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An Assessment of Options to Improve the Removal of Excess Nutrients from European Wastewater

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Abstract In this contribution we quantify the potential to reduce the discharge of excess nutrients with European wastewater, by modelling pollution, greenhouse gas emissions and the costs of measures under various scenarios. We analyse two types of strategies, namely extending the requirement of nutrient removal to broader areas and increasing the removal efficiency of nitrogen and phosphorus. As the former strategy usually entails some infrastructural investment while the latter entails an optimization of the existing processes, the two imply different costs, and increasing efficiency usually shows a better costbenefit ratio. In any case, actions increasing nutrient removal are expected to pay for themselves in terms of the benefits they bring in abating water pollution and (for nitrogen) greenhouse gas emissions. Nitrogen removal becomes even more attractive whenever

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D. Thornberg BIOFOS, Copenhagen, Denmark the infrastructural investments that we assume may be avoided, thanks to the possibility of an optimized use of the existing infrastructure. Phosphorus removal may become even more attractive with the progress of technologies for its recovery from wastewater and sludge, and with the increase of its demand for fertilizers and other industrial uses.

Keywords Wastewater treatment · Nutrient removal · Nutrient recovery · Nitrogen · Phosphorus · Europe

1 Introduction

Nutrients (nitrogen, N, and phosphorus, P) are discharged in excess to most European surface water bodies, and N from agricultural fertilisers often leaches to groundwater causing significant pollution (Grizzetti et al., 2021). Loads and flows of N and P in the European Union (EU)'s water bodies have been studied for long (Grizzetti et al., 2012, 2021; van Puijenbroek et al., 2019), highlighting that urban wastewater is a significant source, although often not the dominant one. While agricultural and other diffuse sources of N and P vary from year to year depending on the weather, emissions with wastewater are rather constant. Usually they represent about 25% of the total pollution load that reaches the seas from Europe in terms of N, and about 50% in terms of P (Grizzetti et al., 2021). Better nutrient management is clearly identified as a priority in the European Green Deal (European Commission, 2019), and particularly in the Farm to Fork Strategy (European Commission, 2020a), Biodiversity Strategy (European Commission, 2020b) and Zero Pollution (European Commission, 2021) policy initiatives. These propose that the EU and its Member States develop an Integrated Nutrient Management Plan (INMAP), in which a better management of nutrients from wastewater treatment plants (WWTPs) may play a significant role.

The removal of nutrients from sewage treatment plants has been required for thirty years in the EU according to the Urban Wastewater Treatment Directive 91/271/EEC (UWWTD). This sets an obligation to remove at least between 70 and 80% of total P and total N at WWTPs of capacity above 10,000 population equivalents (PE) when discharging in specifically designated "sensitive areas", with a maximum effluent concentration of 15 and 10 mg/L for total N, and 2 and 1 mg/L for total P, for WWTPs with a capacity below and above 100,000 PE, respectively, based on a 24-h composite sample.

Even when the UWWTD is properly implemented, though, many European water bodies still fail to achieve a good ecological status due to nutrient pressures (EEA, 2018; Grizzetti et may reduce the suspended so al., 2021; Nikolaidis et al., 2022). In order to reduce nutrient pollution, more stringent N and/or P removal requirements at WWTPs can be an easier option than addressing diffuse sources of pollution, because process control at a point source can be relatively quick and effective. On the contrary, reducing diffuse pollution usually entails distributed measures such as buffer strips or wetlands, typically implemented through a number of small-scale interventions, often on individual farms and with highly decentralized management, making the removal processes difficult to control.

In addition, the removal of nutrients at WWTPs may be also desirable from the perspective of the plant operator. For instance, denitrification is beneficial anyway as it improves sludge settleability (Metcalf & Eddy, 2014), reduces electricity consumption and greenhouse gas (GHG) emissions (Parravicini et al., 2022), and makes the removal of micropollutants more cost-effective (Pistocchi et al., 2022). The removal of P could be associated with the recovery of this critical raw material (EC, 2020a), and may reduce the suspended solids in the effluent thanks to the improved design of settlers it entails. Modern municipal sewage treatment plants in

proper operation usually remove more than 90% of total P and 80% of total N, with a potential to reach a removal close to 90% for N and above 95% for P in the best cases (DWA, 2020).

Nutrient removal requirements stricter than the UWWTD are already being set. For instance, in the German catchment area of lake Constance, plants larger than 40,000 PE must attain annual mean total P concentrations in effluents below 0.3 mg/L and P removal efficiency of 95% (BW, 2005). The German federal state of Rhineland-Palatinate has requested operators of WWTPs discharging in water bodies failing to reach a good ecological status to reduce limit values for total P mean annual concentration in effluents between 0.7 mg/L, for smaller plants, and 0.4 mg/L, for larger plants, and even below 0.2 mg/L in certain critical cases (Münch et al., 2020, Svenskt, 2016).

In this paper we examine the implications of tightening the requirements on nutrient removal from wastewater in the EU. After introducing the methods followed and data used in the paper, we quantify N and P discharges from the EU's WWTPs to the receiving water bodies under present conditions. Then we explore how we could reduce these discharges by increasing removal efficiency, extending the areas where removal is required, or a combination of the two. We quantify the reduction of N and P loads that we can achieve, the expected change in nutrient concentrations in the European stream network, the costs entailed and the balance of greenhouse gas emissions. Moreover, we tentatively quantify the associated benefits. Our results provide a basis for the appraisal of options for a possible revision of the regulation of N and P discharges in the EU.

2 Materials and Methods

Our analysis includes the following steps:

- A) Quantification of N and P loads from the EU's WWTPs under current conditions;
- B) Definition of policy scenarios and estimation of the reduction of loads emitted from WWTPs and conveyed to coastal waters, as well as concentrations in the stream network, under each scenario;

C) Quantification of the costs of implementing each scenario, and the benefits deriving from the reduction of loads.

The paragraphs below describe each step more in detail.

2.1 Quantification of N and P Loads from WWTPs

We use the database of WWTPs reported by the EU's Member States compliant with the UWWTD, made available by the European Environment Agency (EEA: https://www.eea.europa.eu/data-and-maps/data/water base-uwwtd-urban-waste-water-treatment-directive-7). In this contribution, we refer to the data of the 10th UWWTD Implementation Report (EC, 2020c),

reflecting data from 2016, as the most recent official data available at the time of performing the analysis presented here. The reported information includes spatial location, capacity and treated load (expressed as population equivalents, PE), whether the level of treatment is mechanical only (primary), biological without N or P removal (secondary), or biological with N or P removal (tertiary), and compliance with the emission limit values set in the UWWTD. With reference to the discharges of N and P from WWTPs above 2000 PE or anyway reported under the UWWTD, we calculate the cost and benefits of the different scenarios. In total, we account for about 521 million population equivalents (PE) from EU's WWTPs, of which roughly 80% are subject to N and P removal. Table 1 summarizes the PE in each country that undergo or do not undergo

Table 1 Breakdownof reported populationequivalents (PE) byEuropean country,depending on the level oftreatment (EC, 2020d)

