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Phosphorous removal and recovery from urban wastewater: current practices and new directions

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1 **Abstract**

2 Phosphate rocks are an irreplaceable resource to produce fertilizers, but their availability will not be
3 enough to meet the increasing demands of agriculture for food production. At the same time, the
4 accumulation of phosphorous discharged by municipal wastewater treatment plants (WWTPs) is
5 one of the main causes of eutrophication. In a perspective of circular economy, WWTPs play a key
6 role in phosphorous management. Indeed, phosphorus removal and recovery from WWTPs can both
7 reduce the occurrence of eutrophication and contribute to meeting the demand for phosphorus-based
8 fertilizers. Phosphorous removal and recovery are interconnected phases in WWTP with the former
9 generally involved in the mainstream treatment, while the latter on the side streams. Indeed, by
10 reducing phosphorus concentration in the WWTP side streams, a further improvement of the overall
11 phosphorus removal from the WWTP influent can be obtained. Many studies and patents have been
12 recently focused on treatments and processes aimed at the removal and recovery of phosphorous
13 from wastewater and sewage sludge. Notably, new advances on biological and material sciences are
14 constantly put at the service of conventional or unconventional wastewater treatments to increase
15 the phosphorous removal efficiency and/or reduce the treatment costs. Similarly, many studies have
16 been devoted to the development of processes aimed at the recovery of phosphorus from
17 wastewaters and sludge to produce fertilizers, and a wide range of recovery percentages is reported
18 as a function of the different technologies applied (from 10-25% up to 70-90% of the phosphorous
19 in the WWTP influent). In view of forthcoming and inevitable regulations on phosphorous removal
20 and recovery from WWTP streams, this review summarizes the main recent advances in this field to
21 provide the scientific and technical community with an updated and useful tool for choosing the
22 best strategy to adopt during the design or upgrading of WWTPs.

23 **Keywords:** phosphorus removal; phosphorus recovery; sewage sludge; urban wastewater; WWTP.

24

25 **List of abbreviations**

AD	Anaerobic digestion
AGS	Aerobic granular sludge
A ² O	Anaerobic/anoxic/oxic
AOB	Ammonium oxidizing bacteria
AP	Acidification potential
BOD	Biological oxygen demand
CAS	Conventional activated sludge
CED	Cumulative energy demand
CFR	Continuous-flow reactor
COD	Chemical oxygen demand
CSTR	Completely stirred tank reactors
DO	Dissolved oxygen
DPAO	Denitrifying PAO
EBPR	Enhanced biological phosphorous removal
EU	European Union
FBR	Fluidized-bed reactor
GWP	Global warming potential
HM	Heavy metal
HRAP	High-rate algal pond
HRT	Hydraulic retention time
IA	Intermittent aeration
IFAS	Integrated fixed activated sludge
MBBR	Moving bed biofilm reactor
MBR	Membrane bioreactor
MLE	Modified Ludzak Etlinger
MLSS	Mixed liquor suspended solids
MUCT	Modified University of Cape Town
NF	Nanofiltration
NOB	Nitrite oxidizing bacteria
PAO	Phosphorous accumulating organisms
PHA	Polyhydroxyalkanoates

Poly-P	Polyphosphates
PR	Phosphate rocks
RAS	Recirculated activated sludge
S2EBPR	Side-stream EBPR
SBR	Sequencing batch reactor
SCWO	Supercritical water oxidation
SDG	Sustainable Development Goal
SND	Simultaneous nitrification denitrification
SRT	Sludge retention time
SSM	Sidestream MLSS fermentation
SSR	Side-stream reactor
TN	Total nitrogen
TP	Total phosphorus
TRL	Technology readiness level
TSS	Total suspended solids
UASB	Upflow anaerobic sludge blanket
UCT	University of Cape Town
UMIF	Unmixed in-line MLSS fermentation
VFA	Volatile fatty acids
VSS	Volatile suspended solids
WWTP	Wastewater treatment plant

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1. Introduction

Phosphorous (P) plays a key role in the global food supply because, together with nitrogen (N) and potassium (K), is a biolimiting nutrient for plants and, for this reason, is largely used to produce chemical fertilizers. P is a relatively limited resource (Wellmer and Scholz, 2015) and almost 85% of P used in agriculture derives from the extraction and processing of PR (Geissler et al., 2015). To meet the agricultural demand, from 1993 the global consumption of elemental P deriving from PR raised by almost 3-4% per year (Chen and Graedel, 2016). Global PR reserves amount to almost 65×10^3 Tg, and they could reach 290×10^3 Tg hypothetically considering deposits and unprocessed rock of varying quality (Van Kauwenbergh, 2010). In fact, according to the last summary of the U.S. Geological Survey, the worldwide PR reservoirs amount to 71×10^3 Tg, while the excavated PR amount equaled 223 Tg in 2020 (U.S. Geological Survey, 2021). Furthermore, to reach the target 2.3 of the Sustainable Development Goal of United Nations (doubling the productivity of smallholder farms) by 2030, the need for an increase of P application by 39% (126 Tg) has been estimated (Langhans et al., 2021). According to this, the constant increase of P demand provides a growing concern over long-term security of P supply (Desmidt et al., 2015). Indeed, it has been estimated that global PR reserves will last 3-4 centuries, but excluding Morocco, which holds almost 75% of global PR reserves (Jasinski, 2010), all the other countries will deplete their share within the next century (Cooper et al., 2011).

Moreover, the price of PR used to produce fertilizers influences the cost of food, as in the case of cereals, and consequently the world hunger. Consequently, many countries are dealing with this topic to overcome the problems generated by the sudden increase in the cost of PR, as happened in 2008 (Neset and Cordell, 2012). Specifically, the 5Rs strategy (Realign P inputs in agriculture, Reduce P losses in the hydrosphere, Recycle P in bio-resources, Recover P from waste, and Redefine the food system) has been suggested to EU countries in the past few years (Withers et al., 2015). As a result, the EU has welcomed these suggestions and is trying to put them into practice.

71 For instance, the European Parliament has recently proposed the promotion of “large scale fertilizer
72 production in the EU from domestic organic or secondary raw materials” (European Parliament,
73 2016). Ott and Rechberger (2012) highlighted that almost 20% of the P used for food, feed and non-
74 food products, such as detergents, by the EU15 former countries ends up in industrial and urban
75 wastewater treatment plants (WWTPs). As reported by the authors, 70% of P comes out from
76 WWTP in the form of sludge, not always reused in agriculture, while the remaining 30% ends up
77 directly in the hydrosphere. Additionally, part of the P contained in fertilizer and WWTP sludge
78 reused in agriculture is washed away by rain, with a consequent migration of P to surface waters
79 (Ball Coelho et al., 2012). Another study on the EU27 countries highlighted that 27% of P imported
80 during 2005 (655 Gg) was lost after the consumption of commercial goods (i.e., food and
81 detergents) mainly in wastewater (55%), food waste (27%), and pet excreta (11%) (van Dijk et al.,
82 2016).

83 As a result, in 2013 the yearly amount of P from anthropogenic pollution that reaches the sea has
84 been evaluated to be equal to 37.9 Tg, which represents more than the 50% of the global annual P
85 consumption, i.e., 68.7 Tg (Chen and Graedel, 2016). In particular, the accumulation of P, derived
86 from WWTPs and runoff, in small lakes, rivers or backwaters is the main cause of eutrophication
87 (Huser et al., 2016), which generates serious problems to the environment and human activities.
88 Indeed, eutrophication causes turbidity, hypoxia and malodor in surface waters, leading to a serious
89 burden on the aquatic ecosystem (Conley et al., 2009). Moreover, blue-green algae (cyanobacteria)
90 may excrete toxins that are dangerous to humans and animals (Dittmann and Wiegand, 2006).

91 In this context, WWTPs play a key role in the reduction of eutrophication phenomena, since many
92 treatments, some being still under investigation, could be adopted to reduce the P concentration in
93 WWTPs effluents. For instance, under the guidelines of the EU Urban Wastewater Directive, the
94 achievement of lower P concentrations in UK wastewater reduced the average annual total P
95 concentration in rivers from 0.27 to 0.1 mg P/L during the 1974-2012 period (Worrall et al., 2016).

96 Despite this, according to van Puijenbroek et al. (2019), numerical calculation on a global scale
97 showed a discharge in wastewater equal to 1.5 Tg of P in 2010 and estimated values up to 1.6-2.4
98 Tg of P expectable by 2050. Indeed, this clearly highlights the necessity to achieve very high P
99 removal efficiency within WWTPs. However, the increasing efficiency of P removal from
100 wastewater concurrently leads to the nutrient accumulation in sewage sludge. Then, the scientific
101 community is striving to enhance P recovery from wastewater and sewage sludge. By performing
102 both P removal (on the mainstream line) and recovery (on the side streams) within the WWTP is
103 possible to implement a strategy consistent with the principles of circular economy (Geissdoerfer et
104 al., 2017; Sauvé et al., 2016). This also meets the measures foreseen by the EU and other countries
105 to counteract the negative effects of a predictable P scarcity and excessive release in the
106 hydrosphere.

107 In line with this, many studies and patents have been devoted to the development of treatments and
108 processes to remove and/or recover P from urban and industrial wastewater (Desmidt et al., 2015),
109 leading to significant advances on this topic in recent years. Some technologies have already been
110 implemented in WWTPs and are currently being optimized, while many have been tested only in
111 laboratory reactors or pilot-scale installations. The present review provides a critical perspective of
112 the existing P removal and recovery systems from WWTP streams with a focus of full-scale
113 technologies and indicates the new research directions based on recent studies on this topic.

114 **2. Phosphorous removal from municipal wastewater**

115 The average TP concentrations in municipal wastewater typically ranges between 6 and 12 mg/L
116 (Rittmann and McCarty, 2012; US EPA, 2007). The typical phosphorous forms present in urban
117 wastewaters are orthophosphates (PO_4^{3-}) and polyphosphates (Poly-P) which turn to PO_4^{3-} after
118 slow hydrolysis (Park et al., 2016). Secondary treatment involving only aerobic biological treatment
119 displays a TP removal efficiency ranging from 20 to 30%, resulting in effluent concentrations from
120 3-4 to 8-10 mg P/L, being higher than typical TP standard in protected watershed, i.e., 1 mg/L

121 (Daneshgar et al., 2018). Therefore, supplemental TP removal should be provided in WWTPs to
122 avoid eutrophication of the surface water (Lüring and Oosterhout, 2013).

123 Generally, phosphorus removal from WWTP influent is carried out during mainstream wastewater
124 treatment, while recovery is performed via side-stream processes, e.g., in the sludge streamline. In
125 mainstream WWTP treatment, phosphorus can be removed upstream, within and downstream the
126 biological phase. The first and third approaches are commonly carried out via chemical methods,
127 while the second one via biological treatment, although addition of chemical for P precipitation
128 within the biological treatment is also reported in literature (Carolina et al., 2011; Ma et al., 2020).

129 Calcium, magnesium, aluminum and iron ions are commonly employed as precipitators in the
130 primary wastewater treatment (pre-precipitation), in the post-precipitation treatment (after
131 secondary biological treatment) or in the co-precipitation treatment (contemporary to the biological
132 treatment) (Magrí et al., 2020). In the first system, 65-80% removal efficiency can be achieved with
133 a relatively low P concentration in the final effluent (~1 mg/L) and wastewater alkalinity can
134 influence reactant dosage (Metcalf & Eddy Inc. et al., 2014). In the post-precipitation treatment, it is
135 possible to achieve lower P concentration (from <0.1 to 0.5 mg P/L for sludge separation by
136 filtration and sedimentation, respectively) in the final effluent (Kroiss et al., 2011). These final
137 effluent concentrations can be obtained due to a more efficient chemical action ascribed to the
138 conversion of the organic phosphates to orthophosphates during the biological step. The post-
139 precipitation system is more expensive than pre-precipitation as additional treatment basins are
140 required. In the co-precipitation treatment, which is suitable for activated sludge reactors, chemical
141 consumption is reduced because of the combined effect operated by the sludge recirculation and the
142 sorption and precipitation processes, and no additional basins are needed. For these reasons, this
143 system is less expensive than the post-precipitation treatment also because tanks of moderate size
144 are required, but chemical and biological sludges are not differentiated. In the present case,
145 aluminum and iron ions are commonly employed as precipitators and phosphorous removal

146 efficiency can reach values up to 93-99% (Costa et al., 2019; Filali-Meknassi et al., 2005).
147 Although chemical precipitation remains an established and highly efficient solution for phosphorus
148 removal and is applied in many WWTPs, current research is producing significant effort in
149 enhancing biological phosphorus removal with the aim to limit the consumption of chemicals,
150 reducing treatment costs and guaranteeing a higher process sustainability. In the following sections,
151 both well-assessed and innovative biological treatments are critically described.

