Recovery of Phosphorus in Sewage Sludge Treatment

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The recovery of phosphorus brings a significant contribution to the saving of resources. In recent years, a variety of methods have been developed for this purpose. Some of these processes have already been successfully implemented and are in industrial use. The technologies start at different points in the material cycle of the phosphorus. A very good possibility is offered by using accumulating phosphorus compounds in wastewater treatment plants. In municipal wastewater, phosphorus loads occur on a significant scale. Here phosphorus is enriched and can be used again after its preparation. The use of the resulting phosphorus as a secondary raw material can make an important contribution to the protection of global resources [14].

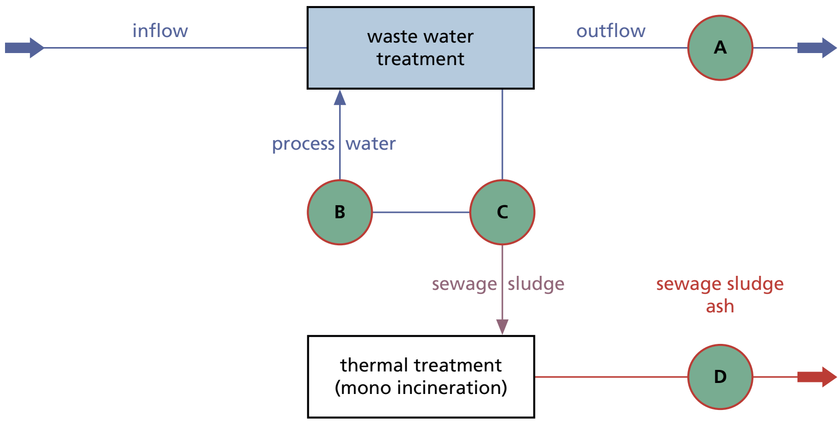
1. Accumulation of phosphorus compounds in sewage sludge treatment

The use and processing of phosphorus compounds lead to the accumulation of phosphorus-containing waste at different locations. Essential for the recovery is the finding of phosphorus streams in which the phosphorus is not returned to the circulation. The largest amount of phosphorus compounds that can be usefully used for recycling is the inflow of municipal wastewater treatment plants [4]. The annual phosphorus freight in Germany is over 61,000 tons/year [5]. Here, a liquid phase (wastewater and water from the sludge treatment), a sludge phase (crude and digested sludge), and a sewage sludge ash phase (from sewage sludge incineration) are distinguished. In the phosphorus streams mentioned here, three main types of phosphorus compounds can be distinguished [14]:

Dissolved phosphorus (PO_4^{3-}): The anion of phosphates is water-soluble and thus represents the most abundant, dissolved form of phosphorus in the water. Phosphorus, which is present as a phosphate ion, can be recovered easily [16].

Biodegraded phosphorus: Here the phosphorus is bounded in the biomass. In the biological stage of wastewater treatment, bacteria absorb phosphorus and grow. The biologically bound phosphorus must first be released for recovery [16].

Chemically bound phosphorus: If phosphorus is bound in metal phosphates, it is called chemically bound phosphorus. In wastewater treatment plants iron or aluminium salts are often used to precipitate dissolved phosphorus as iron or aluminium phosphates. These phosphates are hardly available biologically and show very poor dissolving behaviour [16].



	phosphorus content (relative to inflow)	type of phosphorus compound
A outflow	10 %	dissolved phosphorus
B process water	with precipitation: < 5 % with BioP: -50 %	dissolved phosphorus
C sewage sludge	90 %	dissolved, biological and chemical bounded phosphorus
D sewage sludge ash	90 %	chemical bounded phosphorus

Figure 1: Sites for phosphorus recovery in treatment plants

Sources: Schaum, C.; Cornel, P.: Technologien zur Rückgewinnung von Phosphor aus Abwasser und Klärschlamm, Fachtagung VSA/SVUT, Luzern, 2014

