

Phosphorus Recycling in Treatment Plants with Biological Phosphorus Removal

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1. Introduction

Berliner Wasserbetriebe's sewer network is 8600 km long and covers the 891 km² Berlin city area, with about 3,4 million inhabitants. It has over 4011 km of main sewers, 3133 km of storm water sewers and 1900 km of combined wastewater/storm sewerage. The wastewater and the combined storm/wastewater are led to wastewater-pumping stations in a 1000 km long pressurised main network, directing the water to the 7 wastewater treatment plants (WWTP). The wastewater thus takes around 6 to 18 hours to travel from a house discharge to the sewage works. During this long retention time in the draining system, a biological decomposition process already takes place, and easily degradable low-molecular organic acids are generated which provide a substrate for Bio-P bacteria. Thus, extremely favourable conditions are created for Enhanced Biological Phosphorus Removal (Bio-P EBPR) in the wastewater purification. Following successful R&D work for combined "EBPR/nitrification/de-nitrification" in the WWTP Ruhleben [1]; this process was used for planning and upgrading of Berlin's WWTP's. In the Bio-P process, the phosphorus is stored as polyphosphate in the bacteria's cells and thus transferred to the excess sludge which then contains about 3 - 5 % P (% dry matter). The process engineering technology aims to increase phosphate concentrations in activated sludge to only 3 % P, in order to avoid overloading during stormweather and without using chemicals for phosphate's precipitation finishing.

The P-removal efficiency reached with this process combination allows total phosphorus concentrations < 0.5 mg/l to be achieved in the works discharge, as shown for the year 2001 in the following table 1, for the two large WWTP's Waßmannsdorf and Ruhleben. The Waßmannsdorf treatment plant treated an average of 169 014 m³/day wastewater in the year 2001 using this process and the Ruhleben treatment plant an average of 207 600 m³/day wastewater. This together represented about 56 % of the total wastewater volume for the WWTP's of the Berliner Wasserbetriebe.

In this paper, phosphorus balances are calculated for the wastewater purification and sludge treatment stages for these two large WWTP's. The possible P-recovery potential is then evaluated in different places along the process of wastewater purification and sludge treatment, taking the different physical and biological forms of phosphorus into account. Then the problem of the incrustations in the sludge treatment of the Waßmannsdorf treatment plant is considered briefly and the design of the process engineering technology for incrustation avoidance is presented. This technology enables a precipitation product suitable for recycling to be generated and separated from the anaerobic sludge using a hydro-cyclone.

Table 1: Annual average Phosphorus concentrations in influent and effluent in mg/l as well as removal rates in % in the year 2001 – WWTP’s Waßmannsdorf and Ruhleben

Parameter	Wassmannsdorf			Ruhleben		
	Influent	Effluent	Removal	Influent	Effluent	Removal
Suspended solids	388	6.25	98	630	6.7	99
COD	898	45	95	888	50	94
BOD ₅	393	4	99	314	6.5	98
P _{total}	10.52	0.36	96	9.3	0.3	96
NH ₄ -N	50.5	0.3	99	42.1	1.0	98
N _{total}	69.4	11.6	82	64.3	11.0	82

COD *chemical oxygen demand*

BOD₅ *biochemical oxygen demand*

P_{total} *total phosphorus*

NH₄-N *Ammonium-nitrogen*

N_{total} *total nitrogen*

2. Phosphorus balance for the wastewater purification and sludge treatment processes of the Waßmannsdorf and Ruhleben WWTP’s

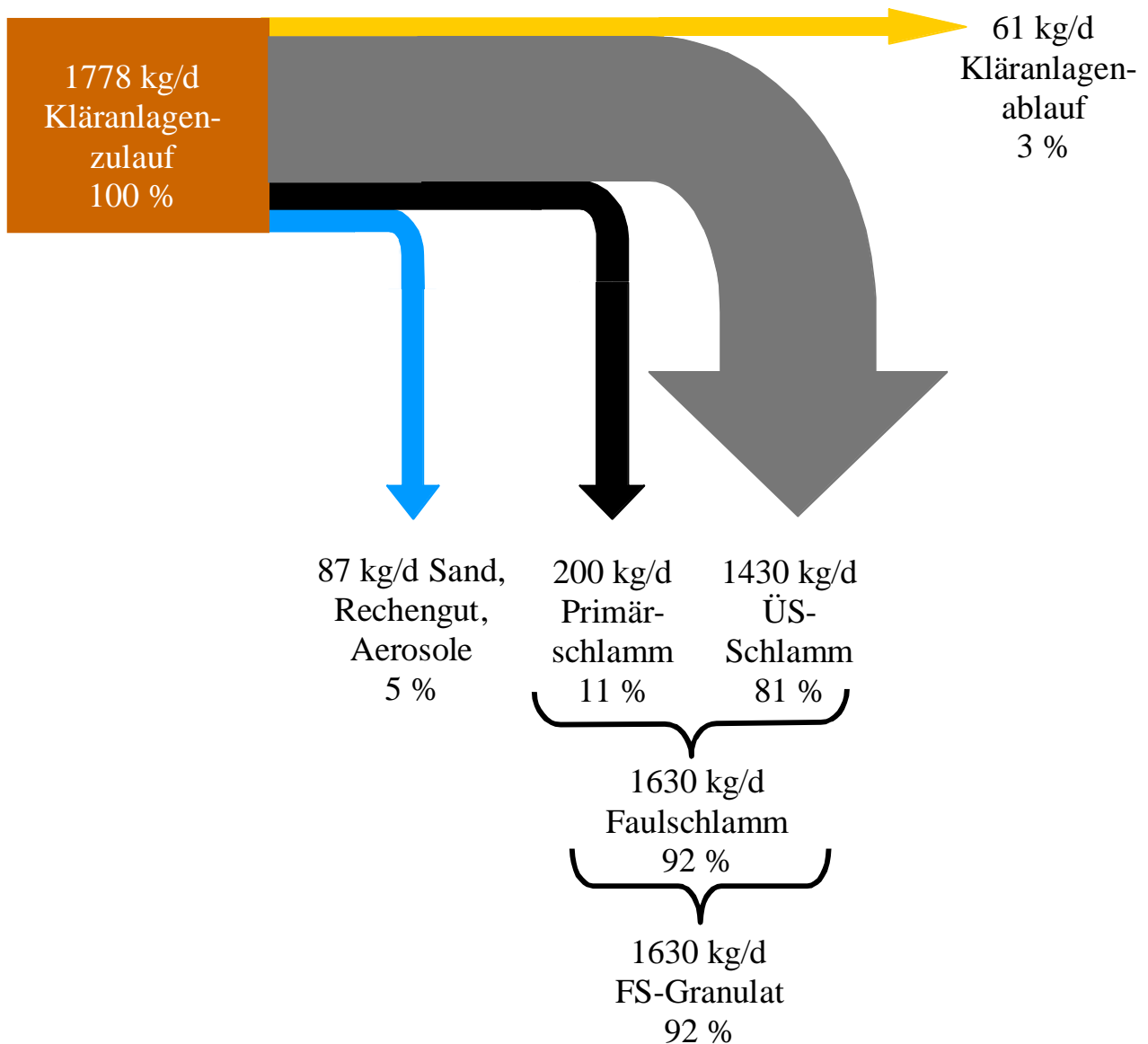
2.1 Sludge treatment in the Waßmannsdorf treatment plant

