% of WWTP influent) and improvement of P plant availability by wet-chemical extraction (Section 3.5.2).
2. Partial heavy metal removal (thermo-chemical treatment) with high recovery potential (85% of WWTP influent) results in a final product with vague plant availability. Improved plant availability is however possible by adding sodium sulfate instead of a chlorine compounds (Section 3.5.4).
3. Nearly complete heavy metal removal (wet-chemical approaches with specific heavy metal removal steps) but lower recovery potential (70% of WWTP influent) and a final product with high plant availability or intended use in the industry (phosphoric acid) (Section 3.5.3).

Direct partial substitution of SSA for raw phosphate ore in the fertilizer or food industry or P_4 production would allow the use of existing infrastructure and sales to existing markets. **EcoPhos®** and **TetraPhos®** deal with almost all types and quality of ashes only minimum P content is limiting. The fertilizer industry is however vulnerable to varying ash quality. When using SSA as a secondary material in the fertilizer industry, regulatory levels for heavy metals need to be met in the final product. Nonetheless, other elements, which are measurable in high concentrations as e.g., Cu or Zn but not considered in mineral fertilizer regulations so far need to be reviewed critically (potentially toxic dependent on the dose). A good ash quality with high P and low heavy metal concentration requires the management of the combustibles (e.g., co-incineration of Cat 1 meat and bone meal that requires thermal treatment) or the implementation of appropriate depollution technologies. From the standpoint of the fertilizer industry, Fe/Al content in the input mixture SSA/phosphate rock shall not exceed 2%. The desired reduction in Fe and/or Al can be achieved by using new alternative precipitants or by enhancing biological P removal.

The final decision regarding a preferred recovery approach should involve close coordination with existing wastewater management facilities and the various requirements regarding recovery potential, decontamination, environmental effects, economic feasibility and characteristic of the final product such as the plant availability and pollutant content.

The technical details and data on P recovery, depollution potential, resource demand, and product quality presented in this review paper are fundamental for an integrated comparative assessment of recovery approaches regarding technical, environmental, and economic criteria. This assessment will be performed for selected technologies in a subsequent paper.

4. Phosphorus recovery from municipal wastewater: An integrated comparative technological, environmental and economic assessment of P recovery technologies


magnesium-ammonia-phosphate (scanning with light microscopy)

4.1 Introduction

Phosphorus (P) is an essential nutrient in the agricultural sector (fertilizer and feed). Phosphate rock (PR) is the raw material for mineral P fertilizer production and feed production and is classified as a critical raw material by the European Commission (EC, 2014). Due to population growth and changes in diet as a result of rising living standards in emerging and developing countries, the agricultural demand for P and consequently its criticality will increase (Reijnders, 2014; Van Vuuren et al., 2010). Furthermore, countries lacking P deposits are entirely dependent on imports and are therefore vulnerable to market fluctuations in fertilizer and mineral P prices (World Bank, 2016). These challenges have been discussed intensely on scientific and various political levels during the last years. One of many measures for reducing dependency is the recovery of P from obviously available but currently often unexploited national P sources, e.g., municipal and industrial wastewater, meat and bone meal (MBM) and other organic wastes (Scholz et al., 2014). This work focuses on technologies designed to recover P from municipal wastewater, sewage sludge (SS), and sewage sludge ash (SSA). The national P budgets in Central Europe show that municipal wastewater contains a P load that could theoretically replace 40 to 50% of the annually applied mineral P fertilizer in agriculture (Zoboli et al., 2015, Egle et al., 2014a; Gehtke-Albinus, 2012; Binder et al., 2009). Due to potential environmental and health risks (heavy metals (HMs), organic micropollutants (OMs) and pathogens), acceptance of direct sludge applications and thus direct P recovery is low or decreasing in several European countries (Ott and Rechberger, 2012). With the current alternative sludge treatment methods, such as co-incineration in the cement industry, caloric power plants and waste incinerators, P is irretrievably lost.

Consequently, numerous new technological approaches have been developed and in some cases implemented at full-scale in recent years to recover wastewater P at different access points in wastewater treatment plants (Figure 19). Additionally, the existing P industries have shown interest and the ability to integrate SSA or recovered P materials, such as magnesium-ammonium-phosphate (MAP), into their processes to replace raw phosphate ore and produce marketable products, such as mineral fertilizers, animal feed, phosphoric acid, and even P in its pure form (P_4). In this work, the term “recovered material” is used for P-containing outputs from the recovery processes, as most of these outputs are not yet classified and marketable products.

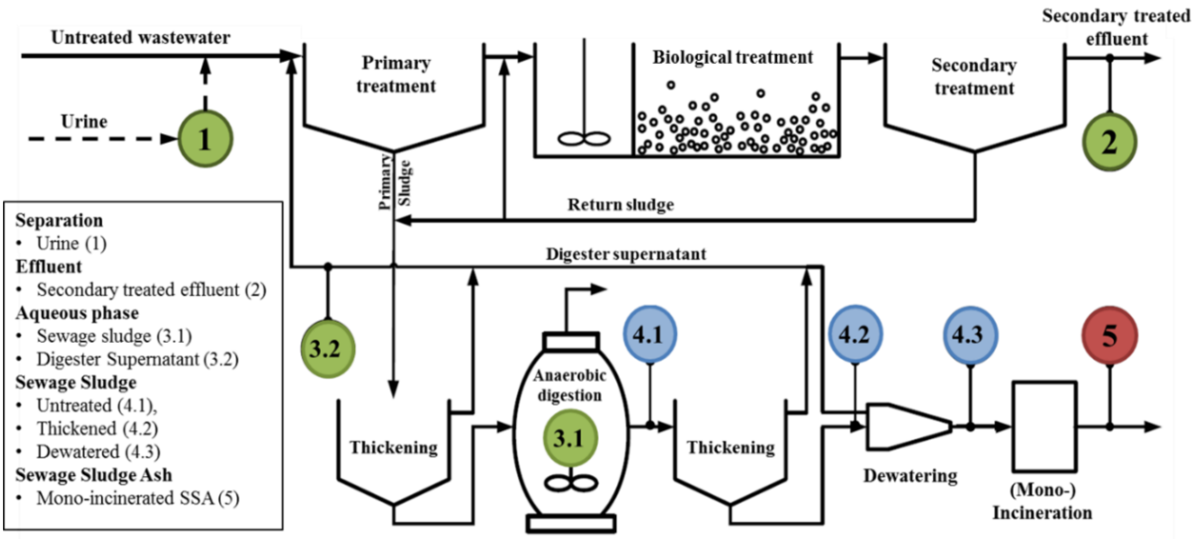


Figure 19: Various possible access points for P recovery approaches during wastewater and sewage sludge treatment or before/after incineration (Montag, 2008; modified sketch).

Considering more than 50 known P recovery approaches is already difficult for (political) decision makers, but comparing them regarding nutrient recovery potential, removal and destruction of potential hazardous substances and assessing the quality of the recovered P-rich materials is even harder. Possible positive or negative effects on the treatment of wastewater are often neglected. Effects on the environment, such as resource demand, wastes that require additional treatment and gaseous emissions, are displayed insufficiently. Furthermore, the realistic and comprehensible costs of the technologies for a society can only be calculated with a reliable and robust database and an appropriate reference system.

The aim of this work is first to develop an appropriate methodological approach for a comparative and integrated technical, environmental and economic assessment of technologies to recover P from different wastewater-related streams. The European Programme P-Rex aimed to achieve similar objectives (P-Rex, 2015). A significant novelty of this study compared to others is the assessment of the technologies within a defined reference system. This allows a comparative assessment along the entire process chain, including the entering wastewater, the treatment plant, thermal sludge treatment, final disposal of resulting waste streams, and the application of the recovered P-rich materials in agriculture or industrial processes. The core of our work is a detailed material flow analysis of P and selected heavy metals to track the paths of P and pollutants from the wastewater treatment plant (WWTP) influent to their final destination (e.g., recovered P material, waste, atmosphere or water bodies). A prerequisite for any meaningful assessment is knowledge of the technical principles and a good resource demand database and a complete substance flow analysis. Fundamental data on the technological background, detailed material flow models, input and output data on resource- and energy demand, recovered material and occurring wastes are provided in Egle et al. (2014b, c), Egle et al. (2015) and P-Rex (2015) and are summarized in Table A 14, A 15, A 16.

Nineteen different approaches, covering the broad field of P recovery technologies, have been selected to apply the developed method (Table 9). The selection criteria for the technologies were access to information and data and the readiness level (full- and pilot-scale implementations were preferable, but unique recovery processes with low technology readiness levels were included to cover a wide range of available technologies). As the P recovery sector is a quite young and dynamic market, some promising technologies could be missing in this selection due to insufficient data to perform this assessment, e.g., the recovery of P as MAP from digester supernatant (**STRUVIA®** (Mêlé et al., 2014), **REPHOS®** (Lebek and Lohmar, 2013), and **PHOSPAQ®** (Abma et al., 2010)), the recovery of phosphoric acid from SSA (**TetraPhos®**; Remondis, 2015), or the SS leaching with CO_2 instead of mineral acids (**Budenheim** carbonic acid process; Stössel, 2013). However, the selected technologies within this work cover most of the technical principles of those that are not considered. The methodology in this work can be applied to these technologies and to newly developed technologies as soon as reliable data are available. Due to the complexity, the methodology and results of a more detailed environmental assessment, including gaseous emissions and cumulative energy demand, is an integral part of a subsequent paper in progress.

Table 9: Considered P recovery technologies from the aqueous phase (green: digester supernatant, dissolved P in anaerobically digested sludge and effluent), sewage sludge (blue) and sewage sludge ash (red). This color code for the different P recovery access points is applied throughout this paper.

aqueous phase	sewage sludge [SS]	sewage sludge ash [SSA]
REM-NUT®[1] [2; ion exchange, precipitation]	**Gifhorn process**[7] [4.1; wet-chemical leaching]	**AshDec® depollution**[12] [5; thermo-chemical, ash depollution, Cl-source: e.g., $MgCl_2$]
AirPrex®[2] [3.1; precipitation/crystallization]	**Stuttgart process**[8] [4.1; wet-chemical leaching]	**AshDec® Rhenania**[13] [5; thermo-chemical, Rhenaniaphosphat, Na_2SO_4]
Ostara Pearl Reactor®[3] [3.2; crystallization]	**PHOXNAN**[9] [4.2; wet-oxidation]	**PASCH**[14] [5; acidic wet-chemical, leaching]
DHV Crystalactor®[4] [3.2; crystallization]	**Aqua Reci®**[10] [4.2; super critical water oxidation]	**LEACHPHOS®**[15] [5; acidic wet-chemical, leaching]
P-RoC®[5] [3.2; crystallization]	**MEPHREC®**[11] [4.3; metallurgic melt-gassing]	**EcoPhos®**[16*] [5; acidic wet-chemical, leaching, P-acid production]
PRISA[6] [3.2; precipitation/crystallization]		**RecoPhos®**[17] [5; acidic wet-chemical, extraction]
		Fertilizer Industry[18*] [5; acidic wet-chemical, extraction]
		Thermphos (P_4)[19*,**] [5; thermo-electrical]

[1]Liberi et al. (2001), [2]Heinzmann (2009), [3]Adnan (2002), [4]Britton et al. (2008), [5]Berg et al. (2007), [6]Montag (2008), [7]Esemen (2013), [8]Weidelener et al., (2005), [9]Blöcher et al. (2012), [10]Stenmark (2003), [11]Scheidig et al. (2013), [12]Nowak et al. (2011), [13]Hermann (2014), [14]Montag et al. (2011), [15]Morf (2012), [16]De Ruiter (2014), [17]Weigand et al. (2013), [18]tenWolde (2013), [19]Schipper (2012); *integration of SSA as secondary raw materials to substitute raw phosphate rock, ** Thermphos, the only P_4–producer in Europe went bankruptcy in 2012 and is therefore actually no relevant solution for Europe.

4.2 Material and methods

Technical principles of P recovery technologies have been frequently published; however, the required information and data to perform an integrated technology assessment is often missing. This work builds upon the work of Pinnekamp et al. (2011), Egle et al. (2015) and P-Rex® (2015), in which the fundamental technological background, detailed material flow models and resource- as well as energy demand were brought together. To achieve a meaningful and robust technology assessment, an extensive review of the literature was performed, technology developers were contacted, recovery plants were visited and laboratory trials were performed to validate the data. The required information and data for this work are subdivided into the following sections: (1) resource demand (e.g., chemicals and energy demand), (2) substance flow data on P, (3) substance flow data on HM, (4) nutrient content, (5) heavy metal pollutant contents in the recovered materials, (6) organic micropollutant contents in the recovered materials, (7) solubility and plant availability, (8) investment cost (capital costs), (9) operating costs, and (10) revenues and savings. Table A 12 and Table A 13 show the origin and quality of the data for different sectors of the investigated technologies.

The information and data gathered originates from several sources, and some assessment criteria data are incomprehensible or not available. Depending on the source of the data, different uncertainties need to be considered. Therefore, a qualitative uncertainty concept (Section 4.2.5) is applied.

4.2.1 Modular reference system

For a robust comparison of the technologies, the data on the processes of the P recovery technologies have to be transferred to a defined reference WWTP for an integrated comparison within a defined reference system. The development of a "modular system" with defined reference processes, including detailed sub-processes, forms the basis for this comparative assessment (Figure 32). The introduction of sub-processes is essential, as recovery technologies address different P sources of a WWTP (e.g., digester supernatant, untreated/treated SS). A reference WWTP with a pollution load of 100,000 population equivalents (PE) (equivalent to a P load of 65,700 kg yr^{-1}), P removal by iron dosing (alternatively, biological P (Bio-P) removal for P recovery from the aqueous phase), and sludge treatment processes, such as thickening (5% DM), anaerobic digestion, dewatering with polymers (30% DM) and co-incineration of sewage sludge (e.g., waste incineration plant or cement industry) have been chosen. These WWTP conditions were chosen because P recovery from combined collected and untreated wastewater is not possible due to its complex composition. Due to the transfer of P from wastewater to sewage sludge (up-concentration) by biological or chemical P removal, which is a typical cleaning step of WWTPs in Central European landlocked countries with sensitive receiving water bodies, P recovery is possible. Detailed WWTP characteristics (e.g., wastewater composition, mass flows, transfer coefficients for P and the selected pollutants) are given in Table A 8 and Table A 9. All assumptions are made in order to have a reference system typical for the Central European situation. Additionally, sensitivity analyses have been performed to estimate how the size of the WWTP impacts the overall results, especially with respect to cost.

As acceptance of direct agricultural SS application is decreasing, especially in Central European countries, thermal sludge treatment, and in particular co-incineration of SS, was chosen as the reference sludge treatment process. The selected reference thermal sludge treatment process is a grate furnace (output: slag), and the resulting flue gas is treated. The resulting outputs are wastewater, filter cake, and treated flue gas. In case of future P recovery from the residues of the thermal processes, mixing with combustibles that are low in P, rich in ash, and rich in heavy metals needs to be avoided (e.g., mono-incineration or co-incineration with selected secondary fuels). The selected reference incineration system for mono-incineration is a fluidized bed reactor creating fine and powdery ash (fly ash). For simplification, it is assumed that the resource demand, flue gas treatment, and transfer of P and heavy metals are equal for mono- and co-incineration systems (Table A 10; Table A 11).

For the waste management system, an immobilization/stabilization process is integrated for non-directly disposable waste occurring from P recovery processes. Disposable waste is either landfilled or transported to an underground waste site (e.g., filter cake from flue gas treatment).

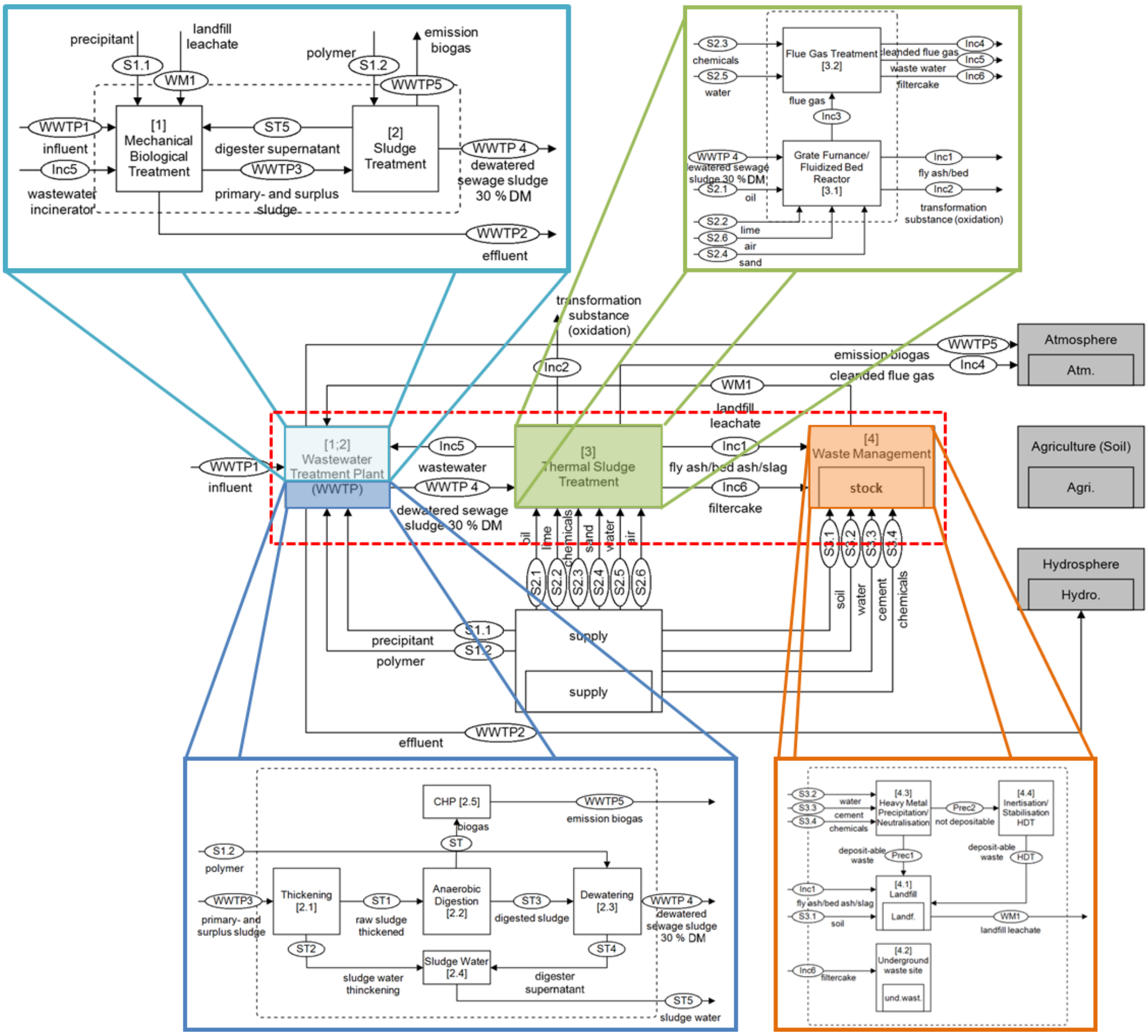


Figure 20: Process scheme for the reference substance flow model (STAN-model) with sub-processes for WWTP (blue), thermal sludge treatment (green), waste management (orange), a supply process (red line: system boundaries) and final receiving processes for emissions (atmospheres, agriculture/soil and hydrosphere).

4.2.2 P recovery potential and pollutant removal

The methodology of material flow analysis (MFA; Brunner and Rechberger, 2004) is applied to track the path of P and the selected heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) from the source (WWTP input) to the final recovered material, wastes or other emissions (output). In the MFA, input and output flows, possible stocks and changes in stocks are balanced in the defined system for a defined period of one year. The goal is the identification of transfer coefficients for the selected processes (Table A 9). MFA is the appropriate method, especially with respect to P recovery and depollution potential. Additionally, direct emissions to the atmosphere, agricultural soil, and water bodies can be illustrated.

Due to substance transformations, this method cannot be applied to organic micropollutants (OMs) and pathogens. Therefore, the path of OMs and pathogens is assessed by the comparison of the load in the reference sewage sludge and the load in the recovered material, if data are available. The considered OMs are adsorbable organically bound halogens (AOX), polycyclic aromatic hydrocarbons (PAH) and dioxins and furans (PCDD/F). For P and HMs, the outcome is a percentage distribution from the WWTP influent to the terminal receivers, including soil (agriculture), waste management, atmosphere and hydrosphere.

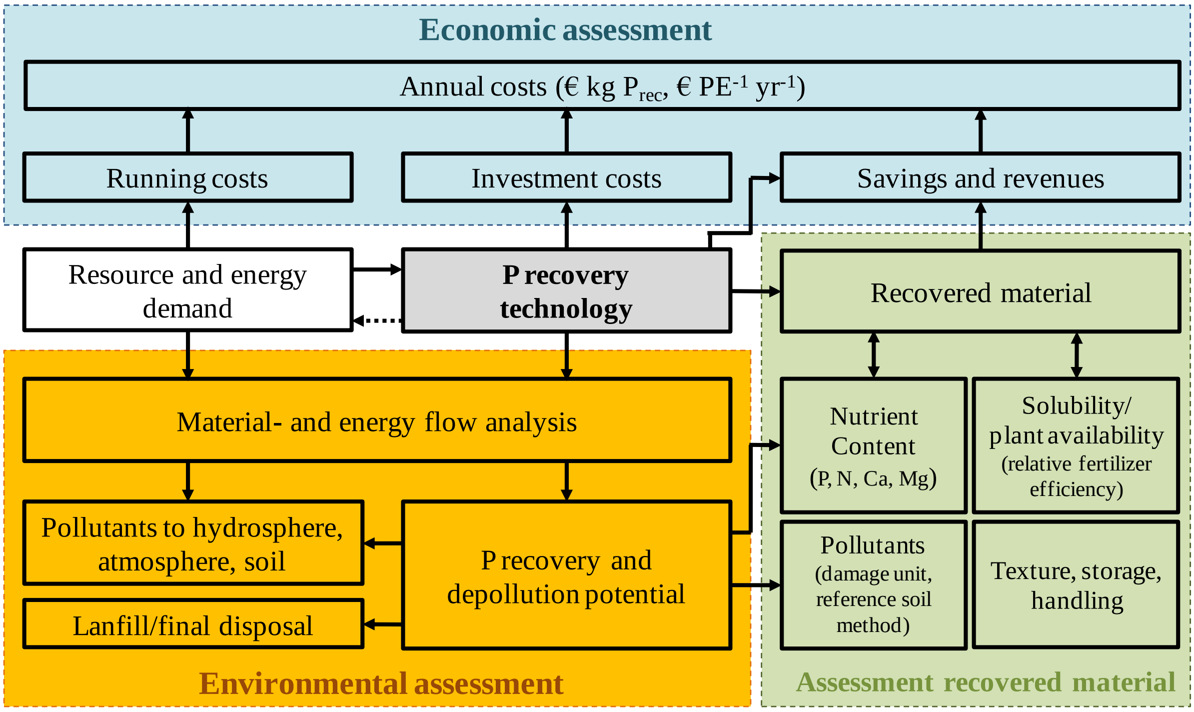


Figure 21: Applied methods to assess the selected P recovery technologies with regard to environmental and economic aspects and to assess the quality of the recovered materials.

Figure 22 shows the methodology used to assess P recovery and depollution potential for a given technology (Steps 1 and 2) and within the whole process chain (Steps 1–4). Step 1 and 2 is the creation of the MFA for a recovery technology (example in Figure 22: wet-chemical leaching approach from sewage sludge ash - PASCH). For this example the elements P (Step 1) and Cd (Step 2) are displayed. Step 3 is the integration of the recovery process into the defined reference system (the example of P is displayed). For this recovery technology the process “incineration” has to be changed to “mono-incineration”. Then the “mono-incineration” output flow “sewage sludge ash” is input to the material flow model of PASCH. Resource demand for the process “recovery approach PASCH” and the process “waste management” is provided by the process “supply”. The P-rich PASCH output “calcium phosphate” is an input to the process “agriculture soil”. Occurring solid waste flows of the recovery process need proper treatment and are input into the process “waste management”. The flow “neutralised process water” from this technology is a backflow to the process “WWTP”. Result (step 4 in Figure 22) is a percentage share distribution of P and selected heavy metals from the influent of the reference WWTP to the atmosphere, hydrosphere, agriculture (soil) or the waste management sector (e.g., landfill or underground deposit).

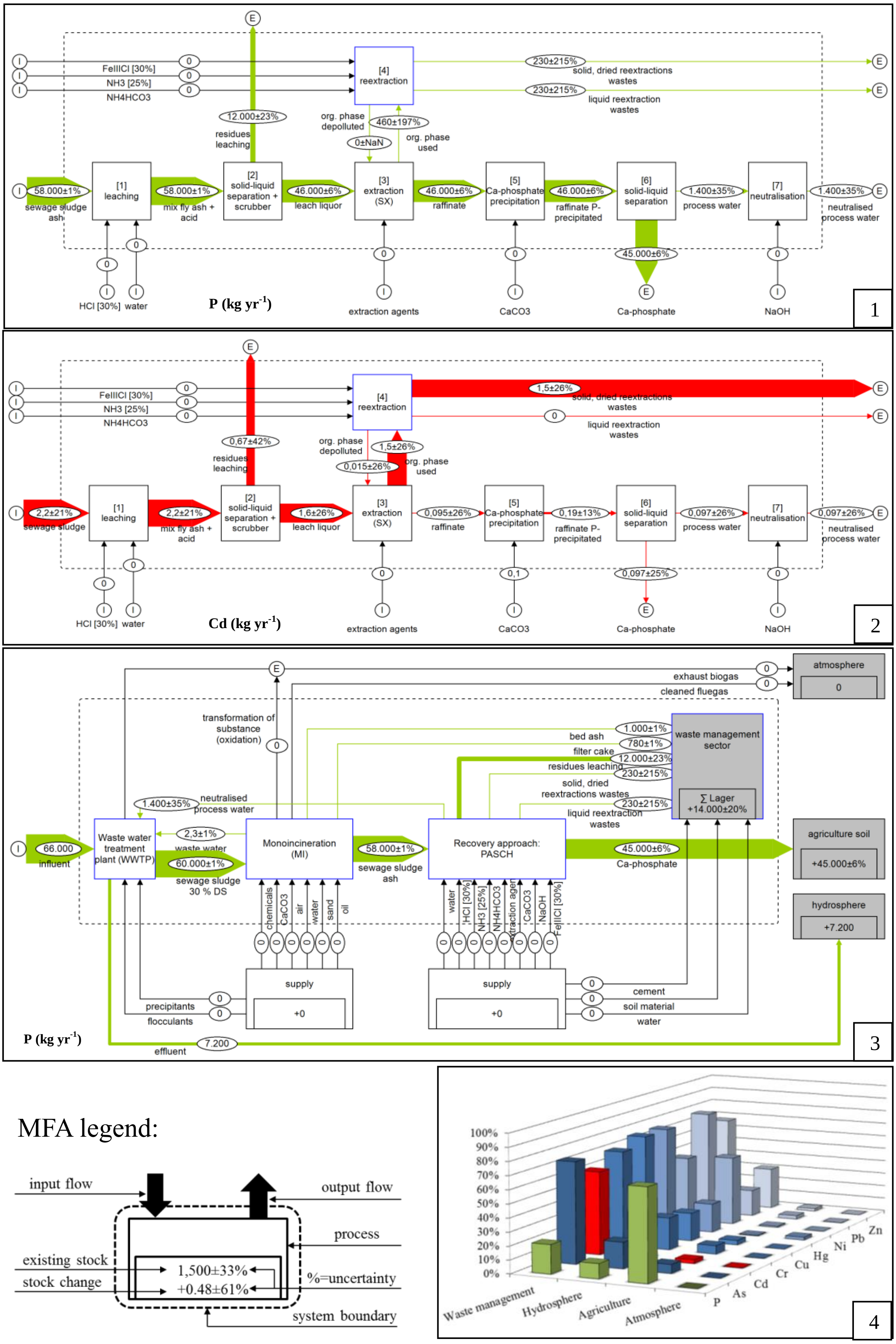


Figure 22: MFA methodology applied to a wet chemical leaching technology to recover P from SSA for P (1) and Cd (2), integration of the recovery process (considering the element P) into the reference model (3) and final percentage distribution of P and heavy metals to the receiving processes waste management, hydrosphere, agriculture and atmosphere (4).

4.2.3 Characterization of the recovered materials/products

The characteristic of recovered materials or products, in particular with regard to nutrient and pollutant content, plant availability and handling, is essential because agriculture and industry demand products with certain specifications. However, the recovered materials and products vary significantly in terms of these criteria.

4.2.3.1 Nutrient content, solubility and plant availability

To assess the recovered material with regard to nutrient content, solubility and plant availability, an extensive literature research was conducted. The literature sources are presented in the Appendix (Table A 12 and A 13). As the recovered materials may be suitable for agricultural use, their macro-nutrient contents (P, N, Ca, and Mg) are presented in Figure 27. In addition to the nutrient content, the plant availability of P is crucial. State of the art extraction tests (e.g., water, citric acid, neural/alkaline ammonium citrate, mineral acids) for mineral fertilizers are critically discussed with respect to their suitability for predicting the availability of newly recovered P materials for plants (Weinfurtner, 2011). Therefore, results from pot or field trials are more meaningful. If data are available, the plant uptake or fertilizing efficiency of a recovered material was assessed in relation to the efficiency of a commercial Single Superphosphate fertilizer (relative fertilizer efficiency, RFE). If there was a lack of data on certain materials, data from a similar type of material is used. For recovered products with direct industrial applications, such as phosphoric acid or P_4, a qualitative assessment with regard to their suitability for industrial use was performed.

4.2.3.2 Pollutant content

To apply a recovered product, the most important criteria with regard to pollutant content are the limit values of the applicable national fertilizer ordinances (HM, OM, and microbiological-hygienic parameters). This is the prerequisite for direct use of recovered products as fertilizers. Thus, as a first step, the recovered material is compared to Austrian limit values for mineral P fertilizers. However, even in cases where the limit values are met, different recovered materials exhibit distinct differences in their pollutant contents. To assess the purity and ensure the comparability of these recovered materials with regard to heavy metals, two methods – damage unit (DU) (Brans, 2008) and reference soil method (RSM) – are applied to compare the recovery products with respect to their heavy metal contents. The heavy metals As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn are considered here.

The principle of the DU method is to express the pollution load as a harmfulness coefficient. A DU value is calculated by taking into account a certain limit value for heavy metals (e.g., compost class A+, Compost Regulation Austrian, 2000) and calculating the quotient by dividing each heavy metal content of a final product by the defined limit value. The quotients are summed up and are related to the P content of the product (Formula 1; example see Table 10). The result is a dimensionless value (DU_P). Low DU_P values correspond to low pollutant contents.

$$C_{DUP} = \frac{\sum_{i=1}^{n} \frac{C_i}{C_i^{reference}}}{P\ concentration}$$

C_{DUP}: Concentration of the damage unit related to the P content; C_i = Concentration of a heavy metal in the recovered material; C_i reference: Concentration of a heavy metal in the reference material (e.g., compost class A+)

Formula 1: Calculation of the damage unit.

Table 10: Calculation method of the damage unit related to the P content (DU_P) for a commercial Single Superphosphate (SSP) heavy metal content based on Kratz et al. (2016).

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Compost Class A+ (mg kg DM^{-1})	0.7	70	70	0.4	25	45	200
SSP (mg kg DM^{-1})	31	89	20	0.03	24	64	161
Quotient DU	14.9	1.3	0.3	0.06	1.0	1.4	0.8

∑ DU kg DM^{-1}	19.7
P (g kg TS^{-1})	88
DU g P^{-1} (DU_P)	**0.22**

The reference soil method calculates the maximum number of years of application of a recovered material to a defined reference soil (one hectare, closed system, 20 cm soil depth, with defined heavy metal content; Smidt, 2010, Klik, 2001) until a tolerable or critical heavy metal concentration is reached. The reference soil is considered a closed system (e.g., no output via leaching). The annual applied P load is 40 kg P ha^{-1}. This method considers two factors: 1) the relevance of a possible harmful effect (lower tolerable load means higher priority) and 2) the heavy metal content related to the relevant nutrient (one kg of P; DU_P). A commercial mineral fertilizer (SSP with 88 g P kg TS^{-1}), a reference sewage sludge and a reference SSA with defined heavy metal contents are given as references. For organic micropollutants and pathogens in the output material of the recovery processes, the dataset is not as complete as for HM, and data are missing for some materials (Table A 12). If data are available, a qualitative assessment regarding the removal of OM and pathogens is carried out (very good depollution (+), moderate depollution (o), no depollution (-), no depollution necessary (*)).

4.2.3.3 Texture and handling

For agriculture applications, the quality criteria for the recovered material are high. Important aspects include storage properties (e.g., no dust, no humidity, and no reactivity), a certain grain size (2–5 mm) and strong enough grains for use in modern application equipment (spreaders). For categorization, the recovered materials are classified as “directly applicable” if they already possess a certain grain size (2–5 mm) or are coarse grained. Otherwise, the recovered materials are classified as “not directly applicable” if their texture is crystalline/powdery and therefore need further treatment steps, such as classification or granulation. If P is recovered by industrial processes in the form of a subsequently marketable product, the classification is “marketable”.

4.2.4 Cost calculation

Economic calculations are based on the settings of the reference WWTP (100,000 population equivalents (PE)) for recovery from supernatant and sewage sludge, as these technologies have to be implemented directly at the WWTP where the sewage sludge is produced. Recovery technologies can also be applied at larger than 100,000 PE WWTPs, and investment costs do not increase linearly with the plant size (economy of scale). Therefore, cost calculations are also performed for a WWTP with 500,000 PE to evaluate the sensitivity of the cost calculations to the plant size. To recover P from SSA, transport to and treatment at centralized plants are feasible. As centralized plants with a high throughput are required (otherwise these plants are not economic), the economic calculations for the recovery of P from SSA are based on a capacity of 15,000 t of SSA per year, which corresponds to ~1.75 million PE^{-1} yr^{-1}. Transport of sewage sludge from a WWTP to a centralized incineration and recovery plant is considered.

4.2.4.1 Annual costs for recovery technologies

Annual costs consist of capital and operating costs. Capital costs are calculated with the annuity method, whereby the investment costs are multiplied by an annuity factor (AF; Formula 2). The data on investment costs originate from the literature, feasibility studies or direct contact with operators of pilot- or commercial-scale plants (Table A 18). The calculation of the annuity factor includes the rate of interest (5% if no information is given by the plant operators) and the expected typical depreciation times of the plant components. The expected useful life is, unless otherwise known, 15 years for construction engineering.

$$AF = \frac{i * (1 + i)^n}{(1 + i)^n - 1}$$

i = rate of interest, n = expected useful life

Formula 2: Calculation of the annuity factor.

Detailed Material and Energy Flow Analysis is used to calculate the operating costs by multiplying the resource demand (Table A 14, A 15, A 16) by its market price (Table A 17). The details on the origin of the data (and therefore their uncertainty) are considered (Section 4.2.5.1). The operating costs include maintenance costs (unless otherwise known, 1% of the investment costs), personnel costs (50,000 € per man-year) and the costs of the disposal of the resulting wastes.

4.2.4.2 Savings and revenues

The considered savings include, for example, reduced disposal costs due to improved dewatering (2–5 percentage points; Ewert, 2009) of the sludge (€ per t of sludge reduced) or reduced nutrient back-flow of P and NH_4 in the digester supernatant. Reduced P back-flow corresponds to a lower demand of iron precipitants, and a reduced NH_4 back-flow results in a lower energy demand for aeration (Table A 14).

