Wastewater To Resource: Design of a Sustainable Phosphorus Recovery System

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To enable a more sustainable wastewater treatment processes, a transition towards resource recovery methods that have minimal environmental impact while being financially viable is imperative. Phosphorus (P) is a finite resource that is being discharged into the aqueous environment in excessive quantities. As such, understanding the financial and environmental effectiveness of different approaches for removing and recovering P from wastewater streams is important to reduce the overall impact of wastewater treatment. In this study, a process-systems modelling framework for comprehensively evaluating these approaches in terms of both economic and environmental impacts is developed. Applying this framework, treat-ment pathways are designed, simulated and analysed to determine the most suitable approaches for P removal and recovery. The purpose of this methodology is not only to assist with plant design, but also to identify the principal economic and environmental factors acting as barriers to implementing a given technology, incorporating the impact of waste recovery. The results suggest that the chemical and ion-exchange approaches studied deliver sustainable advantages over bio-logical pathways, both economically and environmentally, with each possessing different strengths. The assessment method-ology developed enables a more rational and environmentally sound wastewater plant design approach to be taken.

1. Introduction

The need to address global grand challenges such as climate change and resource depletion has led to the emergence and continuing evolution of a more resource-efficient bioeconomy.[1] To promote this, an ambitious circular-economy-based strategy was adopted by the European Commission in 2015,[2] within which a critical emphasis is placed on the waste-economic sector. With the rising waste generation brought about by an expanding population and urbanization,[3] the realisation of a sustainable bioeconomy requires a transition in the waste sectors from passive treatment to proactive resource recovery.[4] In particular, carbon-containing and nutrient rich waste such as wastewater or organic fraction municipal solid waste, represent significant opportunities.[5] Globally, out of an average solid waste generation per person of 0.12–4.3 kg/d,[6] 59%–68% is organic.[7,8] The global trajectory of waste growth projected over the next decades calls for resource-circular technology solu-tions, the development of which must be founded on suitable analysis and evidence.[9] Phosphorus (P) recovery from waste-water is very relevant to the problems discussed above.

Among the different recoverable resources in the waste and wastewater sector, P is a notable priority.[10] As a key nutrient for the growth of living organisms and intensive agriculture, phosphorus rock is a non-renewable mineral resource which has undergone progressive worldwide depletion. By 2033, the worldwide P demand is expected to outstrip supply as a consequence of a globally expanding population accompanied by global food insecurity issues due to the dominant concen-tration of P deposits in five nations.[11] As a non-renewable resource with a crucial role in global food production, and no viable replacement,[12] the need for recovery where possible is clear. Further to the loss of P as a resource, its excessive discharge into the aqueous environment due to human activities like agriculture, household sewage, power generation and industrial applications[13] carries the negative impact of aquatic eutrophication, leading to hypoxic dead zones, harmful algal and cyanobacteria blooms and increased toxicity.[14] Removal and recovery of P from wastewaters can reduce these risks.[15]

Despite P recovery technologies receiving considerable attention and being proposed as a way to advance the United Nations' sustainable development goals (SDGs), e.g. food security, sustainable management of water,[16] very little P is currently recovered due in part to unfavourable economics compared with mined P, though, as modelled by Cordell, Drangert and White (2009),[17] the economics may change as depleting reserves lead to increased extraction costs – peak phosphorus could occur by 2030.[18] A stronger economic case can be made by accounting for the "total P recovery value",[14] which includes products, by-products, services, and avoiding or mitigating environmental damage. Assessment of value across multiple domains can be a complex problem however, with partial solutions potentially leading to ill-informed decisions.[19] Therefore, to maximise the potential of wastewater resources, a

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robust and integrative approach is needed to quantitatively compare the economic and environmental attributes of diverse technology options and consider the whole system over the long-term. While environmental impact studies of nutrient removal technologies have been published (for example an assessment of 27 nutrient removal technologies was carried out by Rahman, Eckelman, Onnis-Hayden and Gu (2016)[20]), little research has been published on such a comparative sustainability assessment of both recovery and removal of P from wastewater.[10,21]

A challenge that emerges when attempting to develop transferrable findings from such multi-technology assessments is the varying context in which the technologies are placed, with the need for careful inventories of resource flows emphasised by Heimersson, Svanström, Laera and Peters (2016).[22] Approaches building on static data hinder the transferability of the assessment approach for universal application.[10] Notably, dynamic mathematical modelling research emerged in the wastewater fields in the last two decades, enabling the performance projection of reactors and simulation of process complexity such as biodegradation,[23] crystallisation,[24] adsorption,[25] and filtration.[26] A critical research gap exists to develop a scalable and quantitative approach linking the dynamic modelling and evaluation tools to inform the process design of P removal and recovery from wastewater.

To address this research gap, a process-systems modelling approach at the interface of Chemical Engineering and Environmental Engineering is proposed here. The objectives of this study are twofold. A scalable process-systems simulation-evaluation framework is first proposed to quantify the long term economic and environmental sustainability of a proposed set of treatment technologies in a holistic manner by integrating advanced simulation and evaluation tools. Different process design options are then explored to provide an insight into the most economically viable and least environmentally damaging pathways for the removal and recovery of P from wastewater to enable a transition to more sustainable wastewater treatment. A state-of-the-art wastewater-treatment simulator was employed in conjunction with life cycle sustainability assessment (LCSA) to form a simulation-evaluation framework. In this study, we particularly focus on two sustainability pillars i.e. environmental and economic impact evaluation. The methodological framework presented here provides a transferable approach for informing the technical design of resource recovery from carbon and nutrient-containing wastes in the solid, liquid and gaseous phases. By highlighting the key environmentally damaging steps and setting performance targets, our proposed framework seeks to contribute to a transformation in the waste sectors from passive treatment to proactive recovery.

2. Results and Discussion

2.1. Simulation Evaluation Methodology

The simulation-evaluation framework is generalised in Figure S1 of the Supplementary Information (SI). GPS-X, a state-of-the-art wastewater-treatment simulator (Hydromantis Inc., Canada) is used to develop design configurations, and carry out operational simulations. This software allows for a diverse range of potential effluent sources, operator functions and flow characteristics to be incorporated into the modelling process. Capdet-Works (Hydromantis Inc., Canada) was employed in conjunction with GPS-X to account for economic aspects. The simulation design specifications are based on the publicly available literature, where the ion exchange has been modelled as an emerging P recovery technology in comparison with other technologies with higher technology readiness level (TRL). Such literature data represent the empirical research advances in the P removal and recovery. GPS-X simulation configured using empirical data enables the design and investigation of the scaled-up P removal/recovery technologies. Taking the simulation results from GPS-X, a life cycle assessment (LCA) was performed in SimaPro, to quantify the environmental impacts of each process unit.