The primary sludge starts thickening by gravity to approx. 5 % dried solid matter (DSM). The excess-sludge-thickening occurs in two steps, via flotation and centrifugation. At this point, both centrate - from the centrifugation of the anaerobic sludge and from the centrifugation of the excess sludge - are led through the pipe to flotation. As cationic products used for flocculation remain in both centrates, only 1 kg per ton DSM of the coagulant aid is needed in order to centrifuge the excess sludge. As a result, a thickening of about 3 % DSM is reached through flotation and about 8 % DSM is obtained by the next centrifugation. The thickened excess sludge arrives with the primary sludge into a collecting plant mixing the sludge. From here the mixed rough sludge is pumped into the sludge block heating station, and is brought to digestion. Up to six sludge digestion tanks of 8000 m³ each are available in the digestion area. The sludge was digested for approx. 20 days in 2001, and is then led via pipes to a collecting mains for the mechanical drainage using four centrifuges (approx. 28 - 30 % DSM) and finally de-watered on approx. 94 % DSM in up to four sludge drying facilities.

2.2 Phosphorus balance - Waßmannsdorf treatment plant

The phosphorus balance for the Waßmannsdorf treatment plant is represented in Figure 1. About 11% of influent P are removed with the primary sludge, predominantly as organic phosphorus particles. With the excess sludge, approx. 80 % of the P-influent load is extracted. New processes of fixing, dissolution and transformation happen in the digestion [2]. Thanks to the non-availability of chemical precipitation products, e.g. iron dredging from the wastewater treatment, it is possible for the phosphate released and now in solution to become connected to one of the different calcium phosphates and magnesium ammonium phosphate (MAP), only in the context when alkaline-earth ions and cations (Ca²⁺, Mg²⁺) are present. MAP is transformed during the high-temperature de-watering process (drying) and is present as Magnesiumdiphosphate in the granules of anaerobic sludge. In the course of the drying process a hot gas supply of about 550°C is added. During the drying process the ammonium escapes as ammonia into the vapour condensate. Thus the nitrogen of the MAP compound is lost and hence is missing for a possible later use as fertiliser. Approx. 4.9 % of the P- influent load is not traceable and is presumably lost from the system with the screening, the sand or through the formation of aerosols.

<i>Total Phosphorus (P_{total})</i>		
1778 kg/day	100% of Treatment plant influent P	100 %
61 kg/day	Treatment plant effluent	3 %
87 kg/day	Sand, screening, aerosols	5 %
200 kg/day	Primary sludge	11 %
1430 kg/day	Excess sludge	81 %
1630 kg/day	Digested Sludge	92%
1630 kg/day	Dewatered Sludge Granulates	92%



<i>German</i>	<i>English Total Phosphorus (P_{total})</i>
Kläranlagen-zulauf	Treatment plant influent
Kläranlagen-ablauf	Treatment plant effluent
Sand, Rechengut, Aerosole	Sand, screening, aerosols
Primär-schlam	Primary sludge
ÜS-schlam	Excess sludge
Faulschlamm	Digested Sludge
FS-Granulat	Dewatered Sludge Granulates