To take revenues for recovered materials into account, large uncertainties have to be considered. Currently, many different P-rich materials are produced, but there is no existing market. Therefore, the value of a recovered material is calculated by multiplying the nutrient components (P, N, Mg, and Ca) by their common market value (P: 1.7 € kg^{-1}, N: 1.1 € kg^{-1}, Mg: 0.3 € kg^{-1}, and Ca: 0.1 € kg^{-1}) (World Bank, 2016). This is a weak point of this method, as it assumes that the bioavailability of the total P content is the same for all recovered materials. However, this is not the case (Section 4.3.3.1). To consider the bioavailability from an economic perspective is not possible as the recovered materials are so varied. Unlike a water-soluble mineral fertilizer, some of the materials are not immediately availability to plants, but their yield is similar. Additionally, plants have the ability to take up even minimally soluble nutrients if there is a deficiency of easily available nutrients. Therefore, the total recovered P load is considered in the revenue calculations. The uncertainties related to these assumptions are considered in a sensitivity analysis (see section 4.2.5.2). In addition to the revenues for nutrients, the revenues for producing energy (heat and electricity) during the recovery process are credited in case that they are significant (Table A 15).

4.2.4.3 Integrated cost calculation

In addition to the cost calculation for the operation of a recovery technology, the costs for the whole process chain are calculated, including costs related to the required changes in the reference system (e.g., type of incineration, changes in the treatment schema, and changes in the amounts and routes of waste disposal). The objective is to capture all costs as well as savings, e.g., reduction of flocking agents, reduced energy demand due to NH_4 removal, improved dewatering of the sludge or revenues from selling the recovered material or produced energy, in connection with the implementation of a P recovery technology from a macroeconomic perspective. These calculations are based on the following reference processes: WWTP processes (Haslinger et al., 2015), thermal sludge treatment (co-incineration/mono-incineration; DWA, 2010), disposal of resulting wastes, and P recovery and transportation.

4.2.5 Uncertainty concept

4.2.5.1 Uncertainty in the data on technologies

As the data from the literature review originate from many different sources, the data quality differs strongly. Therefore, the uncertainty in the data is assessed qualitatively depending on the source. In this work, the uncertainty is categorized as low (+), moderate (o), high (-) and very high (--) (Table 11). If no data are available for certain technologies, missing data can be generated based on knowledge of the basic chemical principles (e.g., magnesium demand for precipitation of dissolved P) or knowledge of other similar technologies. The uncertainty concept is also applied to these alternatively gathered data (for examples, see Table 11). In addition to the uncertainty in the data, one particular challenge is the evaluation of possible future savings and revenues. This is a crucial point in the integrated technology assessment and will be discussed in the following section (Section 4.2.5.2.).

Table 11: Assessment of the uncertainty in the data with respect to their origin and the uncertainty associated with missing data for selected examples.

Data source and examples	Uncertainty
Doctoral and diploma theses and peer-reviewed papers. Verifiable data from plant operators (e.g., data from feasibility studies). Personal information and plant visits. Chemical fundamentals (e.g., stoichiometric ratio Mg:P for P precipitation, dissolution rate of P and heavy metals at different pH).	Low (+)
Reports in conference transcripts and conference presentations. Unverifiable data from plant operators (e.g., data from feasibility studies). No data on organic micropollutants or pathogens in recovered materials from SSA (assuming that organic micropollutants and pathogens are almost or totally destroyed during SS incineration). Calculation of the operating cost based on resource demand from laboratory or pilot scale trials.	Moderate (o)
Non-scientific reports. Data from non-conference presentations. Contradictory data for a technology (e.g., different results for bioavailability of the recovered material). No data, only fundamental principles of the technology and consequently rough estimates based on data on other technologies. Determining the resource demand based on knowledge of the operational costs.	High (-)
No data and no estimates possible based on other technologies, as the technology, resource demand, or output material is unique. Estimating the investment costs and capital cost calculation for technologies at, for example, the laboratory stage.	Very high (--)

4.2.5.2 Sensitivity of the cost calculations

In a sensitivity analysis of the cost calculations, the impacts of the considered WWTP size as well as the impacts of the expected savings and revenues on the costs of implementing a P recovery technology into a given waste water and sludge disposal system are analyzed. This task is challenging because, for example, no market for secondary raw materials exists at present and revenues from product sales can therefore vary significantly. Furthermore, P recovery technologies may have beneficial effects on a WWTP, and energy may be recoverable with the simultaneous stabilization of the sludge. However, at the current state of development, it is difficult to predict whether these revenues and savings can be credited to the full extent**.** The following list details the key assumptions for the sensitivity cost calculation:

- Cost calculations are performed with no or maximum achievable revenues for the recovered material. Maximum revenues are calculated by the total recovered load of nutrients and their market price (Section 4.2.4.2).
- For technologies that recover the dissolved P from digester supernatant or digested sludge, the nutrient back-flow of P and N to the WWTP can be reduced. This results in a lower demand for iron precipitants and a lower aeration demand. No or maximum savings of precipitants and energy are considered. Savings due to the avoidance of unwanted struvite encrustations in pipes and pumps are not considered, as maintenance cost cannot be assessed for this reference WWTP.
- Additional benefits result from the treatment of sewage sludge. Certain treatment processes, such as aeration and leaching of the sludge with acids, will lead to a better dewaterability and therefore reduced sludge disposal costs. No or a maximum improvement of the dewaterability by two percentage points is considered.
- The reference WWTP is defined with a pollution load of 100,000 PE. Larger units can operate more economically by reduced investment costs due to the economy of scale. Therefore, cost calculations for the technologies recovering P from the aqueous phase or sewage sludge are performed for a WWTP with 100,000 PE and 500,000 PE. The exception is the **MEPHREC®** technology, as this process is designed for larger sludge quantities. Therefore, the best-case scenario cost calculation is carried out for a 1–1.8 million PE WWTP.
- Certain technologies are capable of recovering P from sewage sludge and recovering energy during simultaneous stabilization of the sludge. However, these technologies have only been implemented at the pilot scale and/or no further development has been observed. Therefore, a high degree of uncertainty is associated with the efficiency of the energy recovery and the form of the recovered energy (e.g., electricity, heat or combustible gas). A wide range of values is therefore expected for technologies with an oxidation or metallurgic process step. The cost calculations are performed by taking no revenues and the maximum revenues from the energy recovery into consideration.

Consequently, this sensitivity cost calculation yields a wide range of values between the absolute worst-case scenario (no revenues for the recovered material, no up-scaling, no consideration of other benefits) to the absolute best-case scenario (maximum revenues for the recovered material, up-scaling, full consideration of other benefits) (Section 4.3.4.2.2.). The variability is a good indicator with regard to the uncertainty of the data. A low fluctuation margin indicates robust data, and a forecast for the expected future economic costs for P recovery can be predicted with low uncertainties.

4.2.6 Technical maturity

In addition to all the above criteria, the practicability of a technology in its environment is also important. To estimate the technological maturity, the method of technology readiness levels (TRL) is applied (DIN, 2013). TRL is a method used to assess the stage of development of new technologies based on a systematic analysis. The scale ranges from 1 (basic principles observed) to 9 (actual system proven in operational environment) (Table A 19). In addition to the actual state of development, an outlook for the future potential of a technology is given. The outlook is a result of the different assessment parameters given in this work (educated guesses) and discussions with experts in this field of research. As an example, a technology tested at the laboratory scale but with an outlook for a pilot plant is classified as TRL 4/5–6 (recent stage of development: 4, technology validated in lab; stage of development to be expected: 5, technology validated in relevant environment; 6, technology demonstrated in relevant environment). A technology that has been tested at the pilot scale but without visible development prospects is classified as TRL 5–6/- (-, no further development expected).

4.2.7 Functional unit

For a comparative assessment, all results are related to 1 kg P recovered (kg P_{rec}^{-1}). The costs and revenues are expressed in € per kg P recovered (€ kg P_{rec}^{-1}) or € per population equivalent and year (€ PE^{-1} yr^{-1}). PE is a reference value for the pollution load in wastewater (1 PE = 120 g COD d^{-1} in the influent load to the WWTP; in Austria, approximately 2 PE of raw wastewater is produced per inhabitant, with 1 PE stemming directly from the population and 1 PE stemming from industrial sources). To consider the recovery potential of a technology and to compare these values with conventionally produced mineral fertilizers, the use of the functional unit kg P_{rec}^{-1} is advantageous. The reference costs for P gained from raw phosphate rock (30% P_2O_5) and for a commercial Triple Superphosphate (46% P_2O_5) are 0.9±0.3 € kg P^{-1} and 1.7±0.5 € kg P^{-1}, respectively (Time range: 2011–2015; World Bank, 2016). Nonetheless, for technologies that can generate a profit, the functional unit kg

P_{rec}^{-1} is not an appropriate indicator, as a higher recovery potential lowers the profit for 1 kg of P, which can distort the results. In this case, it is helpful to consider the results from both functional units.

4.3 Results

4.3.1 Reference sewage sludge and sewage sludge ash

With simultaneous P removal from wastewater, approximately 90% of P is transferred into the sewage sludge. Therefore, the recycling rate with a direct agricultural sewage sludge application is 90% with regard to WWTP influent. The HM transfer from wastewater to the sludge was considered at different rates depending on the considered HM (50–80%, Table A 9). Without a depollution step, all HM and OM are released to the environment when applying sewage sludge directly to agricultural fields (Figure 24a).

Due to slight losses during incineration, the retrieval rate of P in SSA is 87% with respect to the wastewater input (Figure 26). The retrieval rate of HM in the ash is 80–98%, except for Hg, which features a retrieval rate of approximately 5% (Table A 11). OMs are mostly destroyed at temperatures > 850 °C. The formation of dioxins is not expected with a mono-incineration of the sewage sludge (Zeggel et al., 2015). However, if secondary chlorine-containing materials are co-incinerated to improve the calorific value, the formation and transfer of dioxins to the SSA has to be considered. The concentrations of P, HM and OM in the reference sewage sludge and SSA, which are the basis for the ongoing technology assessment, are presented in the following table (Table 12).

Table 12: P, heavy metal and organic micropollutant content of the reference sewage sludge and reference sewage sludge ash.

Element	Reference sewage sludge	Reference sewage sludge ash	Unit
P	39	84	g kg DM^{-1}
As	5.3	11.8	mg kg DM^{-1}
Cd	1.5	3.2	mg kg DM^{-1}
Cr	53.4	97.5	mg kg DM^{-1}
Cu	306	566	mg kg DM^{-1}
Hg	0.9	0.1	mg kg DM^{-1}
Ni	41	74.7	mg kg DM^{-1}
Pb	67	123.6	mg kg DM^{-1}
Zn	1,117	1,944	mg kg DM^{-1}
AOX	150	≤ 1	mg kg DM^{-1}
∑ PAH	7	≤ 1	mg kg DM^{-1}
∑ PCDD/F	3,300	≤ 1	mg kg DM^{-1}

4.3.2 Recovery potential and heavy metal removal

Depending on the applied technology, i.e., slow crystallization (**DHV Crystalactor®, Ostara®, P-RoC®**) or instant precipitation (**PRISA**), up to 85–95% of the formerly dissolved P can be recovered from digester supernatant. The recovery rate related to the

WWTP influent strongly depends on the type of P removal during wastewater treatment. The more P that is removed biologically (enhanced biological phosphorus removal, EBPR), the higher the redissolution rates during anaerobic sludge treatment, resulting in increased recovery rates. Therefore, WWTPs with mainly biological P removal processes are necessary, as the recovery rate has a direct impact on the economic efficiency. However, with 10–30% P recovery related to WWTP influent, the recovery potential is generally low. Heavy metals are present in only minor concentrations in digester supernatant; thus, the use of the resulting material as fertilizer would lead to low transfers of HM loads to agricultural soils (Figure 23). Considering the secondary treated effluent, a recovery of up to 50% is possible, if no specific P removal is applied at the WWTP (Figure 23).

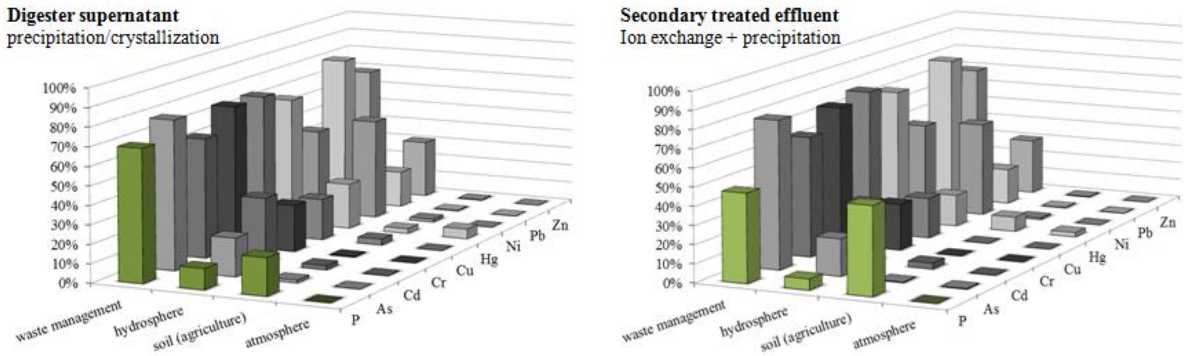
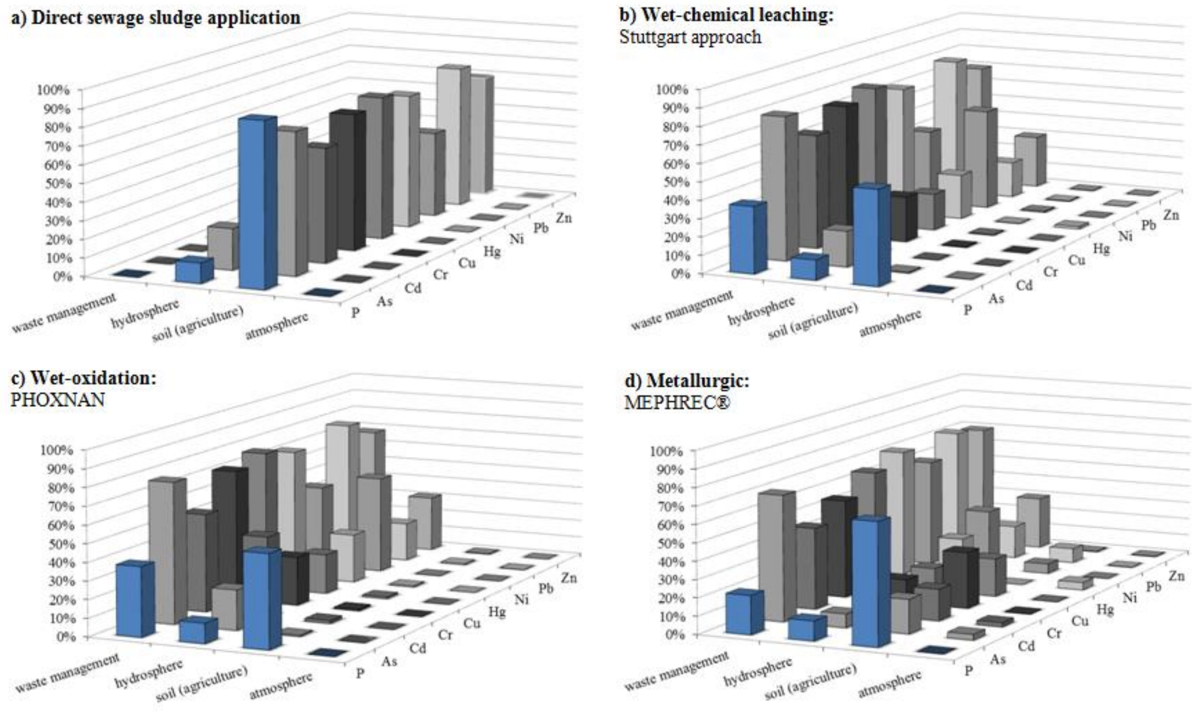


Figure 23: Distribution (% of total) of P and heavy metals from WWTP influents to environmental compartments for technologies that recover P from the aqueous phase (dissolved P in the digested sludge, digester supernatant, and secondary treated effluent).

To recover a greater extent of wastewater P (theoretically up to 90% of the WWTP influent) and to reduce the pollutant transfer to agricultural soils, technologies need to be implemented to process sewage sludge and SSA. In contrast to the aqueous phase, substantial concentrations of heavy metals are present in these two P-rich flows (see reference SS and SSA, Table 12). If depollution technologies are applied, the available technologies show clear differences with regard to P recovery and depollution potential. Wet-chemical leaching (**Stuttgart, Gifhorn and Budenheim process**), wet-oxidative (**LOPROX**) and SCWO approaches (**Aqua Reci®**) show very good depollution potential (up to 98% for all considered heavy metals) for sewage sludge. Therefore, the transfer of heavy metals to the recovered product and ultimately agriculture is low (Figure 24b, c). The recovery potential of these technologies is 45–65% of the P in the sludge and 40–60% of the P in the WWTP influent. The P that is not recovered remains in the treated (acidified) sludge or other inorganic residuals, which have to be disposed of. The P content in these residuals is therefore irretrievably lost. For the metallurgic **MEPHREC®** approach, an inconsistent behavior of heavy metals and high uncertainties of the effectiveness of depollution can be observed during the recovery process. The P transfer from SS is up to 70%, relative to the content in SS, or 65%, relative to the WWTP influent (Figure 24d).


a) Direct sewage sludge application

b) Wet-chemical leaching: Stuttgart approach

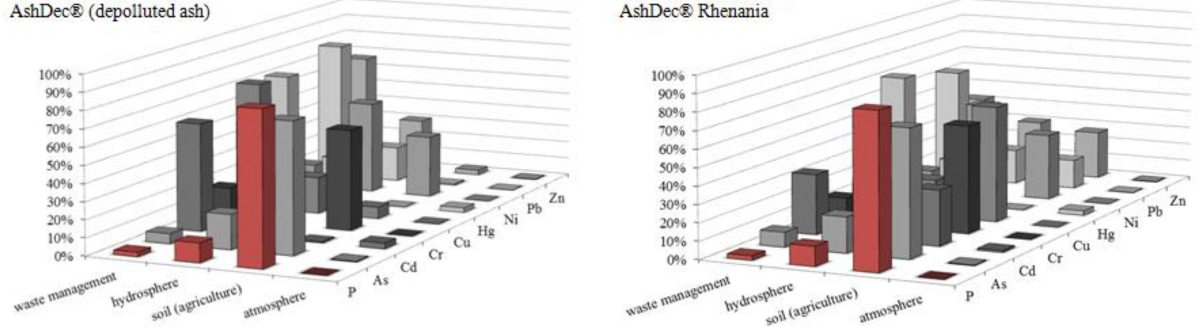
c) Wet-oxidation: PHOXNAN

d) Metallurgic: MEPHREC®

Figure 24: Distribution (% of total) of P and heavy metals from WWTP influents to environmental compartments for technologies that recover P from sewage sludge.

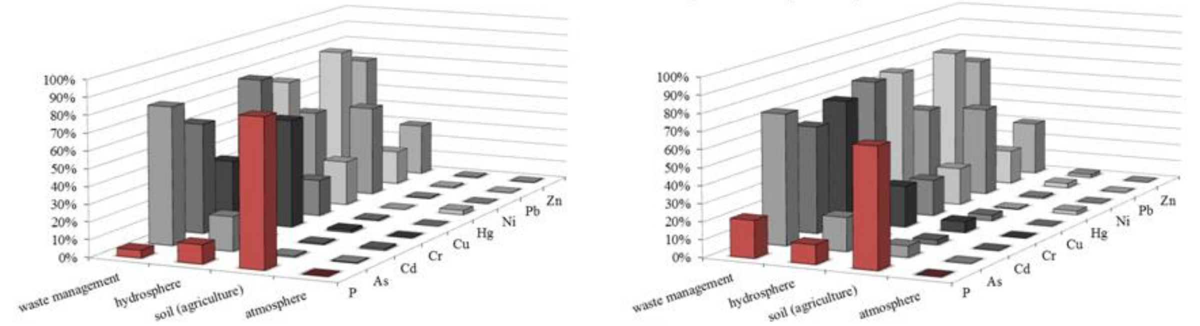
The MFA results for P recovery from SSA differ significantly between the different technologies. Good depollution rates for some HMs and simultaneous high recovery rates of ~98% can be achieved with both **AshDec®** processes. Some heavy metals, such as Cd, Cu, Pb and Zn, can be reduced by up to 90%, whereas others remain in the SSA at high percentages (As, Cr and Ni) (Figure 25a). However, the plant availability is critically discussed (Section 4.3.3.1). To improve the bioavailability, the chloride source can be replaced with sodium sulfate (**AshDec® Rhenania**). However, this substitution reduces the heavy metal removal (Figure 25b). From the input SSA, 95% of the P can be recovered as phosphoric acid with the **EcoPhos®** process. Interfering ions are stepwise removed with selective ion exchangers (~99%) (Figure 25c). Other wet-chemical leaching approaches show recovery rates of 70–80% relative to the ash input and 60–70% relative to the WWTP influent (**PASCH** and **LEACHPHOS®**). Due to the different dissolution properties of P and heavy metals at low pH values, the leaching step alone results in significant depollution with respect to Cr, Fe and Ni (70–90%), but little or no depollution is achieved with respect to Cd and Pb. With a specific depollution step (**PASCH**), recovery rates similar to **LEACHPHOS®** can be achieved but with significantly improved depollution with respect to Cd, Cu, Pb and Zn

(Figure 25d, e). The aim of the wet-chemical extraction approaches is the transformation of non-water-soluble P to water-soluble compounds. Thus, 100% of the P in SSA is “recovered” with **RecoPhos®** and within the **Fertilizer Industry**. However, 100% of the heavy metal load in the SSA is incorporated into the final product (Figure 25f). The industrial **Thermphos®** approach shows a similar recovery potential of ~95% (85% related to WWTP influent) by producing a pure P_4 applicable for multiple industrial purposes.



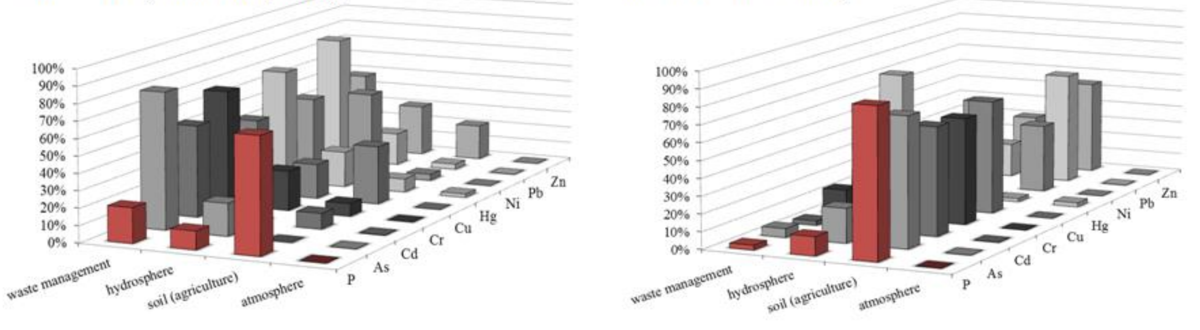
a) Thermo-chemical:
AshDec® (depolluted ash)

b) Thermo-chemical:
AshDec® Rhenania



c) Wet-chemical leaching
EcoPhos® (ion exchanger, P-acid)

d) Wet-chemical leaching:
PASCH (with HM depollution)



e) Wet-chemical leaching:
LEACHPHOS® (without HM depollution)

f) Wet-chemical extraction:
RecoPhos®/fertilizer industry

Figure 25: Distribution (% of total) of P and heavy metals from WWTP influents to environmental compartments for technologies to recover P from sewage sludge ash.

In Figure 26, the P recovery potentials related to the WWTP influent are summarized. In terms of WWTP effluent without specific P removal during wastewater treatment, up to 50% of influent P can be recovered. For technologies recovering P from the digester supernatant, considerable recovery rates of 10 to a maximum of 25%, with the exception of the **DHV Crystalactor®** with recovery rates of up to 40%, are only achievable in WWTPs with enhanced biological phosphorus removal. However, the number of WWTPs with pure EBPR is limited in many European countries. Regarding the extensive use of wastewater P (~90% of WWTP influent), sewage sludge and consequently SSA need to be addressed. Although possessing great theoretical potential, the realistic recovery rates of technologies that recover P directly from sewage sludge are considerably lower. To achieve the highest recover rates, SSA shows the best preconditions. For leaching technologies (**PASCH**, **LEACHPHOS®**), the recovery potential related to the WWTP input is limited to ~65–70%. Distinctly better recovery rates are achievable with the **EcoPhos®** process (> 80% relative to the WWTP input). In processes where SSA becomes a part of the final product (**AshDec®** technologies, **RecoPhos®**, **Fertilizer Industry**), the P recovery rate is in the range of 85% relative to the WWTP input. The advantages of an SSA strategy are the independent location of an incinerator and an inert P-rich ash that allows the implementation of centralized and greater P recovery units (economy of scale, see section 4.3.4.1).

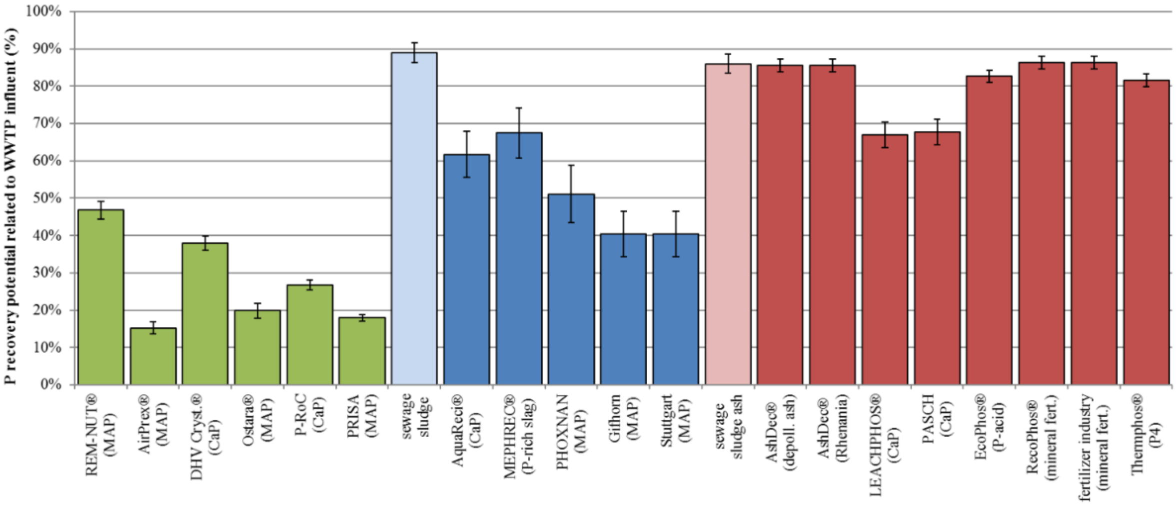


Figure 26: Summery of P recovery potential (%) of recovery technologies relative to the WWTP influent.

4.3.3 Characteristics of the recovered materials

4.3.3.1 Nutrient content and plant availability

The recovered materials from **REM-NUT®, Ostara®**, **PRISA**, **Gifhorn** and **Stuttgart approaches**, and **LOPROX** are MAP ($MgNH_4PO_4*6H_2O$) crystals. The P content of MAP is in the range of 10–12% (Figure 27), and it shows almost the same plant uptake efficiency as commercial fertilizers (RFE: 100%) in acidic soils and partially in alkaline soils, although it is not soluble in water (Kratz and Schnug, 2009, Pérez, 2010, Kratz et al., 2010, Wilken et al., 2015, Kratz et al., 2014). With regard to plant uptake within one or more growing periods, MAP also shows the same performance as commercial fertilizer (Römer, 2013). Calcium phosphate is produced by **DHV Crystalactor®, P-RoC®, Aqua Reci®, LEACHPHOS®,** and **PASCH**, and the P content is approximately 13–17% (Figure 27). Compared to MAP, the plant availability of the recovered calcium phosphate from the different technologies is not consistent. The bioavailability of the recovered calcium phosphate materials is significantly lower in neutral and alkaline soils, although the calcium phosphate material is soluble in citric acid, similar to MAP (Richards and Johnston, 2001). Compared to commercial mineral fertilizer (SSP or TSP), the effectiveness is only moderate in acidic soils and generally poor in alkaline soils (**DHV Crystalactor®, P-RoC®, Aqua Reci®, PASCH;** Weinfurtner, 2011). The calcium phosphate material recovered from the wet-chemical leaching process **LEACHPHOS®** shows good results in terms of solubility and good plant availability in both acidic and neutral soils (RFE: ~90%) (Morf, 2013).The RFE of products from the metallurgic **MEPHREC®** approach is 50–100% in neutral soils but low (0–25%) in an acidic soil (Cabeza et al., 2011; Wilken et al., 2015).

In both raw phosphate rock (PR) and untreated SSA, P is not water-soluble and requires further acid treatment (Krüger and Adam, 2014; Wilken et al., 2015). Untreated SSA exhibits differing fertilizer effects. Wilken et al. (2015) show fertilizing effects for untreated SSA; however, the RFE is < 20% in neutral soils (pH: 7.1) and < 50% in acidic soils (pH: 5.0).

In the materials recovered from SSA, e.g., the depolluted ash from **AshDec®**, a new mineral phase can be observed, possibly implying higher bioavailability of P (Mattenberger et al., 2010, Severin et al., 2013). Nanzer et al. (2014) report RFEs of 70–90% in acidic and neutral soils but only 4% on an alkaline soil. Nonetheless, Römer (2013) reports that the depolluted ash exhibits little improvement over the untreated SSA with regard to plant availability. Wilken et al. (2015) verify these findings in neutral soils but report improved availability in acidic soils (RFE: ~90%). By replacing the chlorine source (e.g., $MgCl_2$) with Na_2SO_4, this thermo-chemical approach provides a partly depolluted ash (Section 4.3.3.2) with significantly improved solubility and plant availability in neutral soils (RFE: 75%) and consistently good plant availability in acidic soils (RFE: 75–90%; Hermann, 2014; Herzel et al., 2015).

The **RecoPhos®** process produces an approved fertilizer with 16.6% P (adding P acid to SSA), and results similar to those of commercial fertilizer have been reported. For this approach, however, it is unclear if the available P originates from the ash or from the added phosphoric acid, which is water-soluble. To use SSA as a secondary raw material in the **Fertilizer Industry**, non-water-soluble P of SSA is partially transformed into water-soluble compounds using sulfuric acid. The iron and aluminum contents, resulting from chemical P removal during wastewater treatment, and the ash/H_2SO_4 ratio affect the conversion of non-water soluble P to water soluble P (water solubility: untreated ash: 0–1%; Fe-ash treated with acid: 40–55%; Al-ash treated with acid: 50–90%; Petzet and Cornel, 2011). Phosphoric acid (**EcoPhos®**) is fully plant available. As P acid, pure P_4 from the **Thermphos®** process is a universally applicable raw material in the fertilizer, food, feed, and chemical industries. Figure 27 gives an overview of the macronutrient contents (P, N, Ca, and Mg) of the recovered materials with the ratio of actually plant available P in relation to the total P content (RFE).

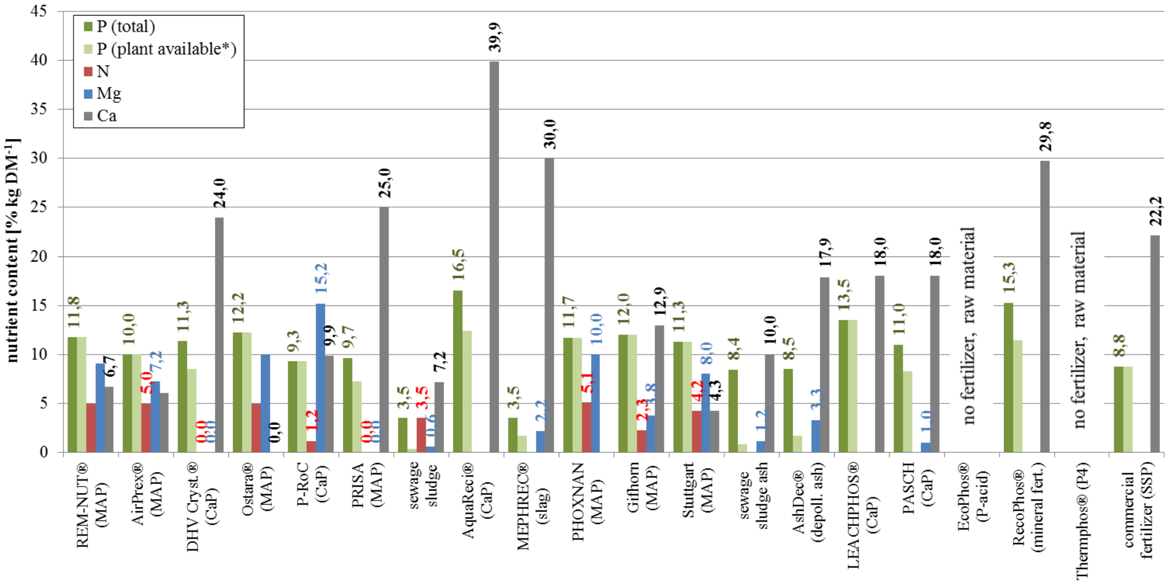


Figure 27: Macronutrient contents (P, N, Mg, and Ca) of the recovered materials and a commercial fertilizer and the plant availability of the P in acidic and alkaline soils.

4.3.3.2 Pollutant contents

4.3.3.2.1 Heavy metals

All the recovered materials exhibit significantly lower damage unit values relative to untreated sewage sludge, ash (SS: DU_P = 0.5, SSA: DU_P = 0.35) and even commercial fertilizer (Single Superphosphate, DU_P = 0.23) (Figure 28). Due to the low evaporation temperature of mercury leading to the transfer of mercury to the flue gas (> 95%), SSA shows lower concentrations and consequently lower damage unit values compared to SS. Heavy metals are present in only minor concentrations in digester supernatants; thus, the recovered materials show very low DU_P (< 0.02). A comparatively higher DU_P for the **PRISA** process results from the high Zn concentration in the recovered MAP material (Montag, 2008). Products from the oxidation and wet-chemical leaching technologies applied to SS exhibit similar damage units to products derived from digester supernatant (< 0.03), even though sewage sludge contains greater pollutant contents. This is a result of the good pollutant elimination rate and the greater P recovery rate, as the damage unit is also based on the amount of recovered P. A DU_P in the range of 0.05 or less for materials derived from ashes is possible with thermo-chemical or the acidic wet-chemical leaching technologies with a specific HM removal step (**PASCH, EcoPhos®).** For the metallurgic **MEPHREC®** approach and technologies that recover P from SSA little or no depollution, the damage unit values are still less than those of a SSP. For the **RecoPhos®** approach, a low damage unit is the result of mixing SSA with phosphoric acid and the consequent enrichment of P. The DU_P for SSA integrated into the **Fertilizer Industry** without a HM removal step and extraction with sulfuric acid is the same as for SSA (DU_P: 0.35).

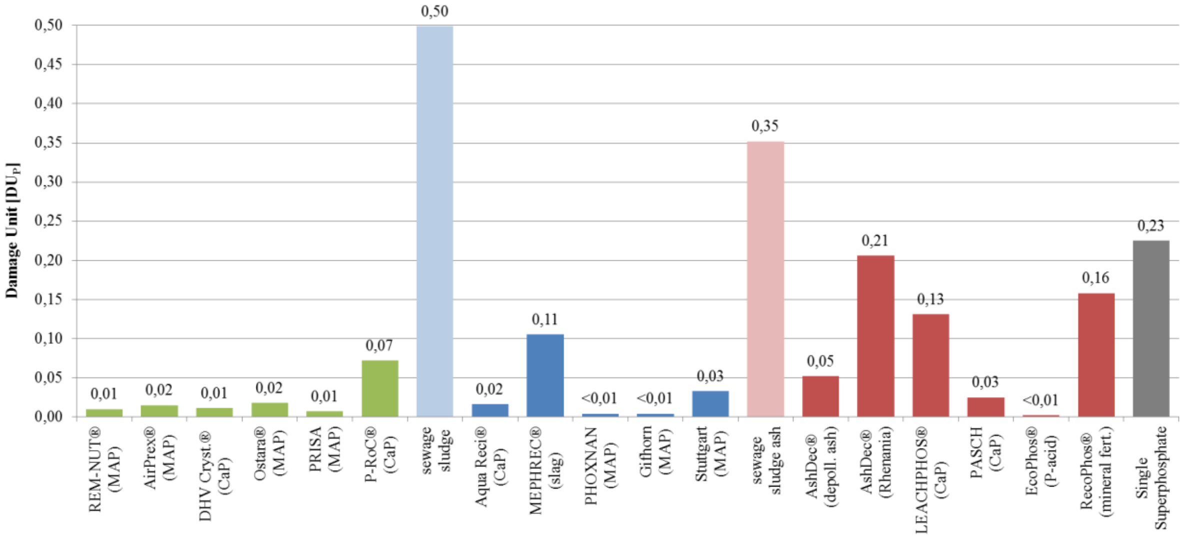


Figure 28: Damage unit (DUP) values for recovered materials, reference sewage sludge/ash and commercial fertilizer (SSP).

