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PAPER

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Exploring resource recovery potentials for the aerobic granular sludge process by mass and energy balances – energy, biopolymer and phosphorous recovery from municipal wastewater†

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Municipal wastewater cannot any longer be perceived as a waste stream because it contains water, energy, fertilizer and other products that can be recovered with innovative technologies in so called 'water resource factories'. Therefore a paradigm shift has been proclaimed to design water resource factories in the future that feed into a circular economy. The rapid development of new resource recovery technologies requires a solid analysis prior to their integration into treatment processes to understand more about their potential to contribute to more circular urban water management practices. Mass and energy balances are an excellent method to model resource recovery potentials of innovative processes at an early design stage because they allow quantifying recoverable resources as well as trade-offs between possible recovery technology choices. We modelled a real wastewater treatment plant which uses aerobic granular sludge treatment and is currently operated with no on-site resource recovery. Then, 5 different possible process designs that would recover chemical oxygen demand (COD) as energy and/or extracellular polymeric substances (EPS), and phosphorous (P) as struvite have been modelled. The integration of anaerobic digestion for subsequent electricity and heat generation from methane provides the possibility to recover on-site a rather small fraction of influent-COD as energy. But if this is combined with chemically enhanced primary treatment (CEPT), almost one third of the influent-COD may be recovered. Simultaneous energy and EPS recovery may lead to trade-offs as CEPT integration for maximum energy recovery may halve the EPS recovery potential but would increase the overall influent-COD recovery rate. Struvite fertilizer recovery integration may only recover a small fraction of influent-P and is therefore questionable when other P recovery options are possible that aim for higher recovery rates. The fertilizer recovery potential may be significantly decreased by EPS recovery since the latter contain P. This study helps to understand how aerobic granular sludge based treatment processes can be designed as water resource factories. Mass and energy balances can be conducted at a very early process design stage and results may be used to identify promising process designs for subsequent more in depth techno-economic or environmental impact assessments.

Water impact

Innovative technologies can recover energy, fertilizers and biopolymers from municipal wastewater. But how can they be integrated into wastewater treatment plants most effectively? Since different technologies may recover different quantities of the same resource while the recovery of one resource may limit the potential to recover another one, a closer look at mass and energy flows of innovative wastewater processes is required.

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Introduction

Wastewater has been recognised as a resource rather than a waste stream for over a decade now.[1] It contains resources that can be recovered with a variety of technologies into reusable water, energy, fertilizers and other valuable

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products.[2] Recovering resources that can be produced in quantities and at costs that match the current market demand and prices[3] and/or tackle projected future resource scarcity[4] enables the transition towards water resource factories (WRFs) instead of wastewater treatment plants (WWTPs).

The aerobic granular sludge process (AGS), also known as the NEREDA® technology was successfully introduced globally at several full scale wastewater treatment plants in recent years and is considered more resource efficient than the conventional activated sludge process (CAS).[5] Due to the granular sludge's excellent settling behaviour and tolerance to high MLSS concentrations the NEREDA process makes large settling tanks and low biomass concentrations in reaction tanks redundant compared to conventional biological treatment processes.[6] The required surface area for AGS processes is therefore roughly 75% lower than for CAS processes that use flocculated sludge.[7] Another advantage of the AGS over the CAS process is biological P removal that requires almost no additions of chemicals as phosphate precipitants. Furthermore, there is no need for energy intensive recirculation of flows between anaerobic, anoxic and aerobic tanks within the plant because these conditions are all established simultaneously in the different layers of each single granule.[8] The higher efficiencies in land consumption, energy and chemical use lead to approximately 25% cost reduction of the AGS process compared to CAS processes.[7]

The AGS process is not only promising in terms of resource and cost efficiency but offers an innovative possibility for product recovery from COD. Extracellular polymeric substances (EPS) are produced by microorganisms to form a hydrogel matrix as a dense network that gives the granular microbial structures their physical stability. EPS are a complex mixture, consisting of polysaccharides, proteins, nucleic acids, (phosphor)lipids, humic substances and some intercellular polymers. They are considered useful polymers for industrial applications as they show unique material properties especially when used as composite material.[9] Biopolymer like materials cannot, in general, be derived from oil-based chemicals and hence their supply relies solely on natural resources.[10] In the Netherlands, a commercial company currently develops a value chain from EPS recovered from AGS to market the polymer under the product name "Kaumera® Gum". A full-scale EPS recovery and down streaming plant has been opened in 2019 in the WWTP of the City of Zutphen.[11]

With growing concerns over climate change, energy saving, energy efficiency and energy substitution have become a common development principle all over the world and are manifested in the 7th UN development goal for more affordable and clean energy.[12] The wastewater sector including academia responds to that goal with exploring energy self-sufficient WWTP designs that reduce net-energy consumption and therefore may achieve carbon neutrality and decrease operational costs.[13] Biogas recoverable from

primary sludge and/or waste activated sludge by anaerobic digestion (AD) and subsequent combined heat and power (CHP) generation is the most widely applied energy recovery route in WWTPs.[14] Biogas production is an established technology for a variety of organic waste streams with growing implementation worldwide. Compared to other feedstock, sewage sludge leads with *ca.* 60–70% to a fairly high methane content.[15–17] It has been shown that combining AD with up-concentration of primary COD in a chemically enhanced primary treatment unit (CEPT) could increase methane recovery while simultaneously minimizing aeration energy consumption in aerobic treatment units, and thus, may reduce greenhouse gas (GHG) emissions and operational costs of WWTPs.[18] In addition to on-site energy recovery, anaerobic sludge digestion also serves the purpose of waste sludge stabilisation[14] which is an important method to decrease waste sludge quantities and thus waste management costs of WWTPs.[19,20]

In addition to energy recovery, phosphorous (P) recovery is another research topic experiencing high interest in academia already since decades and numerous technological recovery routes have been developed.[21] Efforts to recover P have been recently intensified as various EU member states including Germany, introduced legislation to enforce P recovery from WWTPs in the near future.[22] Currently, fractions of the influent-P are predominantly recovered by two technological solutions, namely chemical P extraction from sludge incineration-ash and as struvite mineral from P-rich side streams in WWTPs that apply biological P removal.[23] Struvite precipitation in WWTPs was discovered almost 60 years ago and its removal served initially the purpose of improving plant operations and especially clogging of pipes and equipment.[24] Because it contains both ammonia and *ortho*-P the mineral can be used as a slow release fertiliser applicable to crops in soils with low pH value and is therefore an interesting mineral for recovery.[25] The recovery of struvite by controlled crystallization requires a side stream that contains concentrated orthophosphate and ammonia. Anaerobic sludge digestion plays therefore a key role in struvite recovery as it re-immobilizes fractions of both nutrients.[26]

WWTPs are usually designed according to two major criteria: treatment performance to meet legal effluent standards with a reliable robustness on the one hand and cost effectiveness on the other hand. It has been postulated that resource recovery is missing as an additional but integral part in early-stage process design decisions.[1,27,28] Although the paradigm shift from treatment towards resource recovery and the inclusion of the latter as a central objective in new process designs has been claimed in literature since a decade, little resource recovery technologies seem to have experienced wide implementation yet.[29]