Figure 1: Phosphorus balance of the Waßmannsdorf treatment plant

2.3 Sludge treatment in the Ruhleben treatment plant plant

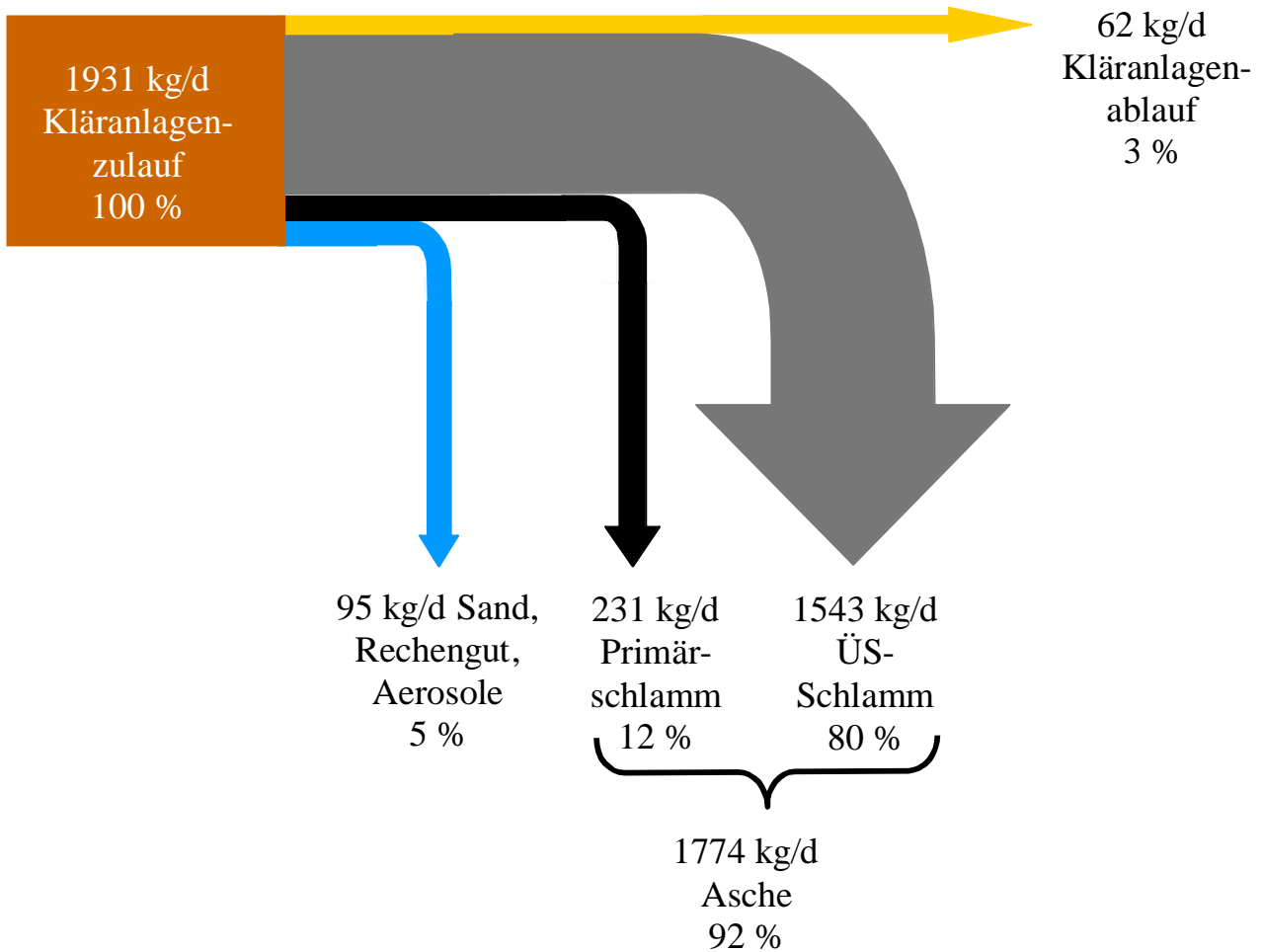
The excess sludge is pumped into primary clarifier, alongside the sewage works influent, and is thus settled along with the primary sludge. Iron-rich precipitation sludge from the phosphate elimination plant Tegel, which treats surface water for the rehabilitation of Tegel Lake, is also pumped into this primary clarifier. The mixed settled sludge from the primary clarifier is transported in a pipe under pressure to the sludge treatment process, which has been operating since 1985 and treats approximately 3000 m³/day with a DSM content of approx. 3.0 - 3.5 %. This uses a two stage process:

- Mechanical thickening in up to six centrifugal thickeners to a DSM content of 24 – 28 %, and
- Incineration in three fluidized bed furnaces with a minimum temperature of 850 °C - ash accumulation about 30 t/day.

2.4 Phosphorus balance - Ruhleben treatment plant plant

The phosphorus balance for the Ruhleben treatment plant plant is represented in figure 2 and gives almost the same proportions of primary sludge and excess sludge as in the phosphorus balance of the Waßmannsdorf treatment plant. Through the iron-holding precipitation sludge of the phosphate elimination plant Tegel, the primary sludge contains a substantial part of ferric oxide hydrates and phosphates. During the incineration, the phosphorus remains in the ashes with a quantity of P-content of only 6 % (with the co-burning of the sewage sludge of the Waßmannsdorf treatment plant, the theoretical P-content amounts to approx. 11 %). The phosphorus may remain, according to [3], as inorganic polyphosphate if Mg and K were present in solution. Further components are SiO₂, CaO as well as ferric oxide, whereas organic pollutants, germs and smell-generating materials have been completely destroyed.

<i>Total Phosphorus (P_{total})</i>		
1931 kg/day	100% of Treatment plant influent P	100 %
62 kg/day	Treatment plant effluent	3 %
95 kg/day	Sand, screening, aerosols	5 %
231 kg/day	Primary sludge	12 %
1543 kg/day	Excess sludge	80 %
1774 kg/day	Ash	92%



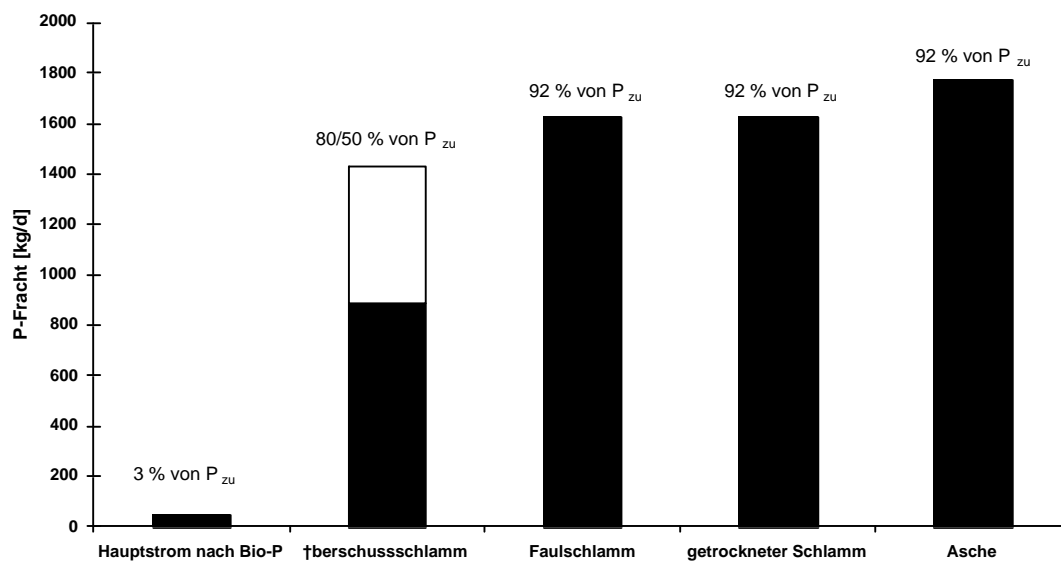
<i>German</i>	<i>English Total Phosphorus (P_{total})</i>
Kläranlagen-zulauf	Treatment plant influent
Kläranlagen-ablauf	Treatment plant effluent
Sand, Rechengut, Aerosole	Sand, screening, aerosols
Primär-schlamm	Primary sludge
ÜS-schlamm	Excess sludge
Faulschlamm	Digested Sludge
FS-Granulat	Dewatered Sludge Granulates