2.1.1 Wastewater Treatment Process Simulation

To simulate P removal and recovery, process models embedded in GPS-X 6.5.1 were implemented. GPS-X is an object-oriented, model-independent and interactive simulation tool, where a variety of process modelling approaches are available (e.g. Activated Sludge Model (ASM), Comprehensive Model (MANTIS) and Anaerobic Digestion Model (ADM)). These process models incorporate the observed biological, physical and chemical processes in wastewater treatment plants and contain a considerable amount of technical information and data developed in last decades. Inspite of the process model advances, the implementation challenges at multiple model interfaces (e.g. state variable mapping) and the simulation complexity (e.g. side stream treatment) hinder the computational tractability of applying ADM, ASM or any other process model directly. Thereby, GPS-X underpinned by an object-oriented approach enables the streamlined simulation of wastewater treatment processes. The GPS-X simulation generates input and output flows, capturing the reaction dynamics, operational configurations and system performances associated with each process. Additionally, the software enables the prediction of effluent quality and the verification of the validity of the operating variables within the realistic operational constraints of the system. Using this software, various technologies and treatment pathways can be compared and analysed in a systematic manner, while long-term cost and environmental impacts can be further analysed.

2.1.2. Life Cycle Environmental and Economic Assessment

Adopting a holistic life cycle approach, two sustainability pillars have been investigated in the current study to assess the economic and environmental performance of different P removal and recovery pathways.

2.1.1.1. Cost Analysis

In conjunction with GPS-X, CapdetWorks 3.0 was used to evaluate the economic performance of each process, using the capital, operational, maintenance and amortization costs associated with the various system components to determine the overall cost. CapdetWorks builds on the industrial well-recognised design algorithms and up-to-date cost database for process units and equipment and is based on research in collaboration with manufacturers, suppliers and consultants; thus, it provides robust preliminary design and cost estimation. The total cost is a function of the capital costs (*CAPEX*), operating costs (*OPEX*) and the discount rate, where the CAPEX and OPEX values were primarily derived from the CapdetWorks database. Time-dependent OPEX values were discounted back to the present value using a fixed discount rate.

2.1.1.2. Scope for Life Cycle Environmental and Economic Evaluation

LCSA consists of analyses of three pillars of sustainability (economy, environment and society).[27] This study focusses on economic and environmental sustainability evaluation and enables one to account for all input-output flows occurring at each life cycle stage throughout the 'cradle-to-gate' wastewater treatment life cycle (see the detailed evaluation framework diagram Figure S1 in SI Section 1). We adopt LCA as formalised by the International Organization for Standardization[28] to derive quantitative insights into the holistic environmental sustainability of P-removal and recovery pathways. The life cycle functional unit was defined as 'per unit (1 kg) of phosphorus removed or recovered from a municipal wastewater stream in a WWTP with a hypothetical capacity of 1000 m³/d and a 20-year life span, where the effluents are below the discharge thresholds' to enable different technology-driven WWTP systems to be compared. A detailed description of the LCA approach used is provided in Section 1 of the SI.

The LCA inventory was developed by using input-output flows derived from GPS-X simulation (Figure S1), supplemented with secondary data from publicly available sources and Ecoinvent database. A problem oriented (midpoint) approach – CML baseline (v 3.02) was applied in the current study as the 'default' Life Cycle Impact Assessment (LCIA) method. Differing from a damage-oriented (endpoint) approach, midpoint LCIA addresses the mechanisms between environmental interventions and associated environmental indicators in a transparent manner and reflects the trade-offs across environmental impact categories without additional damage assessment steps. Under this study, the environmental impact categories at mid-point level under investigation include abiotic depletion, fossil depletion, global warming potential, acidification, eutrophication, ozone depletion, photochemical oxidation and human and eco-toxicities.

2.2. Process Design for Phosphorus Removal and Recovery – LCA Inventory

The importance of ensuring that non-renewable resources such as P are not just removed from waste streams, but recovered for re-use has been highlighted in previous work.[14,16]

In this study, to determine the most economically viable and environmentally benign approaches to removing and recovering P from wastewater, pathways were explored under the three categories of chemical, biological and ion exchange, which are effective at incorporating P into solids and achieving selective recovery.[14]

Within these categories, plant configurations, scales and parameters are determined based on previous empirical studies, with all design choices made to achieve more sustainable operational performance. The design choices and pathway configurations are illustrated in the following sections, and only serve as examples of possible flowsheets. Clearly, a large variety of different technologies can be chosen. In this study, three selective technologies based on the publicly available empirical data were used to demonstrate the applicability of the simulation and evaluation framework. One of the many benefits of our methodology is that many different flowsheets can be evaluated quickly against differing boundary constraints and outputs.

For all processes simulated, identical influent water characteristics are assumed, with chemical compositions shown in the Supplementary Information (Table S1), where for consistency across the different design cases, the flow-rate is set at a constant 1000 m³/d. Below, three scaled-up technology designs based on the published empirical data are presented.

2.2.1. Chemical Technology

2.2.1.1. Technology Overview and Theoretical P Recovery

The chemical pathway is characterised by the addition of a metal salt to remove P from wastewater via precipitation, where commonly applied chemicals include ferric or aluminium cations with chloride and sulphate anions, or lime, aluminium chlorohydrate and poly aluminium chloride (PAC),[29,30] and their characteristics are summarised in the Supplementary Information (SI) (Table S2 and Section 3).