Country	Not undergoing N removal (PE)	Undergoing N removal (PE)	Not undergoing P removal (PE)	Undergoing P removal (PE)
Austria	310,000	20,360,000	13,820	20,660,000
Belgium	229,200	8,986,000	340,400	8,875,000
Bulgaria	2,212,000	4,820,000	2,299,000	4,733,000
Cyprus	38,160	796,600	38,160	796,600
Czechia	344,300	8,430,000	536,300	8,238,000
Germany	1,070,000	109,700,000	2,259,000	108,500,000
Denmark	301,300	11,300,000	57,010	11,540,000
Estonia	16,820	1,564,000	8,156	1,573,000
Greece	397,000	10,300,000	7,866,000	2,834,000
Spain	32,670,000	31,510,000	28,070,000	36,110,000
Finland	2,470,000	2,587,000	-	5,057,000
France	10,480,000	62,010,000	14,010,000	58,480,000
Croatia	2,490,000	159,900	2,490,000	159,900
Hungary	2,378,000	9,271,000	2,640,000	9,009,000
Ireland	4,268,000	627,500	3,641,000	1,254,000
Italy	24,890,000	48,450,000	33,370,000	39,970,000
Lithuania	32,370	2,793,000	32,370	2,793,000
Luxembourg	46,040	589,800	42,600	593,200
Latvia	186,500	1,356,000	186,500	1,356,000
Malta	36,950	752,100	789,000	-
Netherlands	7,920	19,440,000	54,630	19,390,000
Poland	4,158,000	34,010,000	4,158,000	34,010,000
Portugal	10,320,000	1,920,000	10,690,000	1,551,000
Romania	1,038,000	11,680,000	1,042,000	11,680,000
Sweden	2,361,000	10,160,000	-	12,520,000
Slovenia	169,100	1,155,000	169,100	1,155,000
Slovakia	233,100	3,322,000	361,700	3,193,000
EU total	103,162,861	418,016,358	115,173,730	406,005,489

nutrient removal. Larger plants usually include N and P removal processes more often than smaller ones (Table 2), obviously in agreement with the requirements of the UWWTD. For each WWTP in Europe, the load in raw wastewater (sewage) is given by a plant's treated load (PE) times an emission factor. The discharge of N or P with WWTP effluents is calculated as:

$$D_{x} = \sum_{j=1}^{m} \left((1 - \eta_{Ix}) \delta_{I,j} + (1 - \eta_{IIx}) \delta_{II,j} + (1 - \eta_{IIIx}) \delta_{III,j} \right) \varepsilon_{x,j} P_{j}$$

where, for x = N or x = P and m = number of WWTPs in the EU:

- $\varepsilon_{x,j}$ is the emission factor for N or P at the jth WWTP, i.e. the average mass discharge by one PE
- η_{Ix}, η_{IIx}, η_{IIIx} are the removal efficiencies for N and P at primary, secondary or tertiary level of treatment, respectively,
- P_j the waste water load treated by the j-th WWTP in PE

- $\delta_{I,j}$, $\delta_{II,j}$, $\delta_{III,j}$ are Boolean variables equal to 1 if the j-the WWTP operates at primary, secondary or tertiary treatment level, respectively, and 0 otherwise, depending on the assumed scenario.

The emission factor is assumed to vary by country reflecting different lifestyles and diets, as shown in Table 3. On average, one European PE emits 11.18 g/ day of total N and 1.68 g/day of total P including detergents (Malagó & Bouraoui, 2021).

The removal efficiency for N and P is assumed to be constant across the EU for a given level of treatment, consistent with those used in previous EUscale assessments (Grizzetti et al., 2021; Pistocchi et al., 2019; Vigiak et al., 2020), Table 4. The implications of these assumptions are discussed later.

2.2 Options to Reduce Loads and Definition of Scenarios

We focus on the reduction of loads that can be achieved assuming all WWTPs are fully compliant with the UWWTD. Under a "full compliance"

Table 2 N and P removal in European WWTPs by plant size class (EC, 2020d)

Plant size (treated PE)	PE with N removal	PE with no N removal	PE with P removal	PE with no P removal	% Capacity with N removal	% Capacity with P removal
0–2000	1,268,380	2,307,259	791,130	2,784,509	35%	22%
2000-5000	11,816,644	11,193,191	9,557,144	13,452,692	51%	42%
5000-10000	18,880,258	10,872,774	16,791,199	12,961,834	63%	56%
10000-20000	35,674,447	6,503,558	34,551,243	7,626,761	85%	82%
20000-30000	27,182,653	5,052,482	26,299,062	5,936,073	84%	82%
30000-40000	23,515,110	3,905,684	22,562,118	4,858,676	86%	82%
40000-50000	18,711,177	3,183,398	17,951,866	3,942,709	85%	82%
50000-60000	15,743,627	2,083,538	15,299,337	2,527,828	88%	86%
60000-70000	15,480,348	2,524,316	15,014,674	2,989,990	86%	83%
70000-80000	12,700,257	1,870,350	12,331,099	2,239,509	87%	85%
80000-90000	11,071,105	1,456,513	10,654,518	1,873,099	88%	85%
90000-100000	11,211,960	1,405,093	10,933,299	1,683,753	89%	87%
100000-200000	61,957,066	14,374,656	62,107,557	14,224,164	81%	81%
200000-300000	31,203,952	7,539,420	32,295,637	6,447,736	81%	83%
300000-400000	23,100,146	4,840,318	23,822,636	4,117,828	83%	85%
400000-500000	9,694,590	3,555,088	9,209,967	4,039,711	73%	70%
500000-600000	11,502,141	3,808,400	11,434,569	3,875,973	75%	75%
600000-700000	5,790,740	1,356,693	5,790,740	1,356,693	81%	81%
700000-800000	8,926,821	4,484,180	10,387,309	3,023,692	67%	77%
800000-900000	5,866,127	1,683,915	6,676,922	873,120	78%	88%
900000-1000000	7,538,418	922,022	7,549,120	911,320	89%	89%
>1000000	49,180,391	8,240,013	43,994,344	13,426,060	86%	77%

 Table 3
 Emission factors for N and P assumed in this study, based on Malagó & Bouraoui, 2021

Country	N (g/year per capita)	P in excreta (g/year per capita)	P in detergents (g/ year per capita)
Austria	4443	515	95
Belgium	4033	471	82
Bulgaria	3399	430	100
Switzer- land	3923	445	11
Cyprus	3695	457	100
Czechia	3581	421	100
Germany	4235	490	100
Denmark	4653	516	100
Spain	4345	496	100
Estonia	4181	474	100
Finland	4785	541	100
France	4432	521	100
Greece	4232	510	100
Croatia	3623	431	100
Hungary	3511	420	100
Ireland	4498	530	100
Italy	4161	512	60
Lithuania	5066	590	100
Luxem- bourg	4514	514	95
Latvia	4177	491	100
Malta	4596	541	100
Nether- lands	4383	483	82.1
Poland	4189	503	100
Portugal	4596	522	100
Romania	4370	541	100
Slovakia	2803	339	100
Slovenia	3966	481	100
Sweden	4344	484	100

scenario, all WWTPs with a capacity of 2000 PE or more are supposed to have a mechanical and biological level of treatment and, if they discharge in a sensitive area and have a capacity higher than or equal to 10,000 PE, a more stringent treatment (removal of N,

Table 4 Assumed removal efficiency for N and P

	Removal with primary (mechanical) treatment	Removal with secondary (mechanical+biological) treatment	Removal with tertiary (more stringent) treat- ment
N	25%	55%	80%
Р	30%	60%	90%

P or both). A WWTP's spatial location enables classifying the waters where its effluents are discharged as a sensitive area for N, P or both, or a non-sensitive area. Sensitive areas are identified from the published maps available at the EEA (see Bouraoui et al., 2022).

We then consider two types of strategies, namely (1) the extension of more stringent (tertiary) treatment to WWTPs beyond sensitive areas, and (2) the increase of removal efficiency. For strategy (1) the upper limit in the extension of tertiary treatment to the whole territory of the EU, and to all WWTPs regulated by the UWWTD (i.e. with a treated load of 2000 PE or more). Indeed, some EU Member States have already extended the obligation of N and/or P removal to the whole of their territory, and sometimes even to plants treating loads < 10,000 PE.