152 **2.1 Biological P removal in mainstream wastewater treatment**

153 Biological P removal is based on the accumulation of phosphorus in microbial cells forming
154 secondary sludge, which is subsequently removed from the water. In a CAS system, phosphorus is
155 taken up by microorganisms for their metabolic reactions. However, the amount of TP removed in
156 CAS is only 7-18 g per kg of removed organic matter expressed as COD, resulting in a 20-30%
157 reduction of the influent TP concentration (Henze et al., 1997). To increase the efficiency of
158 phosphorus removal, several biological processes characterized by the presence of suspended or
159 attached biomass (chemo- or photo-synthetic) have been developed. The main operating
160 characteristics and TP removal efficiency of several full-scale WWTPs adopting these technologies
161 are reported in **Tables 1-3**.

162

163 **Table 1. Full-scale applications of suspended biomass biological systems for phosphorus removal.**

164 **2.1.1 Biological P removal by suspended biomass processes**

165 In the second half of the seventies, intermittent anaerobic/aerobic conditions and the development
166 of enriched cultures of PAO were proposed to obtain P removal up to 90% (Barnard, 1974). This
167 process, known as EBPR, is mainly facilitated by some specialized microorganisms such as
168 *Tetrasphaera*- and *Accumulibacter*-related bacteria, which respectively constitute the 27-35% and
169 3-24% of the total biomass volume in EBPR systems (Cyzdik-Kwiatkowska and Zielińska, 2016;
170 Stockholm-Bjerregaard et al., 2017). The behavior of these microorganisms is a controversial topic,
171 and it is still being studied. However, the bio-chemical reactions that seem to be recognized by the
172 whole scientific community are reported below and depicted in **Figure 1**. Under anaerobic
173 conditions, *Tetrasphaera*-related microorganisms can assimilate some rapidly biodegradable
174 substrates, such as glucose, glutamate, aspartate and glycine, to ferment them and synthesize
175 glycogen and new cells (Marques et al., 2017). The required energy is obtained from both
176 fermentation of rapidly biodegradable substrates and degradation of Poly-P, with a consequent
177 release of phosphates and low molecular weight organic compounds such as lactate, acetate, and
178 succinate (Nguyen et al., 2011). Meanwhile, the low molecular weight organic compounds, also
179 produced from the metabolism of *Tetrasphaera*-related microorganisms, are taken up and stored as
180 PHA in the cells of *Accumulibacter*-type bacteria thanks to the energy derived from both the
181 hydrolysis of Poly-P and the glycolysis of intracellular glycogen (He and McMahon, 2011). Like
182 *Tetrasphaera*-related microorganisms, also bacteria associated to *Accumulibacter* release
183 phosphates during the anaerobic step due to the hydrolysis of Poly-P. During the subsequent aerobic
184 and/or anoxic phase, due to the scarcity of exogenous energy and carbon sources being already
185 depleted during the anaerobic phase, the stored glycogen and PHA are oxidized aerobically and/or
186 anoxically to provide carbon and energy required for the production of new cells as well as for
187 phosphate assimilation and the consequent restoration of the Poly-P reserve (Kristiansen et al.,
188 2013). Phosphate uptake under aerobic conditions exceeds the amount released during the anaerobic

189 phase, resulting in a net P uptake also defined as *luxury* uptake. Bacteria capable of anoxic
190 oxidation of stored carbon compounds are known as DPAO and include both *Tetrasphaera*- and
191 *Accumulibacter*-related microorganisms, although *Tetrasphaera* has shown lower anoxic P uptake
192 compared to *Accumulibacter* (Marques et al., 2018). *Tetrasphaera* and *Accumulibacter* contributed
193 for the 24-70% of the TP removed, corresponding to 14.84-37.52 mg P/g TSS, in eight different
194 WWTPs performing EBPR, the rest being attributed to the activity of unknown PAO and chemical
195 precipitation (Fernando et al., 2019). P content in the biomass of EBPR systems can reach up to 100
196 mg P/g TSS (150 mg P/g VSS), which is 6.7 times higher than that typically found in CAS sludge,
197 i.e., around 15 mg P/g TSS (20 mg P/ g VSS) (Henze et al., 2008).

198

199 **Figure 1. A brief description of *Tetrasphaera*- and *Accumulibacter*-related microorganisms in an EBPR system.**

200

201 EBPR in WWTPs is commonly achieved in concomitance with N removal. To this purpose, several
202 configurations can be applied (**Figure 2**).

203

204 **Figure 2. WWTP cycles for combined biological P and N removal from wastewater.**

205

206 Among these, the A²O configuration is the easiest, as it simply adds an anaerobic basin, which
207 receives the recycle sludge from the secondary settler, to the classical pre-denitrification system. In
208 literature, some successful A²O applications are reported, although they indicate variable TP
209 removal efficiencies. For instance, TP removal of almost 96% was reported in a full-scale A²O
210 system characterized by an average SRT of 12.7 d and a biomass concentration of 3-4 g/L (Wang et
211 al., 2014). On the contrary, a significantly lower TP removal (43%) was reported for a A²O system
212 operated at a similar SRT (12 d) but lower biomass concentration in the tanks, according to the TSS

213 concentration in the sludge (2.9 g/L) (Lanham et al., 2013). For this application, influent
214 characteristics were almost equal to 253 mg/L for the average COD, 81 mg/L of suspended solids,
215 43 mg-N/L (average N), 33 mg-N/L (average NH_3), 5 mg-P/L (average TP) with a resulting
216 COD:N:P ratio of 51:9:1. A drawback of the A²O configuration is that the NO_3^- contained in the
217 recycle sludge can be used by denitrifying bacteria to oxidize part of the COD destined to the
218 synthesis of PHA (*Accumulibacter*) and glycogen (*Tetrasphaera*) and limit the subsequent aerobic
219 P uptake. Therefore, an alternative configuration avoiding NO_3^- contamination of the anaerobic
220 process should be preferred. At this aim, the 5-stage Bardenpho scheme involves the addition of
221 two further stages (anoxic and aerobic, respectively) downstream the aerobic stage. Specifically, the
222 new anoxic stage is aimed at further lowering NO_3^- concentration from the previous aerobic tank
223 using endogenous carbon. Then, the final aerobic stage is useful for the minimization of phosphates
224 release in the secondary clarifiers. Effluent PO_4^{3-} -P concentrations <0.5 mg/L and P removal
225 efficiencies >95% have been reported in a full-scale WWTP configured with a 5-stage Bardenpho
226 process and treating wastewater containing 240–360 mg BOD_5 /L and 10 mg PO_4^{3-} -P /L (Meyer et
227 al., 2006).

228 The scheme proposed by the University of Cape Town, namely UCT process, is another strategy to
229 overcome high NO_3^- concentrations in the anaerobic tank by recycling sludge to the anoxic basin
230 and mixed liquor from the anoxic to the anaerobic basin (Rittmann and McCarty, 2012). In this
231 way, NO_3^- is mostly reduced under anoxic conditions before entering the anaerobic basin. The
232 MUCT scheme further optimizes this configuration by dividing the anoxic basin in two
233 compartments: the first, which recycles liquor to the anaerobic basin, receives only recycle sludge
234 (which has limited NO_3^- levels), further limiting NO_3^- concentration in the stream feeding the
235 anaerobic bacteria, while the other receives the nitrified liquor from the aerobic phase. As a result, a
236 study by Beer et al. (2006) on real WWTPs (with influent P concentrations ranging from 7.5 to 12
237 mg/L and operating temperatures from 16°C to 25°C) in Australia reported a TP removal efficiency

238 after MUCT treatment in the range of 52-98%. An alternative to MUCT is the Johannesburg
239 process, in which RAS is denitrified in an anoxic tank before being recirculated to the anaerobic
240 basin.

241 Besides the schemes illustrated in **Figure 2**, alternative configurations have been designed with the
242 scope to improve the stability of the EBPR process. Among these, the S2EBPR scheme implies that
243 a portion of RAS or anaerobic mixed liquor is maintained in a SSR for a period of 16-48 h, enabling
244 VFA production via sludge fermentation and consequent PO_4^{3-} release and carbon uptake by PAO.
245 This configuration can be further modified by feeding the SSR with supplemental carbon from the
246 primary sludge fermenter. Other S2EBPR configurations include the SSM, in which part of the
247 anaerobic MLSS is diverted to a SSR, and the UMIF, in which an unmixed zone is introduced in the
248 cycle to favour MLSS fermentation (Onnis-Hayden et al., 2020). An anoxic SSR was also
249 implemented in the BIODENITRO™ process, which consists of two tanks where anoxic and aerobic
250 conditions are alternated. This has the aim of partially degrade the RAS (HRT = 20-30 h) and
251 recirculate the previously fermented stream into the BIODENITRO™ tank operating under anoxic
252 conditions (Lanham et al., 2013; Vollertsen et al., 2006). In the two examples reported by Lanham
253 et al. (2013), high TP removal efficiencies, i.e., 94 % and 96 %, were observed, despite the high
254 differences between of operating conditions, i.e., influent TP concentration equal to 4 and 10 mg/L,
255 influent COD concentration of 198 and 636 mg/L, average flow rate of $49 \cdot 10^3$ and $17 \cdot 10^3$ m³/d, and
256 SRT maintained at 19 and 30 d, respectively. However, since the authors reported that FeCl_3 was
257 added as a polishing step for P removal, it is not possible to determine the contribution of the sole
258 BIODENITRO™ configuration, combined with an anaerobic SSR, in the achievement of the
259 overmentioned TP removal efficiencies. Another interesting side-stream configuration was tested
260 by Wang et al. (2019), who modified a A²O system moving the anaerobic compartment (continuous
261 stirring and HRT = 1.6 h) from the mainstream to the side-stream line with consequent influent inlet
262 into the anoxic compartment. At the same time, RAS was conveyed into the side-stream anaerobic

263 reactor together with primary sludge to provide a supplemental carbon source. With TP and COD
264 concentrations in the influent equal to 3.7 mg/L and 254 mg/L, respectively, a TP removal
265 efficiency equal to 84% was obtained. However, the TP removal efficiency was further enhanced
266 (93%) by adopting an intermittent mixing of the anaerobic compartment. In fact, the intermittent
267 mixing condition led to the sedimentation of a part of the sludge, thus increasing the SRT in the
268 anaerobic compartment and enhancing the hydrolysis/fermentation of RAS.

269 Recent research is moving towards the implementation of more compact systems allowing more
270 efficient and sustainable removal of organic carbon, N and P in one single reactor through the
271 alternation of different oxygen conditions. In this direction, Roots et al. (2020) combined P and
272 shortcut N removal from real municipal wastewater in a pilot-scale (56 L) SBR with a cycle set on
273 an anaerobic period followed by IA and without the addition of exogenous carbon. Aeration was
274 set to target a DO level of 1 mg/L (duration 4-5 minutes), followed by pump shutdown and up to 3-
275 min long anoxic period. Influent characteristics were approximately 2.8 mg N/L (TKN), 1.7 mg N/L
276 (NH_4^+), 1.4 mg P/L (PO_4^{3-}), 2.2 mg P/L (TP), 150 mg total COD/L, 94 mg filtered COD/L, 72 mg
277 TSS/L, and 4.7 meq/L of alkalinity. Average TP, total nitrogen (TN) and COD removal at 20°C
278 were 83%, 81% and 70%, which resulted in effluent TP concentrations ≤ 0.3 mg/L (on average).
279 *Candidatus Accumulibacter* was the most abundant PAO representative in the reactor with relative
280 abundances up to 12%, while *Tetrasphaera* remained always below 0.3%.

281 **2.1.2 Biological P removal by microbial biofilm-based/granular sludge processes**

282 In the last years, research has also focused on technologies able to simplify plant operation and
283 strongly limit the number of basins and recycles required for EBPR while reducing investment and
284 operational costs. In particular, the use of the SBR has gained increasing interest as SBR allows a
285 full control of operational conditions and, by alternating aeration and non-aeration phases, the
286 complete removal of P, N and C in a single basin. To allow the simultaneous growth of AOB, NOB,

287 PAO and DPAO, biofilm processes are advantageous compared to suspended-growth systems, as
288 mass transport limitations within the biofilm create ecological niches allowing the coexistence of
289 different types of bacteria.

290 AGS has been successfully applied in recent years for P removal from both municipal and industrial
291 wastewaters and typically develops in a SBR system. AGS granules have a core which stays
292 anaerobic under the non-aeration phase (feast) and turns partially to aerobic/anoxic during the
293 aeration phase (famine) due to oxygen penetration and nitrifying activity. Indeed, nitrifiers (AOB
294 and NOB) develop on the external layer of the granules where oxygen is more available, resulting
295 in the production of NO_x^- diffusing to the internal layers of the granule (**Figure 3**).