Due to the good depollution rates, the recovered materials can be applied more often than a SSP (except **MEPHREC®**) and SS/SSA until a limiting heavy metal concentration in the soil is reached (Figure 29). For the SSP, Cd, a heavy metal with great damage potential, limits the application to 1,450 a. For SS and SSA, the limiting heavy metals are Cu and Zn, respectively, but Cd is present in both at only micronutrient levels. The DU_P values indicate that most materials based on P recovered from SSA exhibit higher pollution potentials than those derived from most other P sources, and the RSM shows similar results. For most of the recovered products derived from SS, Cu and Zn are the limiting elements, contrary to SSA products, where Ni is the limiting element for two technologies (**DHV Crystalactor®** and **AshDec® depollution**). Figure 29 presents the application factor for the recovered material compared to SSP, for which Cd is the limiting element. This factor shows how many times more the recovered material can be applied compared to a SSP until the defined critical Cd concentration is exceeded.

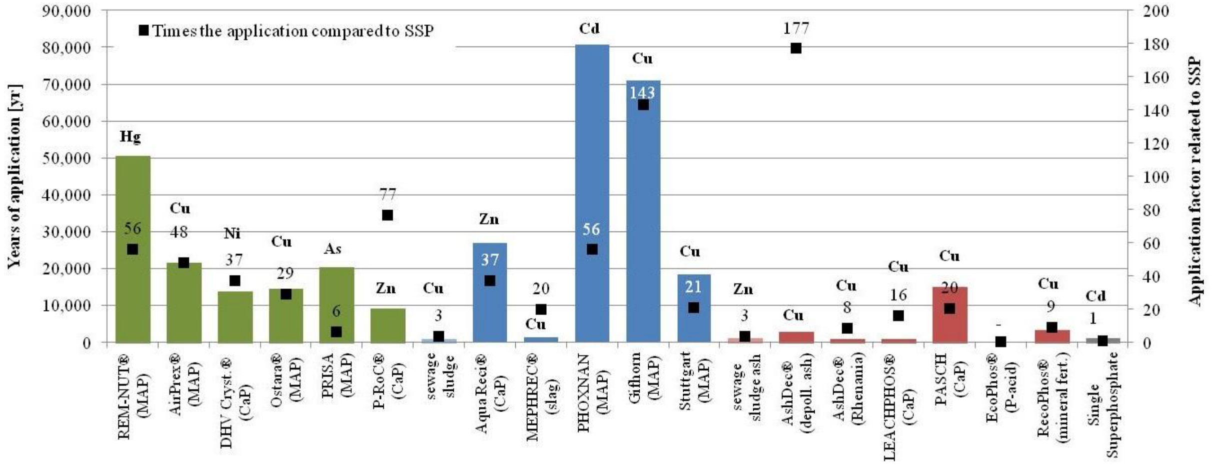


Figure 29: Years of application of the recovered materials, reference sewage sludge/ash and commercial fertilizer until a defined critical concentration is reached, including the limiting heavy metal and the number of times the material can be applied relative to SSP if only Cd is the limiting element (black square + number).

From the existing literature, no material produced by the investigated recovery technologies (except raw SS and SSA) has ever reached the heavy metal limit values in the fertilizer regulations (EC, 2003).

4.3.3.2.2 Organic micropollutants and pathogens

Little or no OMs or pathogens have been detected in recovered material from digester supernatant (Montag, 2008; Pinnekamp, 2011), secondary treated effluent (Ueno and Fujii, 2003) and anaerobically digested sludge (Heinzmann and Lengemann, 2011). This is, in part, a result of the already low concentration of OMs in these P-rich flows. Additionally, during precipitation or crystallization of dissolved P, these pollutants are minimally incorporated into the crystal structure of the precipitants, e.g., struvite (Ronteltap et al., 2007; Uysal et al., 2010; Antakyali et al., 2011).

For the wet-chemical **Gifhorn** and **Stuttgart processes** from sewage sludge, organic micropollutants can be detected but at significantly lower concentrations than in the raw SS (Weidelener, 2010; Günther, 2011; Pinnekamp, 2011). With wet-oxidative or metallurgic technologies, pathogenic microorganisms and OMs are destroyed completely or to a great extent. The pathogens and OMs are destroyed when SS is incinerated properly without mixing with other wastes. The formation of dioxins and furans is possible, but generally these compounds are not detectable or detectable only in small quantities in ashes derived from mono-incineration of sewage sludge (< 1 ng kg ash^{-1}; Krüger and Adam, 2014) (Table 13).

Table 13: Concentration of organic micropollutants in sewage sludge, SSA, recovered materials from P recovery processes and mineral fertilizers (for missing technologies, no OM data are available).

	AOX	B(a)P	LAS	NPE	PCB	Phthalates	Σ PAH 16	PCDD/F
				[mg kg DM^{-1}]				[ng kg DM^{-1}]
SS[1,2]	180	-	2,000	30	0.05–0.1	-	6-16	3,000
SSA[3]	< 10	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	< 1
REM-NUT®[4]	n.d.	n.d.	n.d.	n.d	n.d.	n.d.	n.d.	n.d.
PRISA[5]	84	-	< 0.1	0.5	-	-	< 410	-
P-RoC®[6]	-	0.001	10	-	-	0.02	-	-
AirPrex®[7]	55	-	-	-	0.03	-	-	0.00002
PHOXNAN[6]	-	n.d.	n.d.	-	-	-	-	-
Gifhorn[6]	197	0.005	40	2	-	1	-	-
Stuttgart[8]	-	0.005	0.7	2.1	< 10	-	60	-
PASCH+	-	-	0.3	-	-	0.05	-	-
Min. fertilizer[1]	-	0.009	138	0.03	0.001	1	-	-

n.d. (not detectable), [1]Kördel and Herrchen (2008), [2]Scharf et al., 1997, [3]Adam (2007), [4]Ueno and Fuji (2003), [5]Montag (2008), [6]Pinnekamp (2011), [7]Heinzmann and Lengemann (2011), [8]Weidelener (2010)

4.3.3.2.3 Texture and handling

As all the materials produced by the different recovery technologies are inorganic, storage is possible. Nevertheless, the criteria for direct application with modern spreaders are currently only fulfilled by certain technologies (e.g., **Ostara®, DHV Crystalactor®, RecoPhos®, AshDec®** output after granulation, and the **Fertilizer Industry).** For most other technologies, the recovered material is coarse grained or powdery and dusty. Therefore, further treatment processes, e.g., granulation, are needed (Table 14). The output of the **EcoPhos®** process is a concentrated, marketable, liquid phosphoric acid.

Table 14: Grain size and appearance of recovered P materials/products.

Grain size/appearance	Technology
Pellets/Grain size 2–5 mm (Market ready)	Ostara®, DHV Crystalactor®, AshDec® as granulated PhosKraft® fertilizer, RecoPhos®, Mineral Fertilizer from Fertilizer Industry
Coarse-grained (Market ready)	PRISA, AirPrex®, P-RoC®
Crystalline, powdery (Not market ready)	REM-NUT®, Aqua Reci®, PHOXNAN, Gifhorn, Stuttgart, LEACHPHOS®, PASCH, untreated output from the AshDec® technologies, pure SSA
Liquid (Market ready)	EcoPhos®
Solid (Market ready)	Thermphos®

The results from Section 4.3.1 to 4.3.3 are summarized in Table A 22.

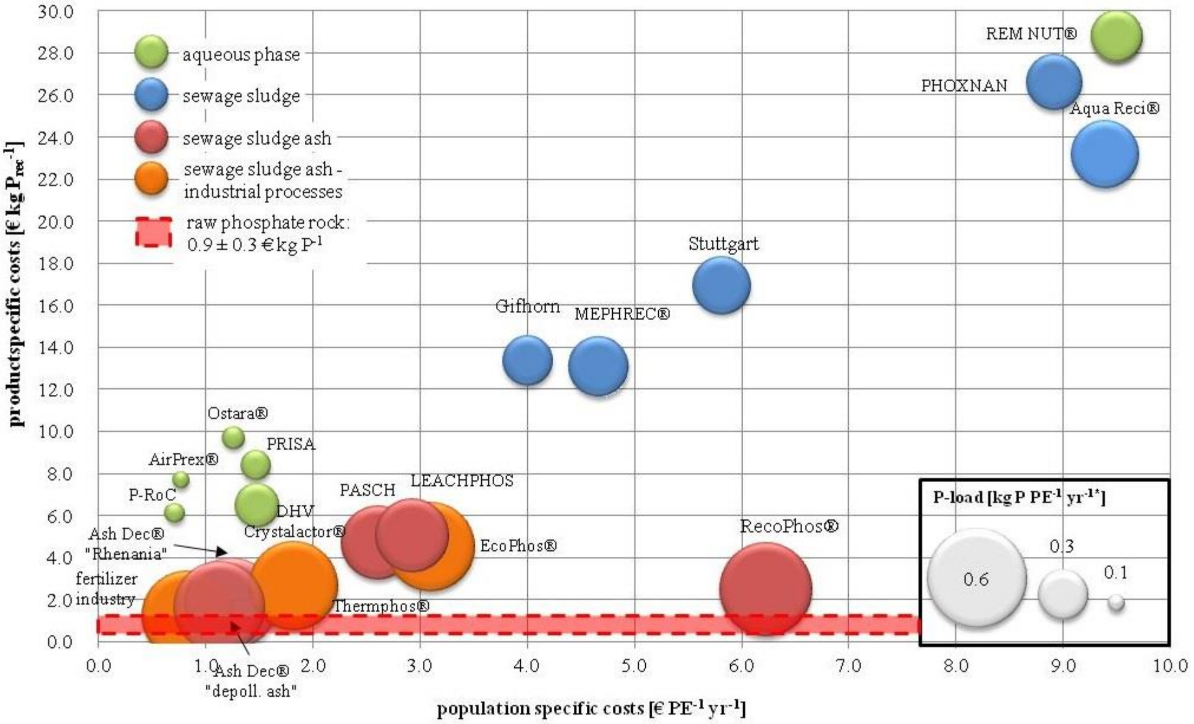
4.3.4 Economic assessment

4.3.4.1 Annual costs of the recovery technologies

With more than 28 € kg P_{rec}^{-1} the **REM-NUT®** approach to recover P from the effluent of a WWTP is the most expensive technology. These extremely high costs result from the great need of resins and chemicals. The annual costs, without savings and revenues, of recovering P from digester supernatant, where P is already present in its dissolved form, are approximately 6 to 10 € kg P_{rec}^{-1} or 0.8 to 2 € PE^{-1} yr^{-1} (**Ostara®**, **DHV®**, **PRISA**, **P-RoC®**, Figure 30). The higher the concentration of dissolved P in the supernatant, the lower the product specific costs. The detailed cost analysis shows that the costs are mainly driven by the investment costs for equipment, such as crystallization reactors. By installing such a reactor in larger treatment plants, the investment costs can be reduced significantly (up to -50% for 200,000 PE) due to the economy of scale. By taking into account possible savings, such as the reduction of P back-flow (reduced demand for flocking agents), avoiding MAP encrustations (and associated maintenance costs) and revenues from the produced fertilizer, these technologies may operate economically. By applying the **AirPrex®** technology immediately after the digester tank, the dewaterability of the digested sludge will be improved due to the aeration of the sludge and the significant reduction of dissolved P by the formation of MAP within the sludge (~90% PO_4^{3-} removal). As disposal costs for sewage sludge are one of the main costs for a WWTP in Central Europe, this technology pays for itself from an economic perspective.

Recovering P from sewage sludge is generally more expensive than recovering P from supernatant. For example, the cost of 1 kg P_{rec}^{-1} produced via the wet-chemical processes is 9–16 € (**Gifhorn** and **Stuttgart processes**). These costs are dominated by the required chemicals, including acids, caustics, and complexation and precipitation agents, and will not be significantly reduced if scaled up. The costs of the wet-oxidation processes, such as **Aqua Reci®** and **PHOXNAN,** are outstandingly high (23–27 € kg P_{rec}^{-1}). However, taking into account revenues, e.g., by using the heat potential of the sludge, converting sludge to an inorganic product and the value of the product, the overall costs will decrease dramatically. Furthermore, in addition to having a recovered material, the output is a disposable inert waste. Thus, further treatment, such as incineration, is not necessary (Figure 31). Regarding the whole process chain, these technologies could be economical. Similar results have been observed for the **MEPHREC®** process.

The costs of the wet-chemical leaching processes that recover P from SSA are approximately 5–6 € kg P_{rec}^{-1} or 2.5–3.0 € PE^{-1} yr^{-1}. These product-specific costs are also valid for the industrial **EcoPhos®** approach. Depending on the scenario (“hot ash” or “cold ash”), the annual cost of the **AshDec® depollution** and **AshDec® Rhenania** technology is approximately 2 € kg P_{rec}^{-1}. Similar results are observed for the **RecoPhos®** process. The high population-specific costs of > 6 € result from the application of expensive phosphoric acid. Due to the enrichment of the ash with P, the product-specific costs are in the range of 2–3 € kg P_{rec}^{-1}. To use the SSA in industrial processes, such as the **Fertilizer Industry** and **Thermphos®**, the assumption is that the ash is used in existing plants; therefore, only the operational costs for the resources were calculated. In this case, the costs of the **Fertilizer Industry** and **Thermphos®** are approximately 1 and 2 € kg P_{rec}^{-1}, respectively.



* Bubble size indicates the recoverable P load in kg P per population equivalent per year. The maximum annual recoverable load of P is 0.66 kg PE^{-1} yr^{-1} or 65,700 kg (reference WWTP).

Figure 30: Product- and population equivalent-specific annual costs for P recovery technologies without savings and revenues from digester supernatant/effluent (green), sewage sludge (blue), sewage sludge ash (red) and recovery of P from SSA in industrial processes (orange).

4.3.4.2 Integrated cost calculation including sensitivity analyses

4.3.4.2.1 Reference system

For the integrated economic technology assessment, the costs for the whole reference system were first identified, yielding an annual cost of approximately 11.1 € PE^{-1} yr^{-1}. Additionally, approximately 1.2 € PE^{-1} yr^{-1} (including capital- and operational expenditure) must be considered for the mono-incineration of sewage sludge compared to co-incineration (12.3 € PE^{-1} yr^{-1} total cost with mono-incineration and disposal of fly ash in a landfill). The details of these costs are shown in Table A 20.

4.3.4.2.2 Recovery technologies

In contrast to the calculation of the cost of the recovery technology itself, the required costs and the possible savings associated with the integration of the recovery technology into the whole wastewater and sludge disposal system are taken into consideration here. Figure 31 presents the economic costs associated with the implementation of recovery technologies into the reference system and displays the margin of fluctuation. Additional costs or savings with regard to the reference system (11.1 € PE^{-1} yr^{-1}) are given as percentage deviations.

The **REM-NUT®** technology is the most expensive technology with regard to one kg of recovered P and one kg PE^{-1} yr^{-1} (Figure 30). Based on the implementation in the defined reference system, additional costs in the range of 50–65% need to be considered. With annual costs of 0.5–1.5 € PE^{-1} yr^{-1}, the **AirPrex®**, **DHV Crystalactor®**, **Ostara®**, **PRISA** and **P-RoC®** processes are significantly less expensive (Figure 30). The implementation of the **AirPrex®** technology to recover dissolved available P directly from digested sludge results in savings due to reduced back-flow of nutrients and improved dewaterability of the sludge. Together with revenues from product sales, this technology is economical, as the revenues and savings exceed the annual costs. For the best-case scenario, the costs can be reduced by 6% compared to the reference system. With the implementation of the recovery technologies **Ostara®**, **PRISA** and **P-RoC®**, the nutrient back-flow via digester supernatant to the WWTP influent can be reduced significantly. Taking into account these savings, the maximum revenues from product sales and the implementation of this technology in a larger WWTP, the revenues and savings exceed the annual costs (Figure 31). In relation to the cost of the reference system, savings of 1–2% are possible. In comparison, due to the high investment costs and great resource demand, the **DHV Crystalactor®** does not operate economically, even with maximum revenues, maximum savings and an up-scaling of the plant to

500,000 PE. Compared to the reference system, additional costs in the range of 25–30% need to be considered.

High annual costs were shown for the **Aqua Reci®** and the **MEPHREC®** processes as standalone processes (Figure 30). However, due to the use of the energetic potential of the sludge and the simultaneous mineralization (incineration can be omitted), reduced costs can also be shown for the whole process chain. Therefore, an economical performance is possible under favorable conditions. However, especially for these technologies, the uncertainty in the cost calculation is particularly high, as the data on the energy yield is subject to high degrees of uncertainty. The wet-chemical sewage sludge leaching technologies (**Gifhorn** and **Stuttgart**) are expensive and increase costs by up to 20–50% compared to the defined reference system. For these leaching technologies, the effect on costs due to possible up-scaling is low, as the costs are dominated by the necessary chemicals. These high costs are particularly noteworthy when compared to the low/moderate recovery rates (40–50% of P with respect to WWTP influent, Section 4.3.2).

Focusing on recovery technologies from SSA, the possible additional costs associated with the mono-incineration of SS were considered (+1.2 € PE^{-1} yr^{-1} or +11% of the total costs of the reference system). The additional costs for both thermo-chemical **AshDec®** (ash depollution and “Rhenania ash”) technologies are in the range of +5–20% compared to the defined reference system. For these technologies, the possible revenues are especially important, as the output is either a depolluted ash with hardly any improved plant availability (revenue: 1 € t ash^{-1}) or a “Rhenania ash” with significantly improved plant availability and consequently higher revenues (≥ 50 € t ash^{-1}). The additional costs compared to the reference system are high (+20%) in association with low revenues, which is the most realistic scenario for the **AshDec®** technology, but lower (+5%) in association with the **AshDec® Rhenania** technology. For recently developed wet-chemical leaching technologies (**PASCH**, **LEACHPHOS®**), the additional costs compared to the reference system are 20 to 35%. Even with the highest possible revenues, the additional costs are +20%. For technologies such as **RecoPhos®** or existing industrial processes where the output is an already marketable product, high revenues are possible. In situations where they can be implemented, these technologies utilized with almost no (**EcoPhos®**, **Fertilizer Industry**) or only small additional overall costs (**Thermphos®**) compared to the reference system. Therefore, by taking the whole process chain, with the necessary and more expensive mono-incineration plants, into account and by assuming maximum revenues for the recovered products of the industrial processes in the best case scenario, no additional costs need to be considered from a macroeconomic perspective. These results are summarized in the overall economic assessment (Table A 23).

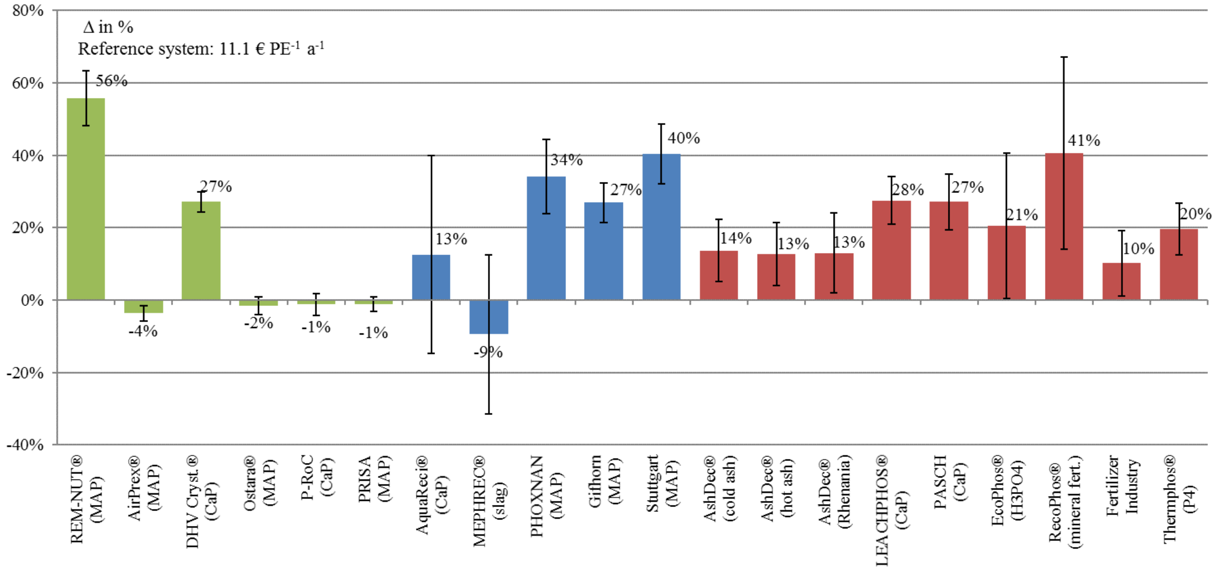


Figure 31: Range of costs in % of the total costs of the reference system for P recovery technologies taking into account possible savings and revenues.

4.3.5 Technical maturity

In recent years, technologies designed to recover the dissolved P from digested sludge and digester supernatant have been successfully implemented in North America, Central Europe and Japan. Three of these full-scale implemented technologies are considered in this work (**AirPrex®**, **DHV Crystalactor®**, **Ostara®**). To recover P from the secondary treated effluent **(REM-NUT®)**, no pilot plant or full-scale operation is currently known.

For technologies with an oxidation and a subsequent P recovery unit that have already been tested at a pilot-scale level (**Aqua Reci®** and **PHOXNAN**), no further development is presently recognizable. In comparison, a pilot plant using the metallurgic **MEPHREC®** approach is currently under construction (Nürnberg, Germany). The leaching technology **Gifhorn** has been implemented at the full-scale in a small WWTP in Gifhorn (50,000 PE), while the **Stuttgart process** is currently implemented at the pilot-scale level (Offenburg, Germany).

No further development beyond the known pilot plant installations in Leoben (Austria) is known for the two thermo-chemical **AshDec®** approaches and the leaching approaches **PASCH** and **LEACHPHOS®** (Zurich, Switzerland). In fact, it remains unclear which technology (e.g., precipitation, solvent extraction, ion exchangers) is the most suitable for removing interfering ions, such as metals and heavy metals, that are dissolved with P in the leaching step. With the beginning of the construction of an industrial-scale plant in Dunkerque (France) and the intended integration of SSA to produce phosphoric acid, **EcoPhos®** seems to be a promising industrial technology for P recovery. The chosen method to remove interfering ions is ion exchange. A similar approach with the application of different ion exchangers is applied by Remondis (**TetraPhos®**; pilot-scale implementation in Hamburg, Germany). Generally, the **fertilizer industry** could be a promising method to integrate SSA into the production process to generate high-quality products, with respect to P and heavy metal contents, (e.g., ICL Fertilizers®, Netherlands). The **RecoPhos®** process, which uses a similar procedural approach as the fertilizer industry (acidic ash extraction with phosphoric acid), has already been implemented at the full-scale with a production capacity of 4.000 t. However, this plant is no longer in operation for unknown reasons. A detailed overview of the technology readiness levels and an outlook on the development potential of the considered technologies are given in Table A 19.

4.3.6 Overall uncertainty in the data on the technologies

The uncertainty and reliability of the used data were critically reviewed, as model results can only be as good and/or true as the input data are. The data originates from sources with differing quality and in some cases no data were available. For most of the technologies, reliable data on the resource demand, MFA for P and HM, nutrient and pollutant contents, solubility and plant availability were available, even for technologies at the laboratory or pilot scale. In comparison, the economic technology assessment was challenging for technologies at the laboratory or pilot scale, as investment costs and therefore capital costs were unknown. Although data from feasibility studies are available, especially for technologies that recover P from SSA, moderate or high uncertainties were considered, as there is no practical experience based on full-scale implementation or long-term studies. In Table A 12 and A 13, the origins of the information and data, along with their uncertainties (classified according to the qualitative uncertainty concept) are described in greater detail. Table 15 provides a comparative overview of the reliability and robustness of the data for each technology based on the qualitative uncertainty concept. The overall uncertainty is the mean value of the summarized uncertainties according to Table A 12 and 13 (+: 1; o: 2; -: 3; and --: 4).

Table 15: Overall uncertainty of the data for the P recovery technologies.

Aqueous phase	Unc. of data	Sewage sludge	Unc. of data	Sewage sludge ash	Unc. of data
REM-NUT®	o/-	Gifhorn	+	AshDec® depollution	+/o
AirPrex®	+	Stuttgart	+/o	AshDec® Rhenania	o
DHV Crystalactor®	o	PHOXNAN	+	PASCH	+
Ostara®	+	Aqua Reci®	o	LEACHPHOS®	+
P-RoC®	+	MEPHREC®	o	EcoPhos®	o
PRISA	+			RecoPhos®	+
				Fertilizer Industry	+/o
				Thermphos®	o/-

As costs are a very important criteria for the implementation of a technology, the uncertainty in the resource and energy demand, the quantity of the recovered material, the yield of energy due to certain procedural approaches and the resulting waste for each technology from the aqueous phase (Table A 14), sewage sludge (Table A 15) and sewage sludge ash (Table A 16) is given in the appendix. Depending on the applied data quality, the robustness and reliability of the assessment results are discussed.

4.4 Discussion

4.4.1 Paths of P and heavy metals

MFA was successfully applied to a defined reference system. This enables the tracking of P and heavy metal pathways along the whole route from the input wastewater, through sludge treatment processes or P recovery technologies to a recovered material or a waste product. With regard to P, many studies have shown similar recovery rate results for the technologies, as well as relative to a specific WWTP (Hermann, 2009; P-Rex, 2015).

The fate of P and heavy metals in the recovery approaches have been investigated to date. The methodology applied in this work is unique as it broadens the perspective to the recovery and depollution potentials for P recovery technologies within the whole wastewater and sludge disposal system. With this approach, we identify the percentages of wastewater heavy metals that end up in the different final sinks are identified (e.g., agricultural soils, landfills, and water bodies). Thus, the different technologies become directly comparable to each other. The results are particularly interesting with respect to the question of the long-term acceptable total load of heavy metals in agricultural soils. This work showed that most of the recovery technologies, with the exception of the wet extraction processes (**Fertilizer Industry, RecoPhos®**) achieve a significant reduction in pollutants compared to the raw SS or SSA input material. In the selected approaches, heavy metals are only diluted in the product compared to the SSA. It can be noted that the data used to construct the material flow models and model the path of P and heavy metals is very good, with the exception of particular technologies (e.g., **MEPHREC®**, **Aqua Reci®**), and reliable results with low uncertainty can therefore be generated for most of the investigated technologies.

However, it must be kept in mind that most of the newly developed P recovery technologies were tested under very limited variations of the boundary conditions e.g., on WWTPs with different properties or only on one type of SS/SSA. P recovery rates, HM loads etc. and therefore the data base (input data) used in the MFA are very limited and do not represent a broad spectrum of applications. Furthermore, from certain recovery technologies wastewater occurs, which could affect a WWTP negatively. Exemplary, heavy metal rich wastewater is generated from a process. The question arises, how higher heavy metal inputs affects transfer coefficients within a WWTP. Even if enrichment is possible within the whole process, it cannot be illustrated, due to the fact, that this MFA was not performed as a time series. Consequently, uncertainties need to be considered.

4.4.2 Quality of the recovered materials and products

As shown with the comparative literature study, compared to commercial mineral P fertilizers, all the recovered materials show poorer or even no solubility in water (similar to untreated raw PR: 1–5%; Weinfurtner, 2011). However, almost all products, even untreated SSA in acidic soils, increase the agricultural yield. Therefore, the results from standard solubility tests (e.g., water, citric acid, and neutral/alkaline ammonium citrate) are often not transferable to the real plant uptake in the field. Field trails demonstrate that struvite and different forms of calcium phosphate, for example, have a relative fertilizer efficiency in neutral soils comparable to a water-soluble commercial SSP, even though the recovered fertilizers show no water solubility (Wilken et al., 2015). However, studies reveal that the plant availability is not solely influenced by the quality of the recovered product. In fact, natural soil properties, such as pH, P supply and type of vegetation, significantly influence the plant uptake (Weinfurtner, 2011). Therefore, further field trials are required to examine their actual fertilizing effect and especially their long-term behavior.

The applied methods to assess the pollution potential of the recovered materials reveals that, with the exception of **RecoPhos®** and the **Fertilizer Industry**, each recovery technology is able to reduce heavy metals significantly compared to the starting input material sewage sludge or SSA. Furthermore, the pollution potential of each recovered material relative to its P content is lower compared to commercial P fertilizers. These findings are confirmed by the comprehensive toxicity and risk assessment of various recovered products within the European P-Rex study (Kraus et al., 2015).

Even in simple lab trials or pilot plants, most of the technologies generated a recovered material with properties similar to those expected for a full-scale implementation. This is due to the fact that the chemical principles do no change due to up-scaling for most of the technologies. The recovered materials from the considered technologies have been tested in many cases and even multiple times by independent laboratories to evaluate the solubility, plant availability, and nutrient and pollutant contents. Therefore, the results from the assessment of the recovered materials feature low uncertainties. These findings are supported by the DU_P and application factor results.

For several recovered materials, data on organic micropollutants and pathogens are incomplete or absent. For some sludge integration methods, such as wet chemical oxidation and super-critical water oxidation, data on the removal and destruction of OMs and pathogens are available in the general literature but not specifically for the recovery technologies. Therefore, greater uncertainties are involved, and further investigations are necessary in this field of research to complete the analysis of the recovered materials. In comparison, the incineration of sewage sludge at > 800 °C almost entirely destroys OMs and pathogens. In the case, although no data are available for the recovered materials from SSA, and it can be assumed with a high degree of certainty that the OM and pathogen contents are very low or even below the detection limit.

4.4.3 Economic assessment

The question of economic P recovery is discussed intensively. This work reveals that recovery of P can be cost-neutral under certain boundary conditions if dissolved P is recovered as struvite or calcium phosphate from the aqueous phase (digester supernatant or dissolved P fraction within the digested sludge) of a WWTP. The positive effects of reduced nutrient back-flow, prevention of maintenance costs, and improved dewaterability account for the largest share of the economic operation of a P recovery technology. Additionally, an economic operation is possible with processes that utilize the energy content of the sewage sludge, destroy the organic content of the sludge, and simultaneously recover a P-rich material. However, for these technologies, the associated uncertainty is very high, as it is very difficult to foresee the actual revenues from heat, electricity or gas generated by technologies with no full-scale implementations currently in existence.

When discussing the cost-effectiveness, the argument is that the recovery technologies should compete with the price of raw PR (0.9±0.3 € kg P^{-1}; World Bank, 2016). However, untreated PR is not water-soluble and therefore not immediately plant available without further treatment. In comparison, recovered materials, such as struvite, have a fertilizing efficiency comparable to a triple superphosphate worth 1.7±0.5 € kg P^{-1} (World Bank, 2016). Within this price range, SSA can be (partly) depolluted with the output of a “Rhenania phosphate” or ash can be treated with phosphoric acid to produce a commercial fertilizer.

The costs of P recovered from SSA are significantly higher than the costs of direct P recycling by applying SS to agricultural fields or in biological sludge treatments, such as composting (Wiebke and Pinnekamp, 2011). This option is not applicable everywhere and is restricted in several European countries (Netherland, Switzerland and likely Germany will set similar regulations). For cities and regions where infrastructure with mono-incinerators (e.g., Austria, Switzerland, Germany, Netherland) is available and a P-rich ash is already generated, little or no addition costs are necessary to recovery of P from SSA compared to disposal routes if maximum revenues are taken into account.

Operational costs can be calculated rather simply, even for technologies with a low technical readiness level. Therefore, the uncertainty for these costs is low. This low degree of uncertainty is based on the profound knowledge of the material flows and resource demands of the recovery technologies. A weak point of the performed economic assessment is the calculation of the capital costs for technologies with a low TRL, as investment costs are unknown at this stage of development.

Possible savings (e.g., reduced nutrient back-flow and improved dewaterability) and additional costs for the disposal of generated wastes could be calculated with low uncertainties, as the detailed material and substance flow analysis forms a good basis. Compared to the savings, the revenues from the recovered materials are highly uncertain, as there is no existing market for these P-rich secondary raw materials. Therefore, the presented results of the sensitivity cost calculation are a recent snapshot and present the range of costs for a technology. The assessment of the technologies in this work is based on a defined reference WWTP. Therefore, variations in the results either in a positive or negative direction are unavoidable. It must be considered that even the characteristics of the wastewater and the quality of SS and SSA can vary significantly with respect to the nutrient and pollutant contents. This variability therefore affects the depollution processes, product quality and costs of the recovery technologies.

4.5 Conclusions

The question of application of P recovery technologies is especially relevant for countries where agricultural reuse of sewage sludge is not currently accepted and/or is restricted by legislation. A large number of technologies have already progressed to commercially working implementations, and others could be implemented without restrictions from a technical perspective. Still, an ideal recovery technology cannot be presented.

As the field of P recovery from municipal wastewater is a young research topic, the evolution is very dynamic. Therefore, changes due to future developments are expected. Nevertheless, the methodological approach presented in this paper is appropriate and applicable to holistically assess P recovery technologies for themselves, but also within existing systems or structures of wastewater and sludge treatment, as well as the disposal of the resulting wastes. This methodology allows already existing and assessed technologies to be compared.

This assessment could be performed with a robust dataset for most of the considered technologies. For 12 out of the 19 technologies, the uncertainty of the dataset can be classified as low or low/moderate. Only two technologies are associated with moderate to high uncertainties. Coincidently, no further development is presently identifiable for these technologies. As an essential precondition to performing this integrated assessment, detailed databases are required for the resource demand, the paths of P and heavy metals, and investment costs. Incomplete or absent data exist for the quality of the recovered materials with regard to organic micropollutants, pathogens and in some cases the fertilizer efficiency. With respect to the economic assessment, investment costs for technologies with a low TRL level are difficult to predict at this stage of development, and the considered capital costs need to be critically reviewed. Generally, due to low TRL values and unknown market situation, the savings and revenues are associated with great uncertainties, especially for technologies that recover P and energy from sewage sludge simultaneously.