Strategy (2) is constrained by the technical limitations on N and P removal efficiency. For N removal, while an efficiency of 80% is considered standard practice for a well-designed and well-operated plant, a higher efficiency may be difficult to achieve due to the need for sufficient carbon sources for denitrification, and difficult operating conditions e.g. due to winter temperature. Pragmatically, we regard 90% as an upper limit for the removal efficiency of N. Higher efficiencies might also entail the need to add external carbon sources, with a risk of disproportionately increasing the costs and climate impacts of wastewater treatment.

P removal is generally more flexible and technically viable than N removal, particularly when occurring through chemical precipitation. The latter uses precipitants (usually aluminium or iron salts, less often calcium hydroxide) whose cations react with the dissolved inorganic orthophosphate yielding insoluble particles eventually removed via sedimentation or filtration, typically after flocculation. Consequently, P removal requires a maximization of the conversion of dissolved phosphate into insoluble form, and its subsequent precipitation. The removal efficiency depends on the process design. We discuss the technical aspects of P removal processes, influencing the achievable removal efficiency, in more detail in the Annex. In most cases, we can achieve a 90% P removal efficiency at reasonable costs with a welldesigned process, while an efficiency higher than 95% requires specific appropriate action (see Annex).

Based on the above considerations, we identify a few scenarios, resulting from various combinations of the two strategies, as summarized in Table 5. For each scenario, we quantify the potential of reducing N and P loads.

2.3 Evaluation of Scenarios in the Broader Context of European Scale Nutrient Balances

Under each scenario, we assess changes in the total load discharged to European freshwater and marine coastal waters. The load to the sea includes the effect of (1) other sources of emission, notably agriculture and atmospheric deposition; (2) the natural attenuation in the stream network before the nutrients reach coastal waters. To account for these two aspects, we make use of the well-established GREEN model (Grizzetti et al., 2021).

The model covers a spatial extent including all river basins draining in European seas, and spanning 44 countries, of which 17 outside the EU. The analysis is performed at the level of connected irregular catchments with an average size of 7 km^2 , each corresponding to a stream segment. Lakes are included in the stream network and provide a specific attenuation of nutrients. The model calculates a steady state yearly mass balance of N and P, and can produce a pseudo-dynamic time series if applied in sequence to a time series of input variables.

Diffuse nutrient inputs to the river network were estimated by spatializing information on sources available at administrative (regional or national) level based on the Corine Land Cover and ESA CCI Land Cover time-series v2.0.7 (CLC, 2021). Point discharges of nutrients from domestic and industrial waste waters

were quantified following the approach of Vigiak et al. (2020) updated with the latest data reported by Member States under the UWWTD (European Commission, 2020d). Annual precipitation, irrigation and water flow are used to describe attenuation and dilution of nutrients in the catchments. The hydrological information was retrieved from the LISFLOOD model (Gelati et al., 2020). The GREEN model was calibrated by marine regions, to account for specific biogeographical

condition, using monitoring data of total N and P available in the EEA WaterBase (https://www.eea.europa. eu/data-and-maps/data/waterbase-water-quality-2) for the period 1990–2018. All details on the model input and calibration are provided in Vigiak et al., 2022.

2.4 Costs and Benefits

The costs of N and P removal are estimated using the expenditure functions of the OECD's FEASIBLE model (COWI, 2010; OECD, 2004). Accordingly, the base-10 logarithm of investment cost per PE of biological (or "secondary") treatment for carbon removal only is $log(C_{sec}) = 3.38 - 0.2632log(Pop)$, where Pop is the population equivalents served by the plant. C_{sec} is constrained to not fall below 115 Euro/PE. The base-10 logarithm of investment cost per PE of secondary treatment for nitrogen $\log(C_{sec,N}) = 3.62 - 0.2612\log(Pop).$ removal is $C_{sec,N}$ is constrained to not fall below 207 Euro/ PE. The base-10 logarithm of investment cost per PE of secondary treatment with P removal is $log(C_{sec,P}) = 3.54 - 0.2808log(Pop)$. The base-10

Table 5 Scenarios analysed in this paper. Keys to short descriptors: "Whole" indicates extension of N or P removal to the whole territory of the EU; "eff" indicates an increase in the

minimum required removal efficiency; "> = 2000 PE" indicates that the provisions are applied to all WWTPs from 2000 PE on

Scenario code	Nutrient addressed	Short descriptor	Strategy	More stringent treatment required in	For plants with capacity (PE)> =	Removal efficiency
PS1	N, P	Full compliance	_	Sensitive areas for N and P	10,000	80% (N), 90% (P)
PS2	Ν	eff	2	Sensitive areas for N	10,000	90%
	Р	eff	2	Sensitive areas for P	10,000	90%
PS3	Ν	Whole	1	Whole EU	10,000	80%
	Р	Whole	1	Whole EU	10,000	90%
PS4	Ν	Whole + eff	1 + 2	Whole EU	10,000	90%
	Р	Whole + eff	1 + 2	Whole EU	10,000	95%
PS5	Ν	Whole + eff, $> = 2000 \text{ PE}$	1+2	Whole EU	2,000	90%
	Р	Whole + eff, $> = 2000 \text{ PE}$	1+2	Whole EU	2,000	95%

logarithm of investment cost per PE of N and P removal is $log(C_{sec.N,P}) = 3.72 - 0.2722log(Pop)$.

Using these equations, we define the additional cost per PE of P removal, C_P , as the average of $(C_{sec,P} - C_{sec})$ and $(C_{sec,N,P} - C_{sec,N})$. The plot of log (C_P) as a function of log(Pop) can be very well approximated by the ordinary least squares (OLS) best fit line:

 $\log(C_p) = 3.18 - 0.3642\log(Pop).$

 C_P is constrained to not fall below 23 Euro/PE. We assume the investment cost per PE to upgrade a secondary plant to a tertiary treatment with N removal to equal half of the costs of a new secondary plant, added to the cost difference between a secondary and an N removal plant:

$$C_N = C_{sec,N} - 0.5C_{sec}.$$

This is always higher than the differential cost, $C_{sec,N} - C_{sec}$, but lower than the cost of a new plant, $C_{sec,N}$, empirically reflecting the fact that upgrading a plant for N removal is more expensive than designing it for N removal from the beginning. However, various parts of the plant can be kept unchanged, allowing significant savings compared to a new plant. The total costs, including investment and operation and maintenance, are calculated on the basis of additional assumptions (Table 6), representative of current European conditions. The total cost per PE including investment and operation is estimated as:

$$TC_{x} = C_{x} \left(\frac{1}{pva(n,r)} + \omega\right) + \varepsilon \left(E_{3} - E_{2}\right)$$

Where x = N or P, ω is the annual operation and maintenance cost as a fraction of the investment cost, ε is the cost of energy, E_2 and E_3 the annual energy demand per PE in secondary and tertiary treatment respectively, and the present value of annuities is:

$$pva(n,r) = \frac{1 - \left(\frac{1}{(1+r)}\right)^n}{r}$$

with r = discount rate and n = years of the investment's lifetime. The values of the parameters used in our calculation are shown in Table 6.