One important reason is the still constantly growing range of technical possibilities and therefore process design complexity increases. More and more innovative treatment and resource recovery technologies become available but

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little is known about how to integrate and combine them effectively.[30–33] One aspect that needs enhanced understanding to design WRFs in the future is that the integration of a resource recovery technology into a process likely implies trade-offs with one or more other possible resource recovery technologies, as influent constituents, like *e.g.* P and COD can only be recovered once.[4] For example, COD recovery as biogas that is combusted to recover electricity and heat may reduce the potential to recover a non-energy carrier product from COD, like *e.g.* biopolymers including EPS.[20] Therefore, to make rational decisions in future WRF process design it is crucial to compare a spectrum of technically possible process designs potentially applicable to a site of investigation at an early design stage.[17] To identify the most feasible process design among possible alternatives requires careful analysis. The quantification of potentially recoverable resources and trade-offs between alternative recovery options enhances the understanding of resource recovery technology integration into a treatment process and therefore can support decision making in WRF design. In addition, estimating how much of a certain product could potentially be recovered on-site of a WRF is essential to create value chains and market the product successfully because too little quantities may not be competitive on relevant markets.[20,34]

This can be achieved through mass and energy balances (MEBs) which allow to model how wastewater constituents are converted in a particular treatment process.[35] In comparison to each other they provide insight on how integrated recovery technologies may influence each other in terms of recoverable products from a process. Therefore, the comparison based on MEBs of different process designs potentially applicable to a municipal wastewater stream helps to find the process that is most feasible from a resource recovery perspective.[36] Since MEBs reveal the concentrations of selected constituents in all streams within a process, also the effluent quality that a process produces can be estimated from the results.

However, optimal resource recovery technology integration always depends on site specific circumstances of the treatment process because influent compositions, treatment technologies, or local market conditions for recovered resources may vary greatly.[35,37] Therefore, this analysis exemplifies selected resource recovery potentials and trade-offs along the case of the Utrecht WWTP in The Netherlands. The plant uses the AGS treatment process and was designed without any resource recovery technology integration. We conducted a MEB for the process and compared it to 5 different theoretical process designs that would recover COD and P on-site at this plant. COD would be recovered either as energy derived from biogas (electricity/heat from methane combustion) or as EPS. P would be recovered on-site the plant as struvite mineral. This way recoverable quantities of these products and trade-offs between certain design choices are revealed. Consequently, the study at hand contributes to the transition towards WRFs by providing insights on potentials of COD and P recovery integration into AGS-based WWTPs. It aims to answer the question: how to integrate COD and P recovery technologies into an AGS treatment process to improve decision making in AGS based WRF design in the future?

Materials and methods

Current process and general assumptions

The current process is based on a WWTP in Utrecht (The Netherlands) which operates six aerobic granular sludge reactors in parallel, each with a capacity of 12 thousand m^3 to treat wastewater of 308 thousand person equivalents in total. The plant recovers no resources on-site, but only off-site at an external sludge incinerator that recovers COD as electricity and P from the leftover ash fraction (ash-P). In the first treatment step, the influent is screened for coarse materials that may cause operational problems before it enters the biological treatment stage. During the following AGS treatment aerobic and anaerobic conversions of constituents take place simultaneously in a three-step cycle of anaerobic fill and draw, aeration and settling. For operational details of an AGS treatment plant we refer to ref. 5. After the biological treatment, the surplus granular sludge is thickened by gravity. Data to model the plant wide conversions of measured influent constituents in different process unit operations have been obtained from literature. A detailed list of parameters applied at each operational unit is provided in the appendices. Modelled process designs and corresponding COD and P flows are depicted by Sankey diagrams where flow sizes are proportional to the influent concentration.

For simplification reasons, several general assumptions have been made for different unit operations and we want to highlight the most important ones: influent COD fractionation values for biodegradable (easily, slowly, inert) and soluble and particulate COD fractions vary considerably in literature and are site specific.[38–40] They have not been measured for the influent and therefore COD conversions are modelled using the total COD value. The energy content of COD is assumed to be 17.8 kJ g^{-1}.[41] The coarse screen, also known as grit removal, is modelled to have no impact on COD and P flows although it has been shown in literature that COD containing solids may be removed.[42] The same applies for the gravity thickening of surplus sludge. Although the excess water flow from sludge thickening is usually redirected into the process[43] and likely contains minor very minor COD and nutrient fractions, those have not been modelled. The energy conversion efficiency of the sludge incineration unit is assumed to be 40% (electric efficiency).[44] Moreover, it is assumed that waste sludge arrives at the incinerator with a dry solids (DS) content of 22% which represents the Dutch average. This implies that over 70% of the sludge COD energy content is needed to evaporate the water until a sludge DS content is reached that is energy positive.[45]

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Oxygen requirements in the NEREDA reactor are determined by COD oxidation and nitrification. The aeration phase is modelled with a 60% COD oxidation where organics are oxidized to CO_2.[46] Total Kjeldahl nitrogen (TKN) inflows into the AGS reactor likely consists to *ca.* 70% of ammonia while the rest is organic N.[40] The latter will almost entirely be converted into ammonia during the treatment process.[47] Consequently the TKN load to the NEREDA reactor consists almost entirely of ammonia which is removed with a rate of over 90%.[48] *Ca.* 20% of the removed TKN ends up in the sludge and the rest is converted to nitrogen gas *via* nitrification–denitrification. The nitrification needs to be accounted for in the estimation of oxygen requirements. The denitrification replaces oxygen for COD removal with a stoichiometry a 2.86 g oxygen per g NO_3–N which lowers the total oxygen requirements.

The recoverable resource quantities are sensitive to the various assumptions made for the different parameters applied in the mass and energy balances. Therefore, a sensitivity analysis has been conducted on those parameters that are reported in literature with a certain degree of uncertainty. It reveals which unit operations need to be optimized to increase yields of a resource recovery pathway and influence its trade-off to another pathway. Parameters most sensitive to recoverable resource quantities are discussed in the results section whereas the detailed results of the sensitivity analysis are available in the ESI.†

Modelled process designs for on-site resource recovery

As explained above, the current process does not recover any resources on site but only off-site from sludge incineration where COD is recovered as electricity and P from incineration ashes. To explore how COD and P could be recovered on-site the treatment plant, the current process was theoretically re-designed into five different configurations Table 1.

On-site COD recovery is integrated by anaerobic digestion (AD) and subsequent combined heat and power (CHP) recovery from obtained methane combustion. The total-COD into biogas conversion rate is assumed to be 50% as measured by[32] under mesophilic conditions. The digestate is handled in a decanter centrifuge from which a liquid supernatant stream is redirected into the AGS reactor as commonly seen in WWTPs.[49] AD has been modelled in combination with/and without integration of polymer based CEPT which diverts primary COD into AD and therefore represents a process for maximized on-site energy recovery. In addition to AD and CHP integration for energy recovery, the second on-site COD recovery integration is EPS extraction from surplus granular sludge. The granular sludge harvested from the AGS reactor consists to 20% of EPS[50] and the EPS downstream processing is assumed to have no losses.