Figure 2: Phosphorus balance of the Ruhleben treatment plant plant

3. Phosphorus recovery potential

From the balance of the phosphorus mass flows we can calculate the P-recovery potential at different places of the wastewater purification and sludge treatment process. The following places were considered (see figure 3):

- Main stream of the wastewater after the Bio-P - processes
- Excess sludge
- Anaerobic sludge
- Dried sludge (anaerobic sludge granulates)
- Ash (Ruhleben treatment plant incinerator).



German	English
P-Fracht (kg/d)	P-load (kg/d)
Hauptstrom nach Bio-P	Main stream after Bio-P
Überschussschlamm	Excess sludge
Faulschlamm	Anaerobic sludge
Getrockneter Schlamm	Dried sludge
Asche	Ash
3% von P zu	3% of P influent
80/50 % von P zu	80/50 % of P influent
92 % von P zu	92 % of P influent
92 % von P zu	92 % of P influent
92 % von P zu	92 % of P influent

Figure 3: Theoretical maximum possible phosphorus recovery potential in different places of the wastewater purification and sludge treatment process

For the determination of the P-recovery potential from the main stream after a biological sewage purification including Bio-P – process, a P total concentration of round 0.3 mg/l was measured, of which the soluble phosphate concentration is < 0.1 to 0.2 mg/l P. The conclusion of this is a maximum P-recovery potential of only approx. 3 %, related to the P- influent load.

The determination of the P-recovery potential of the excess sludge took only in account the relatively readily available portion of phosphorus resulting from enhanced biological phosphorus removal, around 2.5 % of the resulting DSM is phosphorus), that is ignoring the approx 1.5 % of the resulting DSM which is phosphorus contained in cells as nucleic acid, nucleotides and phospholipids. A hydrolysis of the polyphosphates already occurs under anaerobic conditions. The readily available P-recovery potential in the excess sludge thus amounts to approx. 50 % the total sewage works P-influent load. The recovery potential could be increased by a thermal and/or thermally chemical disintegration of the sludge or a total disintegration by acid hydrolysis or by fermentation up to approx. 80 % of the P-influent load, and basic studies are necessary to better evaluate this [4]. For example, trials to disintegrate floating sludge in the Waßmannsdorf treatment plant, showed that substantial phosphorus portions are present in colloidal solution and are difficult to precipitate.

There is a possible access to approximately 92 % of the P-load of the influent, if the anaerobic sludge, anaerobic sludge granulates and ash are taken in account. A direct P-recovery as MAP is here however only possible from the anaerobic sludge. Tried and demonstrated processes for P-recovery from all these sources are not available today. Basic studies are necessary, and there may be a significantly higher cost for the decomposition of ash, than for the recovery from the main stream, from the excess sludge or from the anaerobic sludge.

4. Example of incrustation problems in the Waßmannsdorf treatment plant

Substantial incrustation were found for the first time in Berlin's sewage works in 1994 in centrifugal thickener

s for the de-watering of the anaerobic sludge. These formed as crystalline, white build-ups within the bonnet up to the braking system of the drum. In the Waßmannsdorf treatment plant this led to operational disturbances in 300 mm diameter pipes used for the continuous anaerobic sludge discharge from the sludge digestion tanks to the anaerobic sludge centrifugal thickeners, with incrustations which had obviously developed over several years. Also, downstream of the centrifugation of the anaerobic sludge, crystallisation from the centrate was so significant that the outgoing pipes were fully blocked by incrustations within 2 weeks, obliging mechanical removal of the crystallisations (figure 4). This mechanical clearance had to be repeated every two weeks.



Figure 4: 80 mm diameter bend joint of pump 2 after 319 operating hours and intake aperture after 180 operation hours of the centrate pumping station

As a result of spontaneous release of or degassing of carbon dioxide, oversaturation of the digested sludge and the process water (from the separated centrate of the digested sludge), the pH increases, altering the solubility equilibrium, and thus facilitating crystallisation, and so deposits (incrustations) on the walls. In the Waßmannsdorf treatment plant, the wastewater purification process used, with enhanced biological Phosphorus removal and concentrated streams of digesting sludge, obviously favours the formation of incrustations in the sludge treatment. These incrustations are made of insoluble, crystalline white MAP (struvite = magnesium ammonium phosphate) deposits with small portions of different calcium phosphate compounds.

5. Measures for incrustation avoidance

The following modifications in the process engineering technology were made after extensive investigations, enabling the incrustation problems at the Waßmannsdorf treatment plant to be solved:

- Intensive mixing of the anaerobic sludge by pumping the sludge into the bottom of the digestion tank and re-injecting it at the top.
- In order to avoid a spontaneous and sudden degassing, the anaerobic sludge is pumped through an exhaust pipe at the bottom of the sludge digestion tank and led under pressure to the anaerobic sludge collecting main before the centrifuges.
- Deliberate extraction of the CO₂, e.g. by air injection through a fine-bubble surface aeration system at the bottom of the anaerobic sludge collecting main, just before the centrifuges, causes the pH to rise to approx. 8 and thus to cause controlled struvite precipitation
- Possible addition of magnesium salts as a chemical coagulant improves the dewatering of the anaerobic sludge and therefore the precipitation of the remaining phosphates.
- Dosage of small quantities of anti-incrustation agents to prevent any other incrustations in the pipes and centrifugal thickeners.