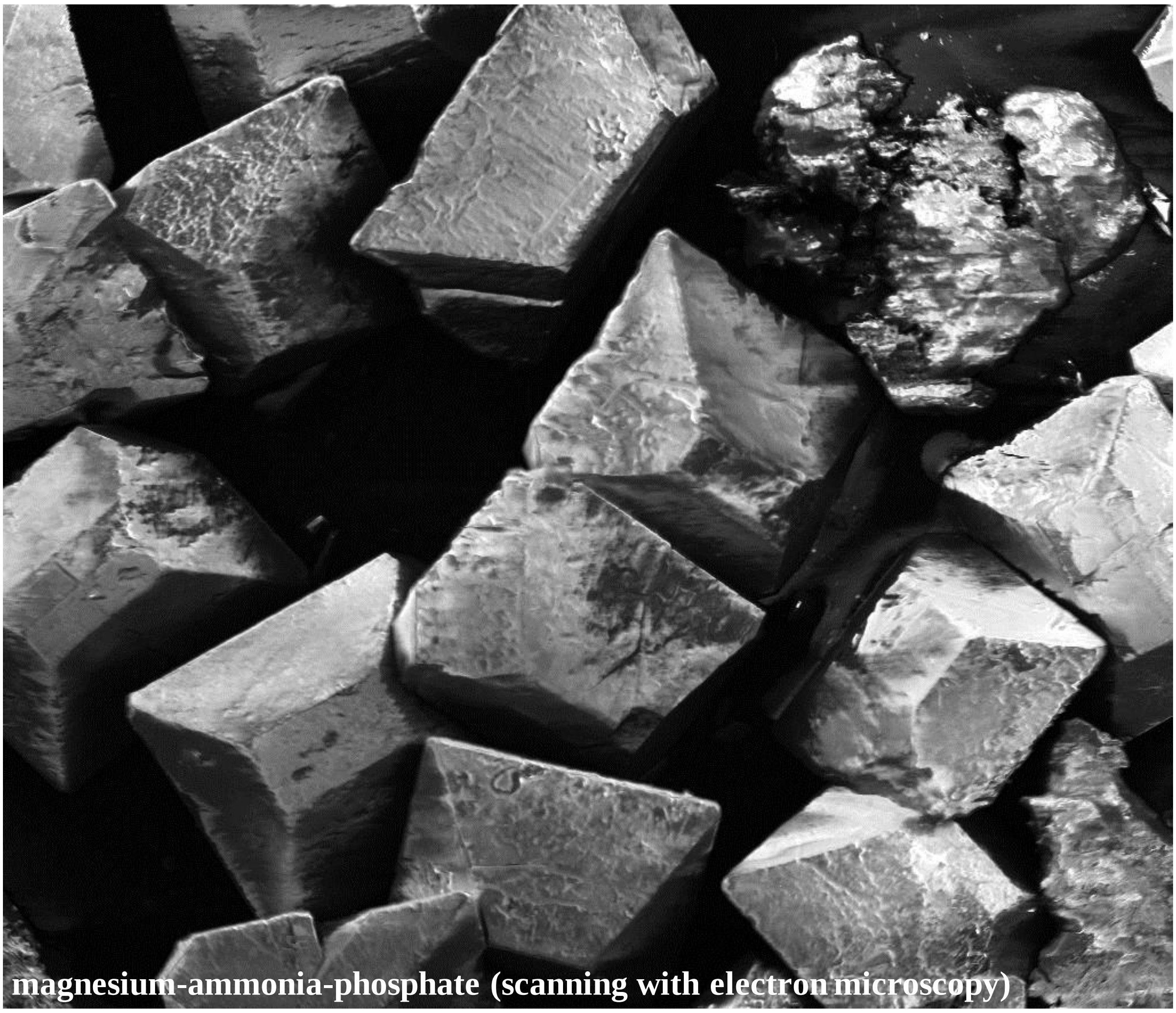
An ideal technology would feature maximum P recovery rates, good removal and destruction of potentially hazardous substances (heavy metals, organic micropollutants and pathogens) and an applicable material with low environmental risks, good fertilizing effects and economic efficiency. However, the results of this paper demonstrate that choosing a certain recovery technology is a trade-off between these criteria.

This work demonstrates that P recovery can be achieved with low costs. In some cases, even financial gains from P recovery can be achieved if dissolved P is recovered from digester supernatant or digested sewage sludge. Nevertheless, the recovery rates are (too) low in this case (< 25% of P in raw wastewater). However, if sewage sludge is incinerated, it is currently possible to recover a high percentage (70–90%) of the P in the wastewater input under specific conditions with little additional costs from a macroeconomic perspective. Nevertheless, there is a tradeoff between the requirements for heavy metal depollution and recovery costs. While recovery with little or no heavy metal depollution effort can already be realized without any significant additional cost compared to a system with sewage sludge disposal in landfills, additional costs associated with significant depollution are estimated to be in the range of 1–2 € PE^{-1} yr^{-1}. This emphasizes that costs are only one parameter when discussing resource recovery. The re-establishment of natural nutrient cycles implies independence from raw material imports from geopolitically unstable regions, independence from fluctuating market prices, development of regional value chains, and simultaneously lower environmental effects. How much a society is willing to pay for these aspects is not covered in this paper but could be the task of a socio-economic investigation.

This integrated assessment reveals that one final parameter for valuation is not constructive, as the different technologies utilize various P-rich sources along the wastewater and sludge treatment processes and achieve various criteria at different levels. In fact, using numerous assessment criteria delivers an overall picture for a particular recovery technology, which can be compared to other technologies and to the future requirements and expectations for P recovery.

To complete the picture for these technologies, especially with regard to the environmental impacts, an assessment considering greenhouse gas emissions, the acidification potential and the cumulative energy demand is a part of ongoing work.

5. Investigations on environmental impacts of technologies to recover municipal wastewater phosphorus



magnesium-ammonia-phosphate (scanning with electron microscopy)

5.1 Introduction

Besides technical and economic criteria, environmental criteria are getting more and more important to assess newly developed technologies and compare their environmental effects with existing state of the art technologies. The purpose of this assessment is to complete the picture of the P recovery technologies which were considered in previous works with respect to environmental impacts (Egle et al., 2016). The aim is on the one hand to make environmental impacts of recovered material directly comparable to environmental impacts of conventional mineral fertilizer production and on the other hand to show the relevance of gaseous emissions and energy demand in relation to the existing wastewater and sludge treatment systems but also in relation to the total environmental impacts of one inhabitant in Austria per year. The chosen method for this environmental assessment is Life-Cycle-Assessment (LCA). Results gained from LCA should give decision makers further arguments when discussing about closing disrupted nutrient loops and especially P recovery from wastewater. For the investigations 17 technologies, covering the broad field of P recovery, were selected (Table 16). To compare the recovery technologies with a commercial mineral fertilizer, data for the environmental impacts of mineral fertilizer production was collected by a literature research. Furthermore, the environmental impacts for a mineral fertilizer will be verified and assessed by own calculation.

Table 16: Considered P recovery technologies from the aqueous phase (green: digester supernatant, dissolved P in anaerobic digested sludge and effluent), sewage sludge (blue) and sewage sludge ash (red). This color code for the different P recovery access points is applied throughout this paper.

aqueous phase	sewage sludge [SS]	sewage sludge ash [SSA]
REM-NUT®[1] [ion exchange, precipitation]	**Gifhorn process**[7] [wet-chemical leaching]	**AshDec® depollution**[12] [thermo-chemical, ash depollution, Cl-source: e.g. $MgCl_2$]
AirPrex®[2] [precipitation/crystallization]	**Stuttgart process**[8] [wet-chemical leaching]	**PASCH**[13] [acidic wet-chemical, leaching]
Ostara Pearl Reactor®[3] [crystallization]	**PHOXNAN**[9] [wet-oxidation]	**LEACHPHOS®**[14] [acidic wet-chemical, leaching]
DHV Crystalactor®[4] [crystallization]	**Aqua Reci®**[10] [super critical water oxidation]	**RecoPhos®**[15] [acidic wet-chemical, extraction]
P-RoC[5] [crystallization]	**MEPHREC®**[11] [metallurgic melt-gassing]	**Fertilizer Industry**[16*] [acidic wet-chemical, extraction]
PRISA[6] [precipitation/crystallization]		**Thermphos (P_4)**[17*] [thermo-electrical]

[1]Liberi et al. (2001), [2]Heinzmann and Lengemann (2011), [3]Adnan (2002), [4]Britton et al. (2008), [5]Berg et al. (2007), [6]Montag (2008), [7]Esemen (2013), [8]Weidelener et al., (2005), [9]Blöcher et al. (2012), [10]Stenmark (2003), [11]Scheidig et al. (2013), [12]Nowak et al. (2011), [13]Montag et al. (2011), [14]Morf (2012), [15]Weigand et al. (2013), [16]tenWolde (2013), [17]Schipper (2012a); *integration of SSA as secondary raw materials to substitute raw phosphate rock

5.2 Material and methods

The investigations on environmental impacts for P recovery technologies are building up on the data sets gathered in Egle et al. (2014a,b), Egle et al., 2015 and P-Rex, 2015 and results from the previous integrated technology assessment (Section 5). Due to missing data on resource demand at this point of investigation, the two technologies **AshDec® Rhenania** and **EcoPhos®** are not considered in this work. No data are available on the resource demand for the construction of the technologies and therefore they cannot be considered.

5.3 Life cycle assessment (LCA)

To assess technical processes and their potential environmental impacts, the methodology of Life Cycle Assessment (LCA) has been developed (ISO standards 14040/44, 2006). LCA is a widely applied method and can be used for wastewater and sludge treatment but also for other industrial processes connected with wastewater (Remy, 2015). The following steps are part of the LCA:

1. Definition of goal and scope (system functions and boundaries, data quality)
2. Life Cycle Inventory (LCI; collection of all input and output data)
3. Life Cycle Impact Assessment (LCIA, environmental indicators, functional unit)
4. Interpretation (discussing of results with regards to limitations of LCA)

With this method potential environmental impacts can be quantified for a single technology and a defined system based on quantitative information on resource needs and emissions to all relevant environmental compartments which are affected by the considered system. Direct effects of a P recovery technology but also indirect environmental effects from production of any utilities or disposal of any wastes are considered and assessed. First step is the development of a material flow model, for which complete data base information is essential. All relevant inputs and outputs across the system boundary from or into the environment are quantified and summarized. This inventory information is then evaluated with a defined set of environmental indicators.

5.3.1 Mineral fertilizer production based on raw phosphate rock

To compare recovery technologies with a conventionally produced mineral fertilizer the environmental impacts of the conventional fertilizer production based on raw phosphate rock are determined by a literature research, from different life cycle data bases as GEMIS and EcoInvent but also by own calculations (see Section 5.3.4.1).

5.3.2 Defined system and system boundaries

A defined reference system is chosen for a robust environmental comparison. This reference system is designed as a modular system with defined reference processes, including detailed sub-processes. Figure 32 shows the modular system including a reference wastewater treatment plant (WWTP) with a pollution load of 100.000 population equivalents (PE) (=P load of 65,700 kg yr^{-1}), a reference co-incineration plant for sewage sludge and a reference waste management process for the treatment and disposal of occurring wastes. For P recovery from ashes, a reference mono-incineration plant is defined. Beside the waste management process, the final receiving processes are the atmosphere, soil/agriculture and the hydrosphere. Detailed description of the processes, resource demand and characteristics of the different flows as e.g., wastewater composition, mass flows, transfer coefficients for P and the selected pollutants as heavy metals, organic micropollutants and pathogens are given Section 5. System boundaries are on the one hand the self-standing recovery technology with its resource supply and on the other hand the whole process chain from the WWTP influent to the application of the recovered material to agriculture and the final disposal of occurring wastes.

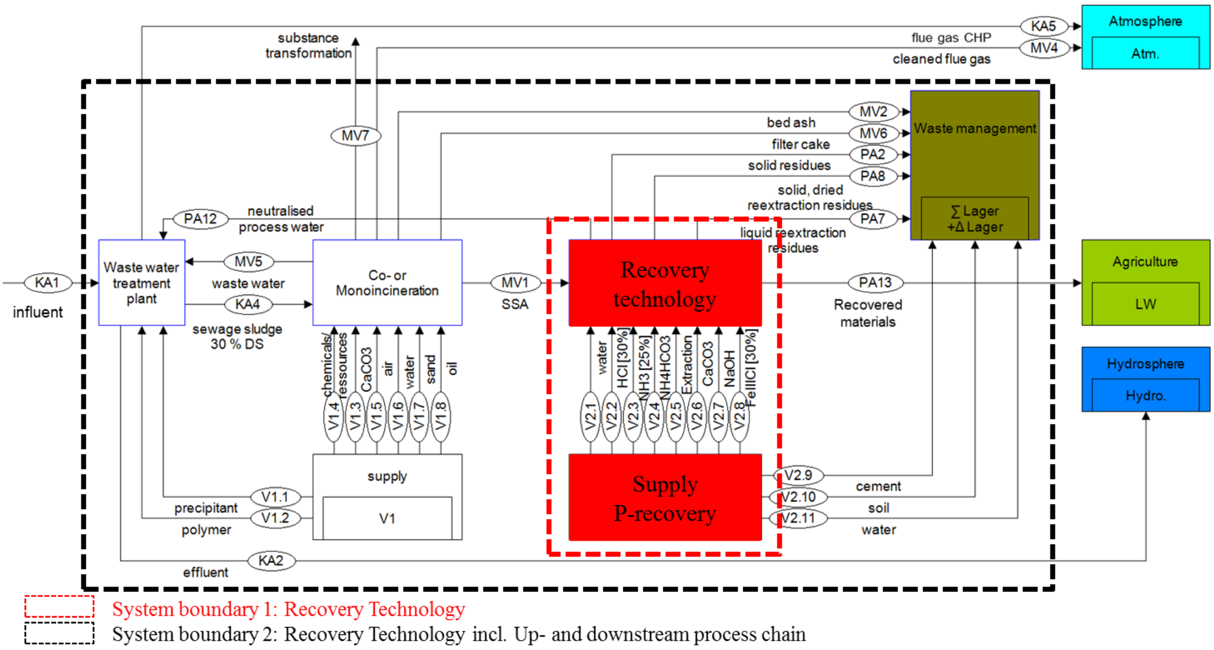


Figure 32: Process scheme for the reference substance flow model (STAN-model) with final receiving processes for emissions (atmospheres, soil (agriculture), and hydrosphere) and system boundaries for the recovery technology (dashed red) and the whole process chain (dashed black).

5.3.3 Life cycle inventory (LCI)

The development of the life cycle inventory is the step to quantify all relevant good or energy inputs and outputs for a single process or the total system. The material flow analysis (MFA; Cencic and Rechberger, 2008) is the chosen method to obtain the input of raw materials and chemicals as well as the output of P rich materials, waste by-products and other direct emissions to environmental compartments. Direct emissions are for example the emerging of CO_2 due to the incineration of primary fuels or emissions of heavy metals through the flue gas. Figure 33 presents the approaches to assess direct and indirect emissions. CO_2 emissions from biogenic combustibles as for example from sewage sludge incineration are not taken into account as these combustibles are considered as CO_2 neutral energy sources. External energy sources and also the energy content which is included in substances as e.g. sewage sludge are quantified with the methodology of energy flow analysis (EFA; Sangwon, 2005). For the processes which enable the recovery of the energy potential in sewage sludge, this energy potential is positively credited within the environmental assessment (Figure 33).

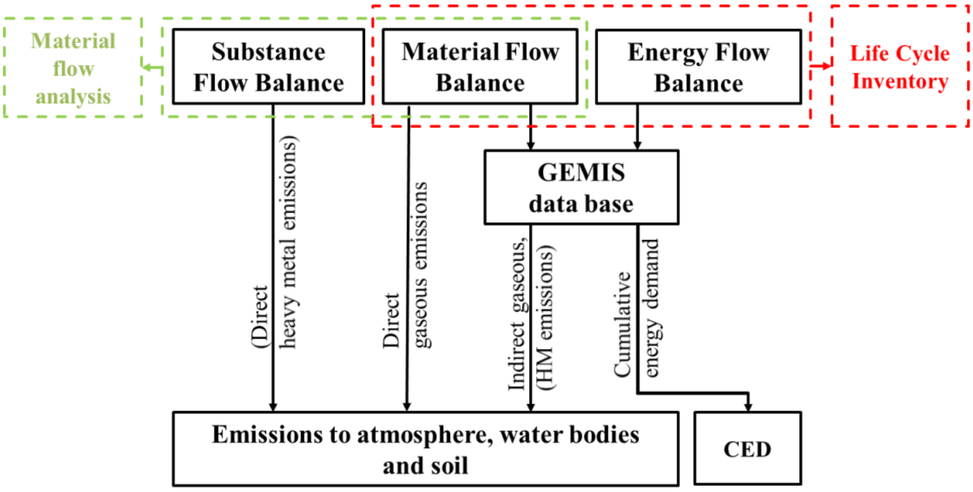


Figure 33: Approach to assess direct and indirect emissions of P recovery technologies and the whole system.

For the integrated assessment of the whole system the transport of sewage sludge, ash or residues is considered (Truck capacity: 20 t; diesel demand: 28 L 100 km^{-1}; diesel density: 0.85 kg L^{-1}; energy content: 11.8 kWh kg^{-1} diesel; specific fuel demand per ton kilometer: 0.012 kg tkm^{-1}). Table 17 gives a detailed listing of the considered transport distances for the Austrian situation based on literature data and own assumptions.

Table 17: Reference transport distances to sewage sludge or ash treatment facilities, agriculture and disposal sites for the Austrian situation.

Transport to processes	Transport in [km] there and back		
	Fehrenbach et al (2002)	Rechberger et al (2007)	Reference transport
Sewage sludge to incineration	160	250	140
Bottom Ash/Fly ash to landfill	-	-	100
Filtercake to underground disposal	-	-	1,000
Ash to centralized recovery technology	-	-	140
Recovered material to agriculture	-	-	40

5.3.4 Life cycle impact assessment (LCIA)

5.3.4.1 Environmental indicators

To perform the environmental assessment the following environmental indicators are considered:

- Gaseous emissions with global warming potential as CO_2, CH_4, N_2O expressed as CO_2 equivalents (CO_2e) (Table 18)
- Gaseous emissions with acidification potential as SO_2, NO_x, HCl, NH_3) expressed as SO_2 equivalents (SO_2e) (Table 18)
- Cumulative energy demand (CED, tool to determining energy requirements during the entire life cycle of a product (VDI, 2012))

The total gaseous emissions are calculated by addition of the direct and indirect gaseous emissions. Direct gaseous emissions result for example from the incineration of heating oil (e.g., 1 L heating oil =3.2 kg CO_2e). Furthermore, the production of the heating oil requires further resources and energy, which also results in the emissions of gases (indirect emissions).

The CED demand is calculated by taking into account the direct energy demand of the recovery processes (e.g., electricity, gas) and the energy demand which is needed to produce the necessary resources for a technology (indirect energy demand). The indirect gaseous emissions and the indirect energy demand are addressed by the life cycle data bases GEMIS (GEMIS, 2011). With the GEMIS data base the whole emissions of a process chain to generate a material is considered. Due to the fact that results from life cycle data bases can vary strongly, the EcoInvent data base (EcoInvent, 2013) was considered to cross-check the dependency of results on the used data base for one selected recovery technology (LEACHPHOS®). An example of the prepared data from the GEMIS data base for natural gas is given in the appendix (Table A 24).

Table 18: Global warming- and acidification potential of selected gases (IPCC, 2007).

Global warming potential	CO_2-Equivalents	Acidification potential	SO_2-Equivalents
CO_2	1	SO_2	1
CH_4	25	NOx	0.7
N_2O	298	NH_3	1.88
		HCl	0.88

5.3.5 Functional unit

To allow a comparative environmental assessment of the different recovery technologies among themselves and a comparison of the environment impacts of the recovery technologies with a conventionally produced fossil mineral fertilizer based on raw phosphate rock, the functional unit 1 kg P_{rec}^{-1} (1 kg P recovered) is chosen. Furthermore, to offset the emissions and energy demand for P-recycling into relation with other human activities, the results of the CO_2e and SO_2e emissions and CED were related to 1 population equivalent and year (1 $PE^{-1}yr^{-1}$). Population equivalent is a common indicator to describe the organic pollution load of a wastewater treatment plant. With this functional unit, the additional environmental impacts or even savings can be compared with the environmental impact of the defined reference wastewater and sludge treatment system (Section 5.3.2). Furthermore, with the conversion factor from PE to inhabitants by the factor of 0.5 (the wastewater production (PE) of the Austrian society is approximately twice as high as the number of inhabitants) the environmental impacts of the recovery technologies are also comparable to the total annual emissions and energy demand per inhabitant in Austria.

5.3.1 Uncertainty of basis of data

As data from literature reviews originate from manifold sources, data quality differs strongly. The uncertainty of the data for the MFA and EFA (Life Cycle Inventory) is assessed qualitatively depending on the source. In this work, the uncertainty is categorized as low (+), moderate (o), high (-) and very high (--) (see Section 4.2.5). If no data is available for certain technologies, missing data can be generated by the knowledge on basic chemical principles or knowledge from other similar technologies. On this basis, high uncertainties need to be considered.

For the assessment of a technology within the defined reference system, a particular challenge is the consideration of additional benefits, as some recovery technologies positively affect up- and downstream processes. In this work, the benefits of a technology on the system are fully taken into account. However, these benefits are in some cases afflicted with great uncertainties.

Beside the LCI data set, uncertainties need to be considered for the life cycle data base. However, the life cycle data base GEMIS does not indicate any uncertainties for the presented ecological backpack of a raw material. Therefore, additional life cycle data bases as EcoInvent are considered to compare these data. The results will be discussed with regard to the robustness of the applied data (LCI), the considered and in many cases positive effects on the whole system and the applied life cycle data base.

5.4 Results

5.4.1 Environmental effects of fossil fertilizer production based on raw phosphate rock

The environmental effects for the production of 1 kg of P as fossil mineral fertilizer gained from raw phosphate rock (PR) are shown in Table 19. These numbers show a great discrepancy, resulting from different sources of data. Main reason for these variable results is the assessment of the sulfuric acid (H_2SO_4) in the different LCA data bases. H_2SO_4 is the essential chemical in the extraction process to transform not water soluble P compounds in raw PR into water soluble and therefore immediately plant available P compounds. The production steps to gain H_2SO_4 are exothermic. That means if external use of energy delivered from this process is assumed, a credit for CO_2e emissions can be acquired. As significant amounts of H_2SO_4 is needed for fossil mineral fertilizer production (0.5–0.6 kg kg raw PR^{-1}) and such a CO_2e credit is assumed, CO_2e emissions for 1 kg of fossil P fertilizer are negative according to the GEMIS data base. However, the EcoInvent data base indicates contrary numbers (Figure 34) as no credits are given for energy release during H_2SO_4 production. Own calculations with literature data for resource demand (Patyk and Reinhardt, 1997; Silva and Kulay, 2003; Silva and Kulay, 2005) combined with emission data from GEMIS show similar results for CO_2e emissions as derived from GEMIS data base, as a credit for H_2SO_4 production has been assumed as well. With the EcoInvent data base and according to Fehrenbach and Reinhardt (2011), significantly higher CO_2e are assumed from the production of a fossil mineral fertilizer.

GEMIS, the own calculations with the GEMIS data base and Fehrenbach and Reinhardt (2011) show similar results for SO_2e emissions in the range of 21–23 g kg P^{-1} for the production of a fossil mineral fertilizer. By comparison the EcoInvent data base shows significantly higher SO_2e emissions by the factor of 4. For the parameter CED the results differ strongly depending on the source, too. EcoInvent indicates CED of 16 kWh kg P^{-1} while Kongshaug (1998) presents lowest CED in the range of -2.4–3.0 kWh kg P^{-1}. The big range of values is a result from the comparison of modern and old technologies for fertilizer production sites in Europe. For the integrated approach to assess technologies within the whole systems, the credits assumed for 1 kg recovered by the technologies are highlighted in grey (Table 19).

Table 19: Gaseous emissions and cumulative energy demand for 1 kg P from fossil mineral fertilizer production based on different sources of data: GEMIS, EcoInvent, Kongshaug(1998), Fehrenbach and Reinhardt (2011) and own calculations based on GEMIS data base. Highlighted grey: Credit for 1 kg P recovered by the recovery technology for the integrated technology assessment (Section 5.4.3).

Parameter	**Source**	**kg kg P^{-1}**
CO_2e	*GEMIS*	*-0.20*
	EcoInvent	2.40
	Own calculation (GEMIS data base)	-0.17
	Fehrenbach and Reinhardt (2011)	1.3–1.8
		g kg P^{-1}
SO_2e	*GEMIS*	*22*
	EcoInvent	84
	Own calculation (GEMIS data base)	21
	Fehrenbach and Reinhardt (2011)	23
		kWh kg P^{-1}
CED	GEMIS	-
	EcoInvent	16,1
	Own calculation (GEMIS data base)	*7.2*
	Kongshaug (1998)	-2.4–3.0

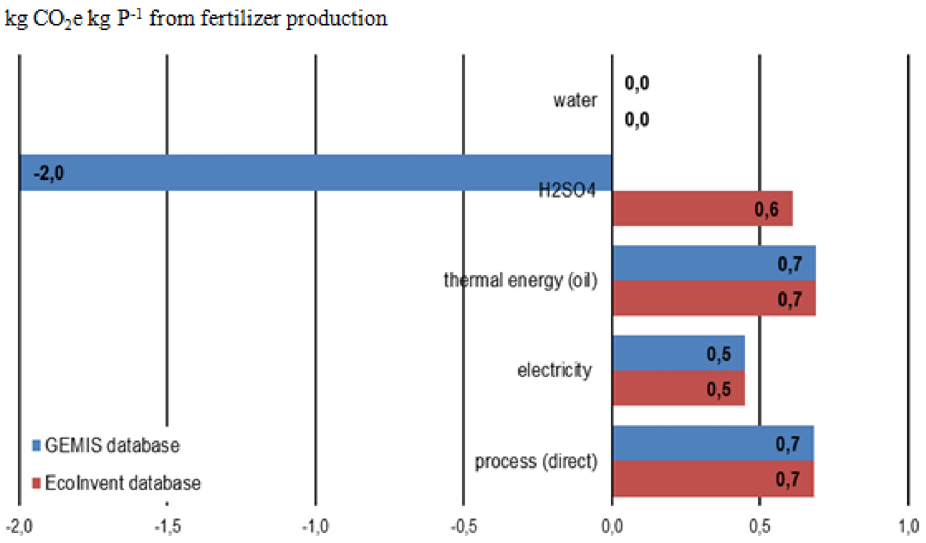


Figure 34: Comparison of CO_2e emission for fertilizer production (1 kg P) based on different data bases (GEMIS and EcoInvent).

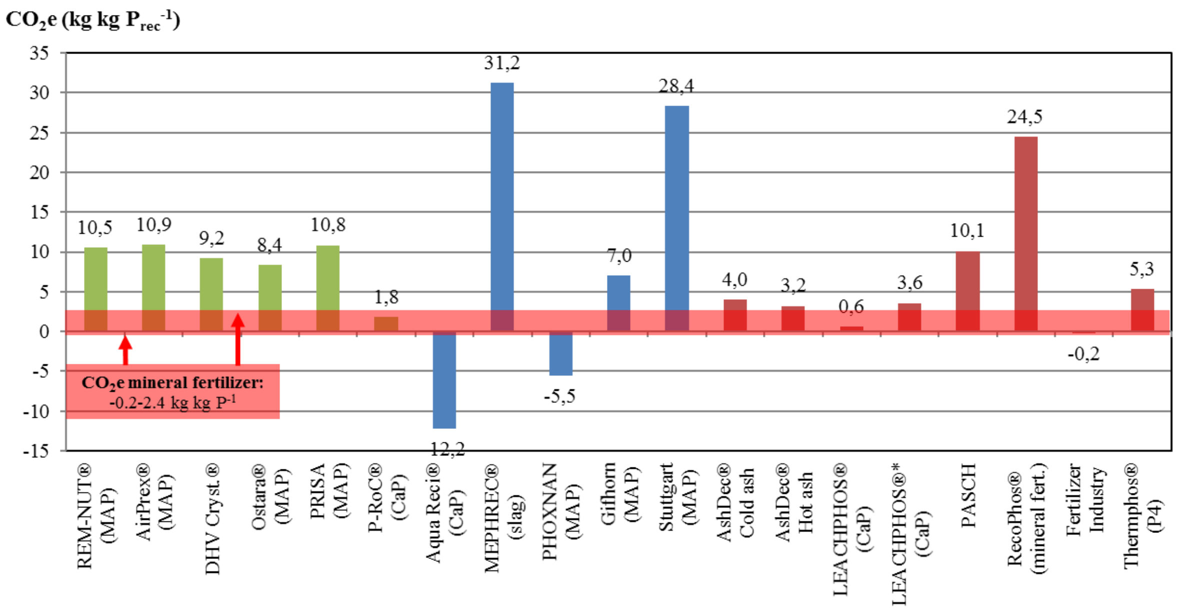
5.4.2 Environmental effects of recovery technologies related to 1 kg P recovered

5.4.2.1 Greenhouse gas emissions (CO2e)

The results for CO_2e emissions for the technologies to recover P from the different P rich flows of a WWTP differ strongly (Figure 35). Surprisingly is the fact, that even though P is already available in its dissolved form in digester supernatant, the CO_2e emissions are, with exception of the **P-RoC®** technology, high compared to many other technologies and considerably higher compared to P gained from raw PR. A reason is the low recovery potential compared to the need for precipitants with a great ecological backpack. The low CO_2e emissions for **P-RoC®** result from the use of a waste by product as crystallization material (Calcium-Silicate-Hydrate). Therefore, no precipitants as $MgCl_2$ or $CaCl_2$ are needed.

The results for technologies to recover P from sewage sludge are very inhomogenic. For the wet-oxidative (**AquaReci®**, **PHOXNAN**) and metallurgic (**MEPHREC®**) technologies the production of energy (heat) and an energy rich gas are included in the assessment. In the best-case scenario it is possible to save CO_2e emissions with the **AquaReci®** and **PHOXNAN** technology, as these two technologies use the energy potential of the sewage sludge. However, high uncertainties need to be considered, as these two technologies are at a low level of development (pilot scale) and the realization of the usage of the produced heat in full-scale implementations is not proven so far. Large CO_2e emissions for the **MEPHREC®** technology arise from the high energy demand (coke) to melt iron and to create a liquid iron bed (~2.000°C) for sludge treatment, even though an energy rich gas is produced. Looking at the resource intense wet-chemical leaching technologies **Gifhorn** and the **Stuttgart** approach, the high CO_2e emissions are surprising, due to fact, that both process use great amounts of H_2SO_4 to dissolve P from sewage sludge. Still, the other necessary chemicals as e.g., citric acid, caustics, and precipitant agents result in the high CO_2e emissions, especially for the **Stuttgart** approach.

For the **AshDec®** technology CO_2e emissions mainly result from the energy demand for heating the ashes up to 850–1,000 °C. However, with 3.2–4.0 kg CO_2e kg P_{rec}^{-1} the emissions are marginally higher than the worst case for a fossil mineral fertilizer. To show the impact of different data bases on the results for the environmental assessment of P recovery technologies, the **LEACHPHOS®** technology has been investigated additionally with EcoInvent data base (Figure 35). H_2SO_4 is the leaching agent to leach P from the ashes. As shown before the data bases GEMIS and EcoInvent differ greatly in respect to assessment of CO_2e emissions from H_2SO_4 production. Therefore, 1 kg P recovered with the **LEACHPHOS®** technology can vary substantially (GEMIS data base: 0.6 kg CO_2e kg P_{rec}^{-1}; EcoInvent data base: 3.6 kg CO_2e kg P_{rec}^{-1}). For the **PASCH** technology higher CO_2e emission can be detected as for **LEACHPHOS®**. This is mainly resulting from the application of HCl as a leaching agent instead of H_2SO_4. By integrating SSA into the fertilizer production process (**fertilizer industry**) negative CO_2e emissions can be identified due to the positive credits for H_2SO_4. Particularly, high CO_2e emissions are shown for the **EcoPhos®** technology, as the applied phosphoric acid has a great ecological backpack.

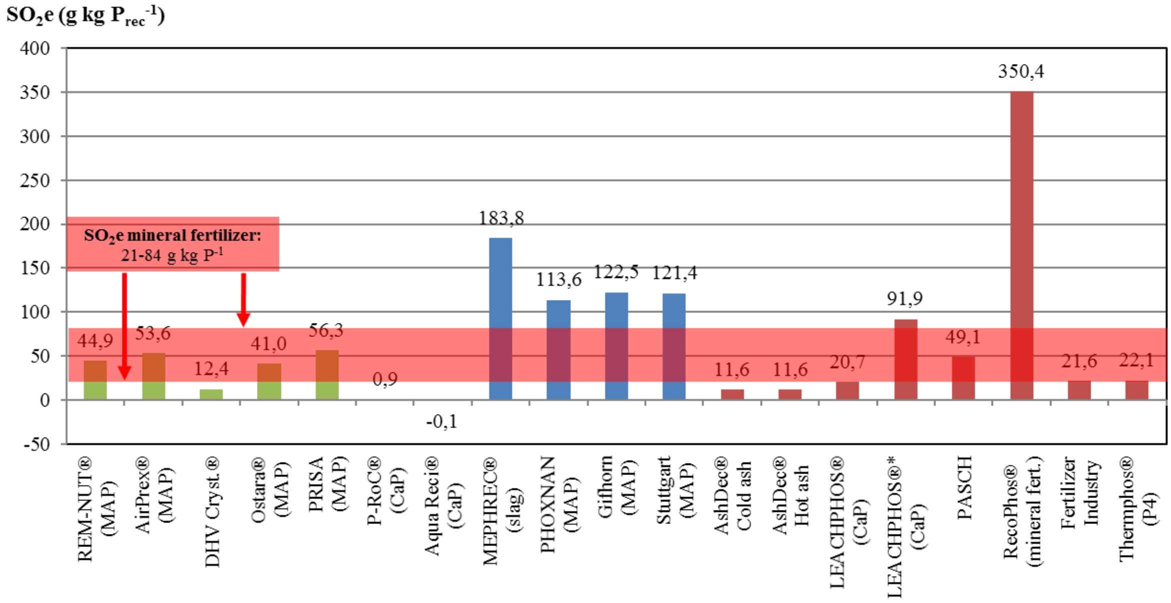


*calculation with the EcoInvent database. Compared to GEMIS database, no credit is considered due to the exothermic production of H_2SO_4.

Figure 35: Greenhouse gas emissions (CO_2e) related to 1 kg of P recovered from wastewater with recovery technologies in comparison to 1 kg of P from fossil mineral fertilizers produced from raw PR.

5.4.2.2 Acidification potential (SO2e)

As sulfuric acid is used to attack raw phosphate rock, SO_2e emissions are in the range or are even higher for mineral fertilizers compared to recovered materials from digester supernatant or sewage sludge ashes. The reason for the good results for some of the technologies recovering P from SSA is the credit for the substitution of great amounts of P. With these technologies recovery rates related to WWTP influent are 70–90% and therefore distinctly higher compared to technologies addressing digester supernatant or sewage sludge. For technologies using H_2SO_4 or phosphoric acid (product from leaching raw phosphate rock with sulfuric acid) as a leaching agent, SO_2e emissions are high (**PHOXNAN**, **Gifhorn**, **Stuttgart**, **LEACHPHOS®**, **RecoPhos®**). The resource intense **MEPHREC®** approach (cement for sludge briquettes, iron and coke) shows high SO_2e emissions even though energy is generated and credited for this process. As already shown for the CO_2e emissions, the SO_2e emissions can vary considerably if different LCA databases are applied (**LEACHPHOS®**, Figure 36).



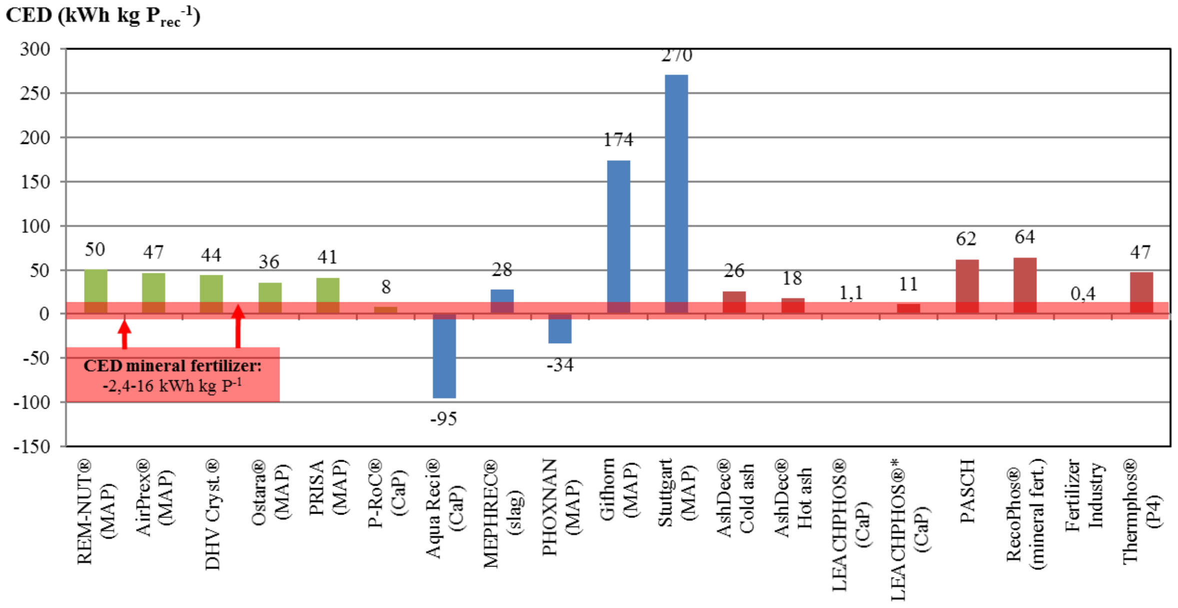
*calculation with the EcoInvent database. Compared to GEMIS database, no credit is considered due to the exothermic production of H_2SO_4

Figure 36: Acidification potential (SO_2e) related to 1 kg of P recovered from wastewater with recovery technologies in comparison to 1 kg of P from fossil mineral fertilizers produced from raw PR.