The recovery of P from the precipitated iron phosphate is analysed, and involves acid and sulphide (H_2S) dosage and precipitation to recover P and pyrite (FeS_2); this can recover approximately 92% P at an S/Fe molar ratio of 2.5.[31] The recovered phosphate in the supernatant can be used directly in fertilizer production (e.g. struvite). Moreover, iron and sulphur
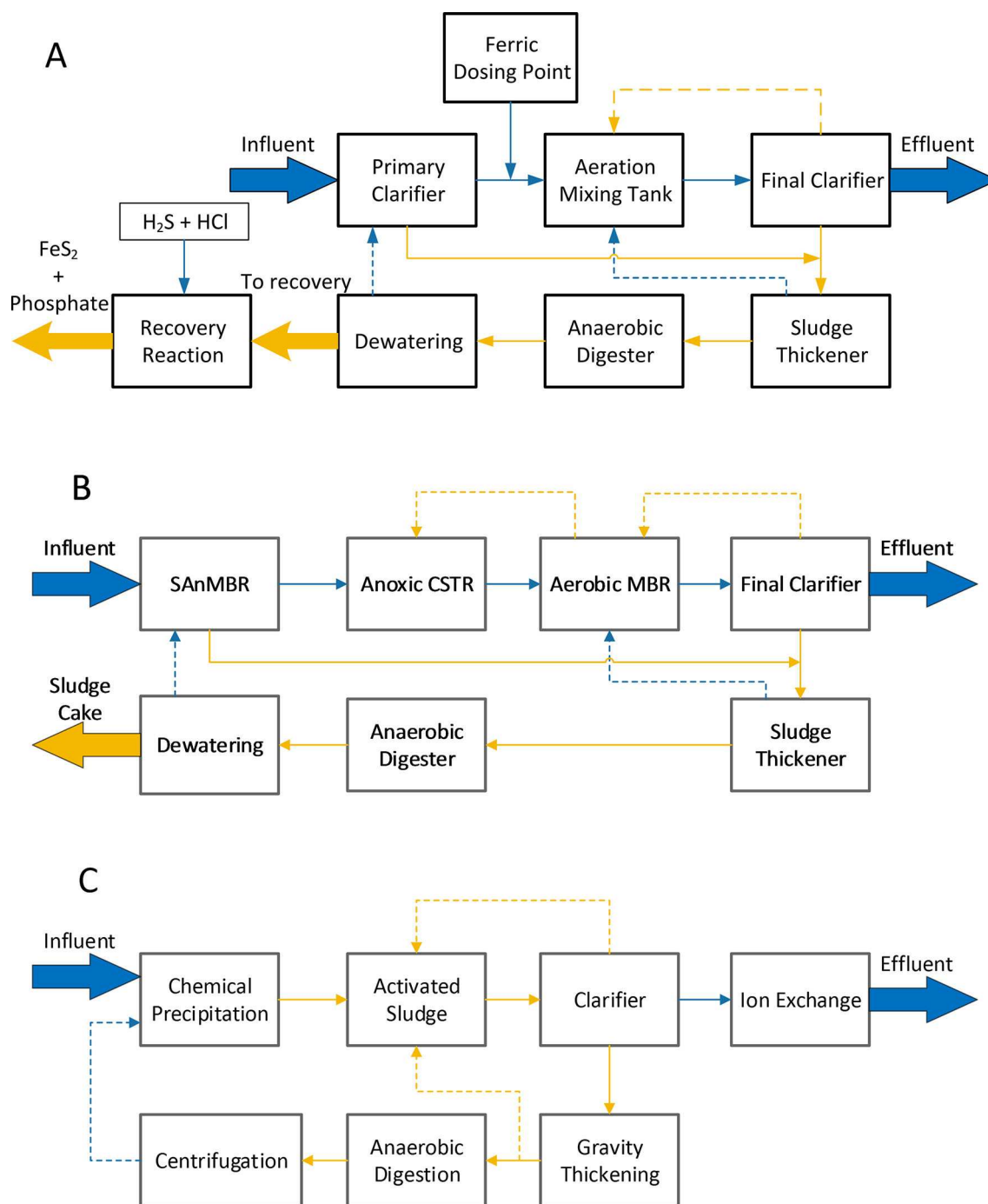
Figure 1. process overview for (A) chemical pathway, (B) biological pathway and (C) ion exchange pathway (water flow and chemical dosing are represented by blue arrows; sludge flow is presented by orange arrows; solid and dash lines represent forward process and recycling loops respectively).

can be simultaneously regenerated via sulphide oxidation of additional iron sulphide (FeS), hence improving the financial viability of WWTP (e.g. ionic reactions demonstrated in previous research[32]). The reaction process for recovering P is summarised in the Supplementary Information (Section 5 and Table S6).