296

297 **Figure 3. Main phosphorus, nitrogen and carbon transformation and layer distribution in an AGS granule during aeration**
298 **and non-aeration phases.**

299

300 Recent applications of the AGS technology have focused on the combined removal of phosphorus,
301 nitrogen and carbon from domestic wastewater. Campo et al. (2020) investigated the C/N/P removal
302 performance of AGS treating real domestic wastewater with low C/N as well as the impact of high
303 SRT on the process in a lab scale reactor. High $\text{PO}_4^{3-}\text{-P}$ removal (96%) combined to C and N
304 removal of 84% and 71% in respective order was obtained when treating wastewater with C/N of
305 3.8, resulting in effluent $\text{PO}_4^{3-}\text{-P}$ concentrations <0.1 mg/L (on average). Moreover, the long SRT of
306 61 d led to low sludge yield ($0.21 \text{ gCOD}_{\text{biomass}}/\text{gCOD}_{\text{removed}}$). Similar P removal (95%) and higher N
307 removal efficiency (87%) were observed in a full-scale AGS reactor in Poland (Świątczak and
308 Cydzik-Kwiatkowska, 2018) operated with a SRT of 30 d (Table 2). Nonetheless, a recent study on
309 a full-scale AGS application displayed a lower P removal efficiency (69%) (Oliveira et al., 2020) .
310 It is worth highlighting that nearly all full-scale AGS systems reported in **Table 2** were operated
311 with mixed wastewater (domestic and industrial), resulting in influent P concentrations often above

312 10 mg P/L. In particular, the industrial wastewater fractions (generally around 30-40% of the
313 influent) originated from different sources depending on the reported study (such as dairy industries
314 and slaughterhouses) (Giesen et al. 2013, Świąteczak and Cydzik-Kwiatkowska 2018, Ortega et al.
315 2021). Despite a significant percentage of industrial wastewater in the plant influents, results from
316 these studies displayed very high P removal efficiency (82-98%), highlighting the good
317 performances achievable through the AGS technology. Further supporting the high AGS efficiency,
318 a case of study from Giesen et al. (2016) reported a P removal efficiency equal to almost 95% for a
319 plant treating a mix of domestic, septic tank and industrial wastewaters and characterized by
320 prolonged periods of industrial peak load.

321 In terms of energy consumption, it was reported that the SBR-AGS system can achieve electricity
322 demand reduction equal to 23%, 35%, and 50-70% compared to activated sludge with EBPR, IFAS,
323 and MBR systems, respectively (Bengtsson et al., 2018). As a consequence, operating costs
324 decrease can be expected through AGS system also highlighting the beneficial investment
325 achievable through its application.

326 The new direction in developing the AGS technology is towards the design of CFRs capable to
327 cultivate the granules, being able to treat higher flow rates and easier to implement in municipal
328 WWTPs than SBR systems, as WWTPs receive wastewater to be treated continuously. The main
329 challenge of continuous-flow AGS systems relates to the stability of the granules, as the absence of
330 a settling phase promotes the retention of filamentous flocs, being commonly washed out in SBRs
331 during the short settling phase. The outgrow of filamentous bacteria in the system can undermine
332 the long-term stability and integrity of the granules. Additionally, the settling phase consists also in
333 a starvation period enabling the feast/famine regime (Kent et al., 2018). To assure granules
334 retention and limit the proliferation of filamentous bacteria in the systems, several continuous-flow
335 AGS configurations providing aerobic, anoxic and anaerobic zones have been tested at laboratory
336 scale, including bubble columns with baffles, serial multiple chambers, and systems coupled to

337 clarifiers or submerged membranes for granules retention. Nevertheless, no full-scale
338 implementation of these technologies has been completed up to date (Rosa-Masegosa et al., 2021).
339 Indeed, removal of filamentous bacteria is not always effective in CFRs especially at high organic
340 loadings (Li et al., 2016), which is detrimental for P removal, as granule destruction would
341 inevitably disrupt the alternation of feast/famine phases providing PAO and DPAO with a
342 competitive advantage for organic uptake over other heterotrophic bacteria. Despite these
343 challenges, further improvement of AGS-based CFRs in terms of granule stability could promote
344 their application in WWTPs in the near future.

345 Similar to AGS, the moving bed biofilm reactor (MBBR) allows the formation of a multi-layered
346 biofilm including nitrifiers, denitrifiers, PAO and DPAO. The simultaneous removal of carbon and
347 nutrients can be achieved by coupling SND to EBPR by operating the MBBR in SBR mode to
348 allow the alternation of aerobic and anaerobic phases (Ferrentino et al., 2018). Recently, lab-scale
349 MBBRs have been applied in continuous-flow mode for the combined removal of P, N and C under
350 stable microaerobic conditions (Iannacone et al., 2019) or by alternating aerobic and microaerobic
351 phases (Iannacone et al., 2021, 2020). The highest P removal efficiency, i.e., 83-86% (on average),
352 coupled to complete C and >80% N removal via shortcut SND was achieved when alternating DO
353 concentrations between 0.2 and 3 mg/L at stable HRT of 1 day and feed C/N and C/P of 3.6 and 11,
354 respectively (Iannacone et al., 2021). Microbial characterization of the MBBR biofilm strongly
355 suggested that P uptake was mainly carried out by bacteria belonging to the genus
356 *Hydrogenophaga*, which dominated the biofilm microbiome under IA conditions. Previous studies
357 reported that bacteria related to *Hydrogenophaga* have the ability to take up P under aerobic
358 conditions at a rate comparable to those observed in full-scale EBPR systems when acetate is used
359 as source of organic carbon (Iannacone et al., 2020). P removal under IA conditions was higher
360 when coupled to shortcut SND than to complete SND under similar operating conditions, which
361 suggests that the organic carbon saved by denitratation (NO_3^- reduction to NO_2^-) can support P

362 uptake under low COD conditions typical of domestic wastewaters (Campo et al., 2020; Chai et al.,
363 2019).

364 The results obtained by Iannacone et al. (2021, 2020) confirm that the anaerobic phase is not
365 mandatory for biological P removal from wastewater. Full-scale experiences had already shown that
366 high-efficiency P removal was achieved in conventional CAS and MLE configurations lacking the
367 anaerobic stage in which P release and denitrification occurred under anoxic conditions in the same
368 basin (Qiu et al., 2019). It was suggested that, if enough organic carbon is available for
369 denitrification, PAO can release P under anoxic conditions as in the anaerobic phase and perform
370 luxury uptake in the subsequent aerobic phase. This was confirmed by the relatively high
371 abundance of *Ca. Accumulibacter* ($\geq 2\%$) and *Tetrasphaera* (0.4-1.8%) in the microbial community
372 of the CAS and MLE sludges (Qiu et al., 2019). Besides, EBPR has been reported also under
373 complete aerobic conditions by using a synthetic wastewater containing 250 mg/L of acetic acid
374 (Pijuan et al., 2006).

375 Although EBPR can occur under complete aerobic conditions, the presence of an anaerobic phase
376 could promote VFA production from complex organic matter typical of real wastewater. VFA
377 levels in the feed of most laboratory-scale studies are not representative of those typically occurring
378 in real WWTP influents (<100 mg COD/L) (Barajas et al., 2002) and, therefore, scale-up of the
379 studied processes and the use of real domestic wastewater is recommended to assess the actual P
380 removal efficiencies. It is worth mentioning a further configuration represented by the IFAS system,
381 which is a hybridization of the MBBR in which carrier-attached and suspended biomass coexist.
382 Some studies on full-scale WWTPs integrating the IFAS technology to EBPR reported TP removal
383 from 48% to >80% for influents characterized by TP concentration ranging from 2.9 to 8.5 mg/L
384 (Kwon et al., 2010; Onnis-Hayden et al., 2011) (**Table 2**). It is worth mentioning that the lower
385 removal percentage (48%) was observed for an influent characterized by 120.5 mg BOD/L, 124.4
386 mg TSS/L, 32.2 mg N/L, 2.9 mg P/L, and a WWTP operated at HRT of 5.9 h and average

387 temperature of 23°C (Kwon et al., 2010). On the contrary, higher P removal efficiency was reported
388 for a WWTP operating at an average mixed liquor SRT of 3.78 d and a recycle flow relative to the
389 influent flow of 40% and 60% for activated sludge and mixed liquor, respectively. Moreover, the
390 WWTP influent was characterized by average concentrations of 8.5, 35.2, and 6 mg/L for TP,
391 ammonia and NO_x, respectively (Onnis-Hayden et al., 2011). A previous study also highlighted the
392 highest cost effectiveness of IFAS-EBPR (93.82 \$/kg of P removed) compared to MBR (135.31
393 \$/kg of P removed) and five-stage Bardenpho systems (102.24 \$/kg of P removed), while a similar
394 value was observed compared to the MUCT process (93.89 \$/kg of P removed) (Bashar et al.,
395 2018).

396

397 **Table 2. Biofilm-based applications for full-scale phosphorus removal.**

398

399 **2.1.3 Microalgae integration into WWTP to enhance P removal**

400 The use of P as a nutrient for photosynthetic organisms has been exploited through the development
401 of microalgae-based P removal bioprocesses. Similar to PAO, microalgae can uptake P in excess of
402 growth requirement (*luxury uptake*) as orthophosphate, which can be also produced by solubilizing
403 organic P via the enzyme phosphatase, and store it inside the cell as Poly-P granules (Bunce et al.,
404 2018). The use of microalgae for P removal from wastewater may lead to several advantages
405 compared to bacterial-based systems. Microalgae can combine efficient P removal and recovery to
406 CO₂ sequestration, energy generation (e.g., through production of biofuels) and production of
407 organic fertilizer (Badgujar et al., 2020; Suleiman et al., 2020). Very high removal efficiencies have
408 been observed in lab-scale studies on microalgae-based wastewater treatment. For instance, Rasoul-
409 Amini et al. (2014) carried out lab-scale tests on the simultaneous removal of N and P from
410 secondary wastewater effluents in 250 mL batch photobioreactors. In this case, *Chlamydomonas*
411 sp., *Chlorella* sp. and *Oocystis* sp were tested by adopting the following experimental conditions:
412 pH=9, constant illumination (4150 lux), T=25 °C, 190.7 mg NO₃⁻-N/L, and 19.11 mg PO₄³⁻-P/L. As
413 reported by the authors, within 14 d the minimum and maximum P removal were 82% and 100%
414 for *Chlorella* sp. and *Chlamydomonas* sp., respectively. Similar P removal efficiencies were
415 observed in a study reporting the treatment of the primary effluent of a WWTP by 11 different
416 microalgae strains (Chawla et al., 2020). Specifically, the batch experiments were performed in 250
417 mL flasks at 25°C and intermittent illumination of 12 h light (50 μmol/m²·s) and dark. In this
418 conditions, effluent P concentrations ranging from 0.1 and 3.2 mg PO₄³⁻-P/L compared to an
419 influent concentration of 15.5 PO₄³⁻-P/L. Compared to the very high removal efficiencies observed
420 at lab-scale experiences, a lower performance for pilot and full-scale applications can be expected
421 due to the difficulty of maintaining stable operating conditions (temperature, irradiation) which can
422 be easily set in laboratory experiments.

423 Although the application of microalgae in WWTP has been shown as a promising opportunity, only
424 a few full-scale microalgae-based systems have been operated so far. The majority of microalgae-
425 based systems applied at pilot and full scale for P removal (**Table 3**) consist in open suspended-
426 growth systems such as the HRAP (Chatterjee et al., 2019) and waste stabilization ponds (Brown
427 and Shilton, 2014). In particular, HRAP are generally characterized by lower electrical energy
428 consumption for mixing (50-110 kWh_e/ML) compared to conventional activated sludge (230-960
429 kWh_e/ML) (Craggs et al., 2011). However, despite the well-assessed technology, further process
430 optimization is fundamental in order to achieve higher nutrient uptake rate as well as cost effective
431 microalgal biomass harvesting (Sturm and Lamer, 2011; Sutherland et al., 2017). Another
432 interesting strategy for microalgae-based treatment is represented by the use of a microalgal
433 biofilms which can allow an easy harvesting from the support material (e.g., flat panels or rotating
434 biological contactors) or by collecting the gel matrixes in which they are embedded. However,
435 microalgae biofilms also present some challenges, as their application requires high surface areas
436 for biofilm development and high capital costs for purchasing the support material (Mohsenpour et
437 al., 2021).

438 The latest research in this field has focused on incorporating microalgae into bacterial systems
439 typically operated in WWTPs. The main advantage of this strategy is that microalgae can contribute
440 to meet the oxygen need of aerobic heterotrophs degrading the influent organic substrates, which
441 can significantly reduce the energy costs for aeration and mixing (Wang et al., 2015). On the other
442 hand, microalgae can capture the CO₂ produced by the oxidation of organic matter which turns into
443 higher production of energy and P-rich microalgae biomass (Cai et al., 2019). Moreover,
444 microalgae-bacteria systems do not require a transition between different operating environments
445 for simultaneous carbon and nutrient removal as microalgae directly assimilate both N and P for
446 cell growth and metabolic functions, thus reducing the complexity of the treatment configuration.