5.4.2.3 Cumulative energy demand (CED)

Compared to a fossil mineral fertilizer production from PR, the cumulative energy demand to recover already dissolved P from digester supernatant is, with exception of the **P-Roc®** technology, considerably higher (Figure 37). For the wet-oxidative (**AquaReci®**, **PHOXNAN**) technologies the production of energy (heat) results in the best-case scenario in possible savings of energy. With the **MEPHREC®** technology also thermal energy can be recovered from sewage sludge, but this approach is dependent from high energy inputs in the form of coke. Furthermore, cement (energy intensive production) is required to form the sewage sludge input briquette to feed the furnace. For the two leaching processes **Gifhorn** and **Stuttgart** as well energy intense chemicals are needed and consequently highest CED demand results for these two technologies (170–270 kWh kg P^{-1}).

Even though the SSA has to be heated up to 850–1,000 °C with the **AshDec®** technology, CED is considerably lower than for technologies to recover P from digester supernatant. For the wet-chemical leaching and wet-chemical extraction technologies **PASCH** and **RecoPhos®** CED is in the range of 60 kWh kg P^{-1} and therefore significantly higher compared to a fossil fertilizer based on raw phosphate rock. With the **LEACHPHOS®** technology, one kg of P can be recovery with an energy demand in the range of a fossil fertilizer. However, the variance of the results for CED when applying different LCA data bases is remarkable. CED for the **LEACHPHOS®** technology is only about 1 kWh kg P_{rec}^{-1} with the GEMIS data base. Appling the EcoInvent data base CED is higher by the factor 10. Therefore, a comparison of technologies is meaningful only, if they are assessed with the same LCI data base.



*calculation with the EcoInvent database. Compared to GEMIS database, no credit is considered due to the exothermic production of H_2SO_4

Figure 37: Cumulative energy demand related to 1 kg of P recovered from wastewater with recovery technologies in comparison to 1 kg of P from fossil mineral fertilizers produced from raw PR.

Detailed results on the environmental effects only for the recovery technologies themselves are given in the appendix (Figure A 8, A 9, A 10).

5.4.3 Environmental effects of the defined reference system and one inhabitant

5.4.3.1 Relevance of gaseous emissions and energy demand

For an integrated assessment of the recovery technologies within the defined reference systems the results for the reference system with respect to CO_2 equivalents, SO_2 equivalents and CED are given in Table 20. The gaseous emissions and CED given in this Table 20 can be seen as fixed values without uncertainties, as these values are the reference values to compare the different recovery technologies.

Table 20: Gaseous emissions and CED calculated for the reference systems.

Parameter	Reference System	Unit
CO_2e	3.7	kg $PE^{-1}yr^{-1}$
SO_2e	12.6	g $PE^{-1}yr^{-1}$
CED	11.3	kWh $PE^{-1}yr^{-1}$

5.4.3.2 Relevance of emissions

Furthermore, to assess the relevance of emissions and energy demand caused by a technology, the environmental impacts are compared to the emissions and energy demand provoked by one inhabitant and year in Austria. The annual total CO_2e emissions per inhabitant are about 10,000 kg (UBA, 2013). As 1 PE corresponds to 0.5 inhabitants the CO_2e emissions from the reference wastewater treatment are 7.4 kg per inhabitant and year. Consequently, the share of WWTP treatment with subsequent sludge treatment and final disposal of occurring wastes is 0.07% compared to the total annual CO_2e emissions per inhabitant. The total annual SO_2e emissions per inhabitant in Austria are 30,100 g taking into account emissions of 17,000 t SO_2 yr^{-1} (SO_2e factor: 1), 162,000 t NO_x yr^{-1} (SO_2e factor: 0.70) and 66,200 t NH_3 yr^{-1} (SO_2e factor: 1.88) (UBA, 2015). For the reference system SO_2e emissions are 25.2 g per inhabitant and year, and therefore the share on the total SO_2e emissions is 0.08%. To assess the relevance of CED, the energetic demand per inhabitant is considered as kg of oil equivalents. In 2013 the per capita consumption was 3,990 kg of oil equivalents (1 kg of oil equivalents = 41,868 kJ or 11.6 kWh) (EUROSTAT, 2015). This corresponds to 46,300 kWh per inhabitant and year. As one PE corresponds to 0.5 inhabitants the CED for the reference system for wastewater treatment is 22.6 kWh per inhabitant and year. Therefore, the CED of the reference system for wastewater treatment compared to the total annual energy demand per inhabitant is 0.05% (Table 21).

5.4.4 Environmental effects of the defined reference system and one inhabitant

5.4.4.1 Greenhouse gas emissions (CO_2e)

In comparison to the reference system, additional CO_2e emissions of 0.6–3.4 kg inh^{-1}yr^{-1} are expected for technologies to recover already dissolved P from digester supernatant and sewage sludge. In case of DHV Crystalactor® around 10 kg CO_2e can be expected (Figure 35). The higher emissions are the results of resources used in upstream processes of the WWTP (acetic acid as a carbon source for BioP) but also the use of the precipitant $Ca(OH)_2$ instead of $MgCl_2$. Related to annual CO_2e emissions per inhabitant and year the additional emissions to recover 10–max. 30% of wastewater P is 0.01–0.03% (Figure 38).

Very inhomogenic the picture for technologies to recover P from sewage sludge. For the assessment of the wet-oxidative and metallurgic technologies the recovery of energy in the form of heat and electricity or a usable gas are included in the assessment. In the best-case scenario CO_2 emissions could be saved with the **AquaReci®**- und **PHOXNAN** technology due to the simultaneous recovery of energy and the inertization of the sewage sludge, which allows a direct landfilling of the occurring ashes. **MEPHREC®** as well a process with a simultaneous recovery of energy shows significant higher CO_2e emissions (Δ to reference system: +330%) as coke and iron are needed to create the iron slag bed. For the wet-chemical technologies **Gifhorn** and **Stuttgart** process, the expected additional emissions vary explicitly as different chemicals are applied to remove or chelating heavy metals, which are dissolved during the primary wet-chemical leaching step (Δ to reference system: **Gifhorn**: +30%; **Stuttgart**: +220%).

To depollute SSA with the **AshDec®** technology, additional CO_2e emissions in the range of 30 to 40% related to reference system need to be considered. If the SSA is integrated into the fertilizer industry, CO_2e emissions can be saved as a results of the application of sulfuric acid which shows negative CO_2e emissions as the production of sulfuric acid is exothermic according to the GEMIS data base (Figure 34). Also a CO_2e positive balance can be shown for the wet-leaching process **LEACHPHOS®** using sulfuric acid. For the ash recovery technologies **RecoPhos®** and **Thermphos®** addition CO_2e emissions of 10–11 kg per PE and year (Δ to reference system: 270–310%) need to be considered. As the high CO_2e emissions for the **RecoPhos®** technology are a result of the great ecological backpack for phosphoric acid, the additional emissions for the **Thermphos®** process are a result of the resources for the process itself but also a results of the necessary changes in the wastewater treatment process. For the **Thermphos®** process ashes poor in iron are required. Therefore, the precipitations agent in the WWTP process needs to be changed from iron to aluminum, which results in greater CO_2e emissions. This example shows clearly how even slight changes in upstream processes effect the environmental impacts for a later P recovery.

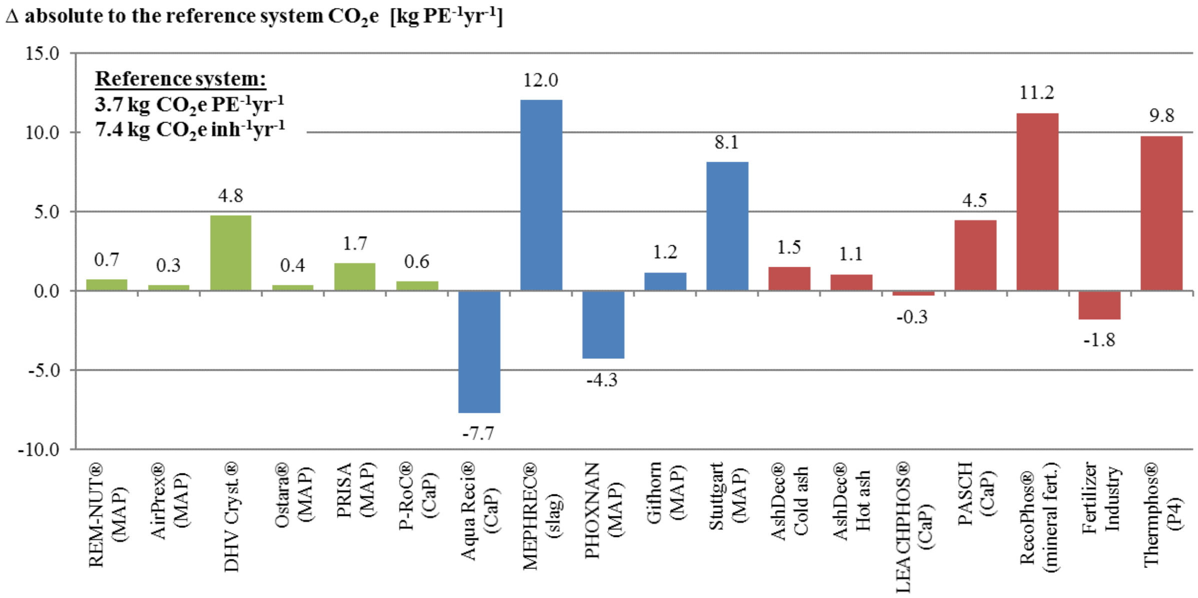


Figure 38: Results in the absolute changes of the greenhouse gas emissions (kg CO_2e $PE^{-1}yr^{-1}$) in relation to the reference system.

5.4.4.2 Acidification potential (SO_2e)

Many technologies are performing well with regard to SO_2e emissions and hardly any additional emissions or even savings of emissions with acidification potential can observed compared to the reference system (Figure 39). As already noticed, SO_2e emissions are high for technologies using sulfuric or phosphoric acid (**PHOXNAN**, **Gifhorn**, **Stuttgart**, **LEACHPHOS®**, **RecoPhos®**). Compared to the reference system high additional emissions can be observed for these technologies in the range of +200% (**PHOXNAN®**) to +1,280% (**RecoPhos®**). However, the relevance with regard to the total annual emission per inhabitant is 0.25 to 1.15% (Table 21).

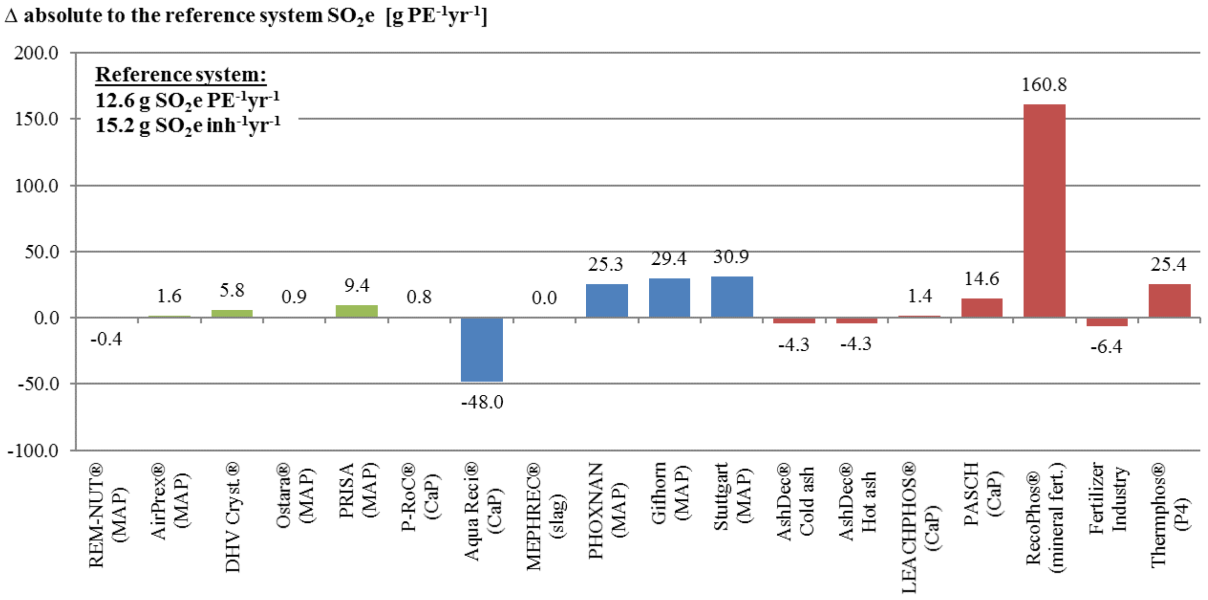


Figure 39: Results in the absolute changes of the acidification potential (g SO_2e $PE^{-1}yr^{-1}$) in relation to the reference system.

5.4.4.3 Cumulative energy demand (CED)

For CED a good correlation with results of the greenhouse gas emissions can be observed (Figure 40). This good correlation relates to the ranking of the technologies amongst each other, the factor of additional or savings in energy related to the reference system and the low level of priority of the additional energy demand compared to the annual per capita consumption (Table 21).

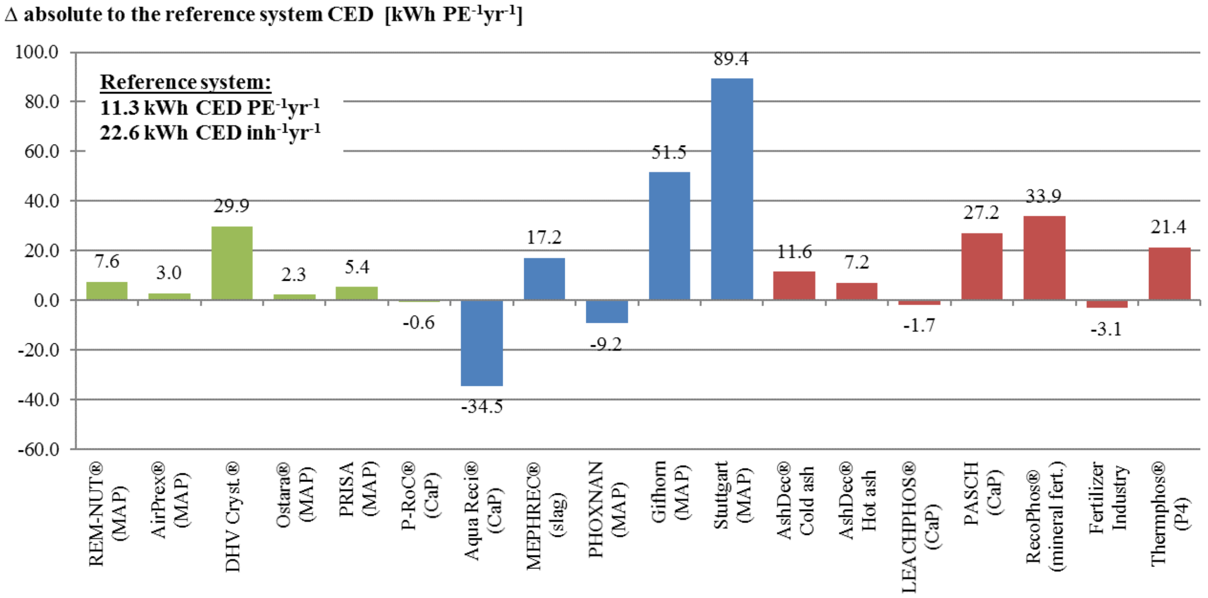


Figure 40: Results in the absolute changes of the CED (kWh PE-1yr-1) in relation to the reference system.

5.4.5 Uncertainty

Even though MFA and EFA provide a robust data base for the life cycle inventory for most of the technologies, certain specific limitations which emerged in this study must be viewed with a critical eye when interpreting the results:

- Different LCA data bases show significantly varying “ecological backpacks” for certain chemicals. Particular worthy of mention is sulfuric acid, as this acid is a basic chemical for fossil mineral fertilizer production and for P recovery technologies. Results based on the GEMIS data base imply that the more H_2SO_4 is used for fertilizer production, the more economically friendly the production is with regard to CO_2e emissions and CED.
- A comparison with a fossil mineral P fertilizer is difficult, as even the environmental impact of a fossil mineral fertilizer can vary significantly depending of the source of data.
- The datasets for fossil mineral P fertilizer production from GEMIS is outdated, as the fundamental data for the fertilizer production process originate from 1997 (Patyk and Reinhardt, 1997)
- For certain technologies but also for the wastewater treatment process particular chemicals are used which are not covered by the LCA data bases. Therefore, similar chemicals are chosen for the assessment, but their environmental impact can vary significantly (e.g., $MgCl_2$ instead of MgO).
- Some important chemicals used as e.g. iron chloride precipitants in the wastewater treatment process are by-products/wastes from industrial processes. LCA data bases include only raw materials which are specially produced to fulfill a purpose. As the full “ecological backpack” is taken into account in our calculations this could be a significant overestimation as the “ecological backpack” could be much smaller for by-products/wastes from industrial processes.

5.4.6 Summary and discussion of environmental impacts

This study reveals for many cases higher environmental impacts of the recovered P compared to fossil mineral fertilizers. Ironically, the more chemicals are used in the form of sulfuric acid, the better the environmental impacts regarding CO_2e emission and CED. In total to preserve the resource phosphorus by recycling additional energy is needed and additional atmospheric emissions are produced.

To understand the dimension of additional environmental impacts right, the additional emissions and energy demand or even savings of emissions and energy of the recovery technologies are related to the annual total emissions and energy use per inhabitant. Table 21 highlights that only minor additional gaseous emissions and minor additional energy demand is necessary compared to the current gaseous emissions and energy use if P is recovered from the different P rich streams of a WWTP or from SSA. In some cases, where P is recovered from sewage sludge with a simultaneous recovery of energy, even savings are feasible with regard to the present wastewater and sludge treatment system. With the following example the relevance of the environmental impacts with regard to the Austrian situation is discusses. By the implementation of a nationwide sewage sludge mono-incineration concept in Austria, 6,000 t of P incorporated in sewage sludge could be transferred to SSA and P could be recovered out of it with an efficiency of 80–100%. As a consequence, 40–50% of the annually applied raw phosphate rock based fossil mineral P fertilizer could be substituted with additional CO$_2$e emissions of 0.09 to a maximum of 0.30% compared to the current system.

Table 21: Gaseous emissions and CED of the reference system and P recovery technologies in relation to the annual emissions per inhabitant.

Technology	CO$_2$e	SO$_2$e	CED	P recovery potential*
Per inhabitant and year	10,000 kg	30,100 g	46,300 kWh	-
Reference system	0.07%	0.08%	0.05%	0%
REM-NUT®	0.09%	0.08%	0.08%	50–60%
AirPrex®	0.08%	0.09%	0.06%	10–max. 25%
DHV Crystalactor®	0.17%	0.12%	0.18%	10–max. 25%
Ostara®	0.08%	0.09%	0.06%	10–max. 25%
PRISA	0.11%	0.15%	0.07%	10–max. 25%
P-RoC®	0.09%	0.09%	0.05%	10–max. 25%
AquaReci®	-0.08%	-0.24%	-0.10%	~60%
MEPHREC®	0.31%	0.55%	0.12%	~70%
PHOXNAN	-0.01%	0.25%	0.01%	~40–50%
Gifhorn	0.10%	0.28%	0.27%	35–50%
Stuttgart	0.24%	0.29%	0.44%	35–50%
AshDec® cold ash	0.10%	0.05%	0.10%	85%
AshDec® hot ash	0.09%	0.05%	0.08%	85
LEACHPHOS®	0.07%	0.09%	0.04%	~60–70%
PASCH	0.16%	0.18%	0.17%	~60–70%
RecoPhos®	0.30%	1.15%	0.20%	87%
Fertilizer Industry	0.04%	0.04%	0.04%	87%
Thermphos®	0.27%	0.25%	0.14%	~85%

*Related to the P input of WWTP

5.5 Conclusions

This work builds up on an integrated and comparative technology assessment and delivers additional environmentally relevant assessment parameters to complement the overall picture of the considered P recovery technologies.

With regard to 1 kg P recovered, this study reveals for many technologies higher environmental impacts (CO_2e, CED) compared to fossil mineral fertilizer production. Paradoxically, the good performance of the fossil mineral fertilizer production results from the great amount of sulfuric acid needed for the fertilizer process. As the ecological backpack of sulfuric acid is assessed differently by various life cycle data bases, even the environmental impact of 1 kg P gained from raw phosphate rock has a wide degree of variation.

However, the outcome of this LCA study is affected by certain limitations due to the choice of definition, data availability (LCI), uncertainties on the effects on up-and downstream processes and the chosen life cycle data bases.

Based on these results, we could assess the environmental impacts of a certain technology within a certain magnitude for three relevant parameters. With this study it can be shown, that if a technology is implemented within the defined system, the expected additional environmental impacts are marginal from an ecologically point of view compared to the great P potential retrieved from wastewater.

6. Summary and conclusion

6.1 P recovery potential in Austria

Austria, like all other European countries except Finland, has no mineable raw phosphate rock deposits and is therefore entirely dependent on imports. As a consequence, the import of raw phosphate rock and marketable P mineral fertilizers represents the major flow of the entire national P balance. Currently, most of the P in Austria and other European countries is used in the sense of a linear economy. P recycling in Europe, with the exception of manure (slurry, dung) used for local agricultural purposes, is still at a rudimentary state. Municipal wastewater offers a P potential of ~ 1 kg P cap^{-1} yr^{-1}, but is currently a mostly overlooked source of phosphorus. As P-removal from waste water is required in Austria at all treatment plants with > 1,000 PE, sewage sludge, the P-sink and waste by-product from the wastewater treatment process, constitutes a P potential of 0.8 kg P cap^{-1} yr^{-1} which corresponds to ~40% of annually applied mineral fertilizers in Austria.

In order to calculate the real potential of P recovery in Austria, it is crucial to analyze the characteristics of Austrian wastewater treatment plants (WWTPs) with respect to their size, design capacity, and distribution. In Austria, 55% of municipal wastewater is treated in only 30 plants with design capacities of ≥ 100,000 PE. Consequently, more than 55% of the total sewage sludge created annually, is coming from 30 of the existing 1,837 WWTPs. Taking into account all WWTPs with capacities of > 2,000 PE (1/3 of all WWTPs in Austria), 98% of wastewater-P can be addressed. This impressive concentration of wastewater, and consequently sewage sludge, could help to implement centrally located P recovery units.

If animal meat and bone meal is taken into account as other alternative P sources, up to 70% of annually applied mineral P fertilizers could be theoretically substituted. Due to potential environmental and health risks as well as legislative restrictions, direct sewage sludge applications, and therefore direct P recycling, is low or even further decreasing in many European countries. Co-incineration in waste incinerators, coal fired power plants, and cement kilns are common sludge treatment options. However, as a consequence of this kind of thermal sludge treatment, P is irretrievably lost as sewage sludge is diluted with other wastes and coal, or is incorporated into cement. Once P is diluted with other wastes, a recovery is impossible with regard to technical feasibility and economic efficiency.

Especially in countries with a great P potential in municipal sewage sludge and little direct agricultural reuse of sewage sludge, the application of P recovery technologies should be considered. Over the past few years, manifold procedural approaches have been developed to recover P from various streams at a WWTP.

An ideal technology for P recovery would include maximum P recovery rates, good removal and destruction of potential hazardous substances (heavy metals, organic micropollutants, and pathogens), an applicable material with low environmental risks, good fertilizing effects, and economic efficiency. However, by choosing a certain recovery technology a trade-off between these criteria needs to be considered. Due to complexity of these technologies, an integrated assessment can be very useful to support decision makers. This Thesis provides an approach to assess the available technologies on the basis of technical, ecological, and economic criteria. Based on this assessment, legislators and political decision-makers could promote suitable P recovery strategies, including technologies to recover P from municipal wastewater and the reestablishment of broken nutrient cycles.

The integrated assessment of this work reveals that taking into account only one final evaluation parameter cannot be adequate, as different technologies address different P rich sources along wastewater and sludge treatment processes. Furthermore, the currently available technologies pursue various objectives (e.g., improved wastewater treatment processes or the stabilization of the sludge). Finally, it is the sum of numerous assessment criteria that creates an overall picture for a particular targeted P recovery technology from wastewater. This overall picture needs to be compared to the future requirements and expectations for a targeted P recovery from wastewater. Based on this comparison, a decision should be made.

6.2 P recovery from different streams of a WWTP

6.2.1 Aqueous phase

In the aqueous phase, where P is present in a dissolved form (e.g., digester supernatant or dissolved P in digested sludge), the recovery of plant available materials such as MAP or calcium phosphate with a low pollution potentials is simple from a technological point of view. Several technologies for P recovery from digester supernatant are already available and widely implemented on an international scale. Emphasis needs to be put on the beneficial effects of these technologies on wastewater treatment processes, namely the reduced back-flow of nutrients (PO_4 and NH_4), the avoidance of incrustation, or the improvement of sludge dewaterability. A combination of these benefits results in savings for WWTPs and, together with possible revenues, the costs for these technologies can be covered, or even a financial profit gained.

However, P recovery from the aqueous phase is limited to WWTPs with mainly biological P removal, as a certain concentration of dissolved P in supernatant or sewage sludge is required. Nevertheless, even in cases of WWTPs with biological P-removal the recovery of P addressing these internal flows of dissolved P has a limited recovery potential of 10 to max. 25% of the total P load in raw waste water discharged into the WWTP.

In comparison to that, the effluent of a WWTP offers a greater recovery potential (50–70% of WWTP inflow). However, it must be considered that, in case a P recovery is applied to the effluent, this technology would be the final barrier to avoid P emissions into water bodies. Therefore, reliable technologies would be of utmost importance. Necessary P enrichment technologies, such as ion exchangers, need to deal with the complex composition of wastewater, and therefore, with also commonly occurring problems as e.g., low selectivity, unwanted adsorptions, and fouling. As a consequence, the costs are relatively high as compared to other P recovery technologies.

6.2.2 Sewage sludge

In sewage sludge, a theoretical P potential of about 90% related to the WWTP influent is available. Practically recovery rates of up the 70 % are achieved. However, the complex composition of sewage sludge (water content, organic- and inorganic fraction, and heavy metals) demands for complex technologies and/or considerable quantities of chemicals. In some technologies waste streams, such as supernatants or acidified and sulfur rich sludge, are produced and need to be handled. This is most notably reflected in comparatively high costs for these technologies. Especially, for newly developed leaching technologies, the recovery quotes are low compared to the required resources (e.g., mineral acids) and the technological effort. For a nationwide recovery strategy, decentralized recovery concepts would have to be applied, as these technologies require sludge with dry matter contents of 2–4%. The costs for the transportation of sludge with a water content of 96–98% to centralized recovery plants is not feasible.

For oxidation and metallurgic processes, the energy potential of the sludge is used, and the sludge is simultaneously mineralized. Therefore, the costs for incineration can be omitted and the occurring ashes or slags directly deposited. With these benefits and also the revenues from the formed P-rich slag, the metallurgic approach could be economic. With exception of a P-rich slag, the recovered materials from SS as e.g. MAP show low pollution potentials and good fertilizing efficiencies. However, the current data base is weak and further investigations will be necessary to finally assess the oxidation and metallurgic processes with the methodology developed in this Thesis.

6.2.3 Sewage sludge ash

In order to achieve an extensive use of wastewater P (~90% of the WWTP influent), the future focus should be laid on the recovery from sewage sludge ash. This requires the avoidance of SS incineration together with other combustibles, containing low-phosphorus and/or high heavy metal and chlorine contents. The mono-incineration of sewage sludge is a typical option for this strategy. However, the best case-scenario would include the co-incineration of other high caloric materials with even higher P and lower pollutant contents (e.g., meat and bone meal). A mono-incineration strategy would allow the implementation of newly developed recovery technologies and/or the use of SSA as a secondary raw material in existing industrial processes. At this time, necessary structures, including mono-incineration plants, still need to be expanded in many European countries.

The advantages of strategies based on mono- or co-incinerations (e.g., with meat and bone meal) for sewage sludge are:

- Central incineration units to address the sludge, and therefore, the P potential of even smaller wastewater treatment plants.
- SSA is storable and can be transported considerably cheaper than dewatered sludge (no organic matter and no water).
- Recovery technologies can be implemented directly at the incinerator or at other central facilities, and are therefore more cost-effective.
- Possibility to store the ash for future recovery (creation of an Austrian “phosphate mine”). However, temporal storage should be the last option as already several new technologies and existing industrial processes are ready to recover P from SSA.

Nevertheless, additional costs need to be considered for the mono-incineration of sewage sludge, compared to direct agricultural application and co-incineration. The overall assessment illustrates that SSA shows the best preconditions with regard to an extensive use of wastewater P and costs. For SSA it could be important to find synergies with the existing industrial infrastructure (e.g., fertilizer industry, phosphoric acid industry).

If a pure SSA would be available on the market, industrial partners such as the fertilizer- or phosphoric acid industry could integrate these SSA into their processes. Such a development could also be beneficial for incineration companies due to possible revenues instead of disposal costs for ashes. As a consequence, the additional social costs of sludge mono-incineration could be partially compensated. Furthermore, with their demand for marketable products and their already established marketing channels, industrial partners could promote P recovery and recycling from wastewater. However, when ashes are integrated into those processes, questions about depollution arise. A dilution of heavy metals can be achieved by processing ashes with raw phosphate ore or mixing them with phosphoric acid, but the heavy metal load in the products remains the same as in the ashes. By producing phosphoric acid or P_4, which is suitable for a wide range of applications (e.g., feed, food, fertilizer), unwanted substances are concentrated in a small waste stream and removed from recycled P.

For newly developed leaching technologies, a purely economic perspective is not the driving force. The recovery potential related to the WWTP input is limited by ~60–70%, but at the same time, a very good heavy metal depollution can be achieved. To improve the ash quality after incineration with thermo-chemical processes, costs are equivalent to 1 kg P gained from raw phosphate rock.

Generally speaking, costs are only one parameter when discussing resource recovery. The re-establishment of natural nutrient cycles implies an independence from raw material imports from geopolitically unstable regions, independence from fluctuating market prices, development of regional value chains, and simultaneously lower environmental effects. How much a society is willing to pay for these aspects is not covered in this paper, but could be the task of a socio-economic investigation. As the field of P recovery is a very young and dynamic topic, it is likely that new and more economic technologies will be developed in the near future. The implementation of recovery technologies or the integration of ashes in existing processes could be accelerated by various economic and political factors.

6.3 Ranking of the analyzed technologies

This ranking is based on numerous assessment criteria and the given structures of the Central European wastewater management system – including the Austrian wastewater management system. The main result is a grouping of the considered recovery technologies, and consequently, a non-binding recommendation for potential technologies. Technologies with the most positive assessment criteria, where already some experiences with full-scale implementation exist or which have a great opportunity for a near future full-scale implementation are listed in the “priority group” (Section 6.3.1). Nevertheless, even in this “priority group” open technological questions need to be addressed before wide spread a full-scale implementation. Technologies at pilot scale status with promising results and possible middle-term full-scale implementations but still weak points and important questions remaining open are assigned to the “observation group” (Section 6.3.2). Technologies to recover P from side-streams as e.g., digester supernatant are listed to the “Bio-P group”. These technologies have in common that they require enhanced biological P removal during the wastewater treatment process and are implemented full-scale frequently (Section 6.3.3). Technologies with a low probability for a future full-scale implementation are not assigned to any group (Section 6.3.4)

6.3.1 Priority group

Based on the results of the integrated technology assessment, primary technologies which are capable to recover P from mono-incinerated municipal sewage sludge are selected as a priority group. The high recovery potential in combination with a good cost/benefit relation of these technologies was chosen as a main selection criterion. Furthermore, the possibility to simultaneously recover P from other P rich wastes, namely meat and bone meal, was included in the selection. Within this selection a tradeoff between technologies with

- higher costs, significant heavy metal depollution but lower recovery potential (EcoPhos, PASCH),
- technologies with lower costs, no heavy metal depollution and higher recovery potential (Fertilizer Industry) and
- technologies lying in the middle in respect to depollution and recovery potential, but having restrictions in respect to P availability (AshDec, LEACHPHOS) can be identified.

A selection therefore will depend on case specific priorities and requirements. Still, for all methods selected in this group open technological questions need to be addressed before full-scale implementation.

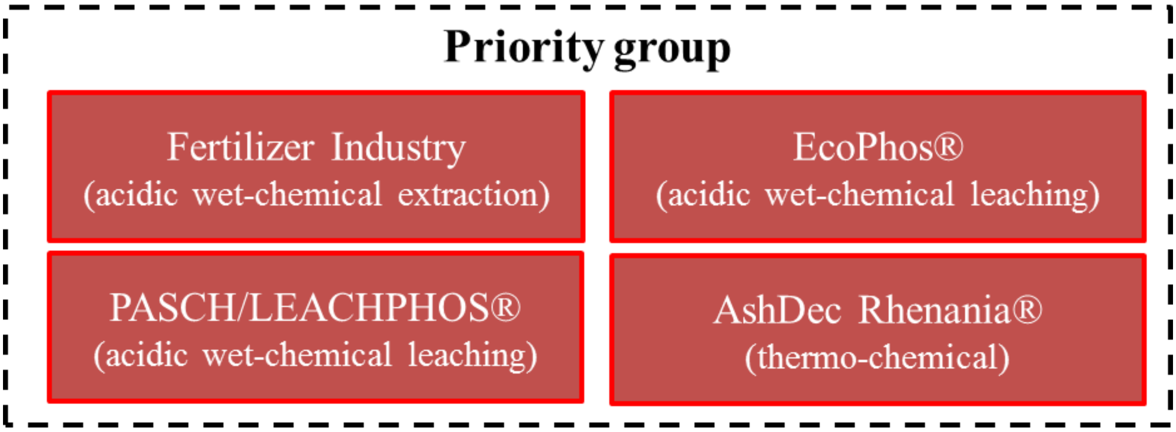


Figure 41: Recovery technologies assigned to the priority group.

6.3.2 Observation group

The observation group contains technologies, which have the potential of full-scale implementation, but still have weak points and/or a demand for further research. These weak points do not allow a classification in the priority group. The considered technologies recover P from sewage sludge ash (Thermphos®) and sewage sludge simultaneously (MEPHREC®). Another already implemented technology, which has been performing on a full-scale for years, could not be analyzed due to a lack of data (project LOTUS, Gifu City, Japan). Shortly before completing this Thesis, new technologies, namely TetraPhos® (Remondis®) and CleanMAP® (Easy Mining/Ragn Sells) appeared on the market and provide promising results. Although these technologies have not yet been assessed, they can be regarded as technologies belonging to the observation group.