Schönberg, A.; Raupenstrauch, H.; Ponak, C.: Verfahren und Produkte der Phosphor-Rückgewinnung. In: Thiel, S.; Thomé-Kozmiensky, E.; Quicker, P.; Gosten, A. (Eds.): Energie aus Abfall, Band 15. Neuruppin: Thomé-Kozmiensky Verlag GmbH, 2018, pp. 679-691

TBF + Partner AG: Studie Phosphorrückgewinnung aus Abwasser und Klärschlamm, Gesamtbericht, 2015

For the removal and further treatment of phosphorus loads, different locations are available in the wastewater treatment plants. Depending on the location of the removal the phosphorus amounts, concentrations and the type of phosphorus compounds differ. Figure 1 shows the basic locations of phosphorus recycling of a conventional wastewater treatment plant. The respective content of phosphorus in the streams is also a guideline for the maximum recovery potential at this point. Since targeted measures for phosphorus elimination are used in conventional wastewater treatment plants, the potential for recovery in the area of the outflow (A) is about 10 %. In a wastewater treatment plant without targeted phosphorus removal, however, a potential of up to 50 % could be made possible here. When recovering via the process water (B), the potential for recovery of phosphorus is influenced by many processes in the wastewater treatment plant and can be between 5 % and 50 % [7]. In order to supply the largest possible phosphorus quantity to a recovery and thus to achieve the greatest theoretical potential, the points C and D are suitable for removal. However, the types of phosphorus compounds presently require more elaborate recovery processes [14].

Since the respective phosphorus streams differ with respect to their parameters (amounts, concentrations and compounds of phosphorus), there are various possibilities to realize a phosphorus recovery.

Wastewater (treatment plant outflow): The requirements of the waste water according to the Wastewater Ordinance (AbwV) limit the amount of phosphorus in wastewater to a low value. Thus, in the process outflow there is a phosphorus amount of about 10 % based on the feed. In the wastewater treatment plant process it is possible to recover the dissolved phosphorus fraction very well by the use of precipitation. For this purpose, using for example magnesium salts, phosphorus compounds can be obtained with low impurities. Due to the low phosphorus concentrations and the very large volume flows to be treated, this possibility is currently presenting an economic problem [5, 14].

Process water from sludge treatment: Process water (also called sludge water) is circulated in wastewater treatment plants and the phosphorus contained in it is concentrated in sewage sludge. The phosphorus dissolved in the sludge water can (as well as in the outflow water) very well be precipitated with e.g. magnesium salts. The phosphorus content of the process water is highly dependent on the operations in the respective wastewater treatment plants and also limited by the return from the sludge (during dewatering). Thus, the potential for recovery generally varies from about 5 % to 30 % (in special cases also to 50 %, e.g. with Bio-P, see Figure 1 and [7]) with regard to the inflow of wastewater treatment [5, 14].

Sewage sludge: A distinction is made here between dewatered and non-dewatered sewage sludge. In the dewatered sewage sludge almost all of the deposited phosphorus is present, but it exists in a biologically and chemically bound form. In the case of non-dewatered sewage sludge, in addition to the phosphorus bound in sewage sludge, the dissolved phosphorus components in the sludge water are also available. In this case the procedures for the recovery from the process water can be used. In order to reach the bound fractions in the sludge phase, a chemical or physical digestion is required.

The volume flow of the dewatered sewage sludge is already considerably reduced. However, the phosphorus is present in biological and chemical compounds. In order to bring the phosphorus into an available form, further treatment is necessary. With the help of chemical and thermal processes, the recovery of phosphorus can be realized in this area. A substantial role is played by pollutants concentrated in the sludge (e.g. organic pollutants and heavy metals). In the recovery process care must be taken to ensure that these impurities do not impair the product quality [5, 14].

Sewage sludge ash: In order to enable meaningful phosphorus recycling from sewage sludge ash, the sewage sludge should be fed to a mono-incineration plant. This not only significantly reduces the volume flow, but also removes the organic components and eliminates the organic pollutants. The phosphorus is present as chemically bound phosphorus in high concentration. With technically and economically more demanding processes, the recovery of phosphorus with high potential is possible here [5, 14].