In addition to COD, also P can be recovered on-site the plant as struvite which can be precipitated from the liquid supernatant that is produced during sludge centrifugation. It is assumed that total-P (TP) in the influent consists of two thirds of solubilized *ortho*-P and one third of P bound to organics.[51] Most organic-P is modelled to be converted into *ortho*-P during the biological treatment[52] and therefore can be potentially recovered as struvite afterwards. *Ortho*-P is reactive and may precipitate due to the presence of Fe, Ca, or Al in the influent. *Ortho*-P binding to inorganic substances will strongly depend on local conditions, like *e.g.* pH, temperature, mineral and P concentrations present in treatment reactors.[6] For simplification reasons, the model accounts only for *ortho*-P bound to Fe because it has been

Table 1 Overview of modelled process designs analysed by mass and energy balances

Site of recovery		On-site recovery		
Resource		COD		P
Recovered product		Electricity/heat	EPS	Struvite
Process unit		AD/CHP	Chemical extraction	Crystallization
Designs	Current	·	·	·
	AD	✓	·	✓
	AD/CEPT	✓	·	✓
	EPS	·	✓	·
	AD + EPS	✓	✓	✓
	AD/CEPT + EPS	✓	✓	✓

shown that it is likely to be present in Dutch influents with an average concentration of 1 mg l^{-1}. Furthermore, is expected that all of this Fe is divalent and therefore leads to vivianite (Fe$_3^{2+}$(PO$_4$)$_2$·8H$_2$O) formation. The *ortho*-P fraction precipitated as vivianite is therefore not recoverable as struvite later in the process.[23] Following these assumptions and stoichiometry, 1 mg of divalent Fe present in the influent would bind 1.1 mg of *ortho*-P that is not available for recovery anymore except as vivianite. However, most *ortho*-P is accumulated in biomass during biological treatment before it is re-mobilized during AD[26] and therefore, the liquid supernatant produced in the decanter centrifuge for digestate handling is rich in solubilized P and ideal for struvite crystallization.[53]

Results and discussion

In the following paragraphs the results of modelled process designs are discussed regarding their resource recovery potentials and trade-offs between resource recovery technology integrations. Despite the growing importance of resource recovery in wastewater treatment the production of clean water for environmentally safe discharge remains also the major objective of WRFs. Therefore it is important to state that all modelled process designs would meet Dutch legal effluent concentrations of COD, P-total, and TKN (see also ESI†). Table 2 summarizes the results of modelled process designs expressed as influent-COD and influent-P recovery rates.

On-site COD recovery as energy

Fig. 1(A) shows COD flows in the current process with an off-site electricity recovery potential from sludge incineration above 9 MW h d^{-1}. The integration of AD and CHP allows to recover a fraction of the influent-COD on-site as electricity and heat. The recovery of energy from COD on-site implies a trade-off with off-site electricity recovery of −60% (Fig. 1(B)). The recovered electricity can directly be consumed on-site the plant to reduce its energy consumption from the grid or can be supplied to it for off-site usage which would imply some losses through *e.g.* the Joule effect in transformers and power lines.[13] The recovered heat can be used on-site for different

purposes, like *e.g.* for heating of the anaerobic digester or for waste sludge drying.[54] In addition to on-site energy recovery, the integration of AD serves also the purpose of waste sludge volume reduction which can be expected to be in the range of 30–50%.[55] Therefore AD integration likely decreases costs for sludge transport to the incinerator. If a CEPT unit is integrated additionally the on-site energy recovery can be more than doubled compared to only AD integration (Fig. 1(C)). To ensure a sufficient denitrification in the AGS reactor it is important to maintain a high enough biodegradable-COD (bCOD) which is according to the model over 4 gbCOD/gN, even in those designs with CEPT integration where a large COD fraction is removed before AGS treatment.

CEPT integration increases not only the energy recovery potential on-site but also off-site compared to AD integration alone. This is due to the fact that much less COD is oxidised to CO$_2$ in the AGS reactor when CEPT is integrated and more COD enters the AD. Since AD converts the COD only up to 50% to biogas,[32] relatively more influent-COD enters the incinerator as sludge if CEPT is integrated compared to AD integration alone. Thus, the total influent-COD recovery rate (on-site + off-site) can be doubled with CEPT integration as elucidated in (Fig. 2).

The total-COD into biogas conversion rate of 50% is generally assumed disregarding the loads of waste activated sludge (WAS) or primary sludge (PS) into AD. In reality, the COD into biogas conversion rate might differ slightly depending on whether WAS or a mix of WAS and PS is loaded to AD because PS has a higher anaerobic COD biodegradability than WAS. The COD biodegradability of the latter largely depends on the solids retention time (SRT) in the aerobic treatment. The higher the SRT, the more recalcitrant the WAS is to further biological degradation and thus, the lower the conversion into biogas may be.[56] Being aware of this, the model may slightly overestimate the biogas yield in the "AD" design. The sensitivity analysis confirms that the total-COD into biogas conversion rate has a high impact on recoverable energy quantities on-site. This is also valid for the assumed methane content of biogas of 65% which has been reported in several studies as a reasonable assumption for sewage sludge digestion.[16,43,57] Nevertheless,

Table 2 Summary of influent-COD and influent-P recovery rates of modelled process designs in %

Influent-COD recovery rates %

	Current	AD	AD/CEPT	EPS	AD + EPS	AD/CEPT + EPS
Electricity (on-site)	—	6.9	14.7	—	5.1	13.9
Heat (on-site)	—	7.3	15.5	—	5.4	14.6
EPS	—	—	—	8.8	9.0	4.1
Electricity (off-site)	4.1	1.7	3.5	3.0	1.2	3.3
Total	*4.1*	*15.9*	*33.7*	*11.8*	*20.8*	*35.9*