2.2.1.2. Process Configuration

The process is illustrated in Figure 1A, where the chemical pathway can be separated into three parts – wastewater treatment, sludge treatment and the P recovery process. The process configuration for wastewater and sludge treatment is detailed in Figure 2A. Effluent from the primary clarifier and ferric ions are then fed into the aeration mixing tank to ensure high ferric phosphate precipitation efficiency. The final clarifier
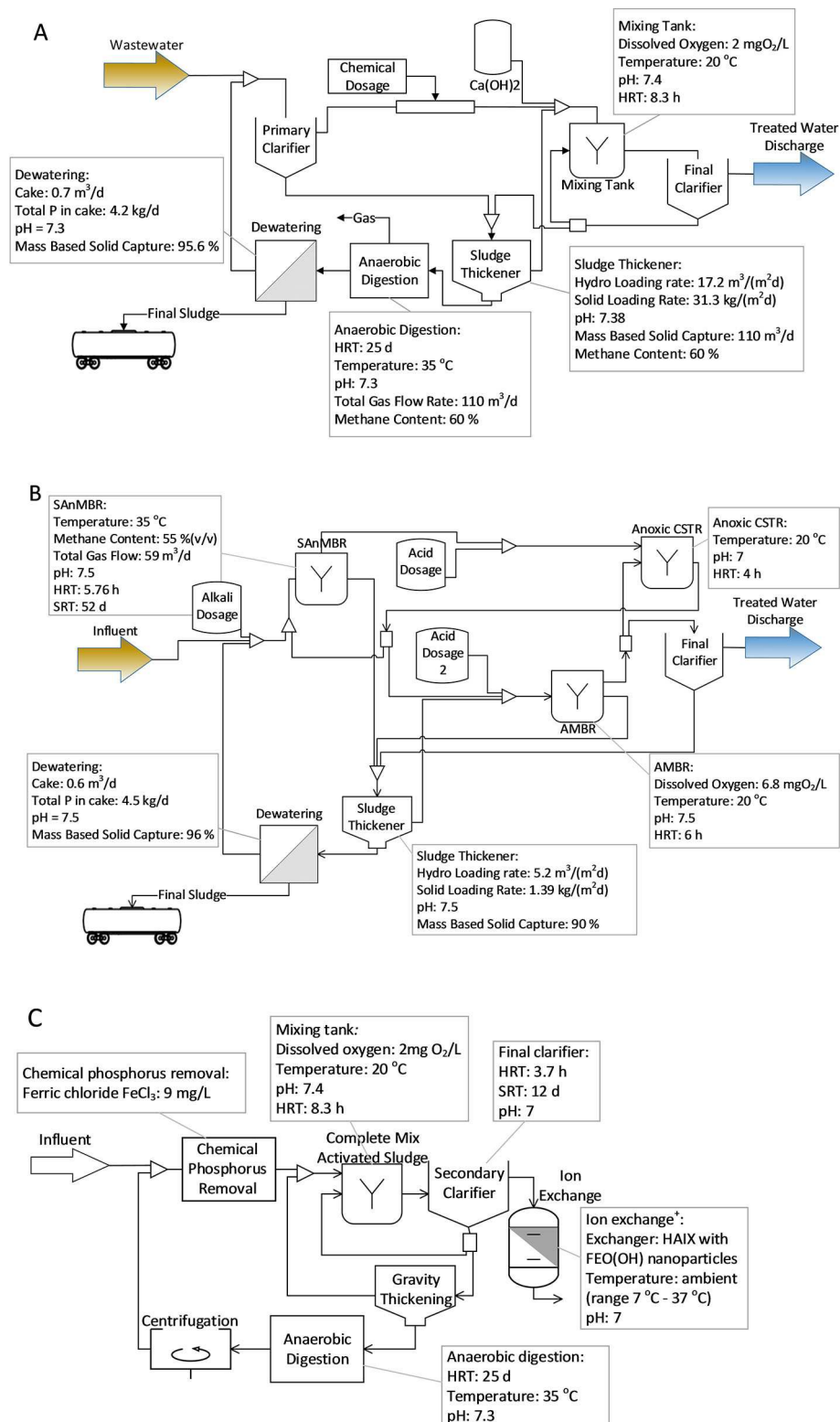
Figure 2. Process configuration and operating parameters of (A) chemical pathway; (B) biological pathway; (C) ion exchange for P removal.

separates ferric phosphate precipitate from the wastewater and discharges treated water meeting environmental standards.

The phosphate enriched sludge from the clarifier is separated into two streams, the first is recycled back to the

aeration tank for further treatment, while the second is pumped to the sludge treatment units. In sludge treatment, sludge from both clarifiers combine and are thickened, and overflow water containing soluble nutrients is recycled and treated in the

aeration tank, while the thickened sludge goes to the digester to produce biogas. Nutrient enriched sludge flows to the dewatering unit where the filtrate contains soluble P and is then recycled back to the primary clarifier. The solids cake is pumped to the reactor for P and iron recovery by dosing with H_2S and HCl solution in a mixed reaction tank. The design parameters are presented in the detailed configuration in Figure 2A.

2.2.2. Biological Technology

2.2.2.1. Technology Overview

As illustrated in Figure 1B, a system with a submerged anaerobic membrane bioreactor (SAnMBR)/anoxic/aerobic membrane bioreactor (MBR) was chosen for investigation as it can be adapted to membrane processes with a conventional anaerobic/anoxic/aerobic sequence for biological P removal. This has been experimentally proven to be energy efficient while reducing sludge production, and membrane processes have the advantage of eliminating suspended solids in the effluent stream.[33]

2.2.2.2. Process Configuration

The system configuration is demonstrated in Figure 2B, where the key operating parameters have been selected based on previous research (a detailed overview of the selection rationale can be found in Supplementary Information Section 3). The presence of the SAnMBR is critical, as the higher P release under anaerobic conditions contributes to a higher P uptake rate in the aerobic reactor.[34] In the overall process flow, permeate from the SAnMBR flows to the subsequent anoxic reactor, where anoxic P-uptake occurs along with denitrification, with nitrate acting as electron acceptor. Such a configuration, with the anoxic process taking place before the aerobic process can reduce the ammonia effluent, overcomes a critical drawback of a typical anaerobic–anoxic/nitrification sequencing batch reactor.

Receiving effluent from the anoxic mixing tank, the AMBR is modelled to further degrade the chemical oxygen demand (COD) and biological oxygen demand (BOD) while removing the remaining ammonia, where the nitrifying bacteria at this step lead to a high ammonia removal efficiency. Like the SAnMBR, the AMBR leads to low sludge production; thus, the biological pathway has been modelled as the route to minimise sludge production, while generating biogas. In the case of presence of the P enriched sludge, P can be recovered for struvite or calcium phosphate production, which is not addressed in this study.

2.2.3. Ion Exchange Technology

2.2.3.1. Technology Overview and Theoretical P Recovery

As shown in Figure 1C, the ion exchange route involves chemical precipitation, activated sludge and a highly PO_4^{3-} selective ion exchanger unit, where a commercialised polymeric anion exchanger (HAIX), bounded with hydrated ferric oxide (FeO(OH)) nanoparticles, was selected because of its high sorption affinity towards phosphate.[35] Under this pathway, an approach to derive struvite ($Mg(NH_4)PO_4$) by adding $MgSO_47H_2O$ and NH_4Cl is modelled. The chemical reaction and resulting recovery rate associated with P recovery through struvite production can be found in Supplementary Information Section 5 and Table S6.