2. Approaches to recover phosphorus

Depending on the location of the phosphorus recovery, different concepts and processes can be applied. There is a large number of processes for this, which differ with regard to their technological concepts and operating parameters. Figure 2 is intended to give a (simplified) general overview of procedural approaches that can be used at the respective locations [5, 14].

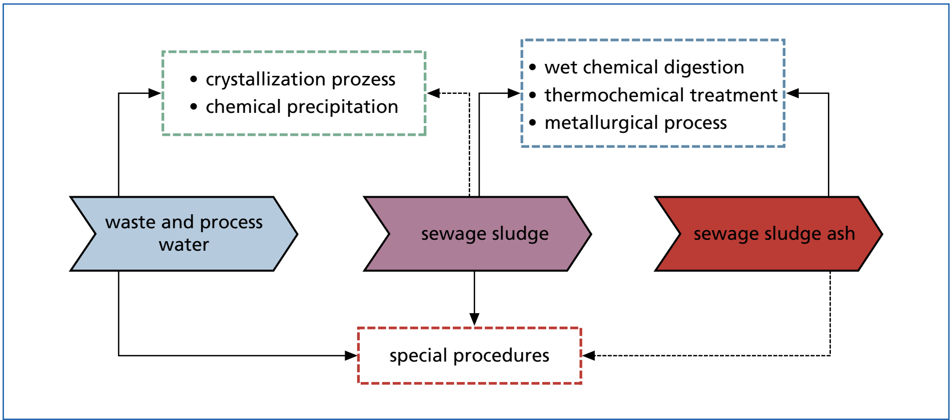


Figure 2: General overview of process approaches to recover phosphorus

Source: Schönberg, A.; Raupenstrauch, H.; Ponak, C.: Verfahren und Produkte der Phosphor-Rückgewinnung. In: Thiel, S.; Thomé-Kozmiensky, E.; Quicker, P.; Gosten, A. (Eds.): Energie aus Abfall, Band 15. Neuruppin: Thomé-Kozmiensky Verlag GmbH, 2018, pp. 679-691

Crystallization process and chemical precipitation: These processes recycle *dissolved phosphorus* (PO_4^{3-}) and are already using industrially proven processes that can recover phosphorus from wastewater, process water and partly from digested sludge [17]. Characteristics are a beginning increase of the pH value by injection of air or leaching by addition of lime or caustic soda. The precipitation or crystallization is usually achieved by adding magnesium or calcium salts. The obtained phosphates (magnesium or calcium ammonium phosphate) can be used as a good plant-available fertilizer after further treatment (e.g. cleaning). Phosphorus which is biologically and chemically bound in the sludge is not captured in these processes, so that the recovery potential is currently limited to about 30 % [5]. In a wastewater treatment plant where P-elimination is carried out, for example with iron and aluminium salts, the phosphorus is chemically bound and thus no longer exists as *dissolved phosphorus* (compare [16]). Great advantages of these methods are not only their relatively simple technical and economic feasibility, but also e.g. the prevention of deposits in pipelines due to spontaneous precipitation processes [14].

Wet chemical digestion – Acid digestion process: With the acid digestion process, it is also possible to convert the biologically and chemically bound phosphorus into a dissolved form. By the use of acids, the pH-value of the sewage sludge or of the sewage sludge ash is reduced, so that the present phosphorus compounds (biologically and chemically bound) dissolve. For this purpose, sulphuric, hydrochloric or phosphoric acid, as well as CO_2 are used. The degree of dissolution depends on the pH value and thus on the acids used. However, the degree of re-dissolution of the phosphorus also increases the re-dissolution of heavy metals which are present in the sewage sludge or in the sewage sludge ash. Before the pH-value of the solution is raised again, any solids are still separated. The increase in the pH-value takes place (as in crystallization and precipitation processes) by adding lime or caustic soda. Subsequently, the phosphorus can be precipitated or crystallized. In order not to generate large amounts of heavy metals in the product, different methods are used, e.g. complexing agents such as citric acid, nanofiltration, solvent extraction or ion-exchangers [5]. The products of these processes, like those from the crystallization and precipitation processes, represent a good plant-available fertilizer. The recovery potential (about 60 %) and the quality of the products depend on the technical and economic effort. An application of the wet-chemical digestion process for the recovery of phosphorus from sewage sludge ash enables a recovery potential of up to 90 % [5, 14].