Influent-P recovery rates %

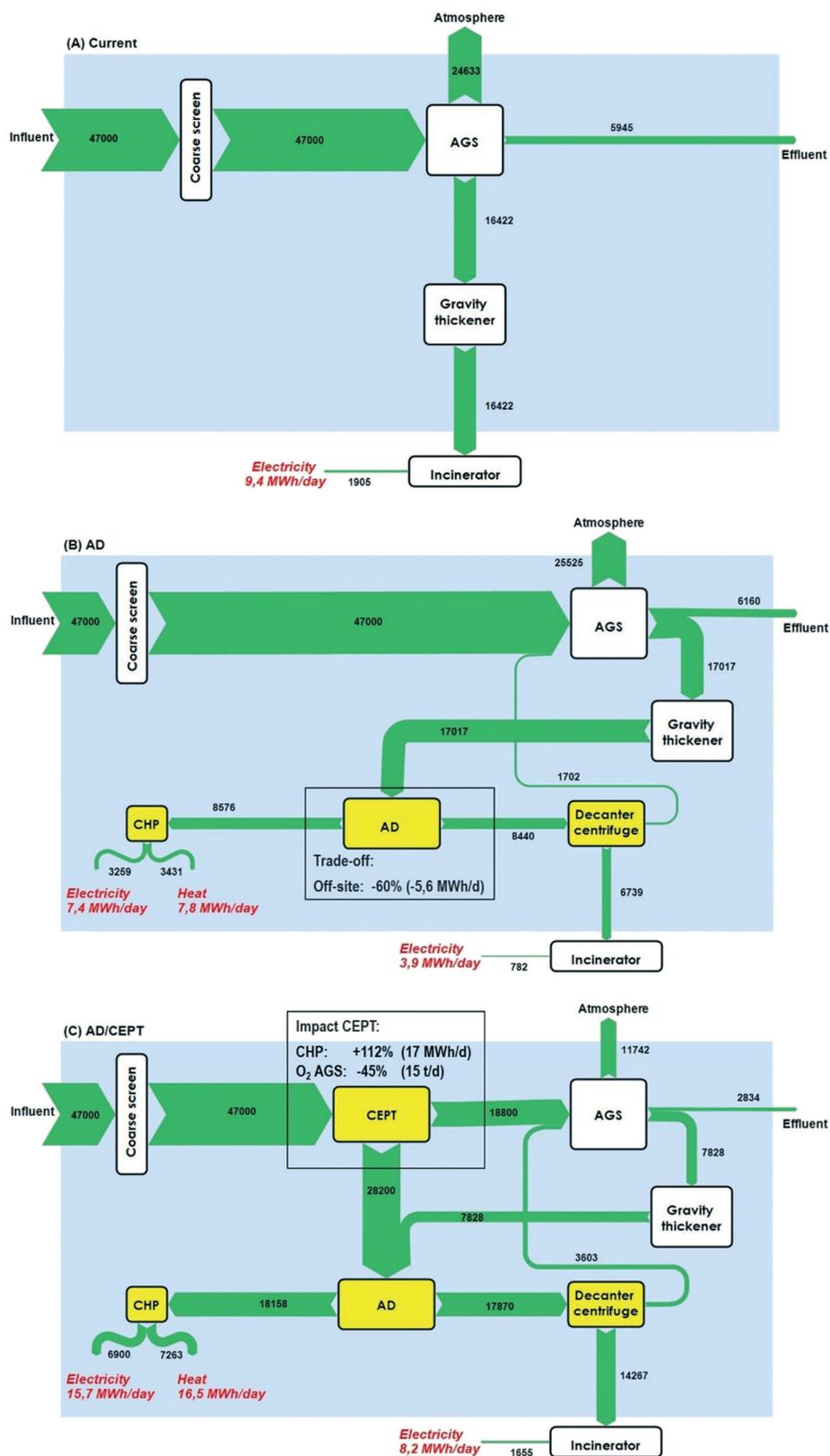|	Current	AD	AD/CEPT	EPS	AD + EPS	AD/CEPT + EPS
Ash-P	73.0	65.0	67.5	58.8	52.0	61.5
Struvite-P	—	9.3	9.6	—	7.3	8.7
Total	*73.0*	*74.2*	*77.1*	*58.8*	*59.3*	*70.3*

Fig. 1 Modelled COD flows in kg COD d^{-1} of (A) current process design with no on-site resource recovery, (B) AD and CHP integration to recover energy on-site, (C) CEPT integration for maximum energy recovery on-site. Yellow unit operations represent integrated units compared to current process design. Recoverable product quantities in red.
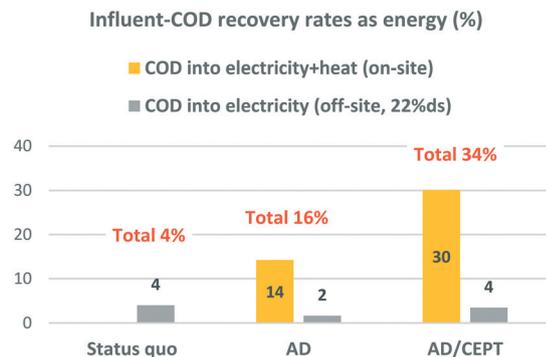
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Influent-COD recovery rates as energy (%)

■ COD into electricity+heat (on-site)
■ COD into electricity (off-site, 22%ds)

Fig. 2 Influent COD recovery rates as energy on-site, off-site and in total.

it is important to keep in mind that this value assumes steady and well-managed anaerobic sludge digester systems and can be lower due to, for example, high temperature variations or overloads.[15] Another assumption that is sensitive to the obtained energy on-site is the electricity/heat conversion efficiency of the CHP unit. The present study assumes a rather conservative efficiency of *ca.* 40% but depending on the size of the unit and its age there are increased efficiencies likely to be obtainable.[15] Finally, the COD removal rate of the CEPT has a significant effect on the energy that is recoverable from the biogas pathway and therefore keeping the assumed 60% COD removal rate steady is necessary for a sustainable on-site energy recovery strategy.

In addition to a higher on-site energy recovery potential, another favourable effect of CEPT integration is a decreased oxygen requirement in the NEREDA reactor by −45% due to lower COD and TKN loads (Fig. 1(C)). On the first glance CEPT integration may be promising in lowering GHG emissions of a process because it increases the methane recovery potential and decreases aeration. Nevertheless, both allegedly positive environmental effects can be severely offset by the necessary consumption of polymers that imply negative environmental impacts like abiotic depletion of elements and fossil fuel resources. This is also valid for cost–benefit calculations of CEPT units as polymers represent an additional cost factor.[35] Furthermore, also AD integration can lead to severe hidden direct CH_4 emissions which may even exceed emissions avoided through energy recovery from biogas combustion.[58] In addition, the integration of AD-based energy recovery technologies leads to higher operational costs as integrated units need regular maintenance like *e.g.* the CHP unit needs to be cleaned frequently from deposits that deteriorate its efficiency.[15] Therefore, the presented results of AD and CEPT integration should be interpreted as an intermediate step towards a more complete process assessment including additional technical, economic, and environmental performance criteria.

Another challenge is that the off-site energy recovery potential from sludge incineration is sensitive to the assumed water content of the sludge. This study assumes the Dutch average value of 22% DS content for waste sludge

which implies high energy recovery losses in the incinerator due to water evaporation requirements. This finally allows to recover less than one third of the energy loaded into the incinerator in form of organic matter.[45] Due to various possible sludge dewatering processes, each resulting in different water contents, DS content of waste sludge is reported with great variations in literature. Because the sensitivity of the assumed DS content to the recoverable electricity from sludge incineration is high, it needs to be adjusted for site specific circumstances before one aims to transfer the results of this study to another case. Another sensitive assumption in the off-site electricity recovery pathway is the electrical efficiency of the incinerator. It is assumed that 40% of the loaded dry solids that remain after evaporation energy subtraction are converted into electricity. But depending on the type and age of the incinerator this efficiency may also be lower.[44,59]

It is also likely that surplus heat from the incineration process can be used to further increase the DS content of the sludge before incineration.[60] Therefore, the estimated off-site energy recovery potential from the incinerator might be an underestimation and is strongly dependent on site specific conditions.