The chemical coagulation and precipitation agent MgCl₂ is added into the anaerobic sludge collecting main before the centrifuges (see figure 5). The extraction of CO₂ from the digested anaerobic sludge, by air injection through a fine-bubble surface aeration system at the tank bottom, raises the pH value to approx. 8. This induces a deliberate and controlled precipitation of larger quantities of MAP and

smaller quantities of calcium phosphates, which are then pumped together with the anaerobic sludge into the centrifugal thickeners. Furthermore, it improves the dewaterability of the anaerobic sludge [see also 5] and clearly shows that the centrifugation needs smaller quantities of cationic flocculant addition.

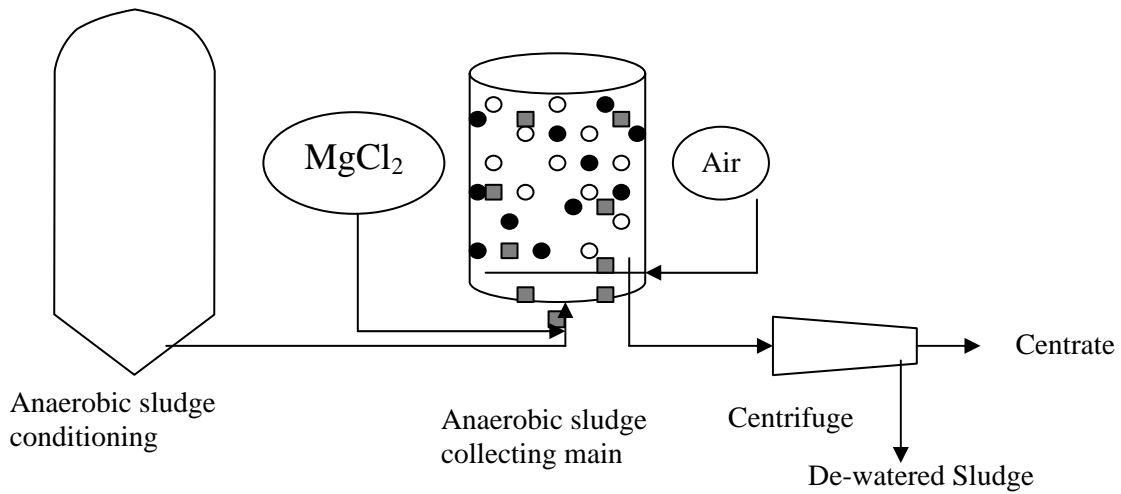


Figure 5: Process scheme for deliberate MAP precipitation during the sludge treatment process

6. Measures for phosphorus recovery

To recovery for re-use of the precipitated MAP, it is necessary to separate the MAP from the anaerobic sludge. As measured in the sieve analysis, the MAP crystal density is of 1.7 kg/dm^3 and the particle size distribution is $95 \% > 0.063\text{mm}$. It was decided to use a centrifugal separation by means of hydro-cyclones to separate the MAP precipitation product from the anaerobic sludge (3 - 4 % DSM). First attempts in a pilot plant were made with different sizes of nozzles in the hydro-cyclones and using three different pressures. The objective was to achieve separation of particles $> 0.05\text{mm}$. It was shown that MAP could be enriched in the base outflow of the hydro-cyclones while phosphorus concentrations were reduced in supernatant outflow of the hydro-cyclones. The P concentration could be nearly doubled with favourable combinations of cyclones and nozzle diameter (left column in figure 6), and the process was effective with significant through flow rates. A subsequent second hydro-cyclone stage allowed a further increase in the P-concentration to approx. 40 kg/m^3 , giving a yield of phosphorus of 20 % (right column in the figure 6). This corresponded to a P-load of approx. 316 kg/day , which could directly be recovered from the anaerobic sludge in the second stage, and amounts to 18 % of the P inlet load in the WWTP Waßmannsdorf. It was shown that this experimental process is in principle suitable for the separation of the MAP precipitation product in the anaerobic sludge. A complete washing process for the raw MAP is not yet installed, but has been tested in practice at pilot level.

German	English
Pges.-Gehalt (kg/m ³), Pges.- Ausbeute (%)	P total content (kg/m ³), P total yield (%)
Pges. Gehalt Zulauf	P total content influent
Pges. Gehalt Unterlauf	P total content base outflow
Pges. Gehalt Ausbeute	P total content recovery
Zulauf	Influent
Überlauf	Supernatant outflow
Ablauf	Base outflow
bester Zyklon	Best Cyclone
Erste Stufe zweistufig	First stage of two
Zweite Stufe	Second stage