Thermochemical treatment: A thermochemical treatment can be applied to sewage sludge as well as to sewage sludge ashes. Especially for ashes from mono incineration plants, these methods can be used well. In high concentrations, the phosphorus is chemically bound after combustion. The type of phosphorus elimination in the wastewater treatment plant (biological or precipitation) no longer has any influence here. The thermochemical treatment is not about extracting the phosphorus, but about leaving it in the matrix (sewage sludge, sewage sludge ash) and convert it into an available form. For this purpose, the sewage sludge or sewage sludge ash is heated to high temperatures (depending on the method of about 500 °C [15] up to 1,000 °C [3]). By this organic compounds are destructed and can be removed via the gas stream. To remove the heavy metals, mineral salts (chlorides, sulphates) are usually added to the process. The resulting heavy metal chlorides can then be separated via the gas phase. Also, other additives promote the formation of magnesium and calcium magnesium phosphates. These phosphates have better plant availability characteristics. In principle, recovery potentials of up to 90 % are also available here. In the case of phosphate products from thermochemical processes, it should be noted that in general, important constituents for the fertilizer effect (for example nitrogen and sulphur) also escape in the process [14].

Metallurgical processes: These processes are used for both dried sewage sludge and sewage sludge ash. For this purpose, the sewage sludge or sewage sludge ash, with or without prior treatment, with the addition of additives is brought up to temperatures of about 1,500 °C [13] to about 2,000 °C. As a result, not only the heavy metals are removed in the reactor (via the gas phase by the addition of chlorides and sulphates), but also an iron alloy can be captured as a possible product. Depending on the process, the phosphorus is then in the resulting slag, or is separated via the gas stream. Depending on the process, the products obtained are phosphorus containing slag and elementary phosphorus or thermal phosphoric acid. When using dried sewage sludge as a feed material its energetic use is a great advantage. Here, too, recovery potentials of up to 90 % are available in principle [10, 13, 14].

In a general consideration of the occurring material flows and the occurring phosphorus compounds in wastewater treatment plants, general relationships between the material flows, the potential for the recovery of phosphorus, the plant availability and the technical and economic effort can be observed. Figure 3 shows these relationships symbolically.

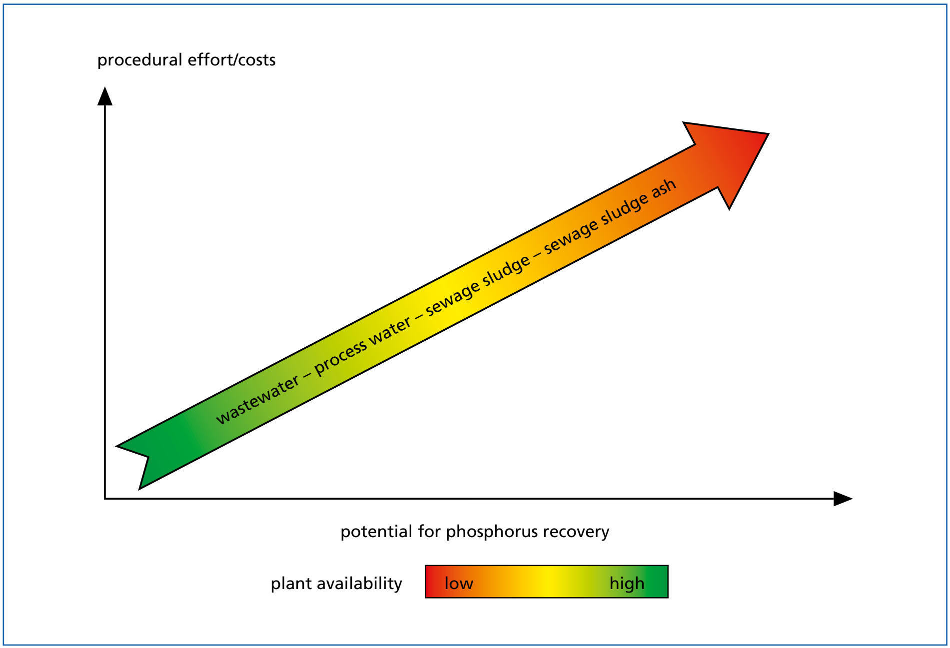


Figure 3: General relation for phosphorus recovery

Sources: LAGA Ad-hoc-AG: Ressourcenschonung durch Phosphorrückgewinnung, Abschlussbericht, 2015