On-site COD recovery as EPS

Energy recoverable from COD has often been referred to as the second most valuable resource in wastewater after water.[17] More recently it has been argued that, following the principles of circular economy and sustainability, COD should be preferably recovered as materials than as energy. This argumentation follows two reasons. First, COD in wastewater contains only a small fraction of the energy because thermal energy recoverable by water sourced heat pumps contains more energy and therefore has a higher potential to save carbon emissions. Secondly, COD has a relatively large exergy content which should be preserved thus converted into carbonaceous materials.[54] The integration of EPS recovery from surplus granular sludge serves this rational. If EPS extraction would be integrated into the current process, more than 3 t of EPS product could be recovered daily, assuming no losses in polymer downstream processing (Fig. 3(A)). In combination with AD integration a trade-off between EPS and energy recovery occurs due to a fraction of COD leaving the system as EPS which is therefore not fed to AD. This is reflected in a decreased on-site energy recovery potential of −26% compared to no EPS recovery integration (Fig. 3(B)). Integrating CEPT additionally to maximize on-site energy recovery implies less COD is fed to the granular sludge which essentially produces the EPS. Consequently, CEPT integration implies that the EPS recovery potential is halved while on-site energy recovery is more than doubled compared to only AD integration (Fig. 3(C)).

When AD is integrated in combination with CEPT, EPS recovery does not necessarily imply a significantly decreased on-site energy recovery potential in comparison to AD
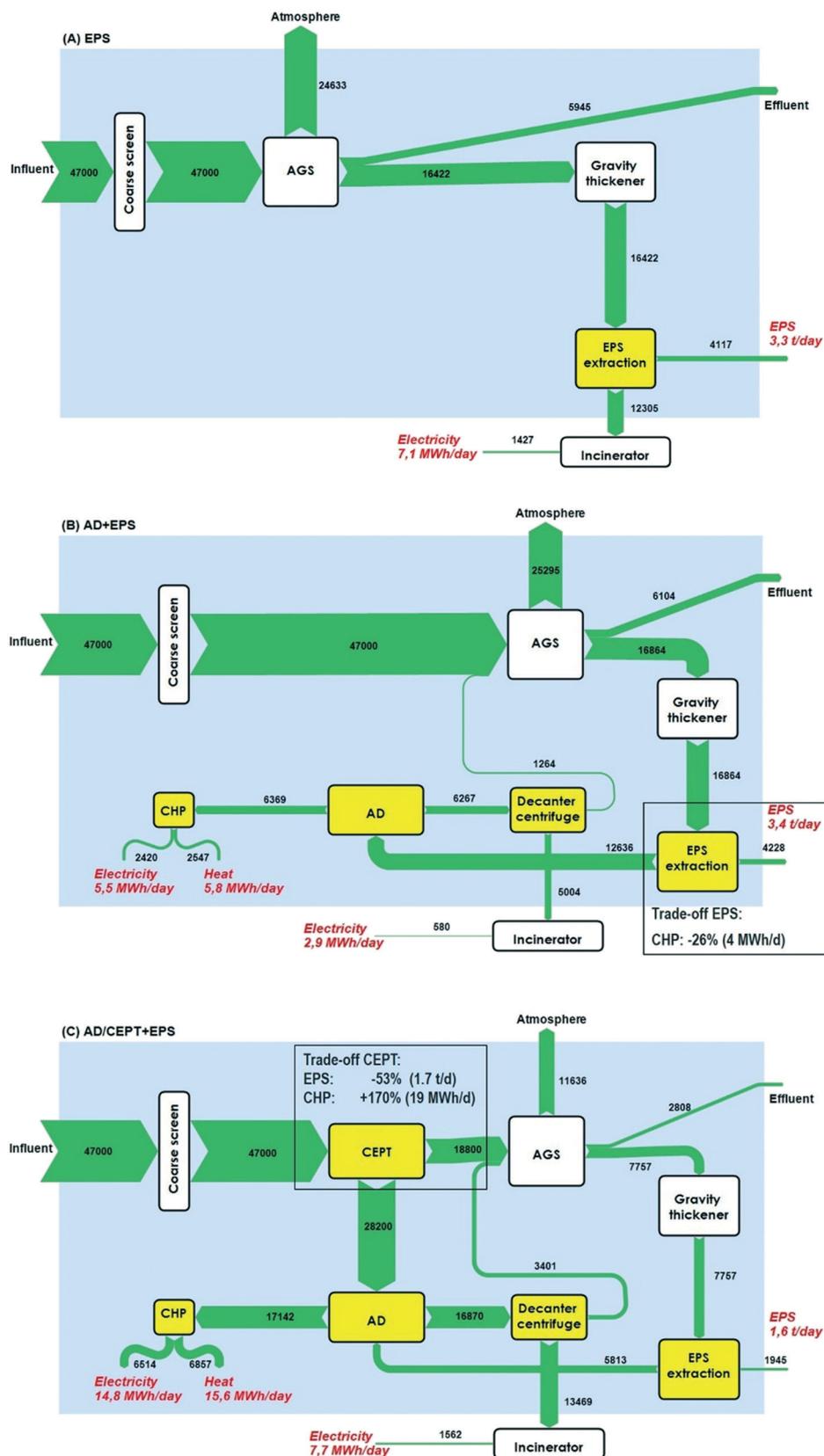
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Fig. 3 Modelled COD flows in kg COD d^{-1} of (A) EPS recovery integration, (B) AD and CHP integration to recover energy on-site and EPS recovery integration (C) CEPT integration for maximum energy recovery on-site and EPS recovery integration. Yellow unit operations represent integrated units compared to current process design. Recoverable product quantities in red.
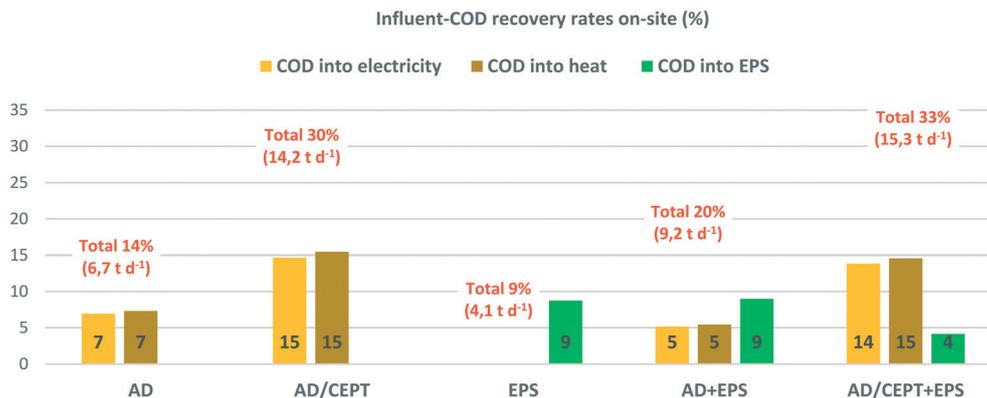
Influent-COD recovery rates on-site (%)

■ COD into electricity　■ COD into heat　■ COD into EPS

Fig. 4 On-site influent-COD recovery rate of analysed process designs in % and in total values (tonnes per day).

integration alone. The difference in energy recovery of the designs "AD/CEPT" and "AD/CEPT + EPS" is just marginal. This is due to the high on-site energy yields obtained with CEPT integration. The elevated energy yields overrule by far the energy losses resulting from COD leaving the process as EPS before it can enter AD. In total, up to one third of influent-COD, which equals *ca.* 15 t COD per day could be recovered on-site in form of energy and/or EPS with the examined COD recovery technology integration (Fig. 4).