447 To establish a successful microalgae-bacteria symbiosis, the microalgae culture and physical-
448 chemical conditions should be accurately selected and maintained inside the system. Microalgae
449 belonging to *Chlorella* and *Scenedesmus*, as well as diversified consortia, have been successfully
450 applied for P removal from urban wastewater after primary treatment (concentrations of 3.8-7.5 mg
451 $\text{PO}_4^{3-}\text{-P/L}$, 147-473 mg $\text{NH}_4^+\text{/L}$, <0.5-0.9 mg $\text{NO}_3^-\text{-N/L}$, 101-147 mg COD/L, and pH ranging from
452 8.21 to 8.34) (Gouveia et al., 2016). In this work, removal efficiencies up to 100% for P and 98%
453 for N were achieved in a pilot-scale photobioreactor (150 L) operating in fed-batch mode over three
454 months (September-November) with average air temperatures from 11°C to 23°C (Gouveia et al.,
455 2016).

456 Regarding the operating parameters, the illumination period and intensity are fundamental to
457 enhance microalgae growth and consequently avoid an increasing bacteria/algae ratio. Although P
458 removal seems less affected, low light intensity (such as 40 $\mu\text{E/m}^2\cdot\text{s}$) may cause the proliferation of
459 nitrifying bacteria, causing NO_3^- and NO_2^- accumulation in the system and reducing TN removal
460 (González-Camejo et al., 2018). The optimal pH range to sustain the growth of both microalgae and
461 bacteria is between 7 and 9. Higher values may occur due to CO_2 consumption by microalgae and
462 impair bacterial growth, resulting in less CO_2 supporting the growth of microalgae and,
463 subsequently, lower P removal from wastewater. Although optimal temperatures of 28-30°C have
464 been indicated for the growth of microalgae (Park et al., 2011), significant P removal from waste
465 liquid streams (90%) has been reported also at temperatures as low as 15°C (Filippino et al., 2015)
466 and 5°C (38%) (Chatterjee et al., 2019) with cultures of *Chlorella* and *Scenedesmus*, respectively.

467 Although promising outcomes have been shown at laboratory and pilot scale, integration of algal
468 and bacterial microorganisms for P removal from wastewater has not been successfully tested at full
469 scale. However, recent studies recently focused on demonstrating the economic feasibility of
470 applying microalgae-bacteria consortia in open systems for the treatment of WWTP streams.
471 Mantovani et al. (2020) showed that the application of a pilot-scale open raceway pond for the side-

472 stream treatment of the centrate (244 mg $\text{NH}_4^+\text{-N/L}$, 0.2 mg $\text{NO}_3^-\text{-N /L}$, 0.4 mg $\text{NO}_2^-\text{-N /L}$, 5.7 mg
473 $\text{PO}_4^{3-}\text{-P/L}$, 83 mg TSS/L, 112 mg COD/L, pH of 8.2 and conductivity of 1492 $\mu\text{S/cm}$) produced
474 from a WWTP sludge streamline led to P and N removal >70% (on average) and nearly satisfied
475 oxygen demand for nitrification. Moreover, the process results in extra energy production by
476 sending additional biomass to AD. Overall, integrating a side-stream raceway pond to the WWTP
477 would result in a net energy saving of around 0.4 W/m^2 .

478

479 **Table 3. Algae-based biological systems for phosphorus removal at full and pilot scale.**

480

481 **3. Phosphorus recovery from WWTP streams**

482 As biological and chemical P removal WWTP treatments have the primary goal to reduce P
483 concentration of the influent wastewater within the mainstream treatment by incorporating it into
484 biomass or precipitates, all removed P ends up in the sludge streamline. Consequently, phosphorous
485 recovery is typically performed from liquid/solid side streams of the WWTP (**Figure 4a**), and it is
486 expected to improve when EBPR or chemical P removal are implemented in the WWTP
487 mainstream line. The presence of a hydrolysis/AD step in the sludge streamline can also help to
488 enhance the subsequent P recovery by promoting mineralization of organic P contained in the
489 sludge and the biological P release from PAO. A fundamental aspect of this practice lies in the
490 potential benefits derived from the recovery of removed phosphorous and its consequent reuse.
491 Many patented treatment systems for phosphorous recovery are nowadays reported in the literature,
492 and some of them have reached a technology readiness level (TRL) of 9 (**Table 4**), meaning
493 successful implementation at full scale. These processes mainly differ from one another according
494 to the matrix representing the P source. In the following sub-sections, P recovery systems will be
495 reviewed by distinguishing the treatments related to the recovery from sludge-derived aqueous
496 streams (digester supernatant, sludge dewatering liquor) and (semi)solid phases (i.e., digestate and
497 sludge ash). Only technologies sufficiently tested with real municipal WWTP streams are discussed.

498

499 **Figure 4. Schematics of phosphorus recovery in the WWTP sludge streamline (a) and of applicable patented technologies for**
500 **P recovery from digester centrate (b), dewatered sludge (c) and sludge ashes (d).**

501

502 **3.1 Systems for phosphorous recovery from liquid streams**

503 P-loaded liquid streams within a municipal WWTP typically refer to the liquid fraction of
504 anaerobically digested or hydrolyzed sewage sludge obtained post dewatering, e.g., centrate or
505 filtrate. P recovery in the liquid fraction can be enhanced by applying sludge disintegration
506 technologies prior to AD, e.g., thermal hydrolysis with Cambi (Di Capua et al., 2020), which
507 enhance P solubilization and subsequent recovery in the sludge liquor.

508 **3.1.1 Crystallization technologies**

509 One of the main processes applied for phosphorus recovery from the liquid phase is represented by
510 crystallization, which commonly occurs in fluidized systems (Chripim et al., 2019). An example is
511 provided by the Crystalactor[®] water treatment technology of Royal HaskoningDHV, which was
512 initially developed to remove hardness from drinking water (Desmidt et al., 2015). This system
513 provides the crystallization of a determined component in a FBR. During the crystallization process,
514 the size of the material constituting the fluidized bed increases, and the resulting heavier material
515 settles to the reactor bottom. Suitable dosage of reagents (e.g., lime and $Mg(OH)_2$) should be
516 provided during the process for adequate crystallization rate in order to achieve phosphorus
517 recovery in the final forms of calcium phosphate, magnesium phosphate or struvite
518 ($NH_4MgPO_4 \cdot 6H_2O$), the latter being an appreciated slow-release fertilizer (Di Capua et al., 2020).
519 Indeed, the relative fertilizer efficiency of struvite can be 100% for acidic soils but lower for
520 alkaline ones (Amann et al., 2018). The latter values are more beneficial when compared to calcium
521 phosphate providing lower P uptake when used in slight acidic and neutral soils (Johnston and
522 Richards, 2003). The main advantage of Crystalactor[®] is the combination in one stage of the

523 conventional steps constituting the precipitation process (i.e., coagulation, flocculation, separation
524 of the produced sludge and its dewatering). Water-free pellets can be produced leading to the
525 elimination of dewatering treatment and related costs. P recovery from the digester supernatant
526 through Crystalactor[®] can reach values up to 40% with respect to the WWTP influent, while typical
527 ranges are from 10% to 25% of the influent P for recovery technologies from the digester
528 supernatant (Egle et al., 2016). However, the higher efficiency observed with the Crystalactor[®]
529 technology can be achievable if enhanced biological phosphorous removal process is applied in the
530 WWTP.

531 Recovery percentage from 10% to 30% are also reported for another crystallization system
532 represented by the patented Ostara Pearl[®] technology (Chrispim et al., 2019). In this case,
533 phosphorous is recovered solely as struvite in the “Pearl” reactor, a FBR in which crystallization of
534 struvite is controlled through the addition of magnesium salts and, if required, NaOH to adjust the
535 pH. The system recycles part of the treated effluent from the top to the bottom of the reactor to
536 control the product size (**Figure 4b**). Once the struvite seeds have reacted the desired size (>0.5
537 mm), they are commercialized under the name of Crystal Green[®] (Gysin et al., 2018). In the
538 Multiform Harvest[™] process, which has been acquired by Ostara in 2019 and already applied in
539 small WWTP and farms, a cone-shaped FBR is used for struvite crystallization through the addition
540 of NaOH and MgCl₂.

541 As an alternative to the fluidized systems, the application of the fixed-bed reactor and CSTR also
542 proved feasible P recovery from supernatant digester. The P-RoC[®] process is based on a fixed-bed
543 configuration (Berg et al., 2007) where crystallization is achieved using calcium silicate hydrate or
544 synthesized pellets of tobermorite. The obtained product displayed a P total yield of 10-11%, which
545 is comparable to the common values for a commercial NPK 12-24-12 fertilizer (TP equal to 10.5%)
546 as well as natural P-rocks (Green, 2015). Differently, the NuReSys[®] process is carried out in two
547 CSTRs, i.e., a stripper for pH and scaling control and a reactor with MgCl₂ addition to promote

548 active formation of struvite pellets (Bio-Stru[®]). NuReSys[®] has been successfully applied at full
549 scale selectively on centrate and digested sludge (5-6% TS) of municipal WWTP as well as on both
550 streams sequentially through a hybrid configuration, resulting in P recovery >86% (Dewaele, 2015).
551 The PHOSPAQ[™] process, developed by Paques, combines biological COD removal to chemical P
552 removal in an aerated CSTR. Aeration provides oxygen for biological COD oxidation to CO₂,
553 which strips and raises the pH to 8.2-8.3, promoting struvite formation through the addition of
554 magnesium oxide (MgO). Struvite pellets (0.7 mm) are harvested from the bottom of the reactor by
555 means of a hydrocyclone followed by a screw press (Driessen et al., 2018). The process has been
556 successfully applied for the removal of phosphorus on sludge dewatering liquors at municipal
557 WWTPs and on industrial effluents, showing reliable full-scale performances. Additionally, this
558 process has been successfully combined with the anammox process in a municipal WWTP for
559 simultaneous removal of PO₄³⁻-P (80%) and NH₄⁺-N (90%) from the effluents of an UASB reactor
560 treating potato processing wastewater. The overall process produced biogas from COD conversion
561 into the UASB and fertilizer-grade struvite, and allowed the WWTP to respect the effluent
562 standards for nutrients (Remy et al., 2013).

563 An example of mainstream phosphorous recovery is represented by the REM-NUT[®] technology. In
564 this case, phosphate is removed from the effluent of secondary treatment through a selective ion-
565 exchange, and then recovered from the spent regeneration liquid of the ion-exchange with chemical
566 precipitation as struvite. The REM-NUT[®] has been developed only at pilot scale but never been
567 applied at full scale, which could be related to the selection of proper ion-exchangers for selective
568 removal of phosphorus and to the high cost of ion exchangers. Besides REM-NUT[®], other studies
569 focusing on combined P removal and recovery from WWTP mainstream line have been carried out
570 at laboratory scale. For instance, Zou and Wang (2016) tested an EBPR coupled with a
571 crystallization step aimed at recovering P as hydroxyapatite from synthetic wastewaters (SRT = 20
572 d; T = 16-21 °C; 5 mg PO₄³⁻-P/L). Results showed that PO₄³⁻-P removal efficiency was 87.5% and

573 59.3% of P compared to the influent concentration. In another study, concurrent P removal and
574 recovery was achieved combining anammox biological treatment and hydroxyapatite precipitation
575 in an expanded bed reactor maintained at 33°C (Ma et al., 2018). In particular, TP removal
576 efficiency equal to 66%, with an influent P concentration of 13 mg/L, was achieved in a 5 L reactor,
577 while a volumetric removal capacity equal to 0.14 kg P/m³·d was estimated.

578 Despite the majority of P recovery processes have the goal of recovering phosphorus as struvite,
579 which can be used as slow-release fertilizer, it is noteworthy to mention that P recovered in the form
580 of other compounds, such as hydroxyapatite or amorphous calcium phosphate, can have a broader
581 scope than fertilization. Indeed, calcium phosphates are the main constituents of PR, which are
582 mined not only to produce fertilizers, but also for the production of detergents, food additives,
583 phosphoric acid and much more (Chen and Graedel, 2016). Recently, the production of calcium
584 phosphates was attempted from the effluent of a UASB reactor. Specifically, by introducing a
585 nitrification step of the UASB effluents, the production of struvite is by-passed due to the
586 conversion of ammonia to nitrates, and the concentration of dissolved inorganic carbon, which both
587 inhibits calcium phosphate precipitation and leads to the contamination of the phosphate with
588 carbonate, is reduced (Monballiu et al., 2018a). This strategy was tested with the UASB effluents
589 collected at a potato processing plant in a two pilot-scale CSTRs connected in series (HRT of 25-27
590 h): the first was operated as a nitrification reactor, while the second was used for the precipitation of
591 calcium phosphates. The influent to the first reactor was characterized by approximately 293 mg
592 N/L (as NH₄⁺), 50 mg P/L (as PO₄³⁻) while Ca²⁺/P and Mg²⁺/NH₄⁺/P molar ratios were equal to
593 0.7/1.0 and 0.8/13.0/1.0, respectively. Accounting for the influent of the second reactor,
594 concentrations equal to 9 and 55 mg/L were respectively observed for NH₄⁺-N and PO₄³⁻-P, while
595 values of 0.6/1.0 and 0.7/0.4/1.0 were reported for the Ca²⁺/P and Mg²⁺/NH₄⁺/P molar ratios. In this
596 study, a satisfactory phosphate recovery of 70%, mainly in the form of hydroxyapatite, was reached
597 at neutral pH (Monballiu et al., 2018b).