Schönberg, A.; Raupenstrauch, H.; Ponak, C.: Verfahren und Produkte der Phosphor-Rückgewinnung. In: Thiel, S.; Thomé-Kozmiensky, E.; Quicker, P.; Gosten, A. (Hrsg.): Energie aus Abfall, Band 15. Neuruppin: Thomé-Kozmiensky Verlag GmbH, 2018, S. 679-691

In addition, there are further technological concepts, which are assigned here to the special procedures. Some of these approaches should be briefly pointed out here:

Ion exchange process: Here, phosphates are adsorbed from the wastewater by the use of ion exchangers and then separated in a re-dissolving process. The ion exchange medium used contributes significantly to the function of this process [16, 14].

Magnetic separator process: This process aims at recovering phosphorus from the treatment plant effluent and thus provides an alternative to biological and chemical phosphorus removal. In the process, micro particles are composed of super-magnetic Fe_3O_4 nanoparticles embedded in an amorphous quartz matrix. Dissolved phosphorus in the wastewater is adsorbed on these particles, which are then deposited in a magnetic field. In a basic salt solution, the phosphorus adsorbed on the particles can finally be used to recover a phosphate [16, 14].

Forward osmosis membrane distillation: In this process, not only phosphorus is recovered from the sludge water, but also water is treated. Forward osmosis is combined with membrane distillation to provide a solution for areas of water scarcity [16, 14].

Thermal pressure hydrolysis: Here it is attempted to achieve a resolving of the phosphorus from the sludge and to recover it in a subsequent precipitation from the process water. Depending on the process, the sewage sludge is brought under high pressure (up to 220 bar) to a temperature of over 375 °C. Subsequently, the recovery of the phosphorus is carried out by a crystallization or precipitation process [16, 14].

3. Products of phosphorus recovery

In the respective process approaches of phosphorus recovery different phosphorus recycling products are obtained. In most cases, these products are used as phosphorus-containing fertilizers or fertilizer additives. Essential criteria for their suitability as fertilizer must be fulfilled. In addition to legal requirements for use as fertilizer, the fertilizer and environmental properties are particularly important. The properties of the products play a decisive role in the large-scale implementation of recovery processes. A method for the characterization and evaluation of secondary fertilizers is shown in [2], compare [14].

At the beginning, a classification of the end products into 6 classes is carried out:

- magnesium ammonium phosphates,
- calcium phosphate,
- iron and aluminium phosphates,
- slag,
- defecated ash,
- other.

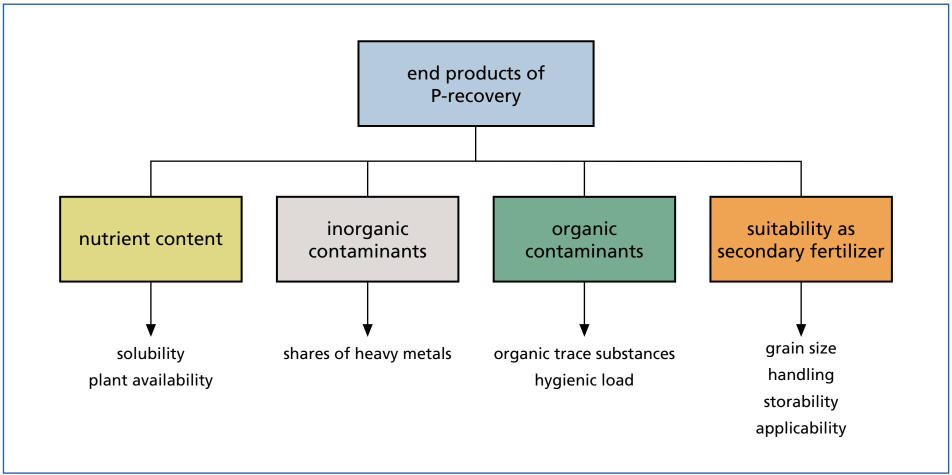


Figure 4: Evaluation method of secondary fertilizer (simplified)