When increasing P removal efficiency, we assume an incremental cost equal to 10% of the total cost of

 Table 6
 Additional assumptions on costs and benefits

Item	Value	Units
Cost of energy	0.10	€/kWh
Energy demand for secondary treatment	25	kWh/PE/year
Energy demand for tertiary treatment	40	kWh/PE/year
Discount rate	2.5%	-
Lifetime of investments	30	years
Value of avoided GHG emissions	90.00	€/t CO2e
Shadow price of removed N	20.00	€/kg
Shadow price of removed P	30.00	€/kg

P removal only, reflecting an increase in use of metal salts for precipitation and minor adjustments to the process. The total cost of increasing P removal efficiency is given by:

$$C_{P,eff} = 0.1 \left(C_P \left(\frac{1}{pva(n,r)} + \omega \right) + \varepsilon \left(E_3 - E_2 \right) \right)$$

The costs of increasing N removal efficiency owe to a better operation of the removal process, possibly including e.g. the reuse of organic carbon from the primary settler, and an adjustment of the treatment processes for denitrification e.g. through instrumentation, control and automation (ICA). Unlike for P, this may entail a more substantial revision of the process. Therefore we assume a total cost for increased N removal efficiency equal to 10% of the total cost of a biological plant for the removal of N:

$$C_{N,eff} = 0.1 \left(C_{sec,N} \left(\frac{1}{pva(n,r)} + \omega \right) + \varepsilon E_3 \right)$$

The costs of N and P removal need to be compared with the benefits under the various scenarios. Benefits considered here include the value of improved water quality as a consequence of N and P removal, and the value of the avoided greenhouse gas (GHG) emissions.

The value of improved water quality is quantified through an assumed shadow price for pollution in line with the approach suggested in UNEP, 2015 (Table 6). GHG emissions for N and P removal scenarios are quantified with the approach presented in Parravicini et al., 2022. The assumed shadow prices and value of avoided GHG emissions are summarized in Table 6.

A third, potentially relevant benefit is related to the recovery of nutrients from wastewater and sludge.

While sludge application in agriculture is also a potential way to recover N and P, some EU countries are already restricting this practice based on concerns for their content in metals and other pollutants. This makes alternative approaches to recover nutrients attractive.

The recovery of N from wastewater is problematic. Higher N removal reduces the potential for recovery, as nitrification and denitrification turn ammonia and nitrate to gaseous N_2 or N_2O . At present, the best option to recover the fertilizing value of N in wastewater is to limit N removal in wastewater treatment, and reuse the effluent for agricultural fertilization-irrigation. This option could be valid in some cases, but not when effluents could contaminate surface- and groundwater.

The recovery of P, on the other hand, is technically feasible and can help reduce the demand of mineral P usually sourced from phosphate rocks, with significant benefits in terms of avoided impacts of mining, transport and processing of mineral P fertilizers. Although the current recovery processes entail relatively high costs, in the long term P recovery may yield potential savings and mitigate geopolitical risks in the global supply chains. P removal could be in principle designed in order to enable some recovery of P as well. However, the technologies for simultaneous P removal and recovery are not yet fully marketready. Increased P removal necessarily leads to more P retained in the plant (as sludge or precipitated crystals), but its recovery is still quite problematic. We present a more technical discussion of P recovery in the Annex. As the benefits of nutrient recovery are largely site-specific, in this exercise we deliberately ignore them.

It is worth stressing that our costs and benefits are assessed on the basis of expenditure models and shadow prices from the years 2010–2015, with the exception of the shadow price of GHG emissions. These assumptions could be justified until short ago by the simultaneous conditions of low inflation, moderate technical progress and slow rise in awareness about the degradation of the environment. In recent times, a series of geopolitical and economic shocks, including the COVID-19 pandemic and the war in Ukraine, have caused a sizable increase in the price of raw materials, industrial products and energy, making our costs too low. At the same time, the urgency of climate change mitigation and pollution control may have raised the awareness of the value of wastewater treatment, which could reflect in higher shadow prices. Moreover, the trend towards higher temperatures and technological developments could in principle increase the removal efficiency of nutrients at lower costs than assumed in our model. Given the uncertainty entailed, and considering that our assessment aims at a comparison of scenarios and not at an absolute economic quantification, we decided to maintain the costs and shadow prices as described above.

3 Results

We quantify the loads of N and P discharged to European water under current condition (referred to the year 2016) and different scenarios of nutrient reduction (Table 5). These include full compliance with the UWWTD, increase of nutrient removal efficiency ("eff"), extension of the sensitive areas ("Whole"), and simultaneous increase of removal efficiency and sensitive areas for agglomerations with different capacity ("Whole +eff" if applied to plants above 10000 PE, and "Whole + eff, >=2000 PE" if applied to all plants above 2000 PE) (Fig. 1). Both N and P emissions are apparently progressively reduced, with the most stringent measures (scenario "Whole + eff, >=2000 PE") almost halving nutrient discharges from WWTPs to surface water.

Point source emissions from UWWTPs can represent an important share of nutrient load in surface waters. This holds particularly in regions that are densely populated and/or where the wastewater treatment level is still inadequate (Fig. 2). Generally, the contribution of point sources from UWWTPs discharges to the total load in surface water is more important for P than for N, since the latter is more mobile and a large part of total N load originates from diffuse agricultural sources (Fig. 2). Consequently, the effect of measures of the different scenarios on the total nutrient load discharged to the European sea varies depending on the marine region considered (Fig. 3). The full implementation of the UWWTD would produce a slight decrease of nutrient load to the sea, with a possible reduction of about 1-2% for N load and between 2-8% for P load. Nutrient loads to the sea are reduced by the additional measures under the various scenarios.

Fig. 1 Loads of nitrogen (above) and phosphorus (below) in Europe, under baseline, full compliance and N, P removal scenarios. The loads from urban WWTPs (UWWTPs) are reported along with those from individual household and other appropriate treatment systems (IAS), those of the population in smaller agglomerations (below the threshold of 2000 PE for reporting under the UWWTD- "unreported population") and, for the current conditions, those from agglomerations above 2000 PE not yet treated.





The most ambitious measures (scenario "Whole + eff, >=2000 PE") could lead to a significant decrease of N load in the Bay of Biscay and Iberian Coast (-11%), Baltic Sea (-8%), Black Sea (-7%), North Sea (-6%), and Mediterranean Sea (between -2% and -22%). Similarly, they could considerably reduce P load to the different seas: Bay of Biscay and Iberian Coast (-16%), Baltic Sea (-11%), Black Sea (-14%), North Sea (-15%), and Mediterranean Sea (between -3% and -50%) (Fig. 3). While the effect of the measures on the total nutrient load to the sea is sizable, the estimated change in

concentrations of N and P in the stream network is less evident (Fig. 4): overall, the model estimated a 2% increase of the stream network length with N concentration below 2 mg N/L, and a 4% increase of the stream network length with P concentration below 0.1 mg P/L (Fig. 4).

The removal of N and P (reduction of discharges compared to the full compliance scenario) can be plotted for all scenarios (Table 5) as a function of the corresponding costs, estimated under the assumptions made above. The results of this cost-effectiveness analysis are shown in Fig. 5 for the



Fig. 2 Share of the total nutrient load conveyed in surface waters accounted for by point source (PS) emissions (including WWTPs): N (above) and P (below)

Fig. 3 Total nitrogen (above) and phosphorus (below) load by European Marine Region estimated by the model GREEN (average 5-years period 2014-2018) under current conditions and the different scenarios of domestic waste water emissions. ABI=Bay of Biscay and Iberian Coast; ACS = Celtic Seas; ANS = Greater North Sea; BAL=Baltic Sea; BLK=Black Sea; BLM = Black Sea and Sea of Marmara; MAD = Adriatic Sea; MAL = Aegean Levantine Mediterranean Sea; MIC = Ionian Sea and Central Mediterranean Sea; MWE = Western Mediterranean Sea





whole EU, and in Tables 7 and 8 for the individual countries. In the following, we refer to the short description of scenarios as per Table 5.