598 **3.1.2 Adsorption**

599 Adsorption represents another possible strategy that can be applied for P recovery from urban
600 wastewaters and liquid fraction of sewage sludge. This process is driven by Van der Waals and
601 electrostatic forces, and is favored by low temperatures. Also, it is reversible and not site-specific
602 (multi-layer formation). Important parameters of the process are the pH of the solution, which
603 controls the absorption capacity, the contact time, the affinity to the sorbent surface, and the
604 increase of sorbent dosage (Ajmal et al., 2018; Kumar and Viswanathan, 2018). Phosphate ions can
605 be effectively removed from water and wastewater through a double step mechanism based on
606 sorption and precipitation. Generally, adsorption occurs via an ion-exchange mechanism between
607 calcium and magnesium compounds present at the surface of adsorbent materials, e.g.,
608 cement/concrete, and phosphate in solution, while the partial dissolution of Ca^{+2} and Mg^{2+} ions
609 from the adsorbent triggers phosphate precipitation, which is favored by the increase of pH detected
610 after ion dissolution (dos Reis et al., 2020; El Bouraie and Masoud, 2017).

611 Current research is moving towards the use of industrial by-products as sorbents for the removal of
612 phosphate from wastewater, promoting the reuse of secondary raw materials for recovery purposes
613 (Sellner et al., 2019; Hashim et al., 2021). These sorbents are chemically constituted by oxides,
614 hydroxides, Si, Fe, Al, Mg, Ca, and Ti carbonates and may have different origins, e.g., power plant,
615 mining and steel industry (fly ash, red mud, slags) (Goscianska et al., 2018; Guo et al., 2018; Kim
616 et al., 2000). Phosphate removal can be also obtained by iron oxides from mineral processing
617 industry (Ajmal et al., 2018) or by wollastonite from the mining of this calcium meta-silicate
618 mineral (Obradović et al., 2017). Ligand exchange and surface precipitation are the predominant
619 sorption mechanisms with removal capacities depending on the by-product composition, size,
620 porosity and on the experimental methodology adopted.

621 Among organic sorbents, ion-exchange resins have largely replaced the use of zeolites and are
622 widely used in various chemical engineering applications, particularly in the water demineralization

623 processes also as phosphate sorbent. Different ion exchange resins have been employed to remove
624 phosphate from wastewater such as Purolite A500P, Purolite A520E, Purolite FerrIX A33E,
625 Amberlite IRA910Cl and Amberjet 1200Na, Dowex 21K XLT (Nur et al., 2014, 2012), or magnetic
626 ion exchange and hybrid anion exchange resins (Nur et al., 2012; Sowmya and Meenakshi, 2013).
627 These resins are structurally characterized by a polymeric matrix with grains in the range of few
628 mm and exchangeable ions. The regeneration of the exhausted resins can be obtained by NaCl,
629 NaOH, CaCl₂, HCl, and Na₂CO₃ (Loganathan et al., 2014) or their mixtures. Involvement of
630 calcium or magnesium salts for resins regeneration leads to precipitation phenomena advantageous
631 for possible phosphorus recovery and reuse as fertilizer. As an alternative, activated carbons are
632 characterized by low cost, easy availability, high porosity, high specific surface area, and can be
633 used for phosphate removal from water and wastewater (Khalil et al., 2017; Yao et al., 2018).
634 Agricultural wastes have been proven low-cost biological sorbents to remove phosphate from
635 wastewater. The surface properties and the porosity of these materials can be improved after
636 chemical modification of the structure. Specifically, efficient anion exchangers are obtained by
637 grafting amine groups onto wheat straw (Xu et al., 2010), whereas chemical treatments may
638 increase the porosity and the number of functional groups as in the case of saw dust from
639 woodworking operations (Benyoucef and Amrani, 2011). Organic wastes with quantitative presence
640 of iron showed good retention capacities of the phosphates (Field et al, 2019, Zou et al, 2017). In
641 recent years, biochar obtained by thermal degradation (pyrolysis) of a biomass of animal or
642 vegetable origin has been increasingly applied on municipal wastewater as phosphate sorbent.
643 Modification of biochar surface with functional additives such as AlCl₃, MgCl₂, and Fe₃O₄ can be
644 performed to enhance phosphate removal efficiency (Zheng et al., 2020). Zheng et al. (2019)
645 applied biochar modified with colloidal and nanosized aluminum oxyhydroxide (AlOOH) and
646 produced through direct pyrolysis of wood chips pretreated with Al salt to recover P from real
647 secondary treated wastewater, showing promising results both in terms of P recovery and

648 application of P-loaded biochar as fertilizer. Indeed, (modified) biochar may in the future be applied
649 for nutrient removal and recovery from WWTP liquid streams through a biofiltration step. Trials
650 with real municipal wastewater have shown a 78% TP reduction after passing through a biochar
651 filter with a surface area of 1000 m²/g and loading of 1 g/L (Manyuchi et al., 2018), demonstrating
652 potential for full-scale implementation for phosphorus recovery.

653 **3.1.3 Electrochemical precipitation**

654 Electrochemical precipitation represents a newly explored and economically sustainable solution for
655 P recovery from municipal wastewater, as it does not require chemicals for pH increase and
656 stoichiometric requirements (struvite precipitation), nor for surface modification of sorbent
657 materials (adsorption). In electrochemical systems, water electrolysis at the cathode generates a
658 local pH increase, driving the precipitation of calcium phosphate, e.g., as hydroxyapatite
659 (Ca₅(PO₄)₃OH). Subsequently, the formed precipitate can be harvested from the cathode without the
660 need of a separation step. A recent investigation by Lei et al. (2018) with municipal wastewater
661 showed that the efficiency of selective P precipitation on the cathode strongly depends on the
662 cathode size and Ca/P ratio in wastewater, the latter being generally much higher than the
663 stoichiometric ratio for hydroxyapatite formation in real wastewater (1.67 mol/mol). This leads to
664 the co-precipitation of by-products such as calcite (CaCO₃) and brucite (Mg(OH)₂), which reduce
665 the purity of the obtained P precipitate. However, the extent of by-product precipitation can be
666 limited by reducing the current intensity and/or through the addition of external P (which anyway
667 would offset the advantage of no chemical utilization). Trials with raw domestic wastewater showed
668 that less than 50% P removal could be achieved with this strategy, being significantly lower than
669 those observed with crystallization and absorption technologies. Further development is needed for
670 potential implementation of electrochemical systems for P recovery in municipal WWTP.
671 Additionally, it should be noted that hydroxyapatite is considered a less valuable P-based product

672 for agricultural utilization compared to struvite, although it can be used as a platform chemical for P
673 extraction.

674 **3.2 Systems for phosphorous recovery from raw and digested sewage sludge**

675 Higher phosphorous recovery from sludge can be expected compared to the influent wastewater as
676 up to 95% of P removed from the WWTP influent is estimated to be transferred to the sewage
677 sludge (Cornel and Schaum, 2009). This could promote the strategy of sewage sludge reuse for land
678 fertilization purposes. This practice is allowed in many countries and represents a convenient and
679 sustainable option for P recycling from sewage sludge (Di Costanzo et al., 2021). Indeed, sewage
680 sludge can be spread to land if complies with required quality standards and after appropriate
681 hygienization, e.g., through thermophilic AD in centralized plants collecting dewatered sewage
682 sludge on regional/national scale (Di Capua et al., 2020). This practice enables the maximum
683 exploitation of sewage sludge nutrient potential, as most of the P contained in the sludge is bound to
684 the solid phase. The issue about agricultural reuse of sewage sludge is the potential presence of
685 pollutants (e.g., HMs, pharmaceuticals and pesticides) at high concentrations, including emerging
686 contaminants being nowadays of increasing concern. Also, depending on the pollutant, the AD
687 process can display effective (such as for polychlorinated biphenyl) or ineffective degradation
688 efficiency, consequently not assuring a suitable sludge quality standard for agricultural reuse
689 (Fijalkowski et al., 2017). From this perspective, many countries worldwide adopt different
690 restrictive regulations to discipline sludge agricultural reuse (Christodoulou and Stamatelatou,
691 2016). Besides the presence of organic pollutants, elevated concentrations of HMs within the sludge
692 often limit sludge application in agriculture, and therefore alternative strategies for exploiting the P
693 content of sewage sludge should be undertaken. Accordingly, the technologies applied for
694 phosphorous recovery from sludge also aim at limiting HMs content in the recovered product to
695 provide a safe fertilizer.

696 Strategies for P recovery from sewage sludge include struvite crystallization (e.g., MagPrex[®]), wet-
697 chemical extraction (e.g., Gifhorn and Stuttgart), wet-oxidation (e.g., LOPROX), supercritical
698 water-oxidation (e.g., Aqua Reci[®]), metallurgic smelt gassing process (MEPHREC[®]) and magnetic
699 separation of Fe-P precipitates. These technologies can reach 40-70% recovery of the P contained in
700 the WWTP influent, being significantly higher than that obtained from the sludge liquid fraction
701 and discussed in section 3.1.

702 The MagPrex[®] system (formerly AirPrex[®]) developed by Berliner Wasserbetriebe involves
703 digestate treatment in an air-lift reactor in which air is recycled for CO₂ stripping and consequent
704 pH increase (from 7 to 8). Then, subsequent addition of MgCl₂ allows the formation of struvite
705 crystals that sink to the bottom of the reactor when reaching a certain size. Full-scale plants
706 operating with this system were reported to achieve phosphate recovery from the digestate >80%.
707 For instance, two WWTPs (in Wassmannsdorf and Amsterdam west) characterized by similar
708 influent dissolved PO₄³⁻-P concentrations equal to 387 and 400 mg/L and SRT for all reactors of
709 10.5 and 8 h displayed a PO₄³⁻-P precipitation efficiency up to 87% and 95%, respectively. In these
710 full-scale applications, higher precipitation efficiency was achieved by dosing 1.9 mol Mg/mol P as
711 MgCl₂, while dosing 1.7 mol Mg/mol P resulted in the lower precipitation efficiency (Zhou et al.,
712 2019). However, only 40-50% of the produced struvite is recovered in the air-lift reactor, as the rest
713 ends up in the solid fraction in the following dewatering phase (CNP Cycles GmbH, 2015).

714 The Gifhorn process is carried out by a first wet-chemical extraction of the P bound in the biomass
715 through the addition of sulfuric acid. Afterwards, the dissolved metals are precipitated as sulfides by
716 dosing Na₂S and adjusting the pH with NaOH. Finally, the addition of Mg(OH)₂ leads to
717 phosphorous precipitation and recovery as a mix of struvite and calcium phosphates (Rashid et al.,
718 2020). The wet-chemical extraction step applied in the Gifhorn process is a general key-point for
719 strategies involved in the phosphorous recovery from sewage sludge. Indeed, the Stuttgart process
720 is performed through a first acidification step of the sludge similar to the Gifhorn approach, but

721 metal ion separation is achieved through complexation by means of chelating agent (such as citric
722 acid) addition before the struvite precipitation step (Meyer et al., 2019). The whole process
723 efficiency depends on the chemicals involved and pH conditions for phosphorous dissolution. In
724 fact, the initial acid leaching step at pH 3 can lead to final phosphorous recovery from Stuttgart
725 process higher than 65% in struvite form with low metal content (Meyer et al., 2019).

726 A different approach is displayed by the Aqua Reci[®] process in which a SCWO step is employed
727 before the acid or alkaline leaching phase (**Figure 4c**). The SCWO phase provides both the
728 mineralization of the total organic matter and the oxidation of toxic metals and phosphorus
729 respectively to their maximum valency and P₂O₅ (Stendahl and Jäfverström, 2004). The resulting
730 ash from the SCWO can further undergo alkaline leaching to extract phosphorous as sodium
731 phosphate that can be easily precipitated as calcium phosphate by lime addition. Alternatively, acid
732 leaching could be involved and precipitation can occur through three neutralization steps gradually
733 raising the pH and recovering ferric phosphate, aluminum, iron, and HMs (Stendahl and
734 Jäfverström, 2004). Despite the overall system can lead to sludge volume reduction, which is
735 beneficial in terms of operating costs, it should be also taken into account that previous life cycle
736 analyses highlighted significant energy consumption related to the process (Amann et al., 2018).

737 Low pressure wet oxidation at temperature ranging between 160°C and 220°C, pressure of 11.8-
738 27.6 atm and acidic conditions (pH=1.5) can be used for sludge oxidation in presence of pure
739 oxygen. This oxidation treatment is identified as the LOPROX system, and is followed by two
740 membrane filtration phases to recover phosphorous from sludge (Blöcher et al., 2012). The first
741 membrane step (ultrafiltration) aims at removing suspended solids from the LOPROX effluent,
742 while the second step, i.e., NF, aims at recovering phosphorus in the membrane permeate.
743 Specifically, the recovery rate is strictly related to the ion rejection effects of the NF membrane on
744 phosphorous. Indeed, higher phosphorus recovery is expected with decreasing rejection achievable
745 through an acidic wash of the NF membrane (Blöcher et al., 2012). Finally, the MEPHREC[®]

746 metallurgic process allows, compared to the other recovery processes applied on sludge, the highest
747 recovery percentage in terms of WWTP influent (up to 75%) in the form of a final P-rich slag where
748 P is present as calcium silicon phosphate (Amann et al., 2018; Günther et al., 2018). Nonetheless,
749 some difference in terms of depollution efficiency can be expected during the process due to
750 possible metals behavior variation (Egle et al., 2016).