2.2.3.2. Process Configuration

As configured in Figure 2C, chemical precipitation was modelled as a pre-treatment step for ion exchange. The effluent from the primary clarifier is mixed with ferric chloride (9 mg/L) before flowing to the aerobic mixing tank. Through aeration, poly-phosphorous and other classes of organic phosphorus are hydrolysed to form ortho-phosphorus. Together with the soluble ortho-phosphate components, this reacts with ferric ions efficiently to form iron phosphate under adequate mixing, which ensures uniform dispersion and intimate contact between chemicals. In the next step, the precipitation of ferric phosphate occurs in the secondary clarifier and is then removed as sludge for the P recovery at the ion exchange step. The relationship between the total exchange capacity of HAIX (Qe) and the initial concentration of P (Co), was plotted and fitted to the Freundlich adsorption isotherm (Eq.(3)), based on the reaction kinetic data of Sengupta and Pandit (2011).[35] The Freundlich equation is given as follows, where Qe denotes the exchange capacity in mg P/g, and Co represents the initial P concentration (mg/L) at a pH of 8.

$$Qe = 4.754 Co^{0.2975} \tag{1}$$

It was assumed that after 100 bed volumes of 500 mg/L PO_4^{3-} were adsorbed, HAIX ion exchange media FeO(OH) is regenerated with a 1 bed volume of 4% sodium hydroxide and 2% sodium chloride solutions; it was assumed that such regeneration would not cause significant degradation in the exchange capacity.

2.3. Economic and Environmental Analysis

The different removal pathways are assessed in this section. The efficiency of each pathway is evaluated in terms of the % P removal achieved. A full breakdown of the costs associated with each pathway is provided to illustrate the economic variation between the different approaches. Finally, a comprehensive sustainability analysis is carried out in which the environmental

impact of the full lifecycle of each pathway is evaluated across many different domains.

2.3.1. Process Efficiency of Phosphorus Removal Technologies

The full results of the simulation are presented in the SI (Table S7). These suggest that each of the three pathways offer effective COD and total Phosphorus (TP) removal, resulting in a TP concentration in the effluent below the discharge threshold of 1 mg P/L. The biological route results in a superior COD removal efficiency (95.6%) in comparison with the other pathways, whereas ion exchange delivers optimum TP removal performance (100%).

The biogas composition derived from a simulation in this study for ion exchange and chemical routes (approximately 60–63% v/v CH_4) in general agreed with the literature data (above 95% v/v of the biogas comprised of CH_4 and CO_2 with 65% v/v CH_4).[36] However, the biogas compositions (71% v/v CH_4, 5% CO_2 and 23% N_2) simulated for the biological route differ from most of the results reported in previous studies. This can be explained by several factors; the relatively low solubility of CH_4 in aqueous phase (approximately 15 mL/100 mL water at 1 atm and 35 °C) compared with CO_2 (which dissolves in the bulk solution, partially generating bicarbonate ion), led to a high CH_4 content in the gas phase.[37] The biogas was methane-rich due to a favourable balance between methanogenic and acidogenic bacteria,[33] while the low CO_2 content in the biogas was a result of the pH value (controlled at 7.0) and the low alkalinity of the bulk liquor.[38] Additionally, the larger quantities of soluble CO_2 exiting in the effluent and the increased CH_4 content (above 70%) were partially a result of the short HRT. Similar biogas compositions with over 70% or even 80% v/v CH_4 have been reported in several studies.[39] The relatively high N_2 content may be caused by: 1) the sparging gas initially used in the headspace of the reactor,[37] or gas entering with the inlet feed and stripped out in the reactor; 2) possible N2 generation from denitrification even at low NO_3^- concentration in the influent.[38]

2.3.2. Economic and Environmental Comparison

2.3.2.1. Economic Evaluation

To analyse and compare the economic viability of the proposed technology pathways, costs are broken down to include both capital and operating aspects of all modelled system components in each of the proposed configurations. A 20-year lifetime was assumed for each of the plants analysed to determine long-term operational cost projections. This total projected operational cost was then calculated as:

$$TotOp = \sum_{n=1}^{20} \frac{OPEX}{(1+i)^n} \qquad (2)$$

where OPEX denotes the total annual operating cost, and i denotes the discount rate, taken here to be 8%. Operational

costs associated with maintenance as well as the required materials, chemicals and energy, labours are included in the annual OPEX for each component. Capital costs incorporate the construction costs associated with each of the system components as well as the associated amortisation costs.

The capital costs and total operating costs are shown for each of the proposed P-removal pathways in Figure 3A. The chemical pathway achieves the lowest operational and capital costs, followed by the Ion-exchange pathway. The biological pathway is the least economically viable, with significantly higher operational costs.

Across all the P removal pathways, capital cost is a driving factor, contributing to over 50% of the total costs in both the ion exchange and chemical routes, and above 30% in the biological route. A further breakdown of the costs is provided for each pathway in Figure 4 and Supplementary Information section 7. The principal operational costs in the ion exchange pathway are shown to be associated with the ion exchange component and anaerobic digestion, which account for approximately 19% and 22%, respectively.

In the chemical pathway, the anaerobic digestion process accounts for a large proportion of the total (26.5%). However, it can also be noted that components such as the iron feed system and the chemical P removal elements comprise a relatively insignificant proportion of the total cost.

The cost breakdown associated with the biological pathway (Figure 4C) indicates a clear deviation from the other two pathways. The operational costs are dominated by membrane bioreactors (SAnMBR and AMBR) and the anoxic continuous stirred-tank reactor (CSTR, CAPEX driven by chemical inputs), which account for 19.2%, 18.9% and 35.9% of the total, respectively; the operational contributions of other components such as dewatering and sludge thickening are negligible.

The full cost breakdown for each pathway is summarised for a 20-year lifespan in Table 1 and Supplementary Information

Table 1. Cost breakdown of WWTP over 20-year plant lifespan.

	Ion Exchange Pathway (Million USD)	Chemical Pathway (Million USD)	Biological Pathway (Million USD)
Construction	4.860	4.458	5.404
Operation	1.282	1.380	1.380
Maintenance	0.494	0.286	0.366
Material cost	0.143	0.145	0.710
Chemical cost	0.059	0.074	7.753
Energy cost	0.132	0.141	0.246
Amortisation	2.912	1.573	2.089
Total	9.882	8.057	17.948