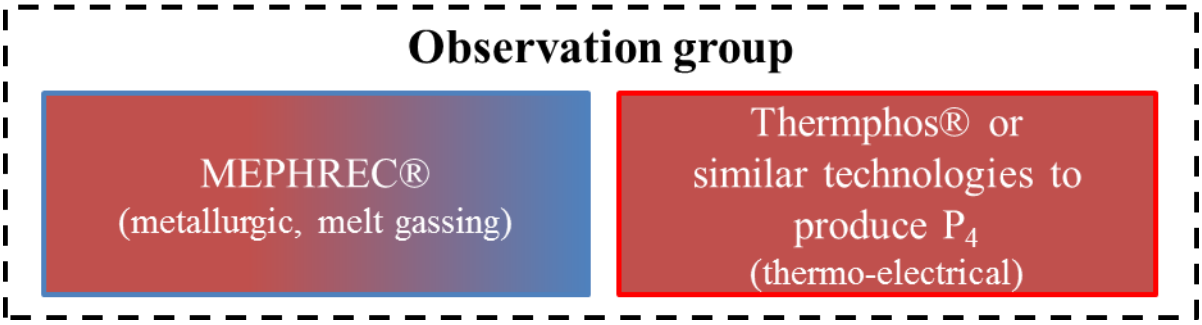


Figure 42: Recovery technologies assigned to the priority group.

6.3.3 Bio-P group

As already presented in Section 6.2.1, all technologies which recover P from digester supernatant or dissolved P directly from the digested sewage sludge, are simple operating processes. They could be (or already are) full-scale implemented in WWTPs with enhanced biological phosphorus removal. Besides of the selected technologies which have been considered in the integrated technology assessment, similar technologies, which recover dissolved P from digester supernatant are operating worldwide (e.g., Phosnix Unitaka, Nishihara Reactor, Sydney Waterboard Reactor, NuReSyS®; Table A 4). In combination with certain pre-sludge treatment options, as e.g., hydrolysis, the yield of P from digester supernatant or directly from the digested sludge can be increased.

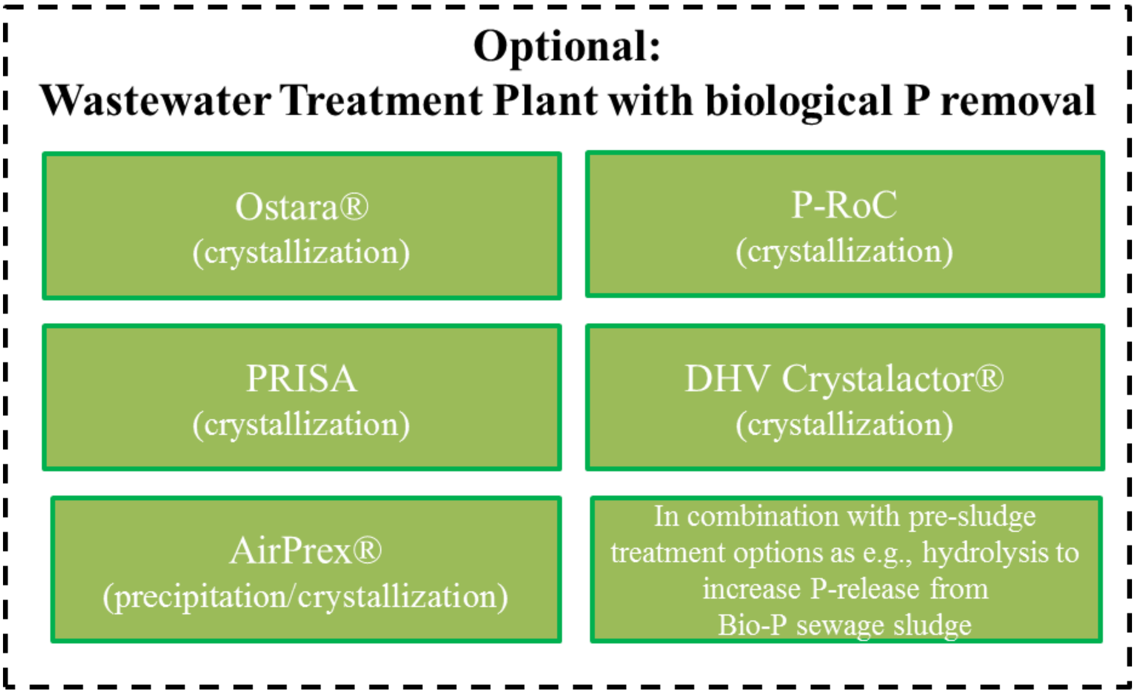


Figure 43: Recovery technologies assigned to the Bio-P group.

6.3.4 No group assignment

Technologies, which perform poorly with regard to the considered evaluation criteria, are not assigned to any group. In these cases, the negative criteria outweigh possible positive criteria. In most cases the main reasons for technologies to be put in this category are a low P recovery potential, but also the high costs in case of an implementation on a national scale.

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List of Abbreviations

Phosphorus compounds

P	phosphorus
P_4	white phosphorus
P_2O_5	phosphoruspentoxid
PO_4^{3-}	(ortho-) phosphate
H_3PO_4	phosphoric acid
MAP	magnesium ammonium phosphate ($MgNH_4PO_4$)
CaP	calcium phosphate
ACP	amorphous calcium phosphate
HAP	hydroxyapatite ($Ca_5(PO_4)_3(OH)$)
HDP	hydroxydicalcium phosphate ($Ca_2HPO_4(OH_2)$)

€ $PE^{-1}yr^{-1}$	euro per population equivalent and year
AF	annuity factor
Bio-P	biological P removal
$cap^{-1}yr^{-1}$	per capita and year
CED	cumulative energy demand
COD	chemical oxygen demand
CO_2e	carbon dioxide equivalents
CSH	calcium-silicate-hydrate
DM*	dry matter
DU	damage unit
DS*	dry substance
EBPR	enhanced biological P removal
EFA	energy flow analysis
$ha^{-1}yr^{-1}$	per hectare and year
HM	heavy metal(s)
IWWTP	industrial wastewater treatment plant
kg P_{rec}	kilogram phosphorus recovered
L	liter
LCA	Life-Cycle-Assessment
LCI	Life-Cycle-Inventory
LCIA	Life-Cycle-Impact-Assessment

MBM	meat and bone meal
MBT	mechanical-biological treatment
MFA	material flow analysis
mg	milligram
MI	mono-incineration
mt	megatons
MWI	municipal waste incineration
MWWTP	municipal wastewater treatment plant
OM	organic micropollutants
PAO	phosphorus accumulating organisms
PE	population equivalents
PG	phosphogympsum
POP	persistent organic pollutants
PR	phosphate rock
RFE	relative fertilizer efficiency
RSM	reference soil method
SCWO	super critical water oxidation
SO_2e	sulphur dioxide equivalents
SS	sewage sludge
SSA	sewage sludge ash
SSP	single superphosphate
STAN	substance flow analysis
TRL	technology readiness levels
TS*	total solid
TSP	triple superphosphate
UL	uncertainty level
UF	uncertainty factor
WAO	wet-air oxidation
WM	waste management
WWTP	wastewater treatment plant

* Due to the publication of this work in different journals, various abbreviations have been used to describe the dry matter content (dry substance (DS), dry matter (DM) and total solid (TS))

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Supplementary tables

Table A 1: Database and calculation of selected P flow: Plant products.

Plant production	Mass flow [t WS yr^{-1}]	UL 1	UF 1	P-conc. [kg P t^{-1} WS]		UL 2	UF 2	P-flow [kg P yr^{-1}]		UF 3	intermediate step
				A1	A2			B1	B2		
soft wheat	1,456,954	1	1.11	3.3	3.5	-0.2	1.03	4,807,948	5,099,339	1.11	293,703,296,450
durum	74,491	1	1.11	3.3	3.5	-0.2	1.03	245,820	260,719	1.11	767,758,706
rye	175,607	1	1.11	3.3	3.5	-0.2	1.03	579,503	614,625	1.11	4,266,785,279
barley	915,865	1	1.11	3.3	3.5	-0.2	1.03	3,022,355	3,205,528	1.11	116,059,331,856
oat	121,088	1	1.11	3.3	3.5	-0.2	1.03	399,590	423,808	1.11	2,028,707,117
corn	2,031,051	1	1.11	2.8	3.5	1.1	1.12	5,686,943	7,108,679	1.16	832,659,290,964
triticale	200,782	1	1.11	3.3	3.5	-0.2	1.03	662,581	702,737	1.11	5,577,848,222
crop	36,268	1	1.11	3.3	3.5	-0.2	1.03	119,684	126,938	1.11	181,997,053
different crop	23,754	1	1.11	3.3	3.5	-0.2	1.03	78,388	83,139	1.11	78,071,162
rice	-										
legumes	97,152	1	1.11	4	5.5	1.4	1.17	388,608	534,336	1.20	6,283,192,458
oilseed	284,160	1	1.11	5	7	1.5	1.18	1,420,800	1,989,120	1.21	91,611,513,190
plant oils	153,887	1	1.11	0	0		1.04	0	0	1.11	0
potatoes	707,308	1	1.11	0.5	1	2.2	1.41	353,654	707,308	1.43	22,936,403,751
potato stark	-										
sugar	436,170	1	1.11	0	0		1.04	0	0	1.11	0
fruits	793,928	1	1.11	0.1	0.2	2.2	1.41	79,393	158,786	1.43	1,155,927,014
wine	257,533	1	1.11	0.01	0.02	2.2	1.41	2,575	5,151	1.43	1,216,281
beer	-										
champignons and mushrums	940	1	1.11	0.7	1.3	2.1	1.36	658	1,222	1.38	62,094
peas	7,312	1	1.11	1	1	2.0	1.33	7,312	7,312	1.35	6,389,470
cucumbers (cornicons	12,992	1	1.11	0.31	0.31	2.0	1.33	4,028	4,028	1.35	1,938,509
cucumbers (salad)	25,189	1	1.11	0.31	0.31	2.0	1.33	7,809	7,809	1.35	7,286,814
cauliflower	6,615	1	1.11	0.44	0.44	2.0	1.33	2,911	2,911	1.35	1,012,413
carrots	78,609	1	1.11	0.35	0.35	2.0	1.33	27,513	27,513	1.35	90,463,437
cabbage, chinese cabbage	37,912	1	1.11	0.39	0.39	2.0	1.33	14,786	14,786	1.35	26,126,128
white and red cabbage	57,205	1	1.11	0.33	0.33	2.0	1.33	18,878	18,878	1.35	42,588,104
melons	445	1	1.11	0.7	1.3	2.1	1.36	312	579	1.38	13,916
pepper, pepperoni	12,415	1	1.11	0.26	0.26	2.0	1.33	3,228	3,228	1.35	1,245,181
tomato	39,459	1	1.11	0.22	0.22	2.0	1.33	8,681	8,681	1.35	9,005,959
red beet	9,678	1	1.11	0.48	0.48	2.0	1.33	4,645	4,645	1.35	2,578,969
salad (icesalad)	51,881	1	1.11	0.35	0.35	2.0	1.33	18,158	18,158	1.35	39,404,445
salad miscellanious	6,040	1	1.11	0.26	0.26	2.0	1.33	1,570	1,570	1.35	294,722
sellery	13,608	1	1.11	0.57	0.57	2.0	1.33	7,757	7,757	1.35	7,190,031
asparagus	2,232	1	1.11	0.39	0.39	2.0	1.33	870	870	1.35	90,555
spinache	10,902	1	1.11	0.48	0.48	2.0	1.33	5,233	5,233	1.35	3,272,557
onions	108,126	1	1.11	0.31	0.31	2.0	1.33	33,519	33,519	1.35	134,269,111
zuccini	4,569	1	1.11	0.26	0.26	2.0	1.33	1,188	1,188	1.35	168,648
other vegetables	55,280	1	1.11	0.7	0.7	2.0	1.33	38,696	38,696	1.35	178,947,073
Total	**8,307,407**							**18,055,593**	**21,228,792**	**1.07**	**1,377,863,687,642**

t WS yr^{-1}, Tons wet substance per year; kg P t^{-1} WS, Kilogram P per ton wet substance; UL 1, Uncertainty level 1 (mass flow); UL 2, Uncertainty level 2 (P concentration); UF 1, Uncertainty factor 1: Uncertainty regarding the mass flow; UF 2, Uncertainty factor 2: Uncertainty regarding the P concentration; UF 3, Uncertainty factor 3: Calculated uncertainty combining mass flow and P concentration; A1/A2, P concentration from (A1) to (A2); B1/B2, P flow from (B1) to (B2); UF 3 Intermediate step.

Table A 2: Database and calculation of selected P flow: Phosphate ore, P raw materials and P fertilizer.

Fertilizer	mass flow [t yr^{-1}]	UL 1	UF 1	P-conc. [kg P t^{-1}]		UL 1	UF 2	P-flow [kg P yr^{-1}]	
				A1	A2			B1	B2
total fertilizer sale	498,000	1	1.11	32	34	-0.1	1.03	15,936,000	16,932,000
fertilizer production	1,400,000	1	1.11	32	34	-0.1	1.03	44,800,000	47,600,000
export fertilizer	1,008,000	1	1.11	32	34	-0.1	1.03	32,256,000	34,272,000

Table A 3: Database and calculation of selected P flow: Animal waste.

Animal waste	mass flow [t WS yr^{-1}]	UL 1	UF 1	P-conc. [kg P t^{-1} WS]		UL 2	UF 2	P-flow [kg P yr^{-1}]		UF3	intermediate step
				A1	A2			B1	B2		
slaugther	267,085	1	1.11	1.5	2	1.3	1.15	400,628	534,170	1.19	5737354683
meat processing	107,235	1	1.11	45	50	0.4	1.05	4,825,575	5,361,750	1.12	3.43238E+11
fallen stock	30,311	1	1.11	45	50	0.4	1.05	249,840	286,138	1.12	920069478.1
milk production	117,765	1	1.11	0.9	1	0.4	1.05	105,989	117,765	1.12	165582400
food trade	40,777	1	1.11	0.9	1.4	2.0	1.33	36,699	57,088	1.35	160956312.1
Kitchen- and food wastes	79,608	1	1.11	0.9	1.4	1.7	1.25	71,647	111,451	1.27	374379619.6
transboundary air traffic	1,537	1	1.11	0.9	1.4	2.0	1.33	1,383	2,152	1.35	228678.4318
Total	**644,318**							**5,691,761**	**6,470,514**	**1.10**	**350,596,143,214**

Table A 4: Overview of approaches for P recovery from the aqueous phase (wastewater, digested sludge (dissolved P), digester supernatant, secondary treated effluent).

Approach	Specifics	Status	Source			Technology				Final product				Efficiency		Reference
			Wastewater	Digester Supernatant	Secondary treated effluent	Crystallisation	Precipitation	Ion-exchange	Other	Struvite	CaP	AlP/FeP	Other	Related to P-flow	Related to WWTP influent	
Adsorption approach		*		x	x		x		x		x			-	-	Pinnekamp et al., 2007
CSIR		*	x	x		x				x				90%	10-25%	Pinnekamp et al., 2007
DHV Crystalactor®	anaerobic tank, acetic acid, cascade	***		x		x					x			85%	max 40%	Brett et al., 1997
Electrochem. approach		*		x		x			x[1]					-	-	Bilbao and Egner, 2012
WWTP Trevisio		***		x		x				x				90%	10-25%	Cecci et al., 2003
Kurita packed bed	raw phosphate as reactor seed	**		x		x				x				90%	10-25%	Pinnekamp et al., 2007
Magnet separator		*	x				x		x			x		-	-	Pinnekamp et al., 2007
Nishihara Reaktor		***		x		x				x				90%	10-25%	Nawamura et al., 2001
NuReSys®		***		x		x				x				90%	10-25%	NuReSys, 2010
Ostara Pearl Reactor®		***		x		x				x				90%	10-25%	Adnan, 2002
PECO	seawater (Mg-source) instead chemicals	**		x		x				x				90%	10-25%	Dockhorn, 2007
PHOSIDIE		*					x	x	x[3]	x				90%	10-25%	Hoffmann and Homa, 2011
Phosnix Unitika		***		x		x				x				90%	10-25%	Ueno and Fujii, 2003
Phospaq®		***		x	x	x				x						Adma et al., 2010
Phostrip®	anaerobic tank, acetic acid	***		x			x				x			99%	-	Kaschka and Weyrer 1999
PRISA	additional retention time and mixing in thickener (raw sludge)	**		x			x			x				99%	max 35%	Montag, 2008
P-RoC		**		x		x					x			85%	10-25%	Berg et al., 2007
REM-NUT®		**			x		x	x			x			99%	50-70%	Liberti et al., 2001
REPHOS®		***		x		x				x				90%	10-25%	Lebek and Lohmar, 2013
STRUVIA®		***		x		x				x				90%	10-25%	Mêlé et et al., 2014
Sydney Waterboard		**		x		x					x			90%	10-25%	Angel, 1999

[1]Crystallization on specific Mg-Kathode, [2]heating, [3]electro dialysis; *Laboratory scale, **Pilot plant, ***Full-scale implementation

Table A 5: Overview of approaches for P recovery from sewage sludge.

Approach	Status	P-elimination		Technology							HM-depollution						Final product					Efficiency		Reference
		Bio-P	chemical P-elimation	Crystallisation	Precipitation	Acidic wet-chemical	Thermal hydrolysis	Wet-oxidation	Super critical water oxidation	Smelt-gassing	No HM-removal required	Sulfidic precipitation	Complexation	Iron bath/flue gas	(Nano) filtration	Process controll	Struvite	CaP	AlP/FeP	Slag	Other	Related to P-flow	Related to WWTP influent	
AirPrex®	***	x[1]		x	x						x						x					80%	10-15%	Heinzmann, 2009
Aqua Reci®	**	x	x						x							x		x				80%	50-70%	Stenmark, 2003
ATZ Iron bath	*	x	x							x				x							x	50%	40%	Mocker et al., 2010
Budenheim	**	x	x			x					x							x				50%	~45%	Stössel, 2013
FIX-Phos	**	x[1]		x							x							x				90%	20-30%	Petzet et al., 2012
HeatPhos	**	x[1]					x				x						x					-	-	Hirota et al., 2010
KREPRO®	**	x[1]				x	x					x					x					-	-	Berg and Schaum, 2005
LysoGest®	***	x[1]		x	x		x				x						x	x				-	-	Ewert, 2012
MEPHREC®	**	x	x							x				x						x		80%	70%	Scheidig et al., 2009
PHOXNAN	**	x[1]	x			x		x							x		x					55%	~50%	Blöcher et al., 2012
Seaborne®/Gifhorner	***	x				x						x					x	x	x			50%	~45%	Esemen, 2013
Stuttgart process	**	x				x							x				x	x	x			50%	~45%	Meyer et al., 2012

[1] BioP wastewater treatment implicitly necessary; *Laboratory scale, **Pilot plant, ***Full-scale implementation

Table A 6: Overview of approaches for P recovery from sewage sludge.

Approach	Status	P-removal		Technology							HM-depollution						Final product						Efficiency		Reference
		Al-precipitation	Fe-precipitation	Wet-chemical leaching	Wet-chemical extraction	Thermo-chemical	Electro-thermal/ reductive	Smelt-gasing	Elektrokinetisch	Bioleaching	No-depollution	Ion-exchange	Sulfuric precipitation	Solvent extraction	Process controll	Unknown	Struvite	CaP	AlP/FeP	Slag	P_4/P-acid	Other	Related to P-flow	Related to WWTP influent	
Ash Dec®	**	x	x			x									x			x	x				~100%	85%	Nowak et al., 2011
BioCon®	*	x	x	x								x							x				-	-	Levlin, 2001
CleanMAP®	***	x[1]		x												x						NH_4-P	-	-	Easy Mining Sweden AB, 2014
Eberhard	*	x	x	x								x	x					x					70-80%	60-70%	Franz, 2008
EcoPhos®	***	x	x	x								x			x	x					x		90%	80%	de Ruiter, 2015
EPHOS	*	x	x						x							x		x					-	-	Sturm et al., 2010
LEACHPHOS®	**	x	x	x							x[1]				x			x	x				70-80%	60-70%	Morf, 2012
LOTUS Gifu City	***	x	x	x											x			x	x				~65	~60%	Takaoto et al., 2010
MEPHREC®	**	x	x					x												x			80%	~70%	see MEPHREC® sewage sludge
PASCH	*	x	x	x										x				x	x				70-80%	60-70%	Montag et al., 2012
P-bac® (Inocre®)	*	x	x							x						x	x	x					-	-	Inocre, 2013
RecoPhos®	****	x	x	x							x												100%	~85%	Weigand et al, 2011
RecoPhos Inducarb	**	x	x				x														x		80%	70%	Rapf et al., 2010
SEPHOS	*	x[1]	x	x							x[1]							x	x				70-80%	60-70%	Schaum et al., 2004
SESAL-Phos	*	x[1]	x	x											x			x	x				70%	~65%	Petzet et al., 2012
TetraPhos Remondis	**	x	x	x								x			x	x					x		90%	80%	Remondis, 2015
Thermphos®	****	x[1]					x								x						x		95%	85%	Schipper et al., 2001
Fertilizer industry	****	x	x	x							x												95-100%	~85%	Ten Wolde, 2013

[1] Al-phosphorus precipitation implicitly necessary; *Laboratory scale, **Pilot plant, ***Full-scale implementation, **** Industrial scale

Table A 7: Nutrient and heavy metal concentrations in selected final products from P recovery approaches, raw phosphate and final mineral fertilizers (SSP, TSP).

	P	N	Ca	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
	g kg DS^{-1}			mg kg DS^{-1}							
Process water											
Ostara®	122	50	-	1	0.5	2.1	67	-	8.7	-	29
DHV®	113	0	240	0.9	0.4	2.1	2.3	<0.1	7.6	2.1	36.6
P-RoC	97	0	250	<5	<0.3	<1-84	<7-22	-	4-37	<5	9-95
PRISA	93	12	99	-	1.8	12	76	0.15	10	13	403
Effluent											
REM-NUT®	118	50	67	0.15	0.25	0.15	0.1	0.25	1	2.25	12.5
Sewage sludge											
Sewage sludge[1]	15-40	25-65	-	4-10	0.5-3	3-60	120-300	0.5-2.3	17-40	20-145	500-1.300
AirPrex®	100	50	61	-	0.25	-	25.5	<0.1	3.6	6.5	50
Seaborne	120	23	129	-	0.1	1.5	11.5	<0.1	17.6	0.5	23.7
Stuttgarter[3]	273	83	-	<2.0	0.8	2.5	<2	<0.1	2.5	<2	7.0
Aqua Reci®	165	0	390	0.7	0.53	0.2	38.8	<0.1	0.9	28.0	141.6
PHOXNAN	117	51	-	-	0.25	1.0	1.7	<0.1	1.1	0.25	6.5
MEPHREC®	35	0	~300	-	0.2	68	123	<0.1	13	2.5	11
Sewage sludge ash											
SSA untreated[2]	50-100	0	-	8-20	1-6	6-120	240-600	<0.1	34-80	40-290	1.000-1.300
Ash Dec®	85	0	100	10.6	0.1	89.6	56.6	<0.1	0.9	12.3	389
Reco-Phos®	166	0	100	9.1	2.2	118	664	0.7	47.4	51.4	1.600
LEACHPHOS	135	0	170	-	1.0	25.0	600	0.5	15.0	12.5	1.500
PASCH[4]	110	0	170	-	0.65	21.5	29.3	-	5.3	13.6	76.5
SESAL-Phos[4]	153	0	29.8	27.9	0.07	-	4.5	-	0.2	3.4	20.7
Fertilizer Ind.	85	0	100	12	3.2	100	560	<0.1	75	120	1.950
Mineral fertilizer (Diettrich and Klose, 2008; Kpomblekou and Tabatabai, 1994;BUWAL, 1991)											
Raw phosphate[5]	90-170	0	-	7-26	2-92	25-637	1-23	<0.1	17-37	1.5-17	204-382
Single Super Phosphate (SSP)	88	0	220	1.2	10.4	89.5	20.1	<0.1	24.4	64.3	161
Triple Super Phosphate (TSP)	210	0	120	1-10	14-52	131-288	5-45	<0.1	17-44	3.5-12	160-490

[1]typical/reference values with possible strong fluctuations; [2]strong fluctuations due to sewages sludge characteristic (heavy metal content, inorganic content) → Assumption of inorganic fraction of 50%; [3]high iron content: 22.000 mg kg DS^{-1}; [4]high aluminium content of ~50.000 mg kg DS^{-1}; [5]Uranium content: 85-191 mg kg DS^{-1} (Rogasik et al., 2007)

Table A 8: Characteristics of the reference WWTP and load of P and heavy metals in wastewater

WWTP		**Element**	**[kg yr^{-1}]**
Influent	200 L PE^{-1} d^{-1}	P	65,700
Sewage sludge	60 g DM PE^{-1} d^{-1}	As	10
Sewage sludge	21.9 kg DM PE^{-1} yr^{-1}	Cd	3
Organic total solid (oDM)	65%	Cr	70
Anaerobic Treatment	47% of oDM	Cu	550
Digested sludge	14.9 kg DM PE^{-1} yr^{-1}	Hg	1.5
Biogas (methane)	7 kg PE^{-1} yr^{-1}	Ni	120
Thickening	5% DM	Pb	90
Dewatering	30% DM	Zn	2,300
P removal	BioP or Fe or Al	AOX	560
Fe precipitant [Fe(II)S]	6.1 kg PE^{-1} yr^{-1}	PAK	14.7
Al precipitant [AlCl$_3$]	4.2 kg PE^{-1} yr^{-1}	PCDD/F	36.5 mg TE*
Polymer	0.07 kg PE^{-1} yr^{-1}		

DM: Dry Matter

Table A 9: Reference WWTP and sludge treatment: Transfer coefficients for P and heavy metals

Input	[WWTP1] Influent		[WWTP 3] Sludge		[ST1] Digested sludge	
Process	(1) Wastewater Treatment		(2.1) Thickening		(2.3) Dewatering	
Output	[WWTP2] **Effluent**	[WWTP3] **Sewage sludge**	[ST4] **Supernatant thickening**	[ST1] **Raw sewage sludge**	[ST5] **Supernatant dewatering**	[WWTP4] **Sewage sludge 30% DM**
P	10	90	5	95	5 (max. 25**)	95 (80**)
As	20	80	2	98	4	96
Cd	35	65	5	95	2	98
Cr	25	75	1	99	2	98
Cu	20	80	3	97	6	94
Hg	25	75	0	100	2	98
Ni	50	50	4	96	6	94
Pb	20	80	3	97	1	99
Zn	30	70	1	99	2	98

*for process [2.2] anaerobic digestion 100% of selected elements is in digested sludge

**for BioP WWTP a higher dissolution rate of P has to be taken into account

Table A 10: Reference co-incineration or mono-incineration of sewage sludge

Input	**[kg PE^{-1} yr^{-1}]**	**Output**	**[kg PE^{-1} yr^{-1}]**
Sewage sludge DM	14	Bottom ash[1]/Fly ash[2]	6.7
Sewage sludge 30% DM	49.7	Filtercake	0.3
Oil	0.3	Flue gas	14.7
Lime	1.1	Wastewater	3.1
Sand	0.2		
Chemicals[3]	0.4		
Water	9.2		

[1]Bottom ash from co-incineration, [2]Fly ash from mono-incineration with a fluidized bed reactor, [3]NaOH (50%): 0.25; NH$_4$ (25%): 0.06; Precipitants (15%): 0.01; FeCl$_3$ (40%): 0.03; Polyelectrolytes: 0.07; HCl (30%): 0.02

Table A 11: Reference co-incineration or mono-incineration and flue gas treatment: Transfer coefficients for P and heavy metals

Input	[WWTP4] Sewage Sludge 30% DM				[MI3] Flue gas		
Process	(3.1) Combustion				(3.2) Flue Gas Treatment		
Output	[MI1] **Fly ash**	[MI2] **Bottom ash**	[MI3] **Flue gas**	[MI7] **Transf. of substance**	[MI4] **Treated flue gas**	[MI5] **Waste-water**	[MI6] **Filter-cake**
P	98.7	98.7	1.3	0	0.3	0	99.7
As	98.2	98.2	0.8	0	5.9	4.6	89.6
Cd	96.9	96.9	3.1	0	5.0	0.1	94.9
Cr	83.5	83.5	16.5	0	0.3	0.6	99.1
Cu	84.5	84.5	15.5	0	0.1	0.0	99.9
Hg	5.0	5.0	95	0	3.5	0.2	96.3
Ni	83.7	83.7	16.3	0	0.1	0.3	99.6
Pb	82.6	82.6	17.4	0	0.8	0.1	99.1
Zn	79.7	79.7	20.3	0	0.3	0.1	99.6

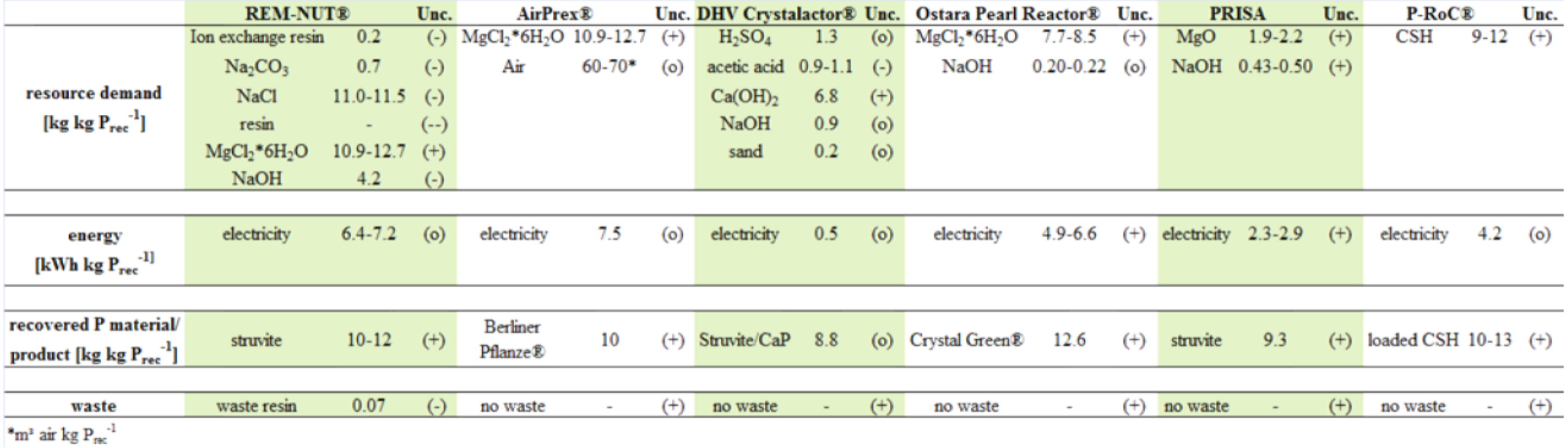
Table A 12: Origin and quality of data (qualitative uncertainty concept). Part A: resource demand, data for SFA of P and HM, nutrient and pollutant content of the recovered material

Technology	(1) Resource demand	Unc.	(2) Data for SFA (P)	Unc.	(3) Data for SFA (HM)	Unc.	(4) Nutrient content	Unc.	(5) Pollutant content (HM)	Unc.
REM-NUT®	Liberti et al. (1986a), Liberti et al. (1986b), Liberti et al. (2001)	o	Liberti et al. (1986a), Liberti et al. (1986b), Liberti et al. (2001)	+	no data (calculated by reference effluent and HM in the final material)	-	Liberti et al. (1986a), Liberti et al. (1986b), Liberti et al. (2001)	+	Liberti et al. (1986a), Liberti et al. (1986b), Liberti et al. (2001)	o
AirPrex®	Bergmans (2011), Heinzmann and Lengemann (2011)	+	Bergmans (2011), Heinzmann and Lengemann (2011)	+	no data (calculated by reference digester supernatant and HM in the final material)	+	Heinzmann (2009), Bergmans (2011), Heinzmann and Lengemann (2011),	+	Kern et al. (2008), Bergmann (2011)	+
DHV Crystalactor®	Gaastra et al. (1998), Adam (2008)	+	Giesen (1999), Pietkeme (2004)	+	no data (calculated by reference digester supernatant and HM in the final material)	+	Giesen (1999)	+	Giesen (1999)	+
Ostara Pearl Reactor®	Fattah (2004), Rieck (2012), P-Rex (2015)	+	Fattah (2004), Rieck (2012)	+	no data (calculated by reference digester supernatant and HM in the final material)	+	Britton et al. (2008), Britton et al. (2009), Crystal Green (2015)	+	Fattah (2004), Britton et al. (2008), Crystal Green (2015)	+
PRISA	Montag (2008)	+	Montag (2008)	+	no data (calculated by reference digester supernatant and HM in the final material)	+	Montag (2008)	+	Montag (2008)	+
P-RoC	Berg et al. (2007), Ehbrecht et al. (2008), Ehbrecht et al. (2011)	+	Berg et al. (2007), Ehbrecht et al. (2008), Ehbrecht et al. (2011)	+	no data (calculated by reference digester supernatant and HM in the final material)	+	Berg et al. (2007), Ehbrecht et al. (2008), Ehbrecht et al. (2011)	+	Schuhmann and Ehbrecht (2010)	+/o
Aqua Reci®	Guibelin (2004), Levlin et al. (2004), Stendahl and Jäfverström (2004), Svanström et al. (2004)	+	Stendahl and Jäfverström (2003), Stendahl and Jäfverström (2004), Levlin et al. (2004), Stark et al. (2006)	+	Stendahl and Jäfverström (2003), Stendahl and Jäfverström (2004), Stark et al. (2006)	+	no data (assessment based on data from other known CaP)	--	no data, calculation based on SFA	-
MEPHREC®	Ingitec (2009), P-Rex (2015)	o	Ingitec (2009)	o	Ingitec (2009)	o	Ingitec (2009), Scheidig et al. (2010)	+	Ingitec (2009), Scheidig et al. (2010)	o
PHOXNAN	Blöcher et al. (2009), Blöcher et al. (2012)	+	Blöcher et al. (2009), Blöcher et al. (2012)	+	Blöcher et al. (2009), Blöcher et al. (2012)	+	Blöcher et al. (2009), Blöcher et al. (2012)	+	Blöcher et al. (2009), Blöcher et al. (2012)	+
Gifhorn®	Günther et al., 2007, Esemen (2013), P-Rex (2015)	+	Günther et al. (2007), Esemen (2013)	+	Günther et al. (2007), Esemen (2013)	+	Günther et al. (2007), Esemen (2013), P-Rex (2015)	+	Günther et al. (2007), Esemen (2013)	+
Stuttgart	Antakyali et al. (2012), Meyer et al. (2012a), Meyer et al. (2012b), Meyer and Steinmetz (2013)	+	Antakyali et al. (2012), Meyer et al. (2012a), Meyer et al. (2012b), Meyer and Steinmetz (2013)	o	no data (calculated by reference sewage sludge and HM in the final material)	-	Meyer et al. (2012a), Meyer et al. (2012b)	+	Meyer et al. (2012a), Meyer et al. (2012b), Preyl (2013)	+
AshDec® ash depollution	Hermann (2013)	+	Hermann (2013)	+	Adam et al. (2009), Mattenberger et al. (2010), Nowak et al., (2011a), Nowak et al, (2011b)	+	Hermann (2013)	+	Hermann (2013)	+
AshDec® Rhenania	Hermann and Reuter (2013), P-Rex (2015)	+	Hermann and Reuter (2013), Hermann (2014)	+	Hermann and Reuter (2013), Hermann (2014)	o	Hermann and Reuter (2013), Hermann (2014)	+	Hermann and Reuter (2013), Hermann (2014)	o
PASCH	Montag et al. (2011)	+	Montag et al. (2011)	+	Montag et al. (2011)	+	Montag et al. (2011)	+	Montag et al. (2009), Montag et al. (2011)	+
LEACHPHOS®	Bühler (2013), P-Rex (2015)	+	Bühler (2013)	+	Bühler (2013)	+	Bühler (2013), Morf (2014)	+	Bühler (2013)	+
EcoPhos®	DeRuiter (2015)	o	DeRuiter (2014), DeRuiter (2015)	+	DeRuiter (2015)	o	DeRuiter (2015)	+	DeRuiter (2015)	+
RecoPhos®	Bohndick (2013)	+	no data necessary (100% of ash in final material)	+	no data necessary (100% of ash in final material)	+	RecoPhos (2011)	+	RecoPhos (2011), Bohndick (2013)	+
Fertilizer Industry	Wiesenberger (2002), Silva and Kulay (2003), Patyk and Reinhardt (1997)	+	no data necessary (100% of ash in final material)	+	no data necessary (100% of ash in final material)	+	Langeveld (2014), Ten Wolde (2013)	+	Langeveld (2014), Ten Wolde (2013)	o
Thermphos®	Hirschberg (1999), Schipper (2012a), Schipper (2012b)	+	Schipper (2012a), Schipper (2012b)	+	Schipper (2012a), Schipper (2012b)	-	no data	--	no data	--

Table A 13: Origin and quality of data (qualitative uncertainty concept). Part B: pollutant content organic micropollutants, solubility and plant availability, capital costs, operating costs, revenues and savings.