751 An interesting research direction that is emerging in the last years is the recovery of the mineral
752 vivianite ($\text{Fe}_3(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}$) from sewage sludge produced in WWTPs performing chemical P
753 precipitation with iron. Indeed, this mineral has been reported as the most abundant P precipitate in
754 digested sludge when iron is dosed in the mainstream line to remove P from wastewater (Wilfert et
755 al., 2018). At this stage, only lab-scale experiments have been carried out on real sewage sludge
756 with magnetic separation concentrating the vivianite by a factor of 2-3, but significant amount of
757 organic matter, quartz and metals remained in the magnetic concentrate (Prot et al., 2019). Indeed,
758 this technology is still at early stage and further research is required towards optimization of
759 separation efficiency and quality of the concentrate. However, vivianite extraction from sewage
760 sludge is promising as it could be applied directly as a slow-release fertilizer in iron-poor soils or
761 used as a platform compound for P extraction, while reducing the amount of sludge to be disposed.

762 **3.3 Systems for phosphorus recovery from sludge ash**

763 Technologies recovering P from sludge ash include wet-chemical extraction (RecoPhos[®] and
764 Fertilizer Industry), wet-chemical leaching (e.g., PASCH and LEACHPHOS[®]), thermo-chemical
765 treatments (e.g., AshDec[®] and Thermophos[®]). These technologies can reach P recovery efficiencies
766 of 70-90% related to WWTP influent.

767 Acidic wet leaching has been widely used as the main step of several systems aimed at phosphorous
768 recovery from sludge ashes. However, sludge ash contains elevated concentrations of HMs,
769 primarily Fe, Zn and Cu, which should be separated from the recovered P for its safe utilization in
770 agriculture (Di Costanzo et al., 2021). Examples of acidic wet leaching for P recovery are provided

771 by PASCH e LEACHPHOS[®] processes which aim at re-solubilizing the P contained in the ashes
772 after the incineration process. For PASCH, a liquid-liquid HMs removal step is also performed in
773 order to achieve a higher purification of the recovered P (Egle et al., 2016). Nonetheless, both the
774 PASCH and LEACHPHOS[®] processes have been applied only at laboratory and pilot scale. In
775 processes such as RecoPhos[®] and Fertilizer Industry, a P recovery equal to 85% of that contained in
776 the WWTP influent can be achieved in the form of mineral fertilizer through acid-wet chemical
777 extraction. Although no selective HMs extraction step is performed, the obtained RecoPhos[®] ash-
778 derived fertilizer has shown compliance with the German Fertilizer Ordinance as well as conformity
779 to conventional fertilizers in terms of HM content and effect on crops (Weigand et al., 2013).
780 Various final recovered products are instead achieved by EcoPhos[®], which has been implemented at
781 full scale despite the plant being not currently in operation (Ploteau et al., 2021). In this case, the
782 technology leads to the production of fertilizer-grade phosphoric acid and/or production of
783 dicalcium phosphate. Ashes are first leached using phosphoric acid and the leach liquor is
784 subsequently decontaminated from HMs by passing through multiple ion-exchange modules. If HCl
785 is used instead of H₃PO₄ as leaching agent, phosphate in the leachate is chemically precipitated and
786 recovered as dicalcium phosphate (Takhim et al., 2019).

787 A further approach for the recovery of phosphorus from sewage sludge ashes is provided by thermal
788 processes. The AshDec[®] system, which has been tested at pilot scale, represents a well-assessed
789 solution in which sludge ashes, mixed with potassium and/or sodium compounds, and a reducing
790 agent (preferably dry sewage sludge), are treated in a rotary kiln at 900–950°C for 15–20 min
791 (**Figure 4d**). Potassium and sodium ions replace calcium ions in the phosphates and form soluble P
792 and PK fertilizers. Simultaneously, HMs evaporate and, as soon as the process gas cools down, the
793 particles condensate and are removed by means of electrostatic precipitators (Hermann and Schaaf,
794 2019).

795 At high temperatures (1500-1600°C) achieved through coke burning or electric heating it is possible
796 to recover phosphorous as elemental white phosphorus (P₄). The latter approach is followed in the
797 Thermphos[®] process, which indeed can be considered as a highly energy demanding strategy.
798 However, the pure (99.99%) product recovered can be suitably reused in the production of several
799 P-based chemicals (such as P₂O₅, NaH₂PO₂, PCl₃, PH₃, etc.), while the calcium-silica slag
800 remaining in the furnace can be employed in road construction (Jupp et al., 2021). In addition,
801 Thermphos[®] produces dust rich in HMs, which is precipitated electrostatically and recycled into the
802 process. However, the presence of high iron levels in the ashes lowers P recovery efficiency, as Fe
803 can be reduced to FeP and FeP₂, reducing P₄ yield. Volatile HMs can partly evaporate, precipitate
804 on dust, and build up in the system through dust recycling, requiring periodical purging. A full-
805 scale Thermphos[®] plant was run in The Netherlands from 2007 until 2012, when the company
806 declared bankruptcy apparently due to excessive market competition for P₄ commercialization
807 (BSR WATER, 2021).

808 **3.4 Environmental impact of P recovery technologies applied in WWTPs**

809 An environmental assessment of the existing technologies applied in WWTP for P recovery has
810 been carried out by Amann et al. (2018) through a life cycle assessment which also considers the
811 impacts of energy production and waste disposal of each technology. The analysis has been carried
812 out based on three indicators: CED, GWP and AP. Technologies recovering P from the liquid
813 fraction of sewage sludge generally feature negative values of the three indicators, indicating low
814 environmental impact. In this regard, an exception is represented by Crystalactor[®], which features a
815 CED of 41 kWh per population equivalent per year due to the high energy demand of this process,
816 as well as positive GWP and AP values. Another positive aspect of these technologies is that the
817 materials recovered from the liquid phase of sewage sludge are generally safe, as they contain
818 extremely low concentrations of HMs, micropollutants and pathogens.

819 Despite the high energy demand of incineration, technologies recovering P from sewage sludge
820 ashes can also feature negative values of the three indicators due to reduced production of P
821 fertilizers using PR and industrial chemicals, being partially replaced by recovered P-rich products
822 such as H_3PO_4 and P_4 . However, materials recovered from sludge ashes can present high levels of
823 HMs and, although incineration destroys pathogens and micropollutants, it can also generate toxic
824 contaminants such as dioxins and furans.

825 In contrast, technologies recovering P from sewage sludge, such as wet-oxidation processes,
826 generally feature positive CED, GWP and AP due to the high demand of energy intense chemicals.
827 Materials recovered from sewage sludge, e.g., through the Gifhorn and Struttgart processes, can
828 feature low contamination levels if decontamination steps for the removal of HMs are carried out.
829 Although some organic micropollutants can be detected, they are generally at lower concentrations
830 than in the untreated sewage sludge (Amann et al., 2018).

831 **3.5 Overall economics of the P recovery processes applied in WWTPs**

832 Annual costs for technologies aimed at phosphorous recovery from post-digestion supernatants are
833 generally around 8-10 € per kg of recovered P (**Table 4**). The NuReSys[®] technology is reported as
834 the cheapest technology on the market, resulting in operational cost of ≤ 3 € per kg of recovered P
835 (Desmidt et al., 2012; Dewaele, 2015), which promotes the use of CSTR as a more convenient
836 alternative to conventional fluidized-bed columns for P crystallization. In comparison, the
837 attractiveness of a fixed-bed technology such as the P-RoC[®] may be limited by higher annual costs
838 (around 6 € per kg of recovered P) (Egle et al., 2016) and by calcium phosphate as the end-product,
839 being less bioavailable to plants than struvite especially in neutral and alkaline soils (Amann et al.,
840 2018). However, some studies suggest the use of refractory calcium phosphates, such as calcium
841 hydrogen phosphate, calcium pyrophosphate, and hydroxyapatite, as substitute of PR (Berg et al.,
842 2006; Monballiu et al., 2020) or for the production of nanostructured P fertilizers (Tang and Fei,
843 2021). Overall, the benefit of implementing these technologies at full scale relates primarily to a

844 reduction of maintenance operations and related costs, as struvite incrustation may severely impact
845 the WWTP piping and pumping systems. Also, struvite precipitation and harvesting significantly
846 reduce P backflow to the mainstream treatment with the digester supernatant, reducing the burden
847 for P removal.

848

849 **Table 4. Full-scale (TLR 9) technologies for P recovery from solid/liquid WWTP streams (Corona Encinas, 2020; Desmidt et**
850 **al., 2012; Dewaele, 2015; Egle et al., 2016; Val del Río et al., 2016).**

851

852 The involvement of chemicals (such as acids and/or basis) for phosphorous recovery can lead to a
853 significant increase of the operating costs. Indeed, the REM-NUT[®] technology, which recovers P
854 directly from the WWTP effluent, results in the most expensive approach with annual costs
855 (excluding savings and revenues) as high as 28 € per kg of recovered P due to the high requirement
856 of resins and reagents (Egle et al., 2016). Additionally, the powdery/dusty struvite obtained by
857 REM-NUT[®] is not directly applicable and marketable, needing a further granulation/classification
858 step prior to be sold. Approaches for phosphorous recovery from sludge through wet-oxidation also
859 result in operating burden due to the high energy demand and annual costs (e.g. 23-27 € per kg of
860 recovered P for the Aqua Reci[®] system) (Egle et al., 2016; Law and Pagilla, 2019). Similarly,
861 considering the recovery systems from sludge ashes, high costs and energy consumption can be
862 expected especially referring to thermochemical treatments. An example is provided by the
863 AshDec[®] system which is significantly energy demanding due to the heat required to separate
864 phosphates and HMs (Ye et al., 2017).

865 Capital and operational costs may be substantially reduced by considering the potential revenues
866 and savings from 1) the commercialization of the obtained P-rich products, 2) reduced sludge
867 disposal volumes, 3) improved dewaterability of the digestate due to aeration and leaching of the
868 sludge with acids and 4) heat recovery from thermal P-recovery processes (Egle et al., 2016).

869 However, some criticalities exist regarding the low market value of the obtained P-rich secondary
870 material (0-100 € per ton) and uncertainties on their safety for agricultural use related to the
871 potential presence of pathogens (Mayer et al., 2016). The latter issue is a further limitation to the
872 use of digestate-derived struvite, since it has been observed to contain more pathogens compared to
873 struvite from the dewatering liquor of digestate (Muys et al., 2021). Despite the high operational
874 costs of thermal treatments for P recovery from sludge, these technologies allow to recover heat and
875 energy and convert the sludge to disposable inert waste, which can considerably decrease the
876 overall treatment cost. However, uncertainties regarding the economic impact of thermal treatments
877 exist, since these technologies have only been implemented at pilot scale (Egle et al., 2016).
878 According to this, the identification of a suitable approach aimed at maximizing the overall benefits,
879 in terms of process economics and valuable product recovery, should consider several aspects. This
880 clearly highlights that specific assessments should be singularly distinguished in order to properly
881 select the optimal technical solution characterized by: (i) significant process efficiency, (ii)
882 economic affordability and (iii) environmental sustainability.

883 **4. Conclusions**

884 Innovations and practices on P removal from WWTP influents are moving towards the
885 implementation of compact and more efficient biological systems such as AGS and carrier-based
886 biofilm systems both in SBR and continuous-flow configurations for the combined removal of P, N
887 and C from urban wastewaters through alternation of aeration phases activating microbial groups
888 with diverse metabolic functions. Although successful operation of these system has been shown up
889 to TRL 9, uncertainties regarding long-term stability remain due to their operational and biological
890 complexity. Therefore, further optimization at full scale is still needed to enhance the stability and
891 promote the application of these systems in modern WWTPs. Similarly, integration of microalgae
892 to bacterial systems might help improving the efficiency and sustainability of P removal in
893 WWTPs, but full-scale experience is required the confirm long-term stability of algae-bacteria

894 consortia and assess their response to changing of influent characteristics, physical conditions,
895 degree of toxicity and microbial community composition. Simultaneous removal and recovery of P
896 is the new trend in modern WWTPs, although incentives from national governments to promote the
897 use of struvite and sludge-derived fertilizers would significantly encourage this practice.
898 Technologies for P recovery from sludge liquors are the readiest for implementation at full-scale, as
899 they offer a high level of technical maturity and are less complex and costly than those recovering P
900 from sludge and sludge ash. Moreover, the high P content of dewatered sewage sludge can be
901 exploited in agriculture when sludge quality and local legislation allow this practice. Coupling P
902 recovery from wastewater/sludge liquors in WWTP and digestate application to land would allow a
903 nearly complete P recycling from waste streams and improve the overall P removal efficiency of the
904 WWTP. In this direction, performing an hydrolysis step in the WWTP sludge streamline may help
905 P solubilization in the sludge liquor (and its subsequent recovery) as well as methanogenesis in
906 centralized AD, being hydrolysis the limiting step of high-solid AD of sewage sludge (Di Capua et
907 al., 2020; Morello et al., 2021). When agricultural utilization is not applicable, thermochemical P
908 recovery from sludge ash offers a promising alternative to P recovery from aqueous streams, based
909 on high recovery efficiencies, safety and the possibility of savings based on heat recovery and
910 cheaper disposal of the inert residue. Direct recovery from the sludge is not yet considered
911 convenient for full-scale implementation due to lower quality of the obtained P-based products, low
912 environmental sustainability, and high costs.