Tables S14–16. The chemical cost associated with the biological pathway emerges as a clear barrier to the economic viability of the biological pathway, while the total costs for the chemical pathway are approximately 20% lower than for the ion exchange pathway.
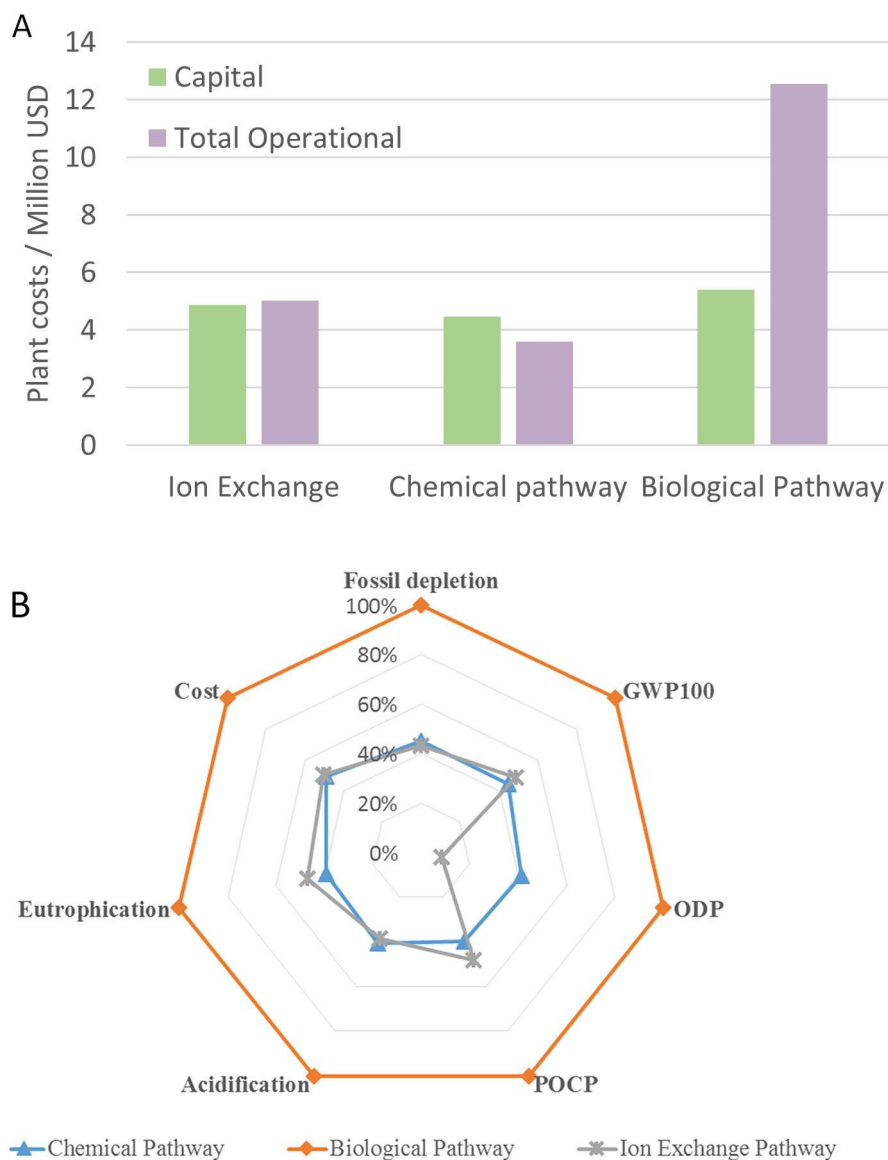
Figure 3. (A) Capital and total operational costs for each set of treatment technologies over a 20-year plant lifespan; (B) Comparisons of the three P-removal pathways (unit: per kg P removed).

2.3.3. Economic and Environmental Evaluation

The input-output flows derived from process simulation and design were fed into the life cycle evaluation model for environmental performance evaluation. The following present and discuss the sustainability performances of the P removal and recovery modelled with the characterised impact assessment presented as normalised comparisons (%) in Figures 3B and 5. The LCIA scores for each individual impact category are given in Supplementary Information Section 5. The LCA results are interpreted below from two perspectives i.e. comparison of P removal and recovery pathways and contributional analyses to highlight the key performance-limiting steps. Please note that this section reflects the LCIA results based on relative mid-point characterisation method and indicates the potential environmental impacts.