Source: Egle, L.; Rechberger, H.; Zessner, M.: Endbericht Phosphorrückgewinnung aus dem Abwasser, Wien, 2014

Compare: Schönberg, A.; Raupenstrauch, H.; Ponak, C.: Verfahren und Produkte der Phosphor-Rückgewinnung. In: Thiel, S.; Thomé-Kozmiensky, E.; Quicker, P.; Gosten, A. (Hrsg.): Energie aus Abfall, Band 15. Neuruppin: Thomé-Kozmiensky Verlag GmbH, 2018, S. 679-691

For the characterization of the nutrient contents, the products are evaluated according to the substance contents of P, N, Ca, Mg and K, as well as the solubility and plant availability of the phosphorus contained in the final product [2]. However, the water solubility of phosphate fertilizer alone should not be taken as an indicator of the fertilizer effect. More suitable for this is the *citric acid and neutral ammonium citrate solubility* [2, 8]. Magnesium ammonium phosphate (MAP) e.g. has a very good fertilizing effect. The most useful fertilizers are MAP (magnesium ammonium phosphate), calcium phosphate, calcium hydrogen phosphate and phosphate-rich slags [16, 14].

The inorganic impurities consist of heavy metal compounds. To comply with legal requirements currently compounds of As, Cd, Cr, Cu, Ni, Hg, Pb and Zn are relevant. With the stated proportions, usually mg/kg dry substance, an evaluation can be made according to different models (heavy metal to phosphorus ratio, pollution unit model, reference soil method [2, 14].

The destruction or removal of organic compounds in the process (e.g., at high temperatures) can ensure safety in terms of organic contaminants and sanitation. In the suitability as a secondary fertilizer, however, other aspects must be considered. The use in handling the product (for example dust formation, use in spreading machines, etc.) and the storability are essential criteria here [2, 14].

Further product options result from the end products of phosphorus recovery processes, which can not only be used exclusively as fertilizer (additive). With sufficient purity, phosphates, phosphoric acid and elemental phosphorus are versatile in their other applications.

4. Thermal and material utilisation of sewage sludge

In order to utilize the entire potential of the sewage sludge, the material and heat utilization of the material stream should be striven for. There are three possible starting points for the recovery of phosphorus:

- Before the thermal treatment, phosphorus recycling can already take place in the wastewater treatment plant. This results in a larger bandwidth for the subsequent thermal utilization of sewage sludge.
- During the thermal treatment, it is possible (even using the converted thermal energy) to bring phosphorus compounds into an available form.
- After a thermal treatment, a phosphorus recovery with very high recovery rates can be carried out from the ash. As a thermal treatment only the mono-incineration is suitable to ensure a high phosphorus concentration in the ashes.

It is obvious that not only a single process for the entire accumulating sewage sludge offers the solution. Decision making is always associated with criteria such as e.g. meaningfulness, associated effort and deductibility of the products obtained.

In the medium term, thermal treatment of sewage sludge in Germany is to be regarded as mandatory. Within the legal framework and based on the current situation, an expansion of the capacities for thermal utilization results [1]. This applies in particular to mono-incineration plants which are necessary for recycling processes from sewage sludge ash.

The wet chemical processes usually allow a recovery of 5 to 30 % of the phosphorus contained in the treatment plant feed, since in most cases only the part of the phosphorus which is available in the sludge water is recovered. These processes are often relatively simple and inexpensive to implement and provide a low-emission recycled material, which is not least because of its usually good plant availability as NP fertilizer (nitrogen-phosphorus fertilizer) or as a raw material suitable for fertilizer production. However, depending on the degree of subsequent purification of the recycled material, the remaining organic content is relatively high [11].