It should be mentioned that the extraction of EPS from the sludge requires chemicals as inputs[9] which need to be accounted for in subsequent economic and environmental impact assessments of those designs which integrate EPS recovery. Furthermore, alkaline pre-treatment of sludge prior to AD has been reported to increase the anaerobic degradability of organics[61] and possibly biogas yields.[62] Since EPS is extracted from the granular sludge with an alkaline process followed by an acid precipitation step,[63] its recovery leaves an alkaline waste stream and therefore may function simultaneously as an effective sludge pre-treatment step. Due to this pre-treatment-like function EPS extraction could lead to increased biogas yields which would reduce the trade-off between EPS and AD-based energy recovery. Maintaining the high pH after alkaline sludge disintegration and applying it also in an alkaline AD may turn the fact that EPS extraction produces a haloalkaline waste stream into another advantage. A few preliminary studies produced biogas with haloalkaline microbial consortia obtained from soda lake sediments.[64,65] Since the CO_2 produced during AD remains solubilized in the broth under alkaline conditions, one of the main advantages of alkaline AD applied to sludge waste from EPS extraction would be a methane rich biogas stream (>95%) that can be used directly as a fuel which would make expensive biogas upgrading redundant.[66] Nevertheless, whether a large scale alkaline AD is applicable and whether the methane yields of such a system are comparable to the ones of lab scale experiments, remains uncertain.

Finally, it should be mentioned that recoverable EPS quantities are sensitive to several assumptions made in the model. Obviously, as already discussed above EPS recovery

can be significantly lowered the more COD is removed in the CEPT unit. Another sensitive parameter is the assumed EPS downstream yield of 100% which is likely to be lower in reality and has been chosen based on lab scale experiments because data from large scale applications are yet missing. Also the assumed parameters related to COD conversions in the AGS reactor determine the EPS recovery significantly. The sensitivity analysis reveals that the assumed total COD removal rate, the fraction of removed COD ending up in the sludge, and the EPS content of the sludge are all positively correlated to the recoverable EPS quantities. This should be kept in mind when transferring the results of this study to other cases where COD conversions taking place in an AGS reactor may differ due to site specific factors. For example, the easily biodegradable COD fraction of the influent influences recoverable EPS quantities and may differ greatly between cases. A key aspect to consider in EPS recovery estimation based on total COD instead of COD fractions is the pre-treatment applied before the AGS reactor as different pre-treatments may retain different rates of slowly biodegradable COD. Especially cellulose may play a vital role in how much of the total COD is converted into EPS because depending on the operational settings it may be hardly degraded during aerobic treatment[67] but still accounted for in a model like the one presented in this study. To sum it up, modelling the mass conversions of particular COD fractions instead of total COD may further improve the accuracy of the EPS recovery pathway.

On-site P recovery as struvite

Fig. 5(A) Shows P flows in the current process in which almost all P ends up in the sludge which is then incinerated off-site. After incineration *ca.* 400 kg P per day can be recovered from the ash fraction. If AD or AD/CEPT is integrated, on-site P recovery of *ca.* 50 kg struvite-P per day from digestate supernatant becomes possible (Fig. 5(B) and (C)). The polymer based coagulation applied in the CEPT leads to a much higher accumulation of influent-P in the obtained primary sludge compared to normal primary
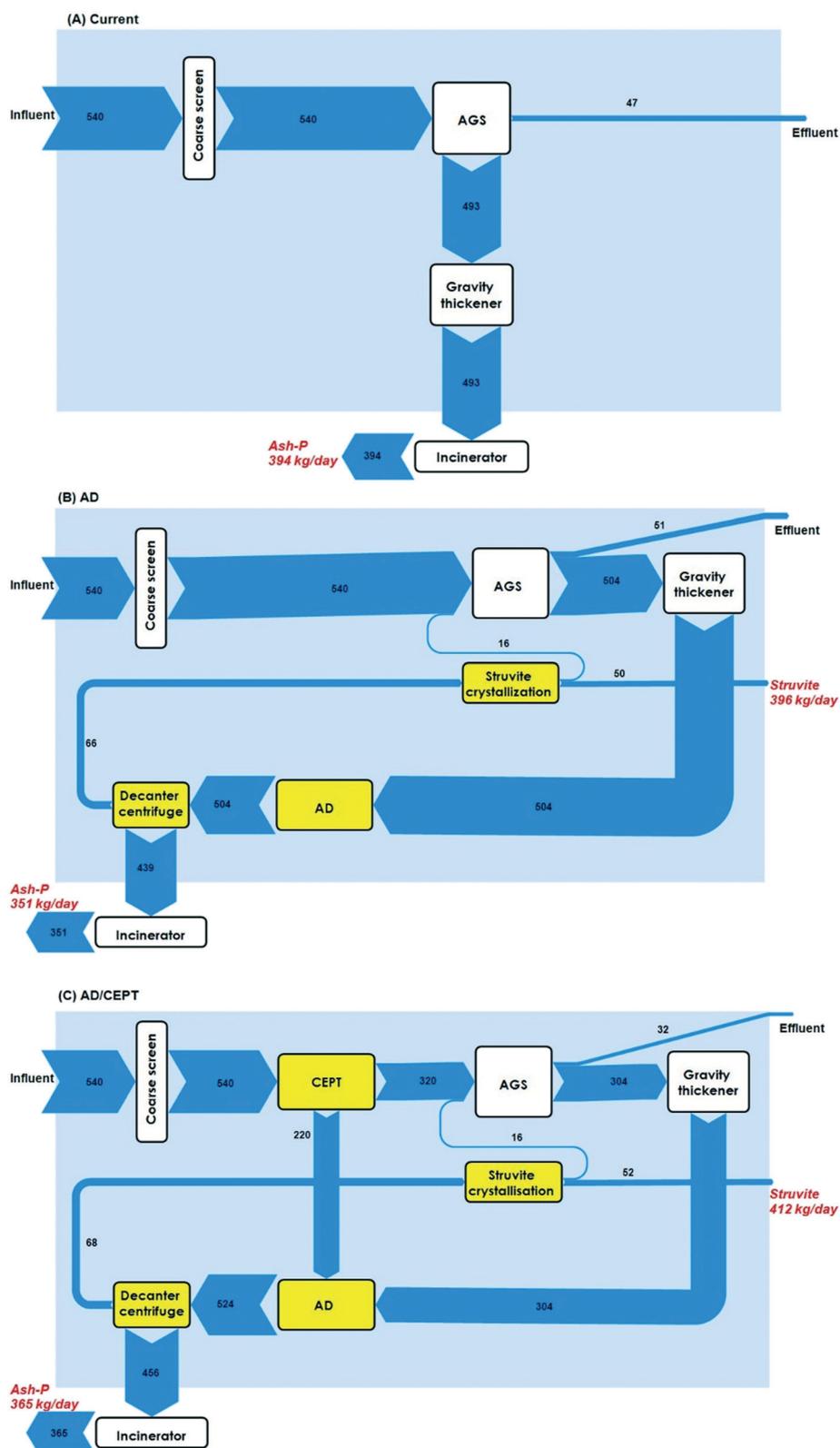
Fig. 5 Modelled P flows in kg P per day of (A) current process design with no on-site resource recovery, (B) AD and CHP integration and subsequent struvite recovery on-site, (C) CEPT integration and subsequent struvite recovery on-site. Yellow unit operations represent integrated units compared to current process design. Recoverable product quantities in red.
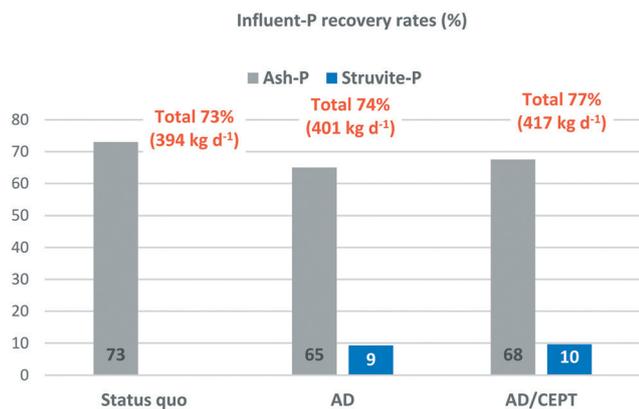
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Fig. 6 Recoverable influent-P rates in % and total P recoverable also in kg per day.