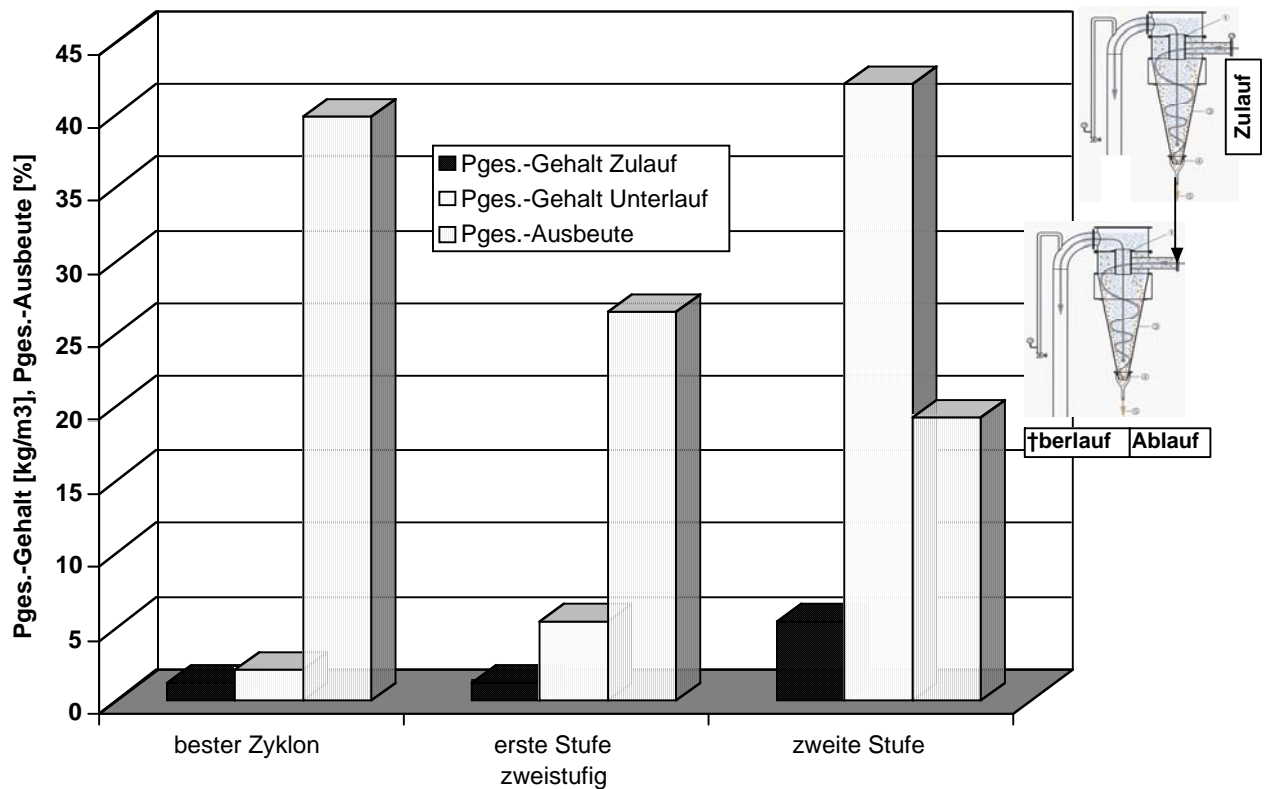


Figure 6: P_{total} content in kg/m³ in influent and base outflow, and P_{total} recovery in % (in the hydro-cyclone base outflow) after a one and two-stage separation with the hydro-cyclone

7. Evaluation of the MAP precipitation products

Figure 7 shows the MAP crystals in the hydro-cyclone base outflow after intensive washing in the laboratory. Both the nutrient contents and impurities were analysed to assess the possible agricultural use of the MAP precipitation product as fertilisers. A comparison with the minimum nutrients contents for different kinds of fertiliser (super-phosphate, dicalium-phosphate with magnesium, NP fertiliser) according to the German fertiliser regulations (DMVO), shows that the precipitated MAP is within the range of other legally certified fertilisers (see figure 8).

In order to assess the pollutant contaminant content of the MAP precipitation product, the heavy metals and organic pollutants (such as PCB and polycyclic aromatic hydrocarbon) were compared with the limit values set by German sewage sludge regulations. The MAP showed contaminant levels 5 times and more below the limit values of the sewage sludge regulation, as shown for some pollutants in the figure 9. To be precise, the pollutants present in the anaerobic sludge are not transferred, or only in very small amounts, into the MAP precipitation product.