Thermal or thermo-metallurgical processes are technically more complex in comparison to wet chemical precipitation processes and thus often more expensive to implement and operate. However, many methods allow high recovery of up to 90 % of the phosphorus contained in the treatment plant feed. Another advantage is the destruction of the organic pollutants contained in the sludge during combustion. The plant availability of the recycled materials obtained can be assessed very differently depending on the method. Essential for an efficient thermal or thermo-metallurgical phosphorus recovery from sewage sludge or sewage sludge ash is the mono incineration of the sewage sludge, since here the phosphorus is present in relatively high concentrations and with a manageable amount of impurities (heavy metals) [11].

Gasification, pyrolysis or carbonation (HTC) processes have also been developed for a number of years with the aim of recycling the phosphorus contained in sewage sludge and tested on a large scale. They have the potential to close a gap in sewage sludge disposal [11, 10].

The following table gives an overview of developed processes for phosphorus recycling from waste water, sewage sludge and sewage sludge ash:

Table 1: Overview of developed processes for phosphorus recycling

Waste water / process water	Sewage sludge / digested sludge	Sewage sludge ash
Adsorption-process	Air Prex / MAP-process	AshDec (SUSAN)
Air Prex/ MAP-process	Aqua Reci	BioCon
CSIR fluid bed reactor	ATZ-iron bath reactor	Bioleaching
DHV Crystallactor	CAMBI	Ecophos
Ebara	Elophos	iron bath reactor (ATZ)
Ekobalans	ExtraPhos (Budenheimer process)	Eberhard-Verfahren
Kurita fixed bed	FIX-Phos	EPHOS
Magnet separator	Gifhorner-process	EuPhoRe
MAP Kristallisation Treviso	KEMIKOND	Inocre
Precipitation / flocculation filtration	Kemira-KREPRO	Kubota
NuReBas-process	KREPRO	LEACHPHOS
NuReSys	Leachphos/ Zar	Mephrec (KRN)
Ostara PEARL™	LOPROX	PARAFORCE
Phosiedi	MEPHREC	PASCH
Phosnix	NuReSys	PhosRec (Koop Schiefer)
PHOSPHAQ	Peco	RECOPHOS (AT)
PHOSTRIP	Phostrip	RecoPhos (DE)/Seraplant
PRISA	POPROX-process	Rhenania
P-RoC (Prophos)	PRISA	SEPHOS
RECYPHOS	PROXNAN	SESAL(-Phos)
REPHOS	Seaborne	TetraPhos
RIM NUT Ion-changer	SESAL-Phos	Thermphos
Struvia	Stuttgarter process	
Sydney Water Board Reactor	Unitika-PhosnixÒ	
	Pyrolyse/HTC-process	

Source: Roskosch, A.; Heidecke, P.: Klärschlammentsorgung in der Bundesrepublik Deutschland, Umweltbundesamt, Dessau-Roßlau, 2018

However, it cannot be overlooked that in Germany until 31.12.2023 a report of the sewage sludge producers to the competent authority is to be brought forward

- on planned and initiated measures for P-recovery,
- for the application or introduction of sewage sludge on or in soils or
- for other sewage sludge disposal according to the Law on Life-Cycle Management [6].

	Wet chemical	Thermal or downstream
Advantages	✓ Cost-effective realization ✓ Easy to retrofit and relatively easy to operate ✓ In most cases good plant availability of the recovery products ✓ Operational advantages (spontaneous struvite formation is prevented, improved sludge dewatering)	✓ High recovery rates of > 90 % (legal requirements are met) ✓ Simultaneous material and energetic use of sewage sludge ✓ Complete destruction of organic pollutants ✓ Flexible use (mostly suitable for all sewage sludge and other substances)
Disadvantages	• Recovery rate currently < 50 % (below legal requirements) • Mostly suitable only for Bio-P systems	• High investment costs • Elaborate procedure (technical staff required)

Figure 5: Comparison of wet-chemical and thermal or thermal downstream process

Source: Roskosch, A.; Heidecke, P.: Klärschlammentsorgung in der Bundesrepublik Deutschland, Umweltbundesamt, Dessau-Roßlau, 2018

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