Technology	(6) Pollutant content (OM and pathogens)	Unc.	(7) Solubility and plant availability	Unc.	(8) Capital costs	Unc.	(9) Operating costs (based on resource demand)	Unc.	(10) Revenues and savings	Unc.
REM-NUT®	no data	--	no data (assessment based on data from other known MAP)	-	Liberti et al. (1986a), Liberti et al. (1986b), Liberti et al. (2001)	o/-	Liberti et al. (1986a), Liberti et al. (1986b), Liberti et al. (2001)	-	Liberti et al. (1986a), Liberti et al. (1986b), Liberti et al. (2001)	o/-
AirPrex®	Kraus et al. (2015)	+	Römer (2006), Kern et al. (2008), Kratz et al. (2014), Wilken et al. (2015)	+	Ewerts (2009), Ewerts (2011)	+	Ewerts (2009), Ewerts (2011)	+	Ewerts (2009), Ewerts (2011)	+
DHV Crystalactor®	no data	--	no data (assessment based on data from other known MAP)	-	Gaastra et al. (1998), Pietkema (2004), Adam (2008)	o/-	Gaastra et al. (1998), Pietkema (2004), Adam (2008)	o/-	Gaastra et al. (1998), Pietkema (2004)	-
Ostara Pearl Reactor®	Kraus et al. (2015)	+	Crystal Green (2015), Wilken et al. (2015)	+	Rieck (2012)	+	Rieck (2012)	+	Rieck (2012)	+
PRISA	Montag (2008)	+	Montag (2008) Römer (2013)	+	Montag (2008)	+	Montag (2008)	+	Montag (2008)	+
P-RoC	Pinnekamp et al. (2011)	+	Römer (2013), Kratz et al. (2014)	+	Wiebke and Pinnekamp (2011)	+/-	Wiebke and Pinnekamp (2011)	+/-	Wiebke and Pinnekamp (2011)	+/-
Aqua Reci®	Veriansyah and Kim (2007)	+	no data (assessment based on data from other known CaP)	o	no data (Using investment costs from PHOXNAN calculation), Wiebke and Pinnekamp (2011)	--	Stendahl and Jäfverström (2004)	+	Guibelin (2004), Stendahl and Jäfverström (2004)	-
MEPHREC®	no data, but expected destruction of organic pollutants at temperature 2.000°C	o	Cabeza et al. (2011), Kratz et al. (2014), Wilken et al. (2015)	+	Scheidig et al. (2010), Scheidig (2012), Scheidig et al. (2013)	o	Scheidig et al. (2010), Scheidig (2012), Scheidig et al. (2013)	o	Scheidig et al. (2010), Scheidig (2012), Scheidig et al. (2013)	-
PHOXNAN	Blöcher et al. (2009), Blöcher et al. (2012)	+	Römer (2013)	o	Wiebke and Pinnekamp (2011)	+/o	Wiebke and Pinnekamp (2011)	+/o	Wiebke and Pinnekamp (2011)	+/o
Gifhorn®	Pinnekamp et al. (2011)	+	Römer (2006), Kratz et al. (2014)	+	Günther et al. (2007), Esemen (2013)	+	Günther et al. (2007), Esemen (2013)	+	Esemen (2013)	o
Stuttgart	Weidelener (2010), Kraus et al. (2015)	+	Kratz et al. (2014), Wilken et al. (2015)	+	no data (Using investment costs from Gifhorn calculation), Wiebke and Pinnekamp (2011)	-	Antakyali et al. (2012), Meyer et al. (2012a), Meyer et al. (2012b), Meyer and Steinmetz (2013)	+	Meyer and Steinmetz (2013)	o
AshDec® ash depollution	no data, but expected destruction of organic pollutants at temperature >850°C	-	Nanzer et al. (2010), Schick (2010), Römer (2013), Hermann (2014), Nanzer et al. (2014), Henzel et al. (2015)	+	Hermann (2013)	+	Hermann (2013)	+	Hermann (2013)	-
AshDec® Rhenania	no data, but expected destruction of organic pollutants at temperature >850°C	-	Wilken et al. (2015)	-	Hermann and Reuter (2013), Hermann (2014)	+	Hermann and Reuter (2013), Hermann (2014)	o	Hermann and Reuter (2013), Hermann (2014)	-
PASCH	Pinnekamp et al. (2011)	+	Montag et al. (2011), Kratz et al. (2014) Römer (2013)	+	Wiebke and Pinnekamp (2011)	+	Wiebke and Pinnekamp (2011)	+	Wiebke and Pinnekamp (2011)	o
LEACHPHOS®	no data, but expected destruction of organic pollutants at temperature >850°C	-	Morf (2013), Wilken et al. (2015), Morf (2014)	+	Bühler (2013)	+	Bühler (2013)	+	Bühler (2013)	o
EcoPhos®	no data, but expected destruction of organic pollutants at temperature 2.000°C	-	Phosphoric Acid = raw material no fertilizer	+	no data	--	DeRuiter (2015)	o	no data	o
RecoPhos®	no data, but expected destruction of organic pollutants at temperature 2.000°C	-	TUM (2011), Weigand et al. (2012), VonTucher (2013), Weigand et al. (2013)	+	Bohndick (2013)	+	Bohndick (2013)	+	Bohndick (2013)	+
Fertilizer Industry	no data, but expected destruction of organic pollutants at temperature 2.000°C	-	Kratz et al. (2008), Wilken et al. (2015), Römer (2013),	+	no data	--	Wiesenberger (2002), Silva and Kulay (2003), Patyk and Reinhardt (1997)	+	Langeveld (2014), Ten Wolde (2013)	+
Thermphos®	no data, but expected destruction of organic pollutants at temperature 2.000°C	-	P_4 = raw material no fertilizer	*	no data	--	Hirschberg (1999), Schipper (2012b)	o	no data	-

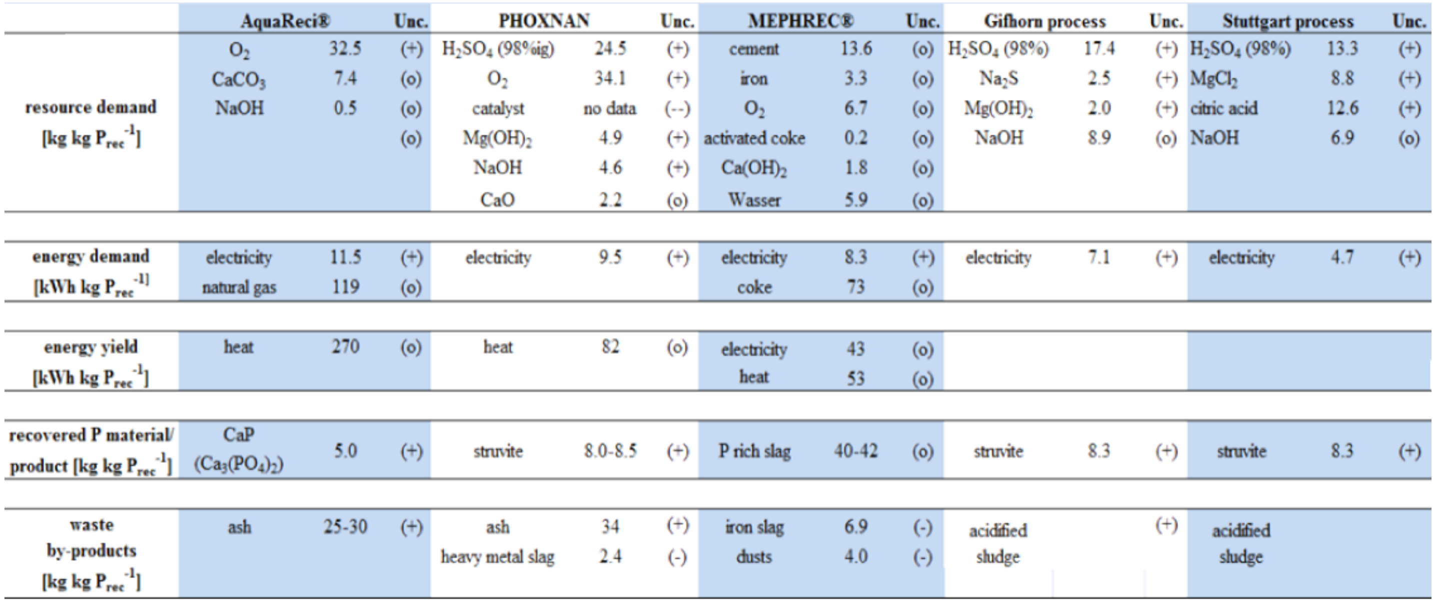
Table A 14: Summary of input and output data on resource- and energy demand, recovered material and occurring waste related to 1 kg P recovered for technologies to recover P from the aqueous phase, including the qualitative uncertainty concept.



	REM-NUT®		Unc.	AirPrex®		Unc.	DHV Crystalactor®		Unc.	Ostara Pearl Reactor®		Unc.	PRISA		Unc.	P-RoC®		Unc.
resource demand [kg kg P_{rec}^{-1}]	Ion exchange resin	0.2	(-)	$MgCl_2$*$6H_2O$	10.9-12.7	(+)	H_2SO_4	1.3	(o)	$MgCl_2$*$6H_2O$	7.7-8.5	(+)	MgO	1.9-2.2	(+)	CSH	9-12	(+)
	Na_2CO_3	0.7	(-)	Air	60-70*	(o)	acetic acid	0.9-1.1	(-)	NaOH	0.20-0.22	(o)	NaOH	0.43-0.50	(+)			
	NaCl	11.0-11.5	(-)				$Ca(OH)_2$	6.8	(+)									
	resin	-	(--)				NaOH	0.9	(o)									
	$MgCl_2$*$6H_2O$	10.9-12.7	(+)				sand	0.2	(o)									
	NaOH	4.2	(-)															
energy [kWh kg P_{rec}^{-1}]	electricity	6.4-7.2	(o)	electricity	7.5	(o)	electricity	0.5	(o)	electricity	4.9-6.6	(+)	electricity	2.3-2.9	(+)	electricity	4.2	(o)
recovered P material/ product [kg kg P_{rec}^{-1}]	struvite	10-12	(+)	Berliner Pflanze®	10	(+)	Struvite/CaP	8.8	(o)	Crystal Green®	12.6	(+)	struvite	9.3	(+)	loaded CSH	10-13	(+)
waste	waste resin	0.07	(-)	no waste	-	(+)	no waste	-	(+)	no waste	-	(+)	no waste	-	(+)	no waste	-	(+)

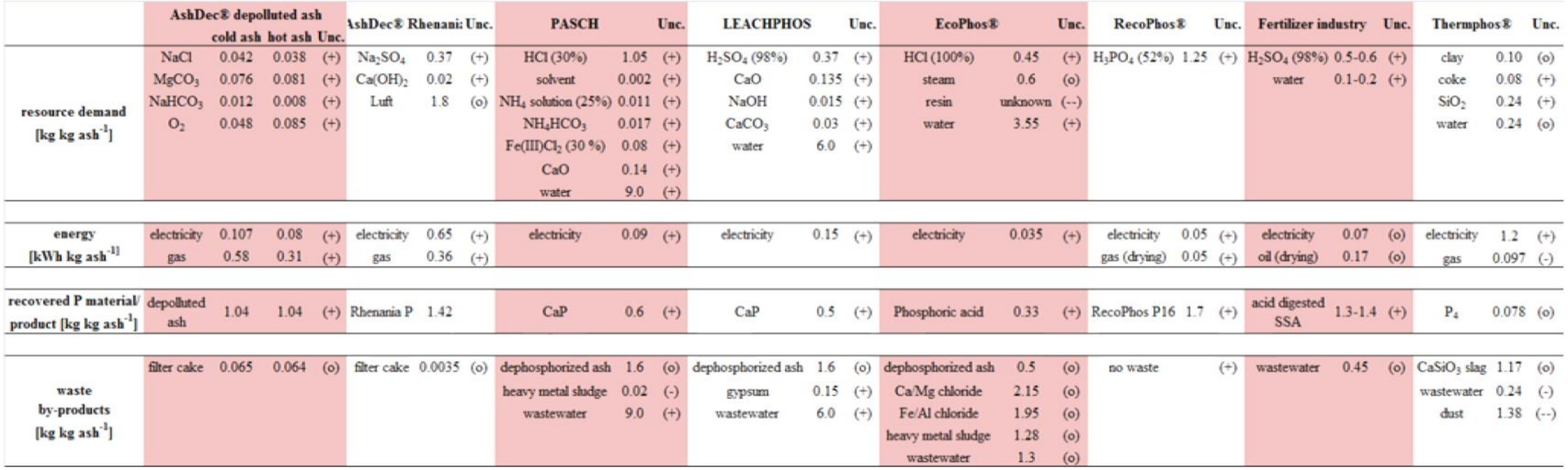
*m^3 air kg P_{rec}^{-1}

Table A 15: Summary of input and output data on resource- and energy demand, recovered material, energy yield and occurring waste related to 1 kg P recovered for technologies to recover P from sewage sludge, including the qualitative uncertainty concept.



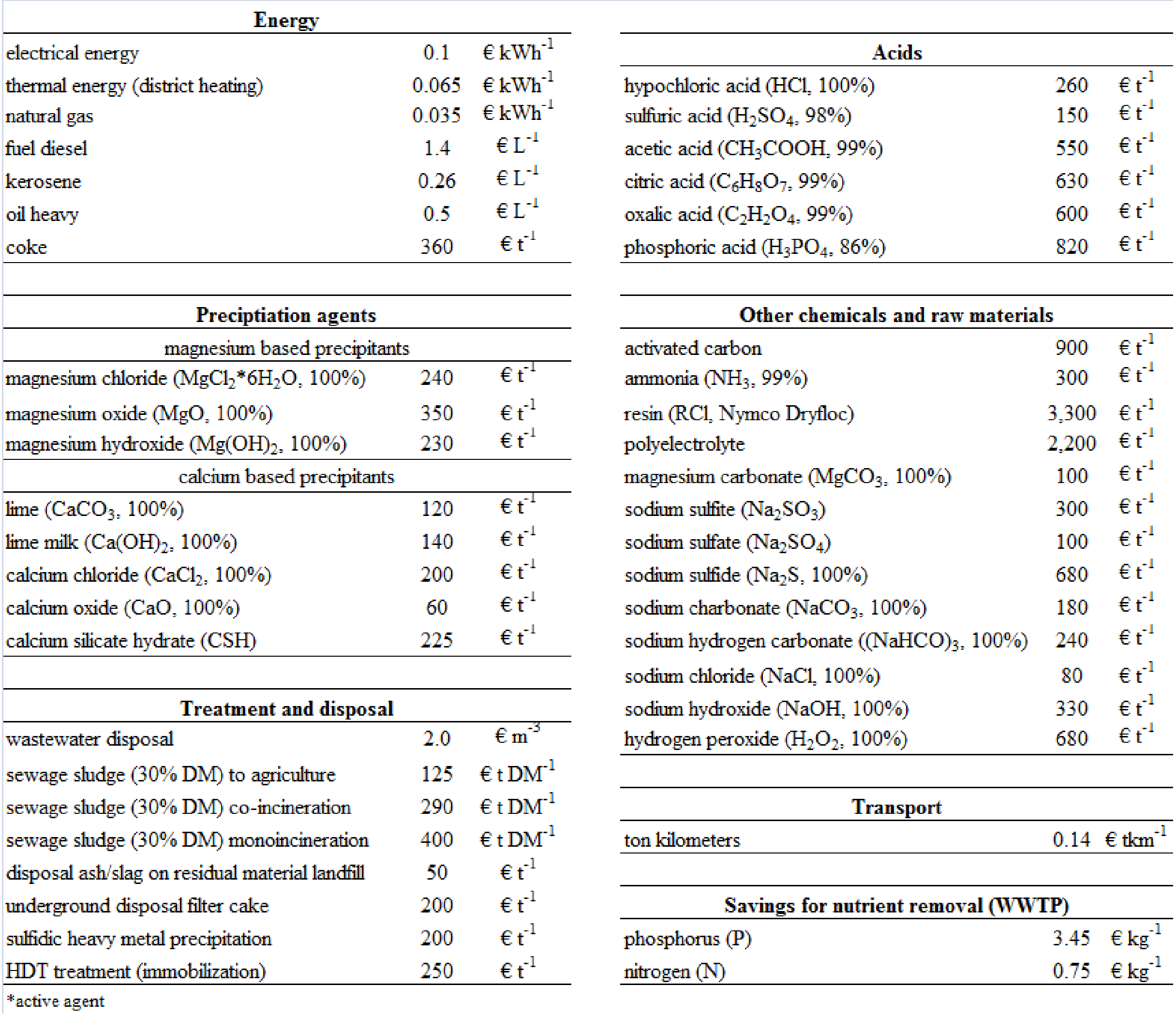
	AquaReci®		Unc.	PHOXNAN		Unc.	MEPHREC®		Unc.	Gifhorn process		Unc.	Stuttgart process		Unc.
resource demand [kg kg P_{rec}^{-1}]	O_2	32.5	(+)	H_2SO_4 (98%ig)	24.5	(+)	cement	13.6	(o)	H_2SO_4 (98%)	17.4	(+)	H_2SO_4 (98%)	13.3	(+)
	$CaCO_3$	7.4	(o)	O_2	34.1	(+)	iron	3.3	(o)	Na_2S	2.5	(+)	$MgCl_2$	8.8	(+)
	NaOH	0.5	(o)	catalyst	no data	(--)	O_2	6.7	(o)	$Mg(OH)_2$	2.0	(+)	citric acid	12.6	(+)
			(o)	$Mg(OH)_2$	4.9	(+)	activated coke	0.2	(o)	NaOH	8.9	(o)	NaOH	6.9	(o)
				NaOH	4.6	(+)	$Ca(OH)_2$	1.8	(o)						
				CaO	2.2	(o)	Wasser	5.9	(o)						
energy demand [kWh kg P_{rec}^{-1}]	electricity	11.5	(+)	electricity	9.5	(+)	electricity	8.3	(+)	electricity	7.1	(+)	electricity	4.7	(+)
	natural gas	119	(o)				coke	73	(o)						
energy yield [kWh kg P_{rec}^{-1}]	heat	270	(o)	heat	82	(o)	electricity	43	(o)						
							heat	53	(o)						
recovered P material/ product [kg kg P_{rec}^{-1}]	CaP ($Ca_3(PO_4)_2$)	5.0	(+)	struvite	8.0-8.5	(+)	P rich slag	40-42	(o)	struvite	8.3	(+)	struvite	8.3	(+)
waste by-products [kg kg P_{rec}^{-1}]	ash	25-30	(+)	ash	34	(+)	iron slag	6.9	(-)	acidified sludge		(+)	acidified sludge		
				heavy metal slag	2.4	(-)	dusts	4.0	(-)						

Table A 16: Summary of input and output data on resource- and energy demand, recovered material, energy yield and occurring waste related to 1 kg ash for technologies to recover P from sewage sludge, including the qualitative uncertainty concept.



	AshDec® depolluted ash				AshDec® Rhenani:		Unc.	PASCH		Unc.	LEACHPHOS		Unc.	EcoPhos®		Unc.	RecoPhos®		Unc.	Fertilizer industry		Unc.	Thermphos®		Unc.
		cold ash	hot ash	Unc.																					
resource demand [kg kg ash^{-1}]	NaCl	0.042	0.038	(+)	Na_2SO_4	0.37	(+)	HCl (30%)	1.05	(+)	H_2SO_4 (98%)	0.37	(+)	HCl (100%)	0.45	(+)	H_3PO_4 (52%)	1.25	(+)	H_2SO_4 (98%)	0.5-0.6	(+)	clay	0.10	(o)
	$MgCO_3$	0.076	0.081	(+)	$Ca(OH)_2$	0.02	(+)	solvent	0.002	(+)	CaO	0.135	(+)	steam	0.6	(o)				water	0.1-0.2	(+)	coke	0.08	(+)
	$NaHCO_3$	0.012	0.008	(+)	Luft	1.8	(o)	NH_4 solution (25%)	0.011	(+)	NaOH	0.015	(+)	resin	unknown	(--)							SiO_2	0.24	(+)
	O_2	0.048	0.085	(+)				NH_4HCO_3	0.017	(+)	$CaCO_3$	0.03	(+)	water	3.55	(+)							water	0.24	(o)
								$Fe(III)Cl_2$ (30 %)	0.08	(+)	water	6.0	(+)												
								CaO	0.14	(+)															
								water	9.0	(+)															
energy [kWh kg ash^{-1}]	electricity	0.107	0.08	(+)	electricity	0.65	(+)	electricity	0.09	(+)	electricity	0.15	(+)	electricity	0.035	(+)	electricity	0.05	(+)	electricity	0.07	(o)	electricity	1.2	(+)
	gas	0.58	0.31	(+)	gas	0.36	(+)										gas (drying)	0.05	(+)	oil (drying)	0.17	(o)	gas	0.097	(-)
recovered P material/ product [kg kg ash^{-1}]	depolluted ash	1.04	1.04	(+)	Rhenania P	1.42		CaP	0.6	(+)	CaP	0.5	(+)	Phosphoric acid	0.33	(+)	RecoPhos P16	1.7	(+)	acid digested SSA	1.3-1.4	(+)	P_4	0.078	(o)
waste by-products [kg kg ash^{-1}]	filter cake	0.065	0.064	(o)	filter cake	0.0035	(o)	dephosphorized ash	1.6	(o)	dephosphorized ash	1.6	(o)	dephosphorized ash	0.5	(o)	no waste		(+)	wastewater	0.45	(o)	$CaSiO_3$ slag	1.17	(o)
								heavy metal sludge	0.02	(-)	gypsum	0.15	(+)	Ca/Mg chloride	2.15	(o)							wastewater	0.24	(-)
								wastewater	9.0	(+)	wastewater	6.0	(+)	Fe/Al chloride	1.95	(o)							dust	1.38	(--)
														heavy metal sludge	1.28	(o)									
														wastewater	1.3	(o)									

Table A 17: Costs for chemicals, raw materials, energy, treatment and disposal of wastes, transport and savings



Energy		
electrical energy	0.1	€ kWh^{-1}
thermal energy (district heating)	0.065	€ kWh^{-1}
natural gas	0.035	€ kWh^{-1}
fuel diesel	1.4	€ L^{-1}
kerosene	0.26	€ L^{-1}
oil heavy	0.5	€ L^{-1}
coke	360	€ t^{-1}

Preciptiation agents		
magnesium based precipitants		
magnesium chloride ($MgCl_2$*$6H_2O$, 100%)	240	€ t^{-1}
magnesium oxide (MgO, 100%)	350	€ t^{-1}
magnesium hydroxide ($Mg(OH)_2$, 100%)	230	€ t^{-1}
calcium based precipitants		
lime ($CaCO_3$, 100%)	120	€ t^{-1}
lime milk ($Ca(OH)_2$, 100%)	140	€ t^{-1}
calcium chloride ($CaCl_2$, 100%)	200	€ t^{-1}
calcium oxide (CaO, 100%)	60	€ t^{-1}
calcium silicate hydrate (CSH)	225	€ t^{-1}

Treatment and disposal		
wastewater disposal	2.0	€ m^{-3}
sewage sludge (30% DM) to agriculture	125	€ t DM^{-1}
sewage sludge (30% DM) co-incineration	290	€ t DM^{-1}
sewage sludge (30% DM) monoincineration	400	€ t DM^{-1}
disposal ash/slag on residual material landfill	50	€ t^{-1}
underground disposal filter cake	200	€ t^{-1}
sulfidic heavy metal precipitation	200	€ t^{-1}
HDT treatment (immobilization)	250	€ t^{-1}

*active agent

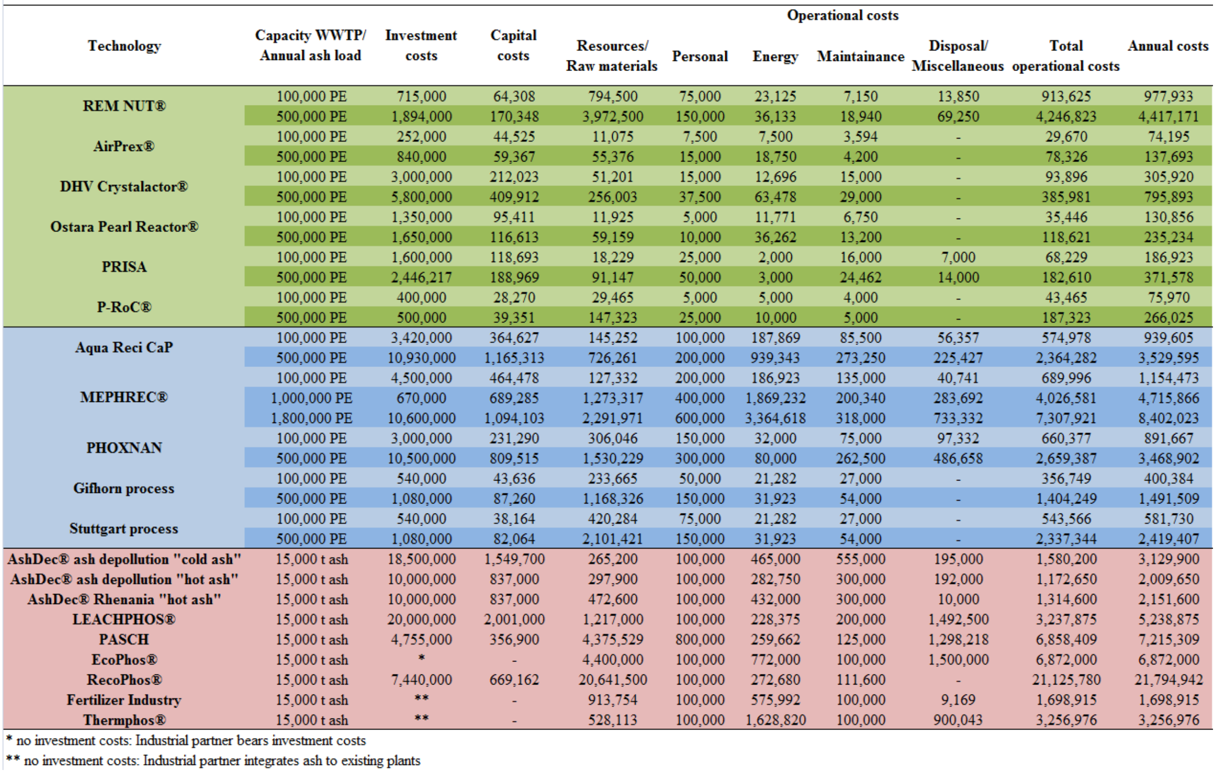
Acids		
hypochloric acid (HCl, 100%)	260	€ t^{-1}
sulfuric acid (H_2SO_4, 98%)	150	€ t^{-1}
acetic acid (CH_3COOH, 99%)	550	€ t^{-1}
citric acid ($C_6H_8O_7$, 99%)	630	€ t^{-1}
oxalic acid ($C_2H_2O_4$, 99%)	600	€ t^{-1}
phosphoric acid (H_3PO_4, 86%)	820	€ t^{-1}

Other chemicals and raw materials		
activated carbon	900	€ t^{-1}
ammonia (NH_3, 99%)	300	€ t^{-1}
resin (RCl, Nymco Dryfloc)	3,300	€ t^{-1}
polyelectrolyte	2,200	€ t^{-1}
magnesium carbonate ($MgCO_3$, 100%)	100	€ t^{-1}
sodium sulfite (Na_2SO_3)	300	€ t^{-1}
sodium sulfate (Na_2SO_4)	100	€ t^{-1}
sodium sulfide (Na_2S, 100%)	680	€ t^{-1}
sodium charbonate ($NaCO_3$, 100%)	180	€ t^{-1}
sodium hydrogen carbonate ($(NaHCO)_3$, 100%)	240	€ t^{-1}
sodium chloride (NaCl, 100%)	80	€ t^{-1}
sodium hydroxide (NaOH, 100%)	330	€ t^{-1}
hydrogen peroxide (H_2O_2, 100%)	680	€ t^{-1}

Transport		
ton kilometers	0.14	€ tkm^{-1}

Savings for nutrient removal (WWTP)		
phosphorus (P)	3.45	€ kg^{-1}
nitrogen (N)	0.75	€ kg^{-1}

Table A 18: Summary of investment costs, capital costs and detailed breakdown of the operating costs into costs for resources, personnel, energy, maintenance and disposal/miscellaneous



Technology	Capacity WWTP/ Annual ash load	Investment costs	Capital costs	Operational costs						Annual costs
				Resources/ Raw materials	Personal	Energy	Maintainance	Disposal/ Miscellaneous	Total operational costs	
REM NUT®	100,000 PE	715,000	64,308	794,500	75,000	23,125	7,150	13,850	913,625	977,933
	500,000 PE	1,894,000	170,348	3,972,500	150,000	36,133	18,940	69,250	4,246,823	4,417,171
AirPrex®	100,000 PE	252,000	44,525	11,075	7,500	7,500	3,594	-	29,670	74,195
	500,000 PE	840,000	59,367	55,376	15,000	18,750	4,200	-	78,326	137,693
DHV Crystalactor®	100,000 PE	3,000,000	212,023	51,201	15,000	12,696	15,000	-	93,896	305,920
	500,000 PE	5,800,000	409,912	256,003	37,500	63,478	29,000	-	385,981	795,893
Ostara Pearl Reactor®	100,000 PE	1,350,000	95,411	11,925	5,000	11,771	6,750	-	35,446	130,856
	500,000 PE	1,650,000	116,613	59,159	10,000	36,262	13,200	-	118,621	235,234
PRISA	100,000 PE	1,600,000	118,693	18,229	25,000	2,000	16,000	7,000	68,229	186,923
	500,000 PE	2,446,217	188,969	91,147	50,000	3,000	24,462	14,000	182,610	371,578
P-RoC®	100,000 PE	400,000	28,270	29,465	5,000	5,000	4,000	-	43,465	75,970
	500,000 PE	500,000	39,351	147,323	25,000	10,000	5,000	-	187,323	266,025
Aqua Reci CaP	100,000 PE	3,420,000	364,627	145,252	100,000	187,869	85,500	56,357	574,978	939,605
	500,000 PE	10,930,000	1,165,313	726,261	200,000	939,343	273,250	225,427	2,364,282	3,529,595
MEPHREC®	100,000 PE	4,500,000	464,478	127,332	200,000	186,923	135,000	40,741	689,996	1,154,473
	1,000,000 PE	670,000	689,285	1,273,317	400,000	1,869,232	200,340	283,692	4,026,581	4,715,866
	1,800,000 PE	10,600,000	1,094,103	2,291,971	600,000	3,364,618	318,000	733,332	7,307,921	8,402,023
PHOXNAN	100,000 PE	3,000,000	231,290	306,046	150,000	32,000	75,000	97,332	660,377	891,667
	500,000 PE	10,500,000	809,515	1,530,229	300,000	80,000	262,500	486,658	2,659,387	3,468,902
Gifhorn process	100,000 PE	540,000	43,636	233,665	50,000	21,282	27,000	-	356,749	400,384
	500,000 PE	1,080,000	87,260	1,168,326	150,000	31,923	54,000	-	1,404,249	1,491,509
Stuttgart process	100,000 PE	540,000	38,164	420,284	75,000	21,282	27,000	-	543,566	581,730
	500,000 PE	1,080,000	82,064	2,101,421	150,000	31,923	54,000	-	2,337,344	2,419,407
AshDec® ash depollution "cold ash"	15,000 t ash	18,500,000	1,549,700	265,200	100,000	465,000	555,000	195,000	1,580,200	3,129,900
AshDec® ash depollution "hot ash"	15,000 t ash	10,000,000	837,000	297,900	100,000	282,750	300,000	192,000	1,172,650	2,009,650
AshDec® Rhenania "hot ash"	15,000 t ash	10,000,000	837,000	472,600	100,000	432,000	300,000	10,000	1,314,600	2,151,600
LEACHPHOS®	15,000 t ash	20,000,000	2,001,000	1,217,000	100,000	228,375	200,000	1,492,500	3,237,875	5,238,875
PASCH	15,000 t ash	4,755,000	356,900	4,375,529	800,000	259,662	125,000	1,298,218	6,858,409	7,215,309
EcoPhos®	15,000 t ash	*	-	4,400,000	100,000	772,000	100,000	1,500,000	6,872,000	6,872,000
RecoPhos®	15,000 t ash	7,440,000	669,162	20,641,500	100,000	272,680	111,600	-	21,125,780	21,794,942
Fertilizer Industry	15,000 t ash	**	-	913,754	100,000	575,992	100,000	9,169	1,698,915	1,698,915
Thermphos®	15,000 t ash	**	-	528,113	100,000	1,628,820	100,000	900,043	3,256,976	3,256,976

* no investment costs: Industrial partner bears investment costs

** no investment costs: Industrial partner integrates ash to existing plants

Table A 19: Technological readiness level with definitions

Technology readiness level	Definition
TRL 1	basic principles observed
TRL 2	technology concept formulated
TRL 3	experimental proof of concept
TRL 4	technology validated in lab
TRL 5	technology validated in relevant environment (industrially relevant environment in the case of key enabling technologies)
TRL 6	technology demonstrated in relevant environment (industrially relevant environment in the case of key enabling technologies)
TRL 7	system prototype demonstration in operational environment
TRL 8	system complete and qualified
TRL 9	actual system proven in operational environment (competitive manufacturing in the case of key enabling technologies)
TRL -	no development visible/expected

Table A 20: Specification of costs for the whole process chain for the reference system

Specification of costs	**€ PE^{-1} yr^{-1}**
Wastewater Treatment	
Wastewater treatment	4.50
Sludge treatment	0.60
Dewatering	0.90
Incineration	
Co-/Mono-Incineration	3.58/4.80*
Disposal ash	0.36
Disposal filter cake	0.06
P recovery	
Technology	-
Revenues	-
Savings	-
Treatment/Disposal of waste	-
Transport	
Sewage sludge to incineration	*0.98*
SSA to landfill	*0.10*
Filter cake to underground disposal	*0.04*
SSA to P recovery technology	-
Wastes from Recovery technology to landfill	-
Costs reference system	***11.12/12.34***

*higher costs for fluidize bed reactors for mono-incineration of sewage sludge (co-incineration: 290 € t DM^{-1}; mono-incineration: 400 € t DM^{-1})

Table A 21: Overview on products and waste outputs, their treatment and final use/disposal from P recovery technologies