At the EU scale, for N the scenario of increased removal efficiency ("eff") is more cost-effective than the scenario of extending removal requirements to the whole territory ("whole"), largely also due to the fact that most of the larger plants have already N removal in place. The cost-effectiveness of the combined scenario if increasing efficiency while extending the requirements ("Whole + eff") is the second highest, while the additional costs entailed by extending removal to plants above 2000 PE are usually less than proportionate to the additional N removal. However, in countries (such as Spain) with relatively few plants performing N removal under full compliance, the extension of removal requirements to the whole territory may yield substantially higher removal of N than requiring higher removal efficiency.

A similar pattern appears also when considering P removal. However, in this case the "eff" and "whole" scenarios are comparable due to the relatively small increase of P removal efficiency that is still possible (95 vs 90%). It is worth noting that extending P removal to smaller plants does not reduce the cost-effectiveness of the measure, due to the relative scalability of the process.

The removal of N delivers potential benefits also when it comes to GHG emissions, while the additional chemicals required for enhanced P removal embed only small GHG emissions in their life cycle (Parravicini et al., 2022). We have used the approach described in detail in Parravicini et al., 2022, to simulate the GHG emissions when N and P removal are extended to broader regions and made more efficient, according to the above scenarios. Figure 6 shows the results of the simulation, highlighting how emissions decrease for all countries under all scenarios of Table 5 for N removal, while



Fig. 4 Share of total river network length in different classes of nitrogen (above) and phosphorus (below) concentration, estimated by the model GREEN (average 5-years period 2014–2018), under current and two scenarios of domestic waste emission (PS1 and PS5). Nitrogen concentration classes: low (<2 mg N/L), medium (2–5 mg N/L), high (>=5 mg N/L). Phosphorus concentration classes: low (<0.1 mg P/L), medium (0.1–0.5 mg P/L), high (>=0.5 mg P/L). (GREEN model extent, including all marine regions except Barents, Norwegian and White Sea)

increases in emissions under all scenarios for P removal are negligible.

Table 9 shows the costs of removing one kg of N or P, depending on the scenarios considered. An increase of N removal efficiency from 80 to 90% yields a reduction of N loads at significantly lower cost than replacing sensitive areas with a "whole territory" approach for the whole EU. Still, combining an increased removal efficiency with a "whole territory" approach delivers a significantly higher reduction of N loads. The "whole territory" approach alone is less cost-effective, and the extension of N removal to plants between 2000 and 10000 PE entails costs disproportionate to the additional removal of N. Assuming again a value of 90 Euro/t CO2e, 20 Euro/ kg N and 30 Euro/kg P (UNEP, 2015), we calculate the benefits for the scenarios considered (Table 9). The costs of N and P removal are always lower than the expected benefits, as captured by the assumed shadow prices of N, P removal and GHG emission reductions.

4 Discussion and Conclusions

We have shown that the extension of N and P removal requirements to the whole territory of the EU, and the increase of N and P removal efficiency yield a benefit/ cost ratio consistently higher than 1 for all scenarios (Table 9).



Fig. 5 The potential

reduction of N and P loads

as a function of costs for

the whole of the EU. Left

y-axis: P; right y-axis: N

Table 7	Additional annu	al costs and N re	emoval under the sc	cenarios of Table 5				
	Costs				Removal			
Country	N, whole	N, eff	N, whole + eff	N, whole + eff., > = 2000 PE	N removal t/y: N, whole	N removal t/y: N, eff	N removal t/y: N, whole + eff	N removal ty : N, whole + eff., > = 2000 PE
AT	E -	€ 41,656,064	€ 41,656,064	€ 56,221,207	1	8,465	8,465	9,524
BE	e -	$\in 18,931,102$	$\in 18,931,102$	ϵ 28,160,791	I	3,307	3,307	3,913
BG	$\in 26,867,076$	$\in 10,327,874$	$\in 41,258,694$	$\in 53,787,648$	1,509	1,609	3,722	4,216
CY	€ 305,922	$\in 1,668,599$	$\in 2,019,801$	€ 2,607,521	14	294	314	340
CZ	e -	$\in 16,375,741$	$\in 16,375,741$	$\in 30,486,648$	I	2,552	2,552	3,349
DE	e -	$\in 228, 516, 783$	$\in 228, 516, 783$	$\in 295, 299, 202$	I	42,807	42,807	47,953
DK	e -	$\in 23, 327, 173$	$\in 23,327,173$	€ 34,639,068	I	4,990	4,990	5,747
EE	E -	$\in 3,144,445$	$\in 3,144,445$	€ 3,930,026	I	613	613	670
EL	$\in 3,808,066$	$\in 21, 150, 364$	$\in 25,540,238$	€ 31,289,624	270	4,191	4,568	4,945
ES	ϵ 389,342,520	ϵ 64,294,851	$\in 515, 155, 891$	€ 637,099,803	31,283	12,616	56,413	63,035
FI	$\in 29,518,813$	$\epsilon 5,499,292$	$\in 39,964,187$	e 49,714,582	2,561	1,230	4,814	5,376
FR	$\in 122,886,964$	$\in 119, 144, 812$	€ 260,925,248	€ 347,289,487	9,461	23,610	36,855	43,168
HR	$\in 37,011,001$	ϵ 355,399	ϵ 42,947,213	$\in 46,993,198$	2,132	52	3,036	3,208
НU	ϵ 30,355,733	$\in 17,973,872$	ϵ 52,933,973	€ 67,551,189	1,759	2,811	5,275	6,086
Ε	ϵ 53,927,747	$\in 1,445,707$	ϵ 63,677,467	€ 76,959,045	4,294	262	6,273	6,972
TT	$\in 273,526,808$	ϵ 99,979,621	$\in 415, 141, 030$	€ 592,492,315	19,291	18,517	45,525	54,720
LT	E -	$\epsilon 5,762,411$	$\in 5,762,411$	€ 6,985,381	I	1,356	1,356	1,457
ΓΩ	e -	$\in 1,257,651$	€ 1,257,651	€ 2,837,891	I	253	253	335
LV	e -	ϵ 3,035,430	$\in 3,035,430$	€ 7,733,074	I	563	563	786
MT	ϵ 611,831	$\in 1,527,175$	$\in 2,230,898$	€ 2,230,898	42	346	405	405
NL	e -	$\in 41, 111, 675$	$\in 41, 111, 675$	$\in 43,038,718$	I	8,311	8,311	8,529
PL	e -	ϵ 74,559,159	\in 74,559,159	$\in 202, 301, 578$	I	14,009	14,009	20,158
PT	$\in 130,947,614$	$\in 3,794,109$	$\in 154,655,170$	€ 190,166,755	10,573	767	15,569	17,459
RO	e -	$\in 24,673,629$	$\in 24,673,629$	$\in 48,548,869$	I	4,999	4,999	6,229
SE	$\in 18,609,694$	$\in 21,853,665$	$\in 43,550,701$	€ 75,272,851	1,391	4,343	6,290	7,989
SI	ϵ 430,868	ϵ 2,363,478	$\in 2,858,560$	€ 7,389,773	23	397	430	660
SK	E -	$\in 6,971,762$	€ 6,971,762	€ 13,781,110	I	840	840	1,088
EU total	1,118,150,656	860,701,842	2,152,182,093	2,954,808,251	84,603	164,110	282,554	328,315