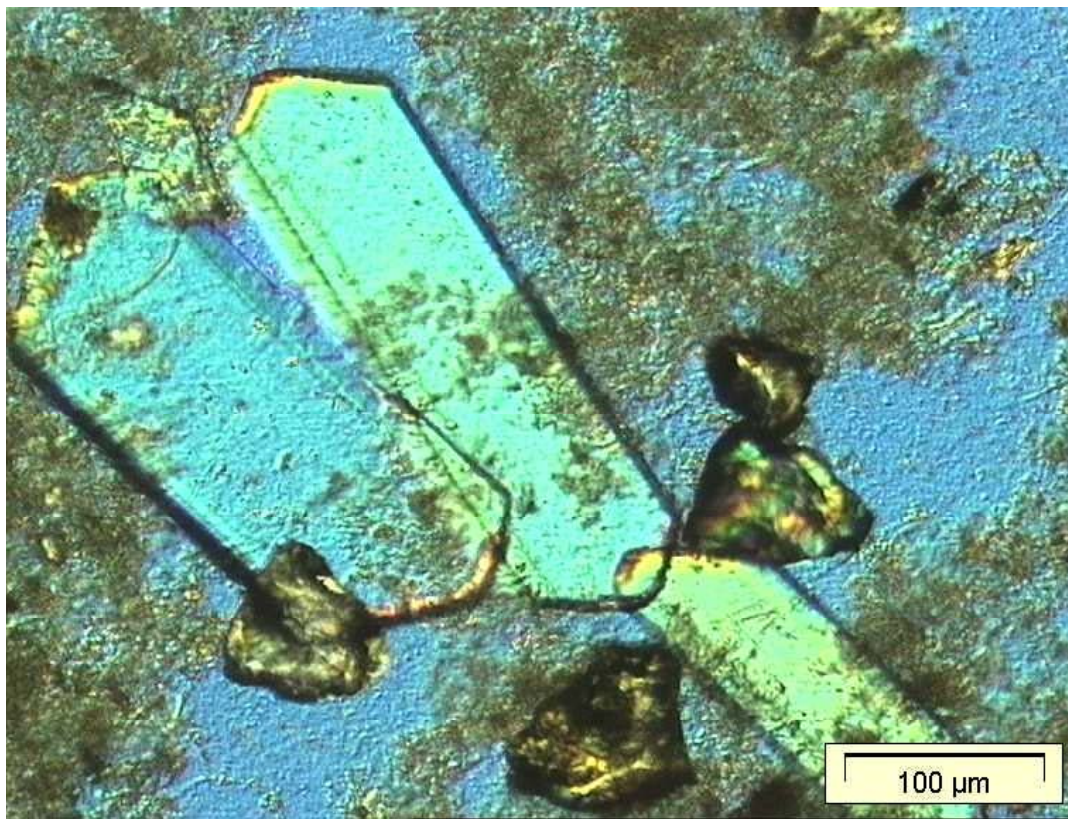
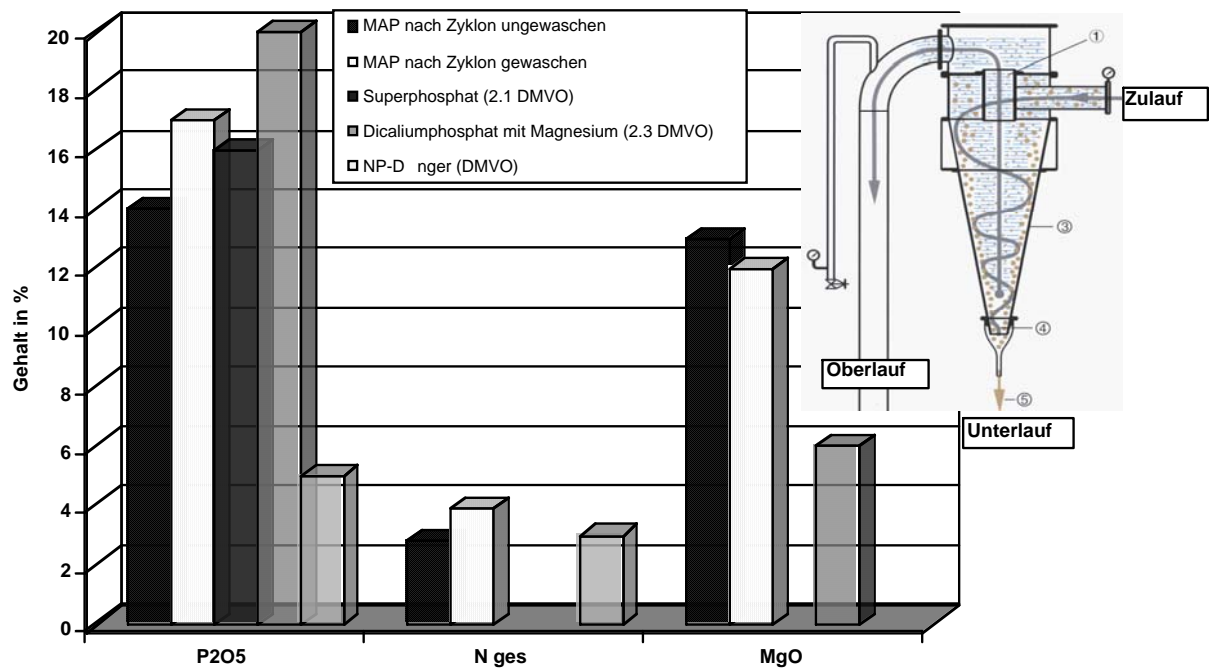
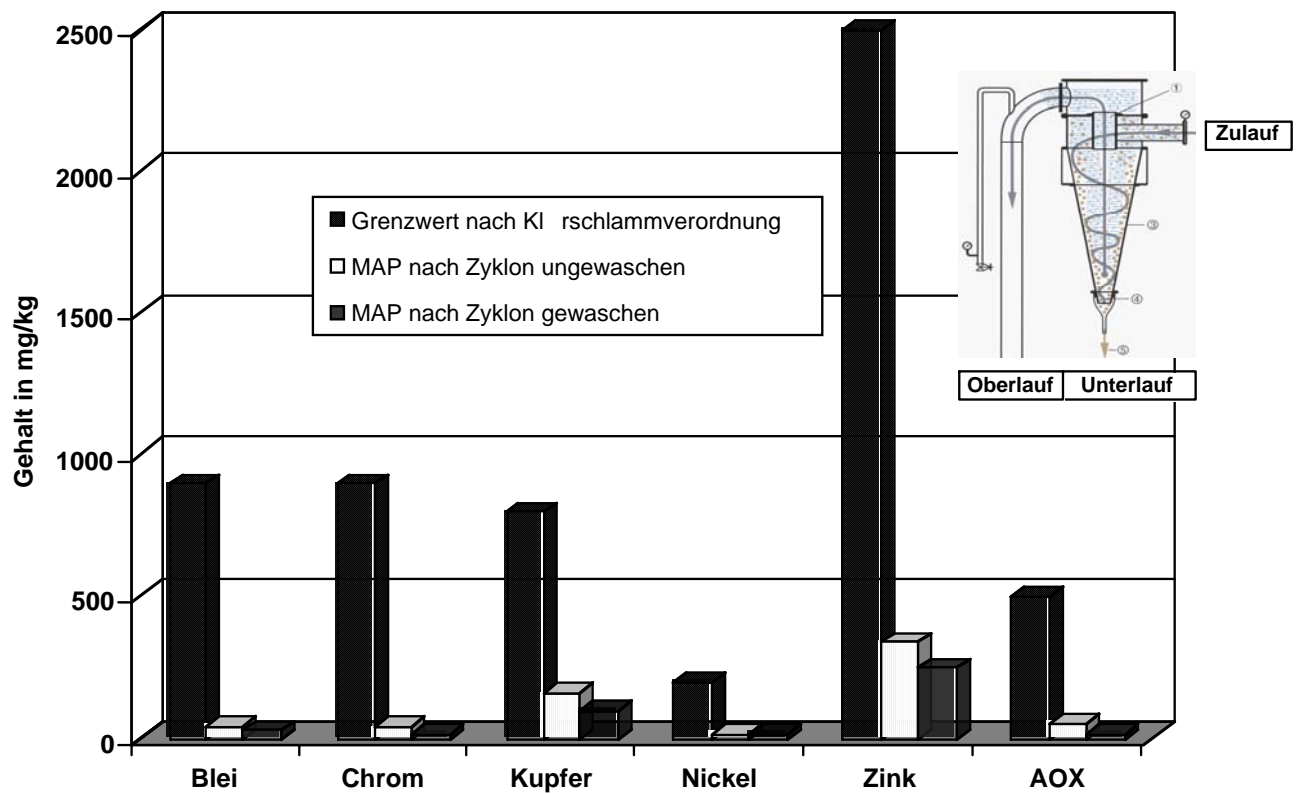