settling.[68] Consequently the integration of CEPT into a process design means that a significantly higher influent-P fraction enters AD instead of the AGS reactor which may also lower P effluent concentration significantly.

Fig. 6 shows that struvite recovery integration increases the total influent-P recovery rates only very minimally because recoverable quantities of struvite-P are very small compared to ash-P quantities. The reason is that only a small fraction of total-P loaded to AD can be expected to end up in the supernatant as soluble *ortho*-P because 80–90% of loaded P remains in the digestate.[32] In addition, 80% of this soluble *ortho*-P can be recovered as struvite crystals.[69] This leads to overall low influent-P recovery rates in the form of struvite-P. In addition, for successful struvite crystallization the molecular ratio of magnesium, ammonia and *ortho*-P (Mg^{2+}: NH_4^+: PO_4^{3-}) should be $1:1:1$.[14] This leads to the required mass of at least 0.8 kg Magnesium per 1 kg P precipitated which represents another cost and environmental factor to be considered for the struvite recovery pathway. For these reasons, it is questionable to invest in a struvite crystallization unit if ash-P recovery from sludge incineration is possible because most influent-P ends up in the sludge and is not available for struvite crystallization.
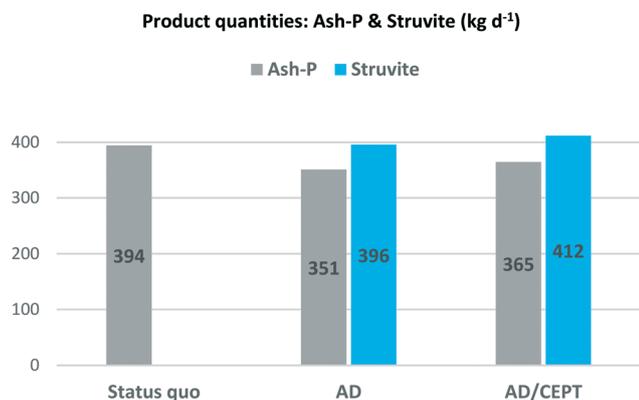

Fig. 7 Recoverable P-fertilizer product quantities in kg per day.

Struvite is a mineral and following its stoichiometry it contains only *ca.* 13% of P while the rest of its mass is ammonia, magnesium and crystal water ((NH_4)$Mg[PO_4]$·$6H_2O$). Considering its total mass, recoverable struvite masses are comparable to recoverable ash-P masses (Fig. 7). One can therefore argue that struvite is equally promising to be marketed as a fertilizer product because it can be supplied in comparable quantities as ash derived P. Since it can be recovered on-site, it may generate revenues for the water utility operating the plant while ash-P revenues are generated externally at the incinerator. In addition struvite recovery may prevent pipe clogging and therefore decrease operational costs.[24] The recovery of struvite from digestate supernatant is mostly sensitive to two process parameters: Firstly, the assumed fraction of P that ends up in the liquid supernatant after centrifuging the digestate (assumed to be 13%), and secondly, the crystallization rate of *ortho*-P during struvite formation (assumed to be 80%). Although assumed values for both mechanisms have been reported in literature,[32,69] there is more research needed to reveal how both parameters can be enhanced in the future to increase struvite precipitation from side streams.

P recovery from sludge incineration ashes requires the realization of dedicated sewage sludge incinerators that are expensive to build.[70] Still, from an overall societal P recovery perspective, the ash-P recovery route has clear advantages because it can bundle the excess P streams from several WWTPs. A standalone sludge incinerator can therefore function as a centralized P recovery unit in a region and may make use of economy of scale to recover high rates of regionally consumed P. There are yet various uncertainties on how to optimize the ash-P recovery route. There is little research published on P recovery rates from sludge incineration ashes and the question remains to what extend the assumed 80% (ref. 71) can be further increased as it is a sensitive value to the results of this study.

1 t EPS contains *ca.* 30 kg P and therefore a trade-off between EPS and P recovery exists leading to *ca.* −20% decreased ash-P and struvite recovery potential in those designs with EPS recovery integration compared to those with none (Fig. 8(A)). Obviously, if CEPT is applied the consequential lowered EPS recovery potential decreases this trade-off as less P is incorporated into EPS polymers and leaves the system as such (Fig. 8(B)). While P-fertilizers are relatively cheap,[72] EPS are a potentially high value product.[50] Therefore it is reasonable to argue that P recovery as EPS may be economically favourable over P recovery as fertilizer. But a reliable statement can only be made if EPS becomes an established product on biopolymer markets and a complete cost–benefit analysis of the investigated process designs is carried out considering site specific circumstances.