	Costs				Removal			
Country	P, whole	P, eff	P, whole + eff	P, whole + eff.,> = 2000 PE	P removal t/y: P, whole	P removal t/y: P, eff	P removal t/y: P, whole + eff	P removal ty: P, whole+eff.,>=2000 PE
AT	e -	ϵ 6,803,625	ϵ 6,803,625	ϵ 8,030,763	. 1	581	581	686
BE	e -	$\in 3,092,686$	ϵ 3,092,686	€ 5,716,858	I	227	227	288
BG	€ 6,873,357	€ 1,652,187	ϵ 9,225,858	$\in 12,242,355$	296	123	468	529
CY	€ 74,769	$\in 271,968$	ϵ 354,214	ϵ 497,961	2	22	25	29
CZ	E -	€ 2,677,056	$\in 2,677,056$	$\epsilon 6,644,095$	1	186	186	270
DE	e -	$\in 37, 342, 813$	ϵ 37,342,813	$\in 57,609,712$	I	2,983	2,983	3,449
DK	e -	$\in 3,810,650$	$\in 3,810,650$	$\in 4,821,516$	1	330	330	413
EE	E -	€ 514,565	$\in 514,565$	$\epsilon \ 629,545$	1	42	42	47
EL	€ 14,662,285	$\in 983,760$	$\in 18,214,808$	$\in 20, 174, 256$	1,389	78	1,698	398
ES	\in 77,063,079	$\in 12, 167, 926$	ϵ 97,630,454	$\in 129,510,346$	4,202	1,024	5,927	7,717
FI	e -	$\in 1,705,334$	$\in 1,705,334$	$\in 1,949,346$	I	151	151	637
FR	$\in 32,889,590$	€ 18,910,882	$\in 55,445,084$	$\in 84,985,388$	1,829	1,616	3,749	4,144
HR	€ 9,113,292	€ 57,739	$\in 10,082,360$	$\in 11,056,205$	375	4	442	466
НU	\in 7,950,004	$\in 2,849,119$	$\in 11,642,293$	$\in 14,870,483$	352	201	612	663
E	$\in 11,751,800$	ϵ 402,031	$\in 13,347,431$	$\in 14,837,078$	654	30	793	957
IT	€ 81,785,296	€ 13,874,766	$\in 104,918,827$	$\in 157, 452, 720$	4,268	1,092	6,071	6,141
LT	E -	$\in 941,800$	ϵ 941,800	$\in 1,208,637$	I	92	92	102
ΓΩ	E -	$\in 204,863$	ϵ 204,863	ϵ 559,504	I	17	17	27
LV	e -	$\in 496,022$	ϵ 496,022	$\in 1,633,055$	1	40	40	71
MT	$\in 1,517,806$	E -	$\in 1,782,400$	$\in 1,782,400$	152	I	177	32
NL	E -	$\in 6,708,302$	$\in 6,708,302$	€ 7,288,238	I	536	536	551
PL	e -	$\in 12, 185, 401$	$\in 12,185,401$	$\in 43,018,847$	I	1,008	1,008	1,877
PT	$\in 33,112,949$	ϵ 515,547	$\in 36,986,366$	ϵ 46,113,621	1,765	44	2,103	2,304
RO	E -	$\in 4,032,762$	$\in 4,032,762$	$\in 9,766,155$	1	367	367	540
SE	e -	ϵ 4,076,459	ϵ 4,076,459	ϵ 4,897,978	I	329	329	778
SI	$\in 104,893$	ϵ 386,736	\in 502,118	$\in 1,566,958$	4	29	34	63
SK	e -	$\in 1, 139, 441$	$\in 1,139,441$	ϵ 3,239,623	I	66	99	98
EU total	276,899,121	137,804,439	445,863,992	652,103,641	15,288	11,218	29,054	33,279



Fig. 6 GHG emissions under the scenarios of Table 5

If we could implement N and P removal in the whole territory of the EU, with a higher removal efficiency than stipulated by the current UWWTD, we would be able to reduce substantially the emissions of N and P corresponding to full compliance. Figure 7, based on the estimated removal of Tables 7 and 8, highlights the marginal shares of the total emissions of N and P that could be eliminated by setting increasingly ambitious requirements for wastewater treatment. The maximum possible reduction would be of more than 60% for N and more than 70% for P. It is worth noting that the removal of N and P with primary treatment may be lower than we assume, and around 10% or less in many practical circumstances. In the absence of enhanced biological processes or chemical precipitation, secondary treatment causes a 30% additional removal of P, making the total removal closer to 40% than to 60%. N removal in secondary treatment can be also lower than we assume. Our assumption of a higher secondary treatment removal efficiency implies an underestimation the benefits of expanding nutrient removal, which is safeside in the appraisal of policy options presented here.

While it is likely that the measures yielding the highest marginal reductions of N and P discharges are always cost-effective, measures yielding a marginally decreasing improvement should be considered more critically. For N, increasing removal efficiency is more cost-effective than extending removal requirements, at lower efficiency, to the whole territory. This finding rests on the assumed cost pattern, whereby increasing removal efficiency entails optimization of the existing processes, while adding N removal where it was not initially present entails also infrastructural development. The former is supposed to come at substantially lower costs than the latter, making efficiency a better strategy, all the rest being equal. Obviously, whenever a plant can be upgraded to N removal without a significant infrastructural overhaul, the two strategies become comparable, making N removal scenarios even more cost-effective.

The case of P is slightly different. As the assumed initial P removal efficiency is already quite high, the advantage of an efficiency strategy is less apparent compared to a strategy of extending removal to the whole territory. An important aspect to address when considering P removal is the possibility to recover P from wastewater or from sludge, as a higher removal may imply a potentially higher recovery as well. P recovery is not common yet in European WWTPs, but is likely to become more and more common also because of stricter regulations being introduced, e.g. in Germany, which in turn trigger technological developments. The economic valorisation of P recovery would further increase the benefit to cost ratio of more stringent P removal.

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GHG emissions.								
	N, whole	N, eff	N, whole+eff	N, whole $+ eff., > = 2000 PE$	P, whole	P, eff	P, whole + eff	P, whole $+$ eff., $> = 2000$ PE
Annual costs	$\in 1,118,150,656$	ϵ 860,701,842	ϵ 2,152,182,093	ϵ 2,954,808,251	ϵ 276,899,121	£137,804,439	€ 445,863,992	ϵ 652,103,641
Removal t/y	84,603	164,110	282,554	328,315	15,288	11,218	29,054	33,279
Benefits from N, P removal	$\in 1,692,066,499$	$\in 3,282,194,366$	$\in 5,651,087,466$	$\in 6,566,292,253$	$\in 458, 641, 200$	ϵ 336,549,007	ϵ 871,630,408	$\in 998, 378, 810$
Cost of removal for 1 kg	€13.22	€ 5.24	€ 7.62	£ 9.00	€ 18.11	€ 12.28	€ 15.35	£ 19.59
Avoided GHG t/year	2,825,764	4,055,120	8,417,005	9,377,841	-347,861	-106,623	-479,226	-509,002
Benefits from GHG emis- sion reduction	$\in 254, 318, 724$	€ 364,960,829	€ 757,530,518	€ 844,005,765	-€ 31,307,557	-€ 9,596,037	-€ 43,130,370	-€ 45,810,263
B/C	1.74	4.24	2.98	2.51	1.54	2.37	1.86	1.46



Fig. 7 Marginal removal of N (below) and P (above) as a consequence of incrementally stringent regulation of WWTPs