Seven indicators: cost, eutrophication, GWP100, ozone depletion potential (ODP), Photochemical Ozone Creation Potential (POCP), acidification and fossil depletion are selected here to demonstrate the economic and environmental comparison for the removal of per unit (1 kg) P. It should be noted that this list of indicators is not exhaustive (a more comprehensive framework for selecting suitable indicators is presented by Iacovidou, Millward-Hopkins, Busch, Purnell, Velis, Hahladakis Zwirner and Brown (2017a)[19]). To illustrate the relative performance of the three different pathways, a normalized spider chart is presented here in which the route with the largest occupied area represents an inferior system. As shown in Figure 3B (and Table S8 in supplementary Section 6), the ion exchange and chemical routes once again outperform the biological pathway for P removal across all KPIs. Overall, the chemical route represents the most environmentally friendly P-removal option
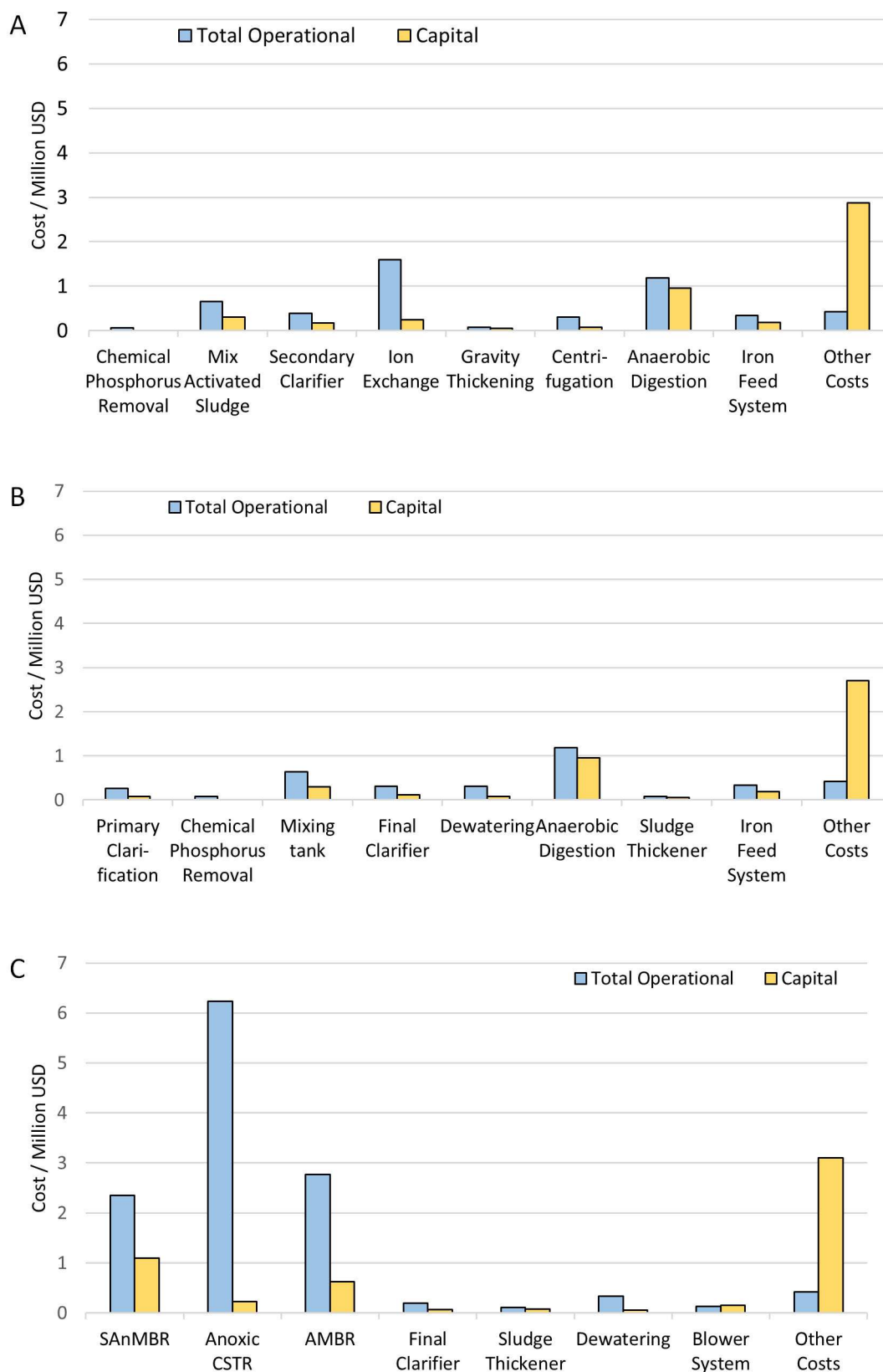
Figure 4. (A) Ion Exchange, (B) Chemical Pathway, (C) Biological Pathway: Capital and operational cost associated with each system component over a 20-year plant lifespan.
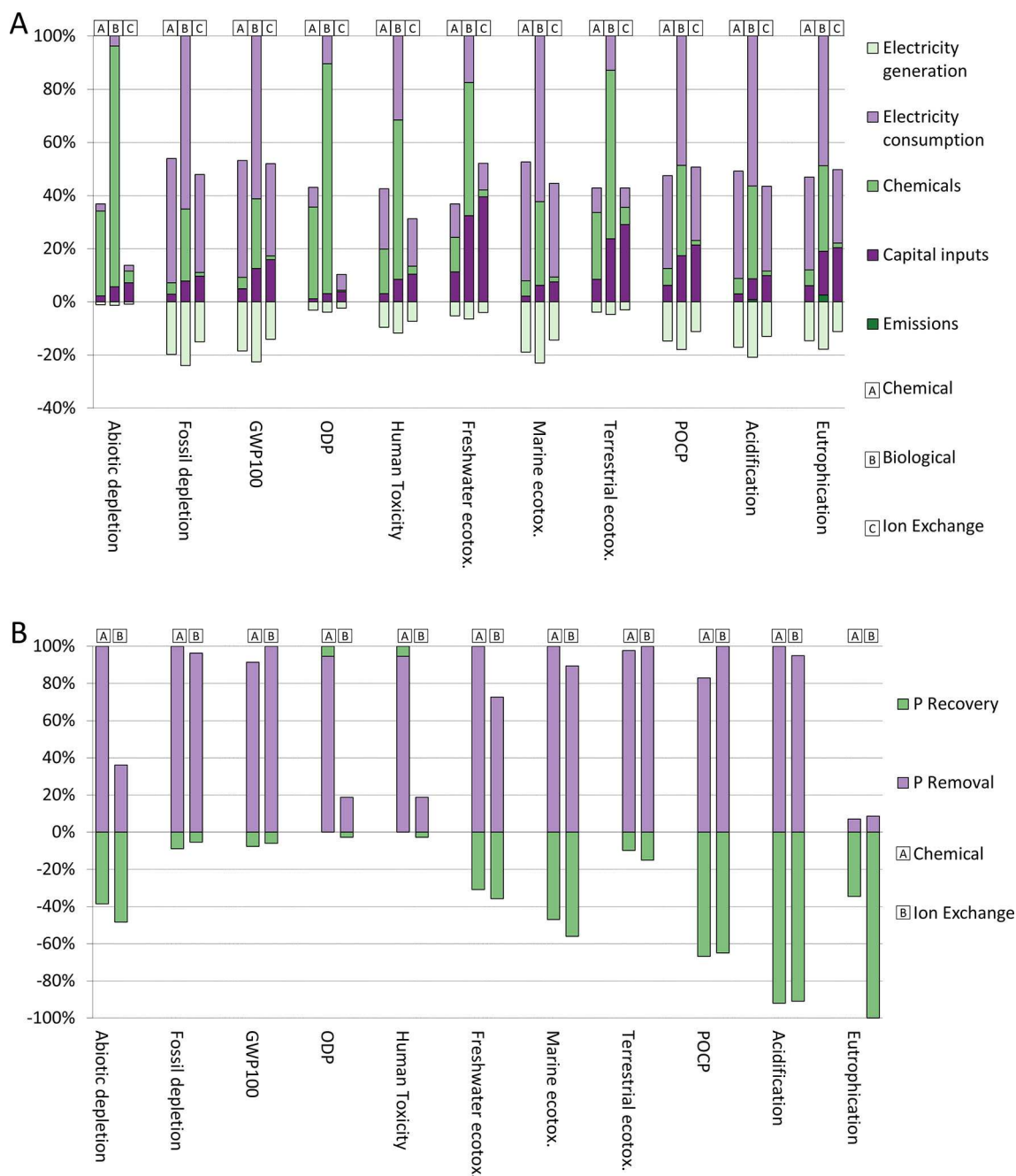
Figure 5. (A) contributional analyses for P-removal pathways (method: CML baseline, unit: per kg P removed) (B) LCA analyses for P-recovery pathways (method: CML baseline, unit: per kg P recovered)

in GWP100, eutrophication and POCP, but cannot compete with ion exchange on ODP; while the ion exchange and chemical pathways incur similar impacts in terms of the fossil depletion and acidification indicators.