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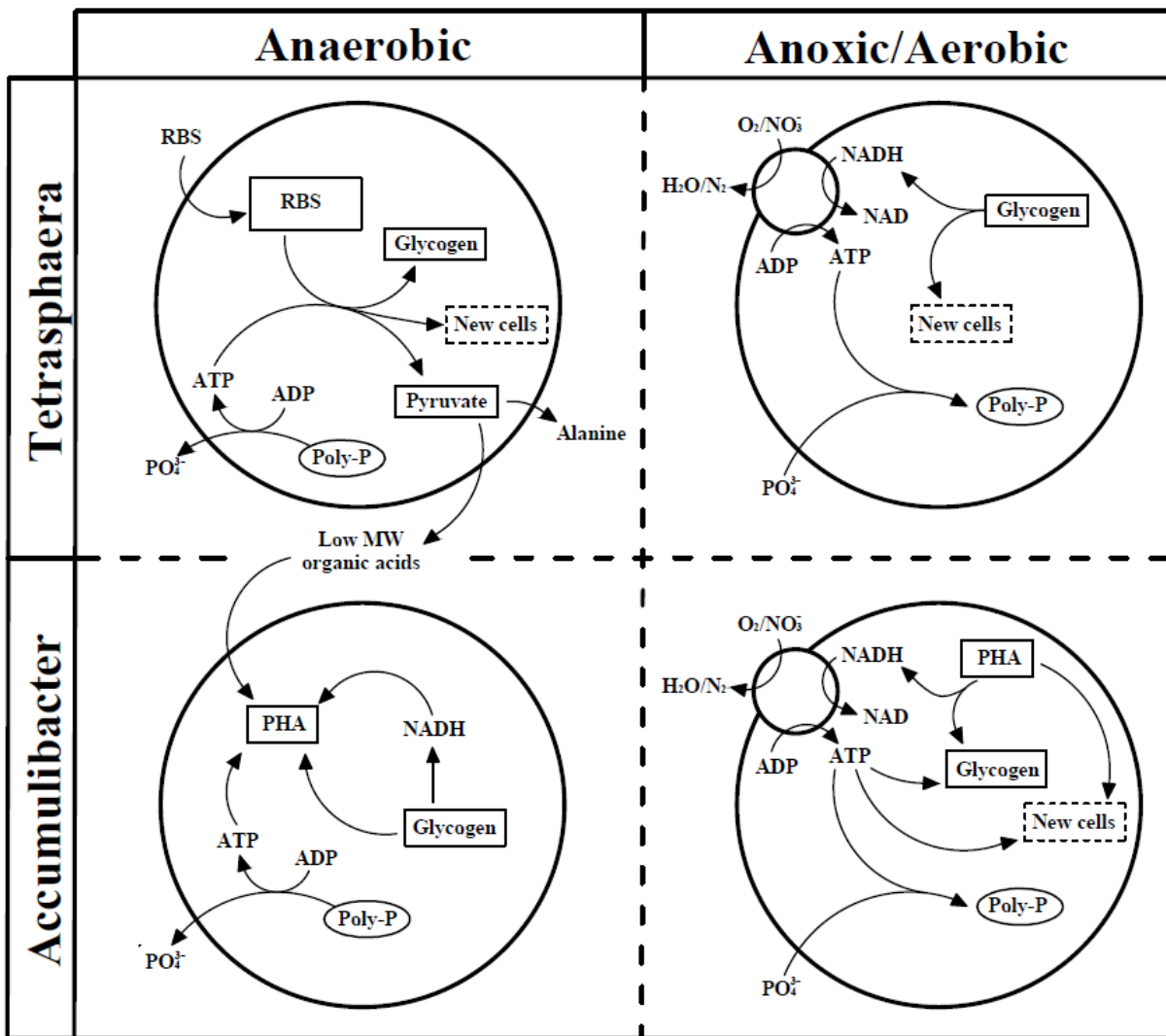
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1468 **List of Figures**

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1471 **Figure 1. A brief description of *Tetrasphaera*- and *Accumulibacter*-related microorganisms in an EBPR system.**

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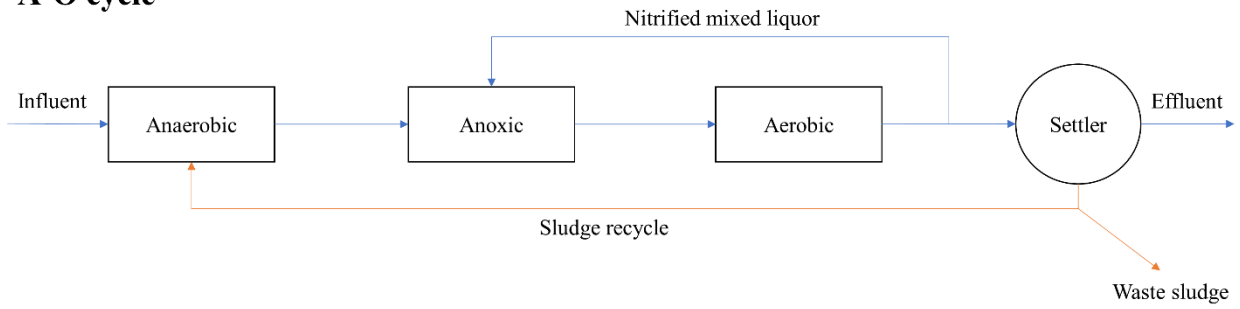
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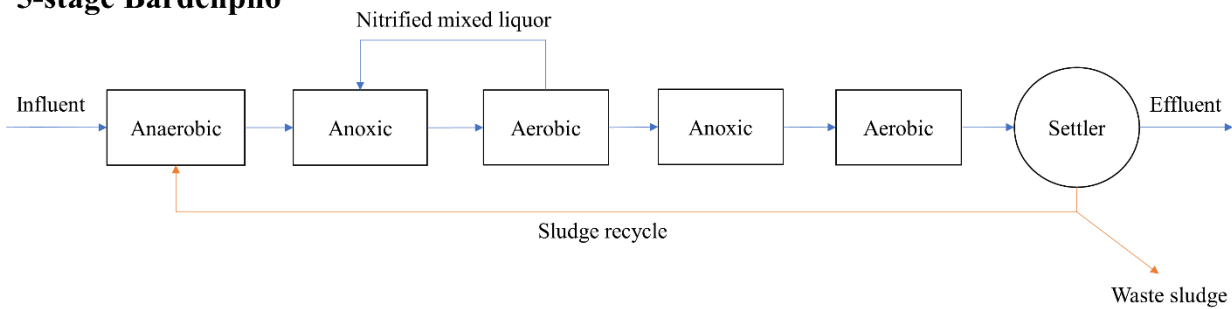
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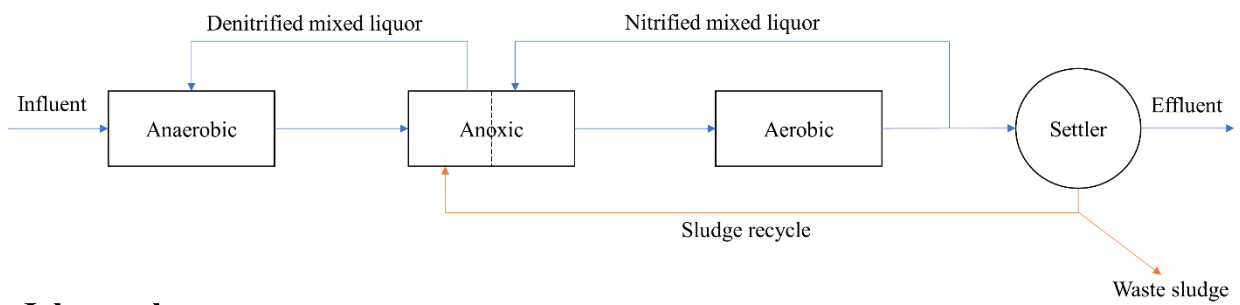
A²O cycle



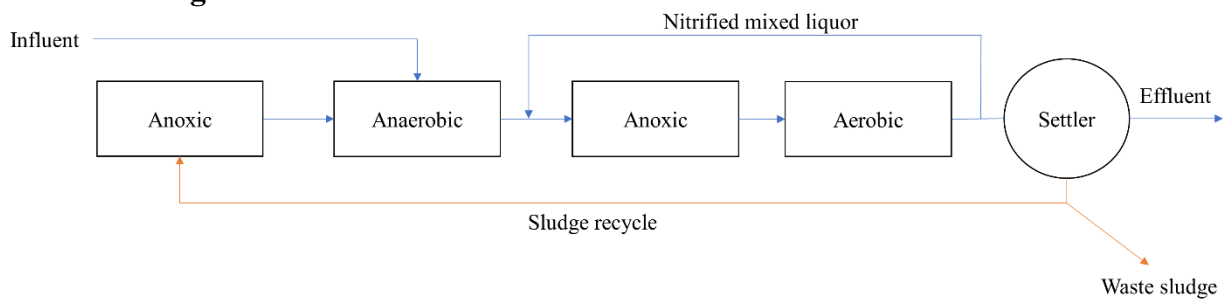
5-stage Bardenpho



UCT/modified UCT cycle



Johannesburg



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1481 **Figure 2. WWTP cycles for combined biological P and N removal from wastewater.**

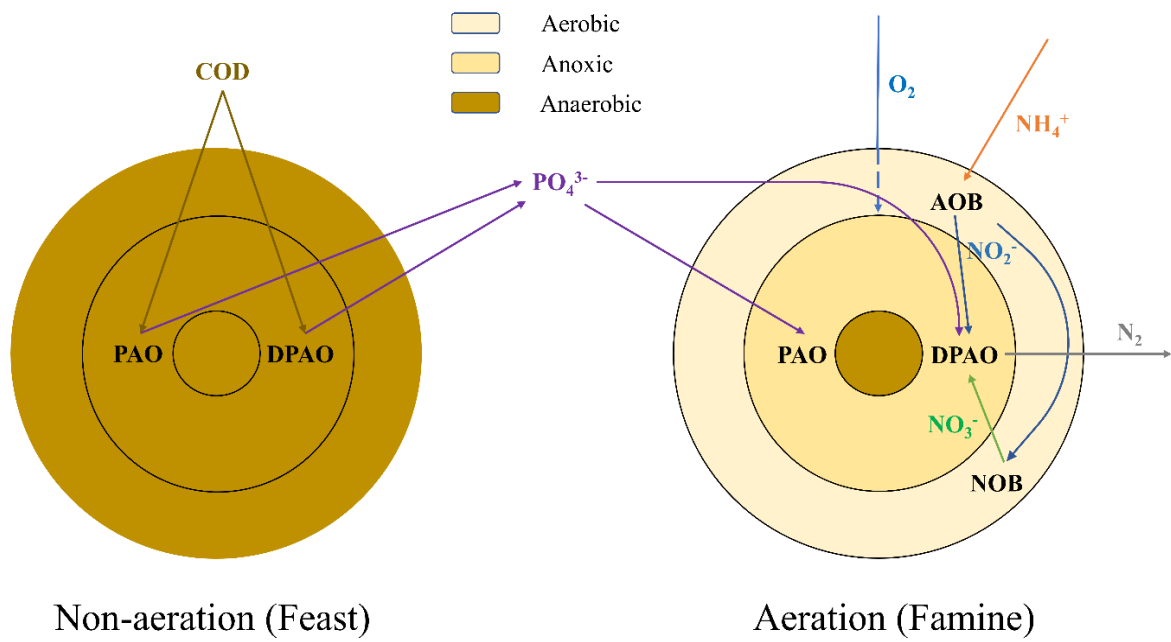
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1488 **Figure 3. Main phosphorus, nitrogen and carbon transformation and layer distribution in an AGS granule during aeration**
1489 **and non-aeration phases.**