	Output	Treatment	(Waste-) Output	Use/Disposal
REM-NUT®	MAP	-	-	Agriculture, raw material for fertilizer industy
	Exhausted resin (Ion-exchanger)	Regeneration (HCl) or unknown disposal	Unknown	Unknown
AirPrex®, DHV, Ostara, P-RoC, PRISA	MAP/CaP	-	-	Agriculture, raw material for fertilizer industy
Aqua Reci®	Calcium or iron phosphate	(Granulation)	-	Agriculture, raw material for fertilizer industy
	Inorganic ash	No	Inorganic ash	Landfill, Construction material, Cement industry
	Exhaust gas	No	Exhaust gas	Atmosphere
MEPHREC®	P-rich slag	Granulation	-	Agriculture, raw material for fertilizer industy
	Waste gas	Gas Cleaning unit, Combustion	Waste water, Exhaust gas	Sewer, Atmosphere
	Iron slag	No	Iron slag	Landfill, Construction material
	Dust containing exhaust air	Filter	Dust	Landfill
PHOXNAN	MAP/CaP/FeP	(Granulation)	-	Agriculture, raw material for fertilizer industy
	Exhaust air	(Bio-) Filter	Exhaust air	Atmosphere
	Sludge with low organic content, (ash)	Drying	Sludge with low organic content, (ash)	Landfill
	Heavy metal sludge	Stabilization/Neutralization	Unknown	Underground waste disposal
	Supernantant	No	Waste water	Sewer
Gifhorn process	MAP/CaP/FeP	(Granulation)	-	Agriculture, raw material for fertilizer industy
	Exhaust air (H_2S)	(Bio-) Filter	Exhaust air	Atmosphere
	Acidified Sludge (rich in Sulfur)	Incineration e.g., cement factory	Ash/Clinker	Landfill/Cement
	Supernantant	No	Waste water	Return to WWTP
Stuttgart process	MAP/CaP/FeP	(Granulation)	-	Agriculture, raw material for fertilizer industy
	Exhaust air (H_2S)	(Bio-) Filter	Exhaust air	Atmosphere
	Acidified Sludge (rich in Sulfur)	Incineration e.g., cement factory	Ash/Clinker	Landfill/Cement
	Supernantant incl. comlexed HM	No, optional HM preciptiation	Waste water	Return to WWTP
AshDec® ($MgCl_2$)	Depolluted ash	(Granulation)	-	Agriculture, raw material for fertilizer industy
	Flue Gas	3-stage flue gas treatment	Filter Cake	Underground waste disposal
AshDec® (Na_2SO_4)	Rhenania ash (partly depolluted)	Granulation	-	Agriculture, input fertilizer production
	Flue Gas	3-stage flue gas treatment	Filter Cake	Underground waste disposal
LEACHPHOS®	Calcium phosphate	No, Granulation	-	Agriculture, raw material for fertilizer industy
	Leached ash	Drying, (Neutralisation)	Dry ash	Landfill, Construction material, Cement industry
	Supernatant (low pH)	Neutralisation (NaOH)	Waste water	Sewer
PASCH	Calcium phosphate	Granulation	-	Agriculture, raw material for fertilizer industy
	Leached ash	Drying, (Neutralisation (lime))	Dry ash	Landfill, Construction material, Cement industry
	Organic solvent (HM removal)	Neutralization/Stabilization	Unknown	Unknown
	Solid residues from reextraction	Neutralisation (NaOH)	Unknown	Underground waste disposal
	Liquid residues from reextraction	Neutralisation (NaOH)	Waste water, Exhaust gas	Sewer
	Supernatant (low pH)	Neutralisation (NaOH)	Waste water	Sewer
EcoPhos®	Phosphoric Acid	Condensation	Condensate	Food-, Feed-, Fertilizer industry
	Leached ash	Drying, (Neutralisation (lime))	Dry ash	Landfill, Construction material, Cement industry
	$Fe/AlCl_3$	No, Condensation	-	Precipitant WWTP
	$Ca/MgCl_2$	No, Condensation	-	e.g., De-icing agend
	Concentrated heavy metal sludge	Neutralization/Stabilization	Filter Cake	Underground
	Exhausted resin (Ion-exchanger)	Regeneration (HCl) or unknown disposal	Unknown	Unknown
RecoPhos®	P-enriched ash	Granulation	-	Agriculture
	Dust	Fabric Filter	No	Recirculation
Fertilizer Industry	Mineral Fertilizer	Granulation	-	Agriculture
	Dust	Fabric Filter	No	Recirculation
	Waste water	Unknown	-	Sewer
Thermphos®	P_4	-	-	Food-, Feed-, Fertilizer industry
	FeP slag	Unknown	FeP slag	Construction material, Landfill
	Waste water	Unknown	Waste water	Sewer
	Flue Gas	Flue Gas Treatment	Filter Cake	Underground waste disposal

Table A 22: Summary results of all assessments criteria

Technology	Recovery potential		TRL	Depollution technology and potential										Recovered material: solubility, relative fertilizer efficiency, pollution potential, handling								
	Recovery process	Related to WWTP influent		heavy metals	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	OM	Recovered material	Solubility			Relative fertilizer efficiceny		Pollution		Handling
															H_2O	CA	NAC	acidic soil	alkaline soil	DU_P	RSM (yr)	
REM-NUT®	90%	50-60%	5-6/-	ion exchange	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[1]	MAP	<1%	90-100%	90-100%	100%	75%	0.010	50,502	+
AirPrex®	85-90%	10-max. 25%	9/9	not necessary	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[1]	MAP	<1%	90-100%	90-100%	100%	75%	0.015	21,510	++
DHV Crystalactor®	85-90%	10-max. 25%	9/9	not necessary	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[1]	CaP	<1%	90-100%	90-100%	75%	50%	0.018	13,681	++
Ostara Pearl Reactor®	85-90%	10-max. 25%	9/9	not necessary	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[1]	MAP	<1%	90-100%	90-100%	100%	75%	0.011	14,514	++
P-RoC®	85-90%	10-max. 25%	5-6/8	not necessary	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[1]	CaP/MAP	<1%	80-100%	80-100%	100%	75%	0.007	20,434	+
PRISA	90%	10-max. 25%	5-6/-	not necessary	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[1]	MAP	<1%	90-100%	90-100%	100%	75%	0.072	9,058	+
Sewage sludge	100%	90%	-	no	🟥	🟥	🟥	🟥	🟥	🟥	🟥	🟥	(-)	sludge	<1%	85%	80-90%	50-90%	70%	0.499	832	-
AquaReci®	~70%	~60%	5-6/7-9	caustic leaching	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[2]	CaP/FeP	<1%	90%	-	75%	50%	0.016	26,875	+
MEPHREC®	~80%	~70%	5-6/7-9	iron slag	🟨	🟩	🟨	🟨	🟩	🟦	🟦	🟦	(+)[3]	P-rich slag	<1%	80-90%	25%	0%	75%	0.105	1,419	++
PHOXNAN	~60%	~40-50%	5-6/-	(ultra/nano-) filtration	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(o)[2]	MAP	<1%	90-100%	-	100%	75%	0.004	80,613	+
Gifhorn	max. 50%	35-50%	9/9	precipitation	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(o)	MAP/CaP/FeP	<1%	50-90%	95%	100%	75%	0.004	71,057	+
Stuttgart	max. 50%	35-50%	5-6/9	complexation	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(o)	MAP/CaP/FeP	<1%	60%	50%	100%	75%	0.033	18,363	+
Sewage sludge ash	100%	87%	-	no	🟥	🟥	🟥	🟥	🟥	🟥	🟥	🟥	(+)[3]	ash	<1%	30-50%	30-40%	25-50%	20%	0.352	1,103	o
AshDec® depoll.	98%	~90%	5-6/9	thermo-chemical	🟧	🟩	🟥	🟩	🟦	🟥	🟦	🟩	(+)[3]	depolluted ash	<1%	30-60%	85%	90%	0%	0.052	2,776	o
AshDec® Rhenania	98%	~90%	5-6/9	thermo-chemical	🟥	🟨	🟥	🟨	🟦	🟥	🟩	🟨	(+)[3]	partly depoll. Ash	<1%	80-90%	-	90%	75%	0.206	762	+
LEACHPHOS®	~70-80%	~60-70%	5-6/7-9	leaching	⬜	🟩	🟩	🟧	🟩	🟩	🟩	🟧	(+)[3]	CaP	<1%	40-90%	90-100%	100%	75%	0.131	878	+
PASCH	~70-80%	~60-70%	5-6/7-9	leaching + organic agent	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[3]	CaP	<1%	80-90%	90-100%	75%	50%	0.025	14,965	+
EcoPhos®	95%	~85%	9/9	leaching + ion exchange	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[3]	Phosphoric acid	100%	100%	100%	100%	100%	0.002	-	++
RecoPhos®	100%	87%	9/9	no	🟥	🟥	🟥	🟥	🟥	🟥	🟥	🟥	(+)[3]	Mineral fertilizer	75%	100%	75%	100%	100%	0.158	3,408	++
Fertilizer Industry	100%	87%	9/9	no	🟥	🟥	🟥	🟥	🟥	🟥	🟥	🟥	(+)[3]	Mineral fertilizer	40-50%	80-90%	20-40%	100%	75%	0.352	1,103	++
Thermphos®	95%	~85%	9/-	silica slag	🟦	🟦	🟦	🟦	🟦	🟦	🟦	🟦	(+)[3]	P_4	-	-	-	-	-	-	-	++
Single Superphosphate	-	-	-	-	-	-	-	-	-	-	-			Mineral fertilizer	80%	100%	100%	100%	100%	0.225	1,450	++

Depollution related to the input flow	
⬜	no data
🟥	<20%
🟧	20-40%
🟨	41-59%
🟩	60-80%
🟦	>80%

Organic Micropollutants (OM)	
+	total destruction of OM
o	OM significantly reduced
-	no OM destruction/depollution

[1] no incorporation of OM during crystallisation process

[2] OM (partly) destruction due to oxidation process

[3] OM destruction due to sludge incineration

Handling	
++	Direct use in agriculture/industry
+	Processing as e.g., granulation necessary
o	Extraction/Mixing and granulation
-	reactive, high water content

Table A 23: Final results table economic assessment

Technology	Economic assessment					
	Annual costs				Δ reference system	
	without revenues and savings € kg P^{-1}	with max. revenues and savings € kg P^{-1}	without revenues and savings € $PE^{-1}a^{-1}$	with max. revenues and savings € kg P^{-1}	Mean Δ in %	± in %
REM-NUT®	28.8	28.7	9.5	8.8	56%	8%
AirPrex®	7.7	-2.0	0.4	-0.2	-4%	2%
DHV Crystalactor®	12.3	3.8	3.1	1.0	-2%	3%
Ostara Pearl Reactor®	9.7	-1.3	1.3	-1.2	27%	2%
P-RoC®	6.1	-0.8	0.7	-0.1	-1%	3%
PRISA	8.4	-1.1	1.5	-0.2	-1%	2%
Sewage sludge	-	-	-	-	-	-
AquaReci®	23.2	-1.1	9.4	0.4	13%	27%
MEPHREC®	13.5	-0.2	4.8	-0.1	-9%	22%
PHOXNAN	26.6	2.7	8.9	0.9	34%	10%
Gifhorn	13.0	5.4	3.9	2.0	27%	6%
Stuttgart	16.6	8.5	5.7	3.2	40%	8%
Sewage sludge ash	-	-	-	-	11%	0%
AshDec® depoll.	1.8	-1.1	1.2	-0.8	14%	9%
AshDec® Rhenania	1.9	-1.1	1.3	-0.9	13%	11%
LEACHPHOS®	5.1	1.7	2.9	1.0	28%	7%
PASCH	4.7	1.6	2.6	0.9	27%	8%
EcoPhos®	4.5	-1.5	3.1	-0.8	21%	20%
RecoPhos®	2.5	0.25	6.2	0.6	41%	27%
Fertilizer Industry	1.2	-1.3	0.8	-0.9	10%	9%
Thermphos®	2.7	-0.35	1.8	-0.2	20%	12%
Single Superphosphate	0.9	-	-	-		

Table A 24: Credits for processes producing electrical- and/or thermal energy and credits for the recovery of nutrients

Credits	CO_2e [g kWh^{-1}]	SO_2e [g kWh^{-1}]
1 kWh electricity	220	0.44
1 kWh thermal energy	350	1.34
1 kg P fertilizer (SSP)	1.253	18.8
1 kg N fertilizer	7.574	28.3

Supplementary figures

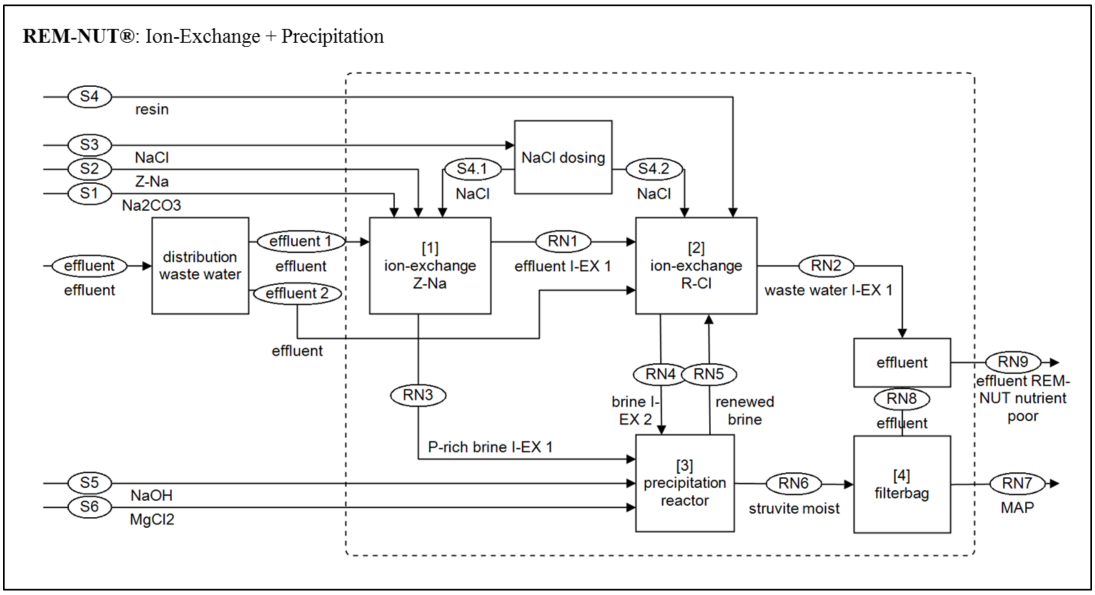


Figure A 1: Material flow models for REM-NUT® to recover P from the secondary treated effluent

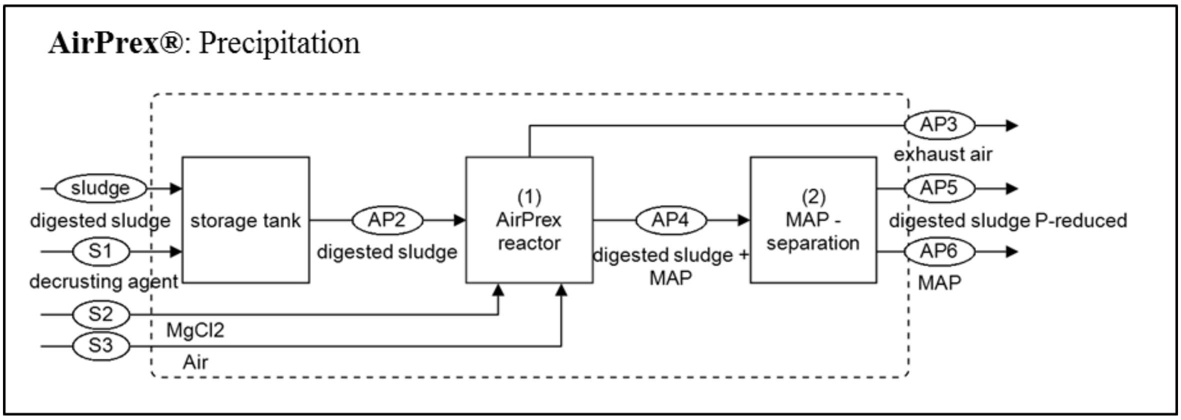


Figure A 2: Material flow models for AirPrex® to recover P from the dissolved fraction of digested sewage sludge

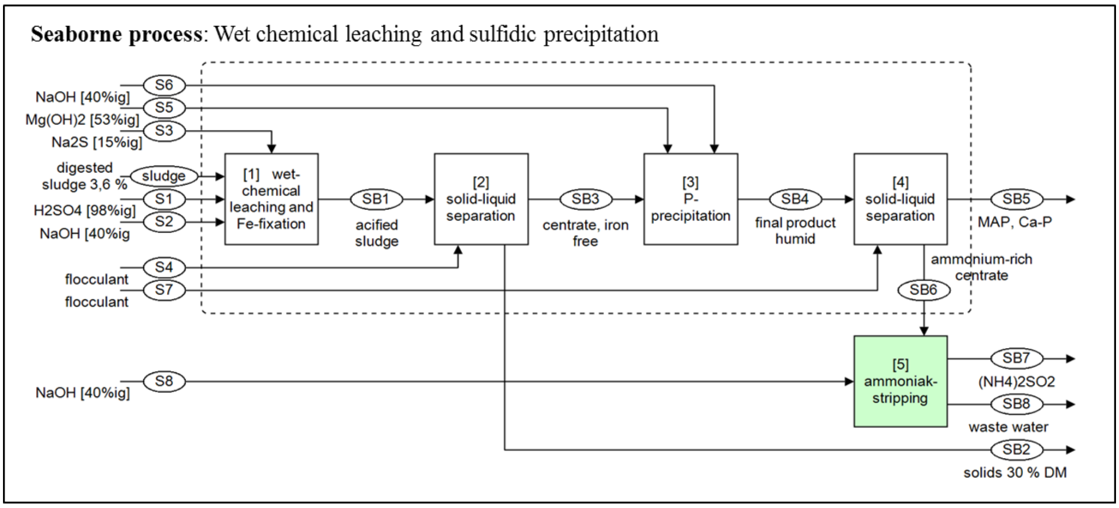


Figure A 3: Material flow models for former Seaborne technologie to recover P from the digested sewage sludge

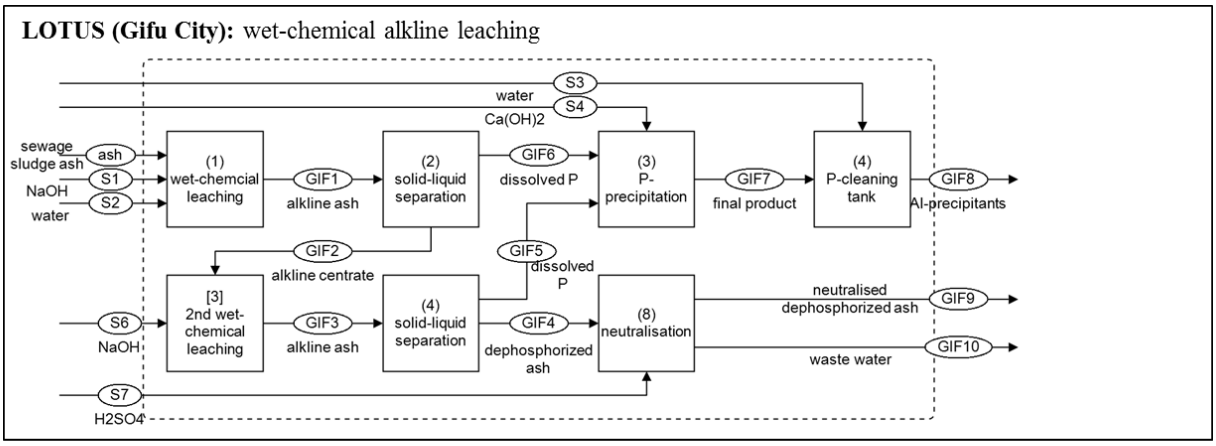


Figure A 4: Material flow model for the wet-chemical leaching technologies LOTUS to recover P from sewage sludge ash

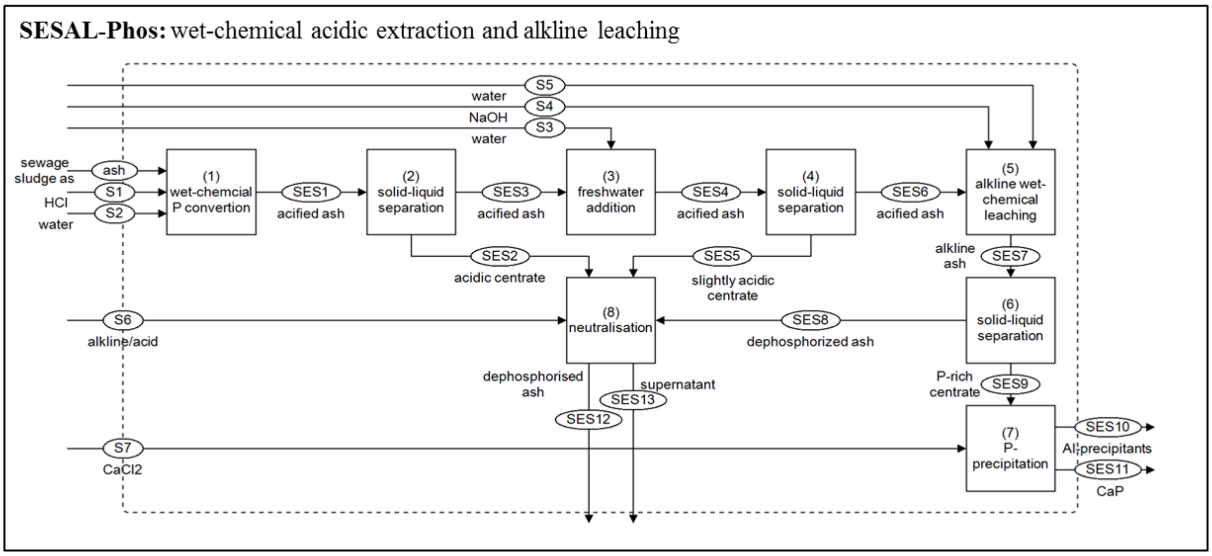


Figure A 5: Material flow model for the wet-chemical acidic extraction and alkaline leaching technology SESAL-Phos to recover P from sewage sludge ash.

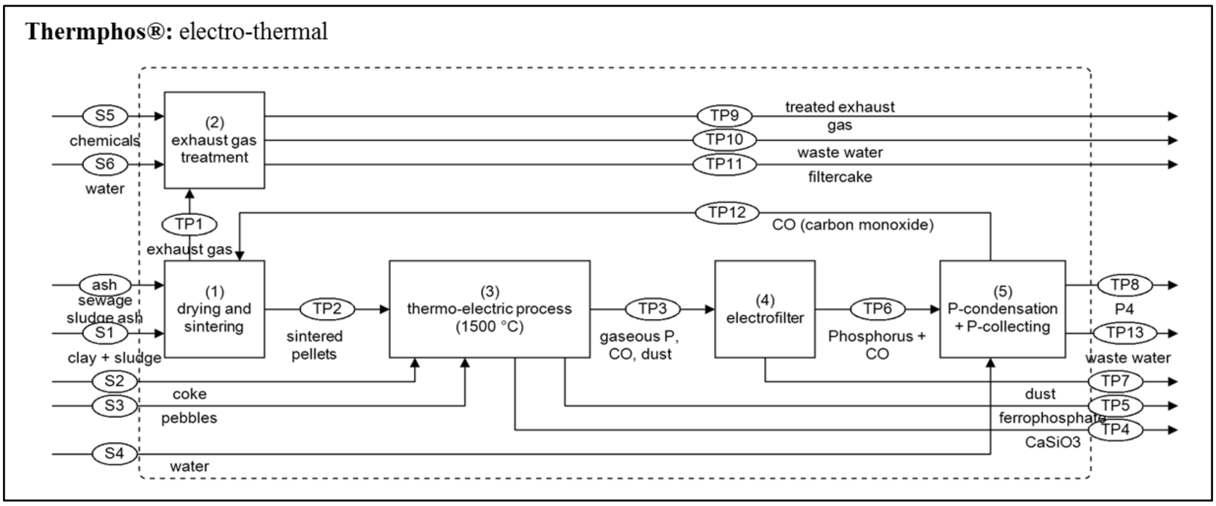
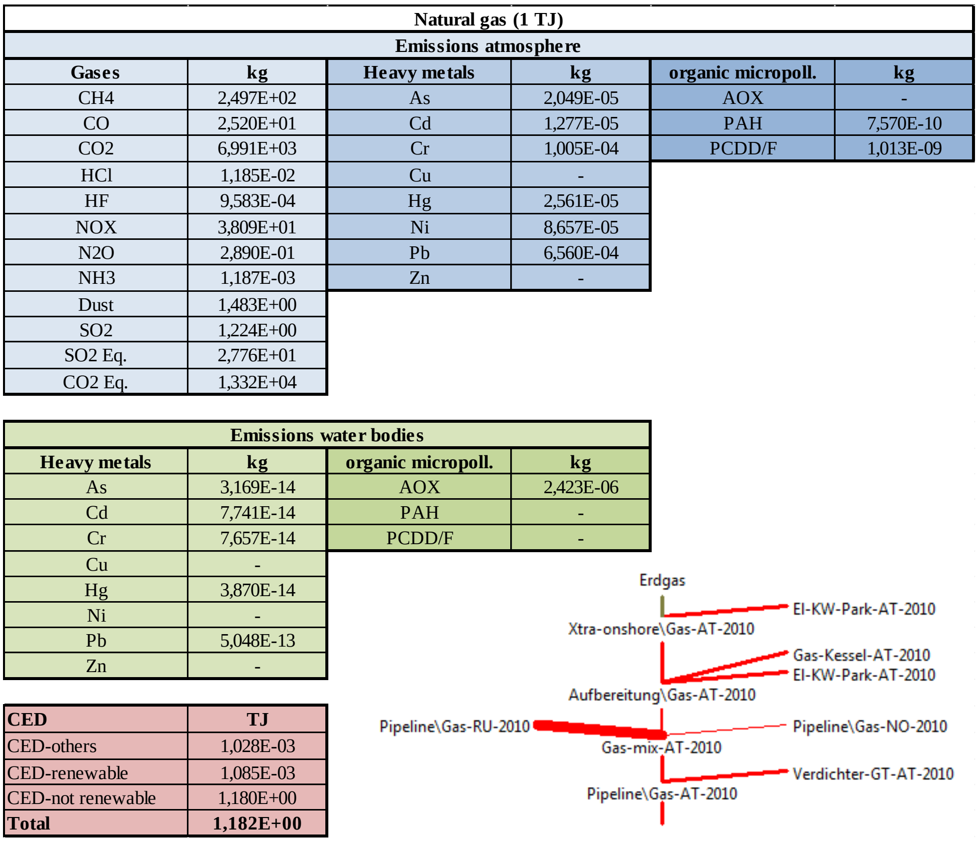


Figure A 6: Material flow model for the thermo-thermal technology Thermphos® to recover P from sewage sludge ash.



Natural gas (1 TJ)					
Emissions atmosphere					
Gases	**kg**	**Heavy metals**	**kg**	**organic micropoll.**	**kg**
CH4	2,497E+02	As	2,049E-05	AOX	-
CO	2,520E+01	Cd	1,277E-05	PAH	7,570E-10
CO2	6,991E+03	Cr	1,005E-04	PCDD/F	1,013E-09
HCl	1,185E-02	Cu	-		
HF	9,583E-04	Hg	2,561E-05		
NOX	3,809E+01	Ni	8,657E-05		
N2O	2,890E-01	Pb	6,560E-04		
NH3	1,187E-03	Zn	-		
Dust	1,483E+00				
SO2	1,224E+00				
SO2 Eq.	2,776E+01				
CO2 Eq.	1,332E+04				

Emissions water bodies			
Heavy metals	**kg**	**organic micropoll.**	**kg**
As	3,169E-14	AOX	2,423E-06
Cd	7,741E-14	PAH	-
Cr	7,657E-14	PCDD/F	-
Cu	-		
Hg	3,870E-14		
Ni	-		
Pb	5,048E-13		
Zn	-		

CED	**TJ**
CED-others	1,028E-03
CED-renewable	1,085E-03
CED-not renewable	1,180E+00
Total	**1,182E+00**

Figure A 7: Prepared data from GEMIS database exemplarily shown for 1 TJ natural gas.

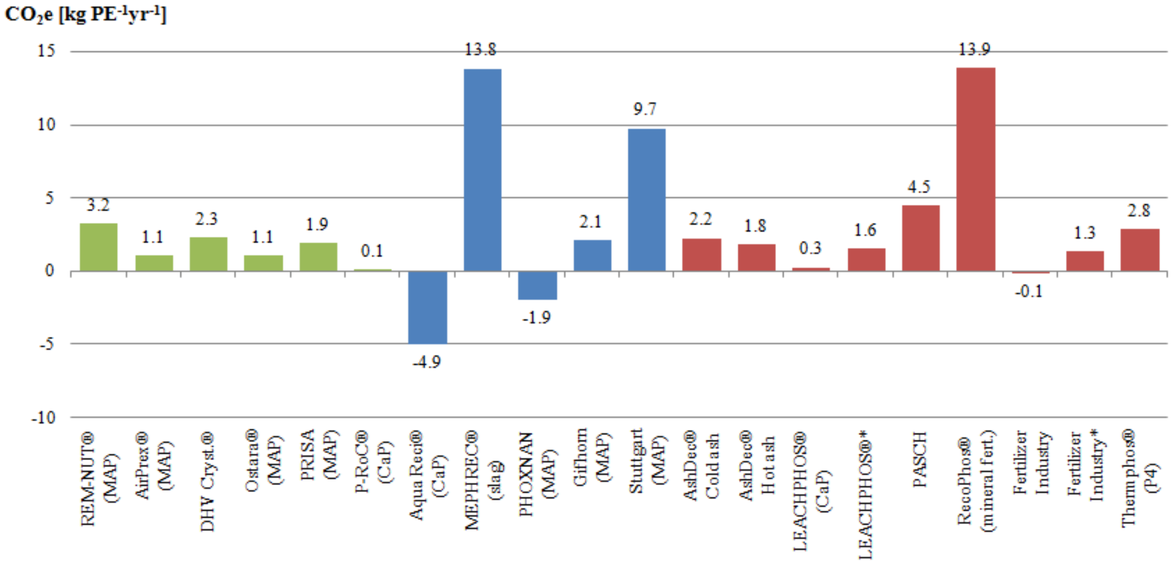


Figure A 8: Greenhouse gas emissions (kg CO2e PE^{-1} yr^{-1}) of the P recovery technologies.

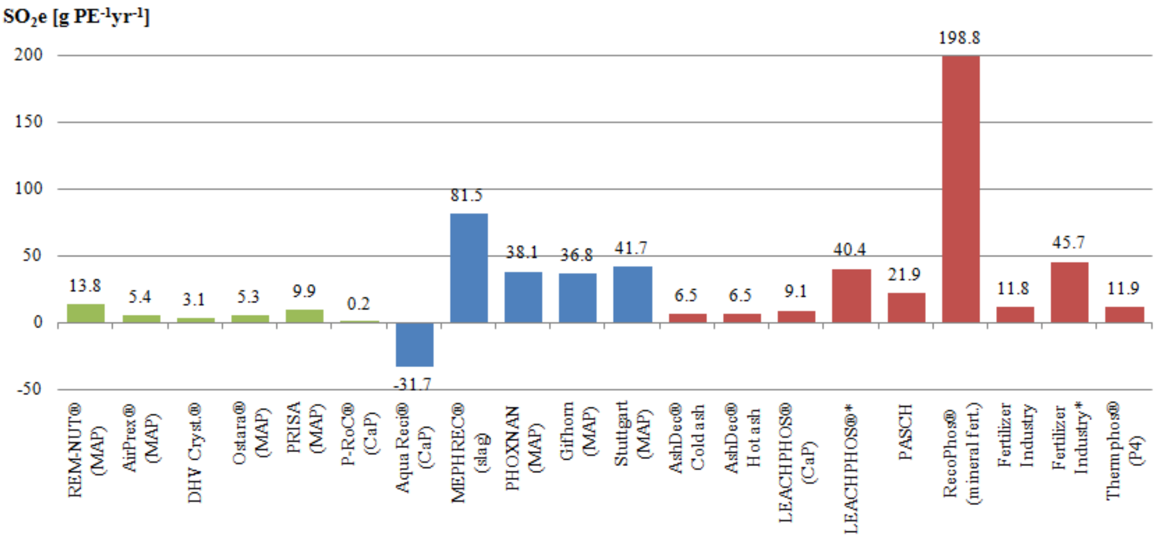


Figure A 9: Acidification potential (g SO2e PE^{-1} yr^{-1}) of the P recovery technologies.

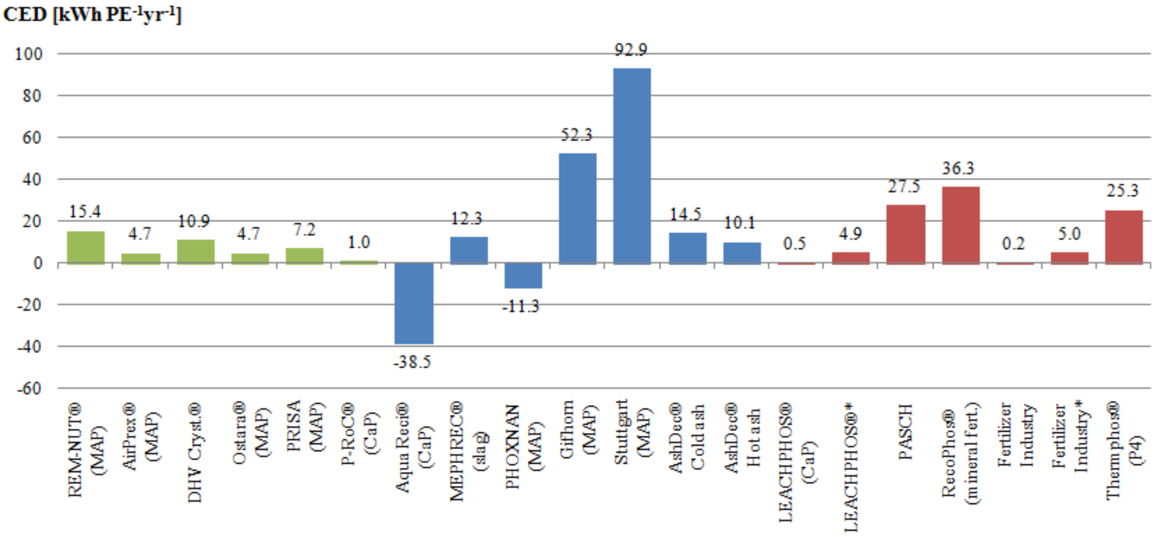


Figure A 10: Cumulative energy demand (kWh PE^{-1} yr^{-1}) of the P recovery technologies.

Autorship

Chapter 2 of this thesis is based on the paper "The Austrian P budget as a basis for resource optimization" by Lukas Egle, Ottavia Zoboli, Helmut Rechberger and Matthias Zessner, Resources, Conservation and Recycling 83, (2014), pp. 152 - 162.

The contribution of Lukas Egle to this paper was:

- concept and method development
- literature research
- scenario development
- data collection and preparation
- performance of the material flow analysis
- paper writing

Chapter 3 of this thesis is based on the paper "Overview and description of technologies for recovering phosphorus from municipal wastewater" Lukas Egle, Helmut Rechberger and Matthias Zessner, Resources, Conservation and Recycling 105, (2015), pp. 325 - 346.

The contribution of Lukas Egle to this paper was:

- literature research, plant visits, interviews, lab tests
- data collection and preparation
- development of the material flow models
- paper writing

Chapter 4 of this thesis is based on the paper "Phosphorus recovery from municipal wastewater: An integrated comparative technological, environmental and economic assessment of P recovery technologies" by Lukas Egle, Helmut Rechberger, Jörg Krampe and Matthias Zessner, Science of the Total Environment (2016), In press.

The contribution of Lukas Egle to this paper was:

- concept and method development
- literature research
- data collection, preparation and evaluation
- performance of the material flow analysis
- paper writing

Chapter 5 of this thesis is based on the paper "Investigations on environmental impacts of technologies to recover wastewater phosphorus by Lukas Egle, Helmut Rechberger and Matthias Zessner (not submitted)

The contribution of Lukas Egle to this paper was:

- concept and method development
- literature research
- data collection, preparation and evaluation
- performance of the Life Cycle Assessment
- paper writing