Conclusion

The results show how to integrate COD and P recovery technologies into an aerobic granular sludge (AGS) treatment

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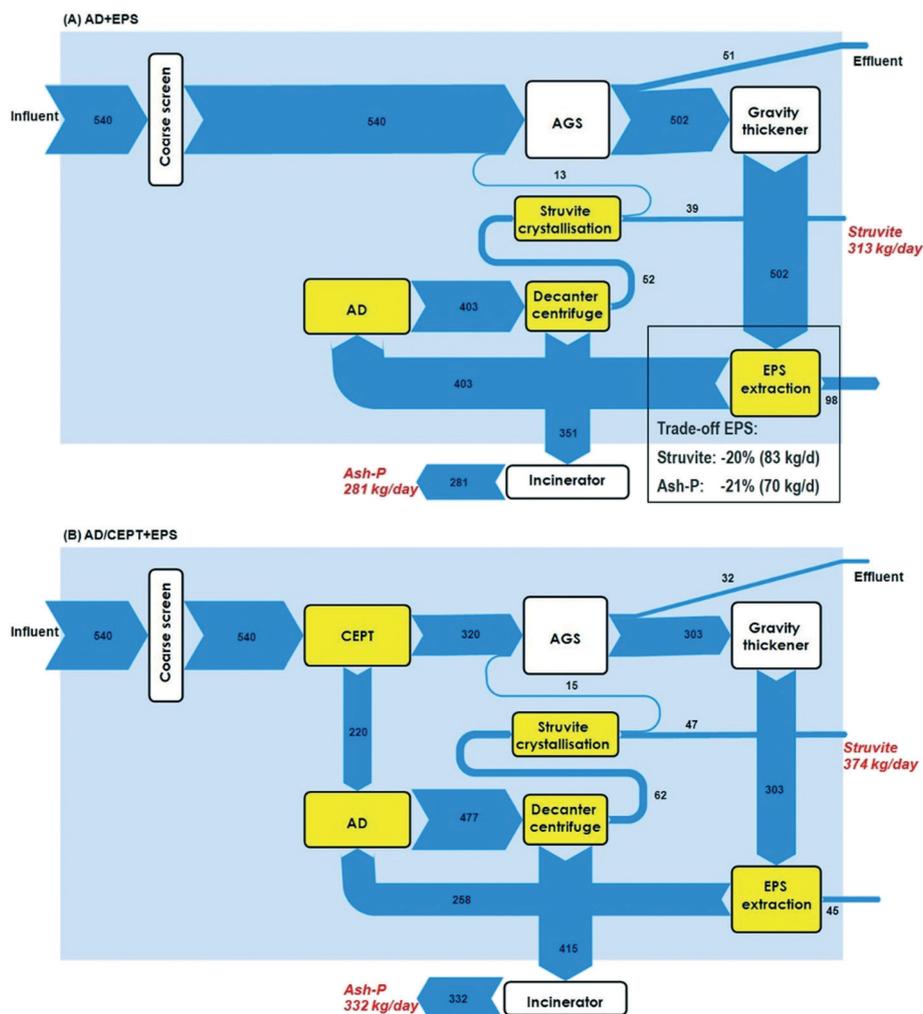This journal is © The Royal Society of Chemistry 2020

Fig. 8 Modelled P flows in kg P per day of (A) AD + EPS design (B) AD/CEPT + EPS design. Yellow unit operations represent integrated units compared to current process design. Recoverable product quantities in red.

process and therefore improve decision making in AGS based water resource factory (WRF) design in the future. Mass and energy balances allow to quantify recoverable products as well as to analyse trade-offs occurring from possible design choices and are therefore an important tool in WRF process design. Significant quantities of resources can be recovered on-site of existing aerobic granular sludge treatment plants. Regarding AD integration, the results of this study confirm previous studies who state that the fraction of influent COD recoverable by AD and CHP integration is rather low.[73,74] Nevertheless, the integration of AD for on-site recovery of electricity and heat may lower the net-energy consumption of a process and also may increase the total energy recovery from COD substantially compared to only sludge incineration. Furthermore, the conducted MEBs show that the integration of COD recovery *via* CEPT for maximum energy and/or *via* EPS recovery can increase the total COD recovery rate significantly. Up-concentrating primary COD by CEPT can significantly increase the energy recovery potential from COD compared to only secondary sludge digestion

because it prevents COD oxidation to CO_2 but leads to a high fraction of influent-COD recovered as energy, either anaerobically or through incineration. Therefore, CEPT integration halves aeration requirements of the plant and since aeration is a major operational cost factor of aerobic wastewater treatment processes[75] it may lower operational costs significantly. Despite these promising results, the overall performance of CEPT integration can only be revealed if required polymers are considered in complete process economic and environmental impact assessments.

The model shows that EPS recovery integration promises to recover several tonnes of biopolymers daily which could generate substantial economic revenues for the plant. The recovery of EPS means that a fraction of the influent COD and P leaves the process as polymers and is therefore not recoverable in other forms any longer. On the contrary, the integration of CEPT for maximum on-site energy recovery decreases the EPS recovery potential significantly. Thus, a clear trade-off between maximum EPS and maximum energy recovery exists. The integration of struvite recovery is

questionable when ash-P recovery is a possibility because the recoverable influent-P rates are much lower for struvite than for ash-P. Therefore, the total influent-P recovery rate is not significantly improved by struvite crystallization from digestate supernatants. Nevertheless, *ca.* 400 kg of struvite could be daily recovered on-site the plant and when marketed successfully could generate additional revenues to the operating utility.

The revealed trade-offs between resource recovery technology integration, show that it is important during the early design stage of a WRF to decide which resources that are potentially recoverable from the wastewater stream should be preferred over others and why. This requires inclusion of case related arguments. The integration of

- AD (including decanter centrifuge and CHP unit),
- CEPT,
- EPS extraction, and
- struvite crystallization

into an AGS-based treatment plant will alter its technical, economic and environmental performance. To identify the most feasible process design each possible process alternative needs to be assessed further in these dimensions. This study shows that mass and energy balances are useful in early stage WRF design because they provide the basis for those subsequent assessment steps. In addition, the balances allow an early estimation about which processes are promising in terms of the quantitative resource recovery potential which provides important insights for the development of value chains for recovered resources. Since mass and energy balances require only data, they are relatively cheap to conduct and are therefore an excellent tool to assess a WRF process design at a very early design stage regarding its resource recovery potential and effluent concentrations of modelled constituents. Thus they provide a useful basis to pre-select promising designs for further in depth and more costly techno-economic and environmental impact assessments. To further improve the precision of predictions from comparable mass and energy balances it is useful to apply COD fractionation into easily and slowly biodegradable and non-biodegradable COD. Another parameter that has to be applied with greater accuracy in the future is the yield of full scale EPS down streaming processes as data is yet unavailable. The first commercial EPS recovery process in Zutphen (The Netherlands) will hopefully reveal more insights on this and other parameters related to successful EPS recovery up-scaling. In addition, this study suggests to investigate sludge incineration further as it is a key process regarding both COD and P recovery potentials. Uncertainties remain on how to optimally decrease the water content of waste sludge to maximize its heating value and what factors determine the recoverability of useful P from incineration ashes.

List of abbreviations

AD	Anaerobic digestion
AGS	Aerobic granular sludge
bCOD	Biodegradable chemical oxygen demand
CAS	Conventional activated sludge
CEPT	Chemically enhanced primary treatment
CHP	Combined heat and power
COD	Chemical oxygen demand
DS	Dry solids
EPS	Extracellular polymeric substances
GHG	Greenhouse gas emissions
MEB	Mass and energy balance
N	Nitrogen
P	Phosphorous
SRT	Solids retention time
TKN	Total Kjeldahl nitrogen
WRF	Water resource factory
WWTP	Wastewater treatment plant

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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