Process	P concentration in effluents (mg/L)	Source	Notes
Pre-precipitation	≤2	Baumann, 2002	grab sample
Simultaneous precipitation	<1	Barjenbruch & Geyer, 2016	
Post-precipitation with sedimentation	≤0,5	Baumann, 2002	grab sample
Two-point precipitation	≤0,5	Barjenbruch & Exner, 2009	
EBPR	< 0,3–0,5	Sabelfeld & Geißen, 2011	with simultaneous precipitation
Membrane process	< 0,3	Pinnekamp & Friedrich, 2003	with simultaneous precipitation
Electrophosphate precipitation	0,29–0,35	Genovese & Hauptmann, 2018	with additional two-point precipitation (pre- and post precipitation)
Flocculation filtration	≤0,5	Barjenbruch & Exner, 2009	
Flocculation filtration	≤0,3	Baumann, 2002	with simultaneous precipitation and flocculation filtration
Flocculation filtration	< 0,2	DWA, 2016	flocculation filtration with fabric filter or continously operated sandfilter in combination with simultaneous precipitation
Flocculation/filtration	< 0,1	Theilen, 2015	
Electrocoagulation and Electroflotation	0,045–0,3	Feng et al, 2003	
Electrocoagulation and Electroflotation	2,04	Tran et al, 2012	
Microalgae	< 0,5	Wawilow & Theilen, 2017	with simultaneous precipitation and EBPR
SuPaPhos	0,05	Drenkova-Tuhtan et al., 2017	New technology, only tested at labora- tory and pilot scale

Table 10 Achievable P concentrations in the WWTP effluents depending on the P removal process adopted

recovered to a significant extent from municipal wastewater and sewage sludge, or the ashes of sewage sludge after incineration separate from other waste ("mono-incineration"). However, P recovery is not common practice, and is only legally required by a few countries such as Germany starting from 2029 (Sewage Sludge Ordinance AbfKlärV, 2017) and Switzerland from 2026 (Ordinance on the Avoidance and Disposal of Waste VVEA, 2016). P can be recovered from wastewater, sewage sludge or ash and recycled. The regulations do not impose a specific technology. P compounds already in particulate form are removed with the solids in the primary or excess sludge. Dissolved P removal from wastewater occurs through chemical precipitation or through incorporation into the biomass (enhanced biological P removal, EBPR).

EBPR exploits the ability of specific bacteria in the bioreactor to store polyphosphates under appropriate operating conditions. The process requires an alternation of anaerobic and aerobic or anoxic conditions, as well as a sufficient amount of biodegradable organic carbon in the anaerobic stage, where it is anabolized with a consumption of polyphosphates (ATP) releasing phosphate. German design standards (DWA A 131, 2016) require a hydraulic retention time (HRT) in the anaerobic stage of 0.5–0.75 h considering the maximum dry weather flow and recirculation of the sludge.

In a following stage, the organic carbon is catabolized by P-accumulating organisms (PAOs) with a synthesis of polyphosphates taking up phosphate. While some PAOs are strictly aerobic, others are compatible also with the anoxic conditions during denitrification. A net elimination of phosphate from wastewater occurs because uptake is higher than release, with P content in the bacterial cells rising from 1–2% up to 10–15% (Lopez-Vazquez et al., 2020). The P accumulated in cells is removed with the excess sludge. The process requires about 500 mg/L of chemical oxygen demand (COD) to remove 10 to 12 mg/L P (Lopez-Vazquez et al., 2020). EBPR is attractive because it avoids the chemicals needed for precipitation, hence it does not alter the saline content of the wastewater. Moreover, it tends to produce smaller amounts of excess sludge. P eliminated through EBPR can be easier to recover than P bound to iron or aluminium as resulting from chemical precipitation However, it may struggle to meet the UWWTD regulatory standards of 1 to 2 mg/L P in the effluents, especially when cold temperatures limit bacterial activity or when a high dilution of the influent sewage due to rain events reduces COD availability. The

Overall, our analysis shows that more stringent and widespread removal of N and P could pay for themselves in terms of the benefits they bring. The main effect of N and P discharge reduction through higher removal at WWTPs is anticipated to be in the loads conveyed to coastal waters, while concentrations in the stream network are not expected to improve very significantly even under the most ambitious removal scenarios. The cost-benefit ratio of P removal would improve further if we could develop technologies for simultaneous removal and recovery. The cost-benefit ratio of N removal, which we assume to depend more strongly on the needs to upgrade the infrastructure, could be much more favourable in several circumstances where denitrification could be achieved in existing plants through non-infrastructural actions, e.g. based on instrumentation, control and automation or on a better use of the existing processes and infrastructure.

Data Availability Data used in this work will be made available upon reasonable request to the corresponding author.

Declarations

Conflict of Interest The authors declare that they have no conflict of interest.

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Annex

A more Technical Discussion of P Removal and Recovery Options

Phosphorus (P) is considered a critical raw material for the EU (EC, 2020d). In principle, it can be process may be improved through control of oxygen and nitrates in the anaerobic stage, enhancement of organic carbon availability, longer HRT in the anaerobic phase, and optimization of the SRT of the aerobic or anoxic phase to stabilize the presence of PAOs (Lopez-Vazquez et al., 2020) while enabling nitrification and denitrification. Even after these measures, though, EBPR may require an additional chemical precipitation stage (DWA A131, 2016). The latter is typically obtained by adding chlorides or sulphates of trivalent Al or Fe. These dissociate and the metal reacts with phosphate ions, forming insoluble Al- or Fe-phosphates. Because of competing acid hydrolysis, the metals also form hydroxydes, so that chlorides or sulphates must be added in excess of the stoichiometric ratio with phosphate. Precipitation occurs in combination with flocculation, enhanced by hydroxide formation from the acid hydrolysis of Fe and Al. The formation of aluminum hydroxide, in particular, may be an essential intermediate step in the precipitation reaction (Bashan & Bashan, 2004). Besides some acidification, the process releases sulphate and chloride ions increasing the saline content of effluents. Bever and Teichmann (1990) estimated a 50 mg/L increase in chloride concentration for a P removal of 10 mg/L with average use of precipitants. Such concentrations may negatively affect the biological wastewater treatment processes, in particular the enhanced biological phosphorus elimination (De Haas et al., 2001). Alternative or complementary precipitants used to avoid this include calcium hydroxide and sodium aluminate. Calcium hydroxyde, however, tends to increase the pH, also not desirable for biological wastewater treatment processes. Moreover, Ca is less effective than Al or Fe at precipitating phosphates (de Haas et al., 2001). Under typical conditions of municipal sewage treatment, a soluble, non-precipitable fraction of P remains in the effluent due to compounds such as phosphonates (Rott et al., 2018). These organic P compounds, as well as polyphosphates, are only partly removed by adsorption on the precipitation products (Baumann, 2002). In principle, the precipitant can be dosed in the area of the primary clarification (pre-precipitation), in the area of the activated sludge tank (simultaneous precipitation) or after biological treatment and secondary clarifier (post-precipitation). Pre-precipitation enhances the removal of solids, including carbon sources that may be needed for denitrification (DWA, 2011), and entails a higher demand for precipitant due to competing reactions. The dosing of the precipitant is also more difficult due to the need of leaving a faction of the influent Page 19 of 23 595

P available for the following biological treatment steps. Hence, the method is not suitable for maintaining low effluent values. Pre-precipitation is particularly suitable for the renovation of existing systems, as it reduces the burden on the biological stage. Simultaneous precipitation is the most commonly used method. The precipitated phosphate flocks are included into the activated sludge and separated in the secondary clarifier. Advantages include the good utilization of the precipitating agents and the positive effects on the settling of the activated sludge. If the precipitant is added at the outlet of the activated sludge basin, its dosing can be easily controlled via an online measurement of phosphate concentrations. This enables achieving low effluent P concentrations while minimizing use of reagents. The additional sludge production is lower than in pre-precipitation.

Post precipitation occurs in a separate treatment step after the secondary clarifier, which requires additional reactors, possibly combining flocculation with sedimentation or including a final filtration. The sludge consists mainly of inorganic precipitation products. Post precipitation can be used in addition to a simultaneous precipitation to achieve very low effluent concentrations.