Figure 7: Microscope photo of MAP crystals in the base outflow of the hydro-cyclone



German	English
Gehalt in %	Content in %
P ₂ O ₅	P ₂ O ₅
N ges	N _{total}
MgO	MgO
Zulauf	Influent
Oberlauf	Supernatant outflow
Unterlauf	Base outflow
MAP nach Zyklon ungewaschen	Unwashed MAP after Hydro-cyclone
MAP nach Zyklon gewaschen	Washed MAP after Hydro-cyclone
Superphosphat (2.1 DMVO)	Super-phosphate (2.1 DMVO)
Dicalciumphosphat mit Magnesium (2.3 DMVO)	Dicalium-phosphate with magnesium (2.3 DMVO)
NP-Dünger (DMVO)	NP fertiliser (DMVO)

Figure 8: Comparison of nutrients in the washed and unwashed MAP precipitation product, after the separation by hydro-cyclone, with minimum contents of different fertilisers according to German fertiliser regulations.



German	English
Gehalt in mg/kg	Content in mg/kg
Blei	Lead
Chrom	Chromium
Kupfer	Copper
Nickel	Nickel
Zink	Zinc
AOX	Adsorbable organic halogens
Zulauf	Influent
Oberlauf	Supernatant outflow
Unterlauf	Base outflow
Grenzwert nach Klärschlammverordnung	Limit value of the sewage sludge regulation
MAP nach Zyklon ungewaschen	Unwashed MAP after Hydro-cyclone
MAP nach Zyklon gewaschen	Washed MAP after Hydro-cyclone

Figure 9: Comparison of the pollutants contained in the washed and unwashed MAP precipitation product after the separation by hydro-cyclone, with limit values of the German sewage sludge regulation.

To be suitable as fertilisers the solubility and the plant availability of the precipitated MAP are crucial. Several investigations on the solubility of phosphorus in a sequential extraction process were accomplished according to Psenner method [6]. The MAP precipitation product showed here a very good to a good solubility in different solvents (from ammonium chloride to hydrochloric acid) (see Table 2).

Table 2 : Investigation of phosphorus availability in a sequential extraction experiment (Psenner)

Substance	P-content	Phosphorus-Solubility (% of product P content)			
		(%)	Very good	good	medium
CaHPO ₄ x 2 H ₂ O	19.6	51.8	23.0	6.1	19.0
MAP – product	21.8	8.1	50.6	40.2	1.0
Fe ₃ (PO ₄) ₂	14.2	0	22.9	76.7	0.2
AlPO ₄	21.9	0.1	0.2	99.6	0
Ca ₃ (PO ₄) ₂	17.7	4.1	5.2	0.9	89.7

This shows the following phosphorus availability for plants :

CaHPO₄ · 2H₂O > MAP precipitation product > Fe₃(PO₄)₂ > Ca₃(PO₄)₂ > AlPO₄

This implies that the precipitation product can be used as a long term fertilizer.

Evaluation of the phosphorus availability of MAP precipitation solids taken from the Waßmannsdorf WWTP in two test soils having a pH value of 6.6 and 7.1 was tested using standard ryegrass, and showed a good phosphorus availability from this material, leading to a good grass growth, in comparison with application of chemical MAP and with monocalcium phosphates [7].

The following procedure would be necessary for the declaration of recovered MAP as a fertiliser in Germany:

- Declaration of the product made by the Berliner Wasserbetriebe with indication of the type and the effectiveness determined by the appropriate analyses
- The National Office for Consumer Protection and Agriculture (LVL) Brandenburg emits a request to the Official Fertiliser Traffic Control
- Examination by the LVL
- Request to the Federal Ministry for Consumer Protection and Agriculture for permission to use as fertiliser
- The Scientific Adviser of the Ministry decides on the permission as fertiliser.

Fertiliser dealers have already stated an interest in marketing the MAP precipitation product. MAP could be an important fertiliser, especially for the soils of Brandenburg region, and also for example for forest areas in Austria [8], which are poor in magnesium.

8. Summary and future plans

The process used in the Waßmannsdorf treatment plant operated by Berliner Wasserbetriebe, combining Bio-P and the digestion of concentrated sludge flows, obviously favours the development of incrustations in the sludge treatment, which are mainly made of magnesium ammonium phosphate (struvite = MAP). Deliberate MAP precipitation not only allows avoidance of incrustations in the sludge treatment, but also to fix substantial phosphorus quantities in the MAP precipitation product. On the one hand, this leads to a substantial reduction of the remaining load in the wastewater purification process in treatment plants and secures the respect of discharge values, because of the reduced P-load in centrates and in process-water returned to the head of the wastewater purification process. On the other hand, the MAP precipitation product can be reused. For this test, hydro-cyclones were used to obtain a direct separation of the MAP precipitation contained in the anaerobic sludge. It was shown that the MAP can particularly be enriched in the base outflow of the hydro-cyclones by a subsequent second hydro-cyclone stage.

The Waßmannsdorf treatment plant is currently testing a two-stage sludge digestion process, with a load of approx. 6 - 12 kg DSM/m³day in the first stage and approx. 5 kg DSM/m³day in the second stage [see also 9]. This intensified sludge digestion increases the mineralization process and simultaneously releases more phosphorus (up to 275 kg/m³ soluble P). An intensive mixing of the anaerobic sludge is here important to avoid the forming of uncontrollable foam. In the second stage, we are considering a dosing of MgCl₂ to induce MAP precipitation. The MAP separation is then achieved using a hydro-cyclone located on the circulation pipeline, downstream of the second sludge digestion tank. With a sludge digestion duration of 5 days during the second stage, the anaerobic sludge will then go approximately 12 times through the hydro-cyclone, increasing the solids separation and the P-recovery.

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