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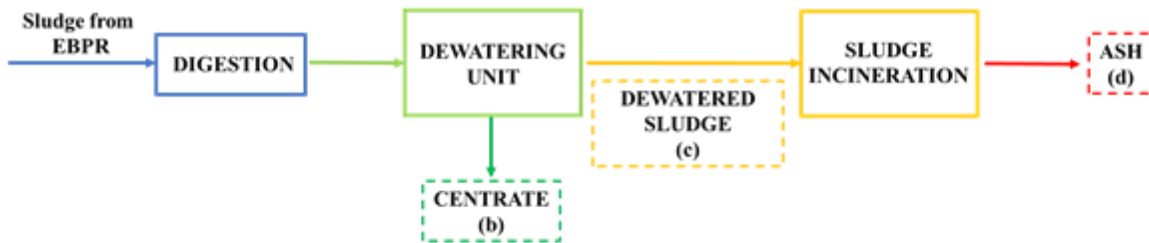
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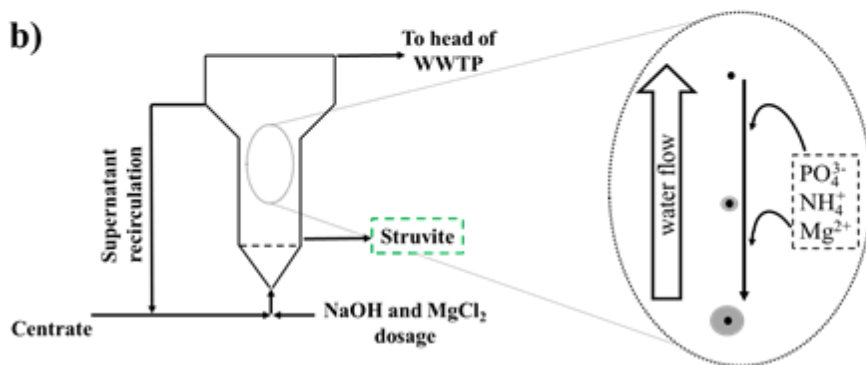
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a)

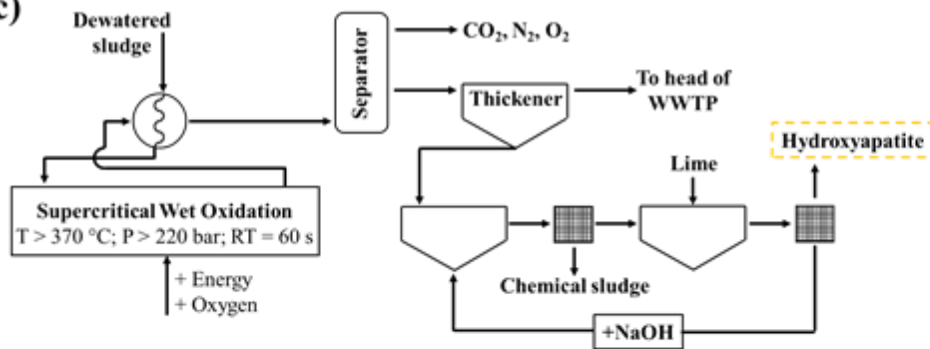


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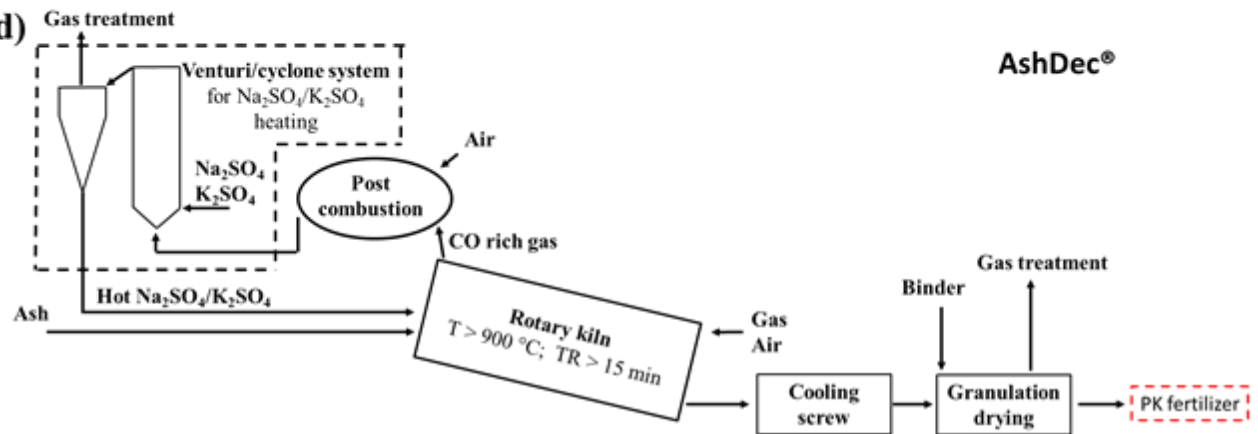
Ostara Pearl®

c)



Aqua Reci®

d)



AshDec®

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1508 **Figure 4. Schematics of phosphorus recovery in the WWTP sludge streamline (a) and of applicable patented technologies for**
 1509 **P recovery from digester centrate (b), dewatered sludge (c) and sludge ashes (d).**

1510 List of Tables

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1512 **Table 2. Full-scale applications of suspended biomass biological systems for phosphorus removal.**

Process configuration	Location	Flow rate (m ³ /d)	Feed P (mg/L)	C:P	Removal efficiency (%)	Reference
A ² O	Moscow (USA)	11,000	2.58-21.54	0-49.23 (mol/mol)	91.2-99.1	Coats et al. (2017)
MUCT	Madison (USA)	158,000	4-7	-	>75	Zilles et al. (2002a)
Orbal	Mazomanie (USA)	2,190	6.1-7.1	34-44 ^a	87.3	Zilles et al. (2002b)
A ² O	Thorneside (Australia)	7,500	11	50 ^b	72.7	Pijuan et al. (2008)
5-stage Bardenpho	(Australia)	12,000	10	24-36 ^a	>95	
A ² O	(China)	225,000	-	-	96-96.8	Wang et al. (2014)
Inverted A ² O	(China)	225,000	-	-	95.4-97	
MUCT	(Australia)	80,000	9.4	-	52	Beer et al. (2006)
3-stage Bardenpho	(Australia)	25,000	13.5	-	96	
Carousel/UCT	(Australia)	60,000	12	-	99.2	
3-stage Bardenpho	(Australia)	32,000	10	-	88	
MUCT	(Australia)	44,000	7.5	-	97.3	
Carousel/UCT	(Australia)	25,000	6	-	75	
MUCT	(Australia)	120,000	10	-	<80	
MUCT	(Australia)	30,000	12	-	98.3	
MUCT	(Australia)	60,000	12	-	83.3	
Side-stream RAS	Apex (USA)	19,700	7.1±0.1	39 ^a	94.4	
Side-stream RAS plus carbon	West Kelowna (Canada)	9,800	6.8±1	38.4 ^a	97.1	
Modified Johannesburg with SSM	Cedar Creek (USA)	13,600	2.7±5.4	102 ^a	67	
Johannesburg with UMIF	Henderson (USA)	11,400	5.7±0.8	46.5 ^a	91.6	
Carousel MUCT	Hardenberg (The Netherlands)	6,000	11±1	25 ^a	94.5	López-Vázquez et al. (2008)
Carousel MUCT	Deventer (The Netherlands)	8,800	7±1	33.3 ^a	87.1	

Carousel A ² O	Katwoude (The Netherlands)	8,200	9±2	25 ^a	95.6	
Carousel A ² O	Hoek van Holland (The Netherlands)	22,000	7±1	16.7 ^a	92.9	
Carousel A ² O	Venlo (Netherlands)	71,400	10±4	20 ^a	97	
A ² O	Waarde (The Netherlands)	10,800	6±3	25 ^a	90	
Sidestream P-stripping	Haarlem Waarderpolder (The Netherlands)	36,900	6±2	11.1 ^a	95	
5-stage step feeding	(Singapore)	361,000	5.53–6.52	5.8-11.2 (g/g)	80.7	
A ² O	(Singapore)	205,000	6.02–8.19	7.8-11.6	100	Qiu et al. (2019)
Inverted A ² O	(Singapore)	68,000	3.48–6.37	10.8-25	76.8	
A ² O	Lisbon (Portugal)	48,000	5±0.9	51 ^b	43	
A ² O	Setúbal (Portugal)	12,000	5±3	206 ^b	85	
A ² O	Hjørring (Denmark)	13,000	4±1	50 ^b	92	Lanham et al. (2013)
Biodenitro [®] configuration with side-stream RAS	Aalborg (Denmark)	49,000	10±5	63 ^b	94	
Biodenitro [®] configuration with side-stream RAS	Aalborg (Denmark)	17,000	10±6	40 ^b	96	
A ² O plus primary sludge fermentate	Hillsboro (USA)	24,628	3.7±0.5	68.6 ^b	82	
Side-stream RAS plus primary sludge fermentate (S2EBPR) with continuous mixing	Hillsboro (USA)	24,453	3.7±0.5	68.6 ^b	84	Wang et al. (2019)
Side-stream RAS plus primary sludge fermentate (S2EBPR) with intermittent mixing	Hillsboro (USA)	20,177	3.8±0.3	76.8 ^b	93	

1513 ^a as BOD₅/P

1514 ^b as COD/P

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Table 2. Biofilm-based applications for full-scale phosphorus removal.

Process configuration	Location	Population equivalents (flow rate)	Feed P (mg/L)	C:P	Removal efficiency (%)	Reference
AGS	Garmerwolde (The Netherlands)	(28,600 m ³ /d)	1.9-9.7	75 ^a	86.6	Pronk et al. (2015)
AGS	Lubawa (Poland)	15,000 PE (3,200 m ³ /d)	19.5±2.1	66.7 ^a	95	Świątczak and Cydzik-Kwiatkowska (2018)
AGS	Epe (Netherlands)	35,000 PE (5,000 m ³ /d)	8-13	101-117 ^a	98	Ortega et al. (2021)
AGS	Gansbaai (South Africa)	(4,000 m ³ /d)	19	66.6 ^a	82	Giesen et al. (2013)
AGS	Ryki (Poland)	38,600 PE (5,320 m ³ /d)	12.6	57.1 ^a	94.7	Giesen et al. (2016)
IFAS-EBPR	Denver (USA)	(17,280 m ³ /d)	4–16.5	8.4–30 ^b	>88	Onnis-Hayden et al. (2011)

IFAS-EBPR	(China)	(50,000 m ³ /d)	2.2–4.5	60.9 ^a	81	Bai et al. (2016)
IFAS-EBPR	Guri (South Korea)	(160,000 m ³ /d)	2.9	41.4 ^c	48.3	Kwon et al. (2010)
biofilm carriers - A ² O	Xi'an (China)	(48,000 m ³ /d)	8.27±2.8	55.2 ^a	97	Xiao et al. (2016)

1518 ^a as COD/P
1519 ^b as biodegradable COD/P
1520 ^c as BOD₅/P
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1529 **Table 3. Algae-based biological systems for phosphorus removal at full and pilot scale.**

Process configuration	Location	Flow rate (m ³ /d)	Feed P (mg/L)	C:P	Removal efficiency (%)	Reference
HRAP	Christchurch (New Zealand)	486-796	0.9-3.9 ^a	28-290 ^b	22-49	Sutherland et al. (2014)
HRAP	Cambridge (New Zealand)	1500	3.8–7.8 ^a	18.1 ^c	32.7-51.9	Sutherland et al. (2017)
HRAP	Rabat (Morocco)	60	7.45	14.8 ^d	66	El Hamouri (2009)
algal pond	Thessaloniki City (Greece)	33-48	3.06	45.5-73.1	26-65	Papadopoulos et al. (2014)
algal pond	Lawrence (USA)	4.7	3.2±0.9	-	<90.6	Sturm and Lamer (2011)

1530 ^a as dissolved reactive phosphorus
1531 ^b as VSS/P
1532 ^c as BOD₅/P
1533 ^d as COD/P

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1544 **Table 4. Full-scale (TLR 9) technologies for P recovery from solid/liquid WWTP streams (Corona Encinas, 2020; Desmidt et**
 1545 **al., 2012; Dewaele, 2015; Egle et al., 2016; Val del Río et al., 2016).**

Technology	Application	Type of reactor	Number of active plants	Annual costs (€/kg P recovered)	Product	Form of product	P recovery efficiency (%)
Crystalactor®	Wastewater/ supernatant (>25 mg P/L)	FBR	4	5-8	Struvite, CaP, MgP	Pellet	<40)
Ostara Pearl®	Digester and thickener supernatants	FBR	8	9-10	Crystal green®	Pellet (0.9-3 mm)	10-25
Multiform Harvest™	Digester supernatant	cone-shaped FBR	2	n.a.	Struvite	Pellet	n.a.
NuReSys®	Digestate, filtrate and centrate of WWTP	CSTR	7	1.31-3	Bio-Stru®	Pellet	n.a.
PHOSPAQ™	Municipal sludge reject liquors	CSTR	11	n.a.	Struvite	Pellet (0.7 mm)	n.a.
MagPrex®	Digestate	Air-lift	7	7-8	Struvite	Coarse- grained	10-15

1546 n.a. = not available.

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