Chemical P removal is simple to install and to operate, hence suitable also for small and very small WWTP. Operation requires a storage tank for the precipitate, a dosing pump and piping that can be easily integrated into existing plants. Precipitation best takes place in an existing turbulent area (power density around $100 - 150 \text{ W/m}^3$) like a siphon or drop structure. The reaction time can be short (about 1 min, DWA, 2011). For flocculation the retention time should be at least 20 min with a power density of around 5 W/m^3 (DWA, 2011). The conditions under which precipitation and flocculation occur significantly influence the efficiency of the process. The pH should be kept between 6.5 and 8.5 for the flocculation process, and can be corrected also with an appropriate choice of the precipitants depending on other aspects of the process (e.g. nitrification). P concentrations of 0.5 mg/L or lower can be achieved with flocculation and filtration, entailing high investments and operational costs. Alternatively, multi-point precipitation (combination of pre-, simultaneous and/or post-precipitation and/ or multiple stages of simultaneous precipitation) and instrumentation-control-automation (ICA) strategies can support the reduction of precipitants needed for the process and achieve lower effluent concentration,

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at a cost that depends on the sophistication of the control strategy. Table 10 below gives an overview of typical effluent values reached with conventional and enhanced or new technologies.

Irrespective of the process adopted, P removal brings a very high percentage of the total P present in the influent to the sewage sludge, from which it can be in principle recovered. Processes of P recovery address sludge water, sewage sludge and sewage sludge ash. Another possibility is the recovery from the effluent, that is in direct combination with P removal. This option has received limited attention so far.

The various P recovery processes implemented so far show different degrees of technological maturity (Egle et al., 2016; ESPP, 2022). Worldwide more than 100 full scale plants are in operation. Most of them are based on EBPR, and consequently recover P from process water, containing soluble P after anaerobic digestion (see ESPP, 2022). As this comprises a small part of the P contained in the sewage sludge, the recovery potential of the influent P is relatively low. Another approach, based on thermal processes (see ESPP, 2022) has just a few full scale applications. Most projects with P-recovery from sewage sludge from plants with chemical P-removal are still in the technological development phase (see ESPP, 2022). Practically all P recovery projects-except the P recovery processes from process water of EBRP-plants-are currently being supported by public funding. Examples include the "Phos4You" project (Interreg NWE, 2022) funded by Interreg North-West Europe, and "Hamburger Phosphorrecycling" with funding from the Ministry of Environment (Germany) for the construction of a large-scale plant for phosphorus recovery in Hamburg (Hamburger Phosphorrecycling GmbH, 2022). In addition, some federal states in Germany have their own funding programs for the development of application-ready and economically feasible processes (BW, 2019) or for the promotion of large-scale demonstration systems and feasibility studies. As part of the BMBF RePhoR funding initiative, seven projects are currently being funded by the German government, the majority of which take thermal processes into account, but three projects also focus on the implementation of processes for P recovery from sewage sludge or process water (KlimaPhoNds, P-Net, Satellite) (BMBF, 2022). This testifies of the need to explore the available processes further in order to achieve market-readiness.

As demonstrated in several full-scale plants, P recovery from the process water of treatment plants

with EBPR can be obtained through anaerobic biological processes with no need for reagents. The phosphate dissolved in the anaerobic reactor is turned into mineral phosphate fertilizer (magnesium ammonium phosphate (MAP), or calcium phosphates) by means of precipitation or crystallization. In order to maximize the recovery of ammonia from the process water, it is possible to add phosphate (e.g. from sludge incineration ashes), thus simultaneously improving N removal.

Examples of P recovery processes directly from the sludge water and from industrial plants are the Crystalactor process implemented in Nanjing and other treatment plants (Royal Haskoning DHV, 2022), the Ostara Pearl process e.g. at the Amersfoort sewage treatment plant (Nutrient platform, 2022) or the AirPrex process, which is used at several sewage treatment plants with EBPR to extract MAP (CNP, 2022). These technologies are fully developed and implemented on a large scale, but are limited to sewage treatment plants with biological P-elimination without Fe- or Al-containing precipitants, in order to avoid contamination of the recovered fertilizer.

However, EBPR alone is not sufficient to achieve very high P removal in the absence of at least some chemical precipitation. For instance, only a few sewage treatment plants use EBPR alone in Germany (DWA, 2016). Chemical P precipitation will continue or even increase in the future for reasons of water protection, which makes the subsequent recovery of P more complex.

Thus technologies for P-recovery from sewage sludge containing iron or aluminium phosphates need further investigation and development, as sewage sludge has a P recovery potential of up to 90%. The complex composition of the sewage sludge makes the processes more demanding in terms of process configuration and use of chemicals, if compared to the crystallization process in process water. Examples of processes applied in sludge are the Stuttgart (Meyer et al., 2018), Gifhorner (Esemen, 2012) and ExtraPhos processes. In principle, these processes can be used in a large part of today's sewage treatment plants, as they are independent of the type of P removal from the effluents. Once P is recovered from the sludge, the latter may undergo any other disposal route. However, the effort required to dissolve the phosphorus bound in the sludge in a first step and then convert it into compounds that are readily available to plants – like MAP—is very high due to the competition of compounds of Fe and Al for phosphate precipitation (Wilfert et al., 2015). The Gifhorner process relies on the sulfidic precipitation of metal ions which, however, poses issues of occupational safety. In the Stuttgart process, the precipitation of the interfering ions is avoided by complexing the metal ions with citric acid. The efficiency of the complexation plays a major role in the efficiency of the MAP precipitation process, product quality and operating costs (Meyer et al., 2018). P recovery with the Stuttgart process at the Offenburg WWTP shows the citric acid added to prevent the precipitation of metal phosphates represented up to 70% of the operating costs (Meyer et al., 2018), calling for cheaper alternatives. Furthermore, the recovery rate of the process does not yet meet the 50% P recovery rate required by the German legislation from 2029 onwards. Experience so far suggests that we still lack a reliable methodology to estimate the required use of chemicals for the dissolution of phosphorus in complex sewage sludge and the potential of P recovery.

P recovery from sewage ash works on the same physico-chemical principles applied to sewage sludge. Most of the processes proposed so far (e.g. EuPhore, Parforce, Tetraphos, Ash2Phos) include a digestion of the phosphorus compounds bound in the ashes and the subsequent recovery of a product from the liquid phase, similar to the dissolution and product recovery from sewage sludge. However, sewage sludge still contains organic substances and not only chemically, but also biologically bound phosphorus, so that the processes are significantly more complex and heterogeneous in comparison with sewage sludge ash. At the same time, P in ashes might be bound in more stable compounds which are harder to dissolve.

So far, to the best of our knowledge, there is no full scale implementation in which phosphorus is recovered from the waste water stream of a municipal WWTP, although some research projects exist. A pilot approach is offered by the RAVITA- Process, where post-precipitation sludge is used as a source for P-recovery. Nevertheless the iron or aluminium rich precipitate needs to be re-dissolved through acidification to produce a fertilizer (ESPP, 2022). However, it would be logical to combine P removal directly with recovery, and new technologies suggest this could become possible in the future. Drenkova-Tuhtan et al. (2017) show that extensive P elimination to extremely low discharge values of less than 0.05 mg / L is possible using tailor-made functionalized particles. With this process, phosphorus could even be eliminated and recovered at low concentrations of approx. 10 mg / L and concentrated by desorption in the side stream. Guaya et al. (2015) demonstrated that zeolites modified with Al for an increased selectivity towards phosphorus show a phosphate absorption more than 10 times higher than natural zeolites. Also, calcium-containing zeolites form superficial layers of (hydroxy) apatite, while struvite is likely to form in saturated solutions in the presence of ammonium. (Ellersdorfer, 2018).

The choice of a technology to adopt for P recovery depends therefore on many aspects of the P removal process in operation at each WWTP.

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