A detailed LCA contributional analysis is presented in Figure 5 (and Tables S9–S11 in Supplementary Section 5), in which the various environmentally damaging steps are highlighted, indicating the most effective opportunities for improvement. Approximately 70–95% of the total environmental burdens of the chemical and biological routes can be attributed to electricity and chemical inputs (e.g. FeCl$_3$ for the chemical

route, and chemical dosing for pH adjustment in biological route); this is due to the energy-intensive processes and pollutants associated with power generation and chemical production. These impacts include greenhouse gas (GHG) emissions (e.g. CO$_2$), acidification and eutrophication emissions (SOx and NOx) resulting from energy consumption (e.g. fuel extraction and combustion), and chemical inputs (e.g. lime for concrete production), as well as the release of toxic compounds (e.g. mercury emitted to air, and the cobalt, nickel, beryllium ions released to water from chlorine production). Energy consumption plays a particularly dominant role in the chemical

route due to the projected high electricity inputs (633.5 kWh/d, which is twice the intensity of the alternatives studied here). Capital input is another environmentally damaging contributor, sharing 5–30% of the environmental impact burden of both the chemical and biological P-removal pathways across all impact categories. Contrasting with the chemical and biological pathways, chemical inputs have relatively little impact in the ion exchange pathway, which is dominated by capital and energy inputs (70–96%). Ion exchange has been simulated as an infrastructurally-intensive route, where chemical precipitation is employed as a pre-treatment, followed by mixing, ion exchange and anaerobic digestion units. The production of the major infrastructure materials needed in these processes – concrete and steel – is energy-intensive with high emission of pollutants. These include the waterborne emissions (e.g. phosphate, nickel, vanadium beryllium) from coal mining and primary steel manufacturing, GHGs and other gaseous emissions (e.g. CO, NO_x) released from building material production (e.g. iron, clinker/cement) and fuel combustion for producing steel and concrete.

Apart from the negative environmental impacts, the LCA results also indicate the environmental benefits of resource recovery. Electricity and chemicals produced in the recovery processes result in less need for electricity and chemical production elsewhere, thus leading to significant savings (illustrated here as negative values in Figure 5A and Figure 5B). A notable impact can be seen on abiotic depletion and eutrophication, leading in some cases to the beneficial effects outweighing the negative burdens associated with fossil/mineral depletion and eutrophic emissions. As demonstrated in Figure 5B (Table S12 in Supplementary Section 6), when beneficial impacts are considered, ion exchange offers a promising route for P recovery, with a superior environmental profile to the chemical pathway in most impact categories despite its high POCP burdens; on GWP100 and fossil depletion, ion exchange and chemical P recovery deliver similar environmental scores. Our results highlight the research challenges and opportunities in process design and optimisation to achieve the best trade-off between resource recovery efficiency, and environmental costs associated with operational and capital resource flows.

3. Discussions

The purpose of the study was to propose a scalable simulation-evaluation framework for comprehensively assessing the economic and environmental strengths and weaknesses of different design options for wastewater treatment technology, and to determine suitable design choices for wastewater treatment plants focussed on removing and recovering P from wastewater using the developed simulation-evaluation methodology. The results demonstrate clear trade-offs between recovery/removal efficiencies and sustainability footprints. It was found that ion exchange is a promising P removal route with the best P selectivity (100% P-elimination); whereas the chemical approach to P removal achieved the best economic performance,

followed by ion exchange. Biological methods, despite exhibiting a strong overall contaminant removal performance (approaching 100% retention of suspended solids and removal of COD, BOD and nitrate/ nitrite) were found to have significantly higher associated costs per unit P removed, with a significantly inferior environmental performance. Chemical and ion exchange approaches had varying environmental strengths and weaknesses. For P recovery, ion exchange with the co-production of green electricity and struvite was demonstrated as the most environmentally sustainable P recovery method. Our cost breakdown and LCA contributional analyses suggest the key performance-limiting steps for P removal and recovery, which inform a future empirical research focus.

The proposed simulation evaluation framework sits at the interface of chemical engineering (process systems engineering) and environmental engineering, which has not been applied in previous research on P removal and recovery. The modelling framework offers a consistent whole systems approach to effectively assess the holistic environmental and economic performances of diverse wastewater treatment technologies and inform the decision-making and improvement 'hotspots' (i.e. performance-limiting steps). We used P removal and recovery as a case study to highlights the insights the modelling framework could provide to support the technical design for resource recovery from carbon-containing and N/P-rich wastes. In future research, iterative simulation-evaluation can be explored, where the scaled-up design can be further refined based on the 'hotspots' identified in evaluation to achieve the trade-offs between conflicting design criteria (e.g. techno-economic and environmental trade-offs). Another future research direction is the model evaluation. Though GPS-X is one of the most advanced wastewater simulation tools, further research efforts are needed to evaluate the simulation model by comparing the model outputs with lab or commercial operational data.

Overall, by incorporating technological advances into a waste-to-resource design framework, our proposed approach links process units and evaluates the whole technology system from a holistic perspective for P removal and recovery. The results demonstrate clear trade-offs between recovery/removal efficiencies and economic/environmental footprints. The overall results suggest that the chemical and ion exchange approaches deliver sustainable advantages over biological pathways, both economically and environmentally. Two key design bottlenecks have been identified in waste-to-resource systems, 1) the inconsistency in the evaluation methods and boundary, which hinder the effective comparison of diverse technologies; 2) the lack of holistic views (environmental and economic), which could lead to problem-shifting in the process design. The simulation-evaluation framework and the insights generated from our study have the potential to unlock the design bottlenecks, targeting key influence factors (e.g. trade-offs between recovery efficiency and economic viability) to catalyse a transition towards more waste resource recovery. Such a transition, characterised by the establishment of a sustainable circular economy, offers a viable way to tackle the UN's ambitious SDG challenges.

Conflict of Interest

The authors declare no conflict of interest.

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