Environmental sustainability of municipal wastewater treatment through struvite precipitation: Influence of operational parameters

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Abstract

The environmental sustainability of wastewater treatment through phosphate (P) and ammonia (N) chemical precipitation (struvite) was examined using the life cycle assessment methodology. Thermally activated (calcined) cryptocrystalline magnesite was used towards struvite formation and four process parameters (contact time, magnesite dosage, initial wastewater pH and temperature) were studied. Raising wastewater's temperature to promote ammonia stripping was found to be environmentally unsound. Magnesite dosage and contact time were identified as environmental hotspots, but not pH. In terms of environmental relevance, when using ReCiPe 2016 life cycle impact assessment method the human health damage category was mostly affected, followed by resource availability, while ecosystems category was affected to a much lesser extent. Environmental impacts were grossly attributed to South Africa's fossil fuel-dependent energy mix, suggesting that renewable energy could largely improve the system's environmental performance. The optimal conditions, from the environmental perspective, were found to be 0.2 g L^{-1} feed dosage and 10 min mixing, at ambient temperature and pH (total environmental footprint 60.9 μPt per treated L of wastewater). To improve N removal efficacy, which is desirable in real-world applications, higher feed dosages and mixing durations are required, albeit at the expense of environmental sustainability (e.g. the 180 min and 16 g L^{-1} dosage environmental footprint sharply rises to 1.87 mPt L^{-1}). The net impact approach revealed that the avoided environmental impacts on the midpoint impact categories freshwater and marine eutrophication grossly outweigh the corresponding environmental impacts of the treatment process. Results suggest that when using a low contact time and magnesite dosage, struvite precipitation could act as a fast, efficient, and environmentally friendly pre-treatment step to practically remove P and grossly reduce N from wastewater. On the other hand, with higher feed dosages and mixing durations struvite precipitation could act as a promising secondary wastewater treatment method with an overall low environmental footprint. Overall, results complement the existing body of

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knowledge on the techno-economic performance of such systems and provide insight to decision- and policy-makers to sustainably scale the process, at village- or industrial-level, in rural South Africa, Lesotho, and further afield.

Keywords: wastewater valorisation; waste beneficiation; low- and middle-income countries (LMIC); magnesium ammonium phosphate (struvite - NH4MgPO4•6H2O); SimaPro; net environmental benefit (NEB) approach.

Introduction

The enrichment of natural water bodies with high nutrient load, typically phosphate (P) and ammonia (N), comprise a growing problem of environmental concern that grossly affects low- and middle-income countries (LMICs). The problem is concomitantly perpetuated by an ever-growing population that proportionally increase nutrient enrichment, typically through untreated or poorly treated wastewater discharges to different receiving aquatic ecosystems. Among others, nutrients encourage the prolific propagation and growth of aquatic plants, which ultimately become a severe problem when they die and start decomposing, since oxygen depletion in water will ensue (Yagi and Fukushi, 2012). An aquatic ecosystem deficient in dissolved oxygen will suffocate and kill aquatic organisms (Masindi et al., 2016b, c). In addition, excessive or dead organisms block light penetration in water, hindering photosynthesis, while the aesthetic value of the ecosystem is also affected by plant infestations and dead organic matter (Peng et al., 2018). However, nutrient discharges to surface water is on the rise, primarily in LMICs (van Puijenbroek et al., 2019).

The above highlight the need to limit the amount of pollutants discharged into receiving aquatic ecosystems. This can be achieved through the proper management of the growing wastewater quantities and a promising method towards this end is P and N precipitation from wastewater, preferably using inexpensive coagulants such as magnesium, with the added benefit of struvite ($NH_4MgPO_4•6H_2O$) formation. Magnesite rock is a relative inexpensive source of magnesium that is readily available in many regions around the world, including LMICs such as South Africa and Nepal where capital and infrastructure for wastewater treatment is weak or non-existent (Krähenbühl et al., 2016; Masindi et al., 2016b, c). Struvite is also a highly effective source of nitrogen, magnesium, and phosphorus that can act as a replacement of chemical fertilisers in the agriculture industry (Luján-Facundo et al., 2019) and particularly as a replacement of rock phosphate which reserves are rapidly dwindling (Ceasar, 2018). This highlights that wastewater sludge can be used for materials recovery in

78 the circular economy context (Gherghel et al., 2019), and especially for phosphorus (struvite)
79 recycling (Jedelhauser and Binder, 2018).

80 The technical and economic perspectives of struvite recovery systems have been extensively
81 studied (Daneshgar et al., 2019; Peng et al., 2018;Yu et al., 2017). However, this is not the
82 case for the environmental perspective, where the body of knowledge is restricted to a small
83 number of studies, primarily focusing on the developed world (Pausta et al., 2018; Zhou et
84 al., 2019). This highlights the need for further research to comprehensively consider the
85 environmental dimension of the process, with focus given on struvite recovery systems
86 outside of Europe (Lam et al., 2020; Sena and Hicks, 2018), and particullarly in LMICs
87 where the sustainable treatment of wastewater is a pressing matter of environmental concern
88 (van Puijenbroek et al., 2019). The environmental dimension of the process can be identified
89 using robust and well-established tools, such as the life cycle assessment (LCA) methodology
90 (Chatzisymeon et al., 2017; Ioannou-Ttofa et al., 2016). In this regard, the net impacts or net
91 environmental benefit (NEB) approach, which considers both the environmental impacts of
92 the wastewater treatment method and the environmental benefits from pollutants removal, i.e.
93 the impacts that are avoided from releasing untreated wastewater (Godin et al., 2012; Igos et
94 al., 2012; Köhler et al., 2012), can also be applied. However, even though this method seems
95 very promising for identifying the environmental benefits arising from pollutants removal
96 from wastewater, it suffers from: i) the need to include as many pollutants as possible (to
97 properly characterize the avoided impacts), and ii) large uncertainties, particularly in the
98 (eco)toxicity assessment (Igos, 2016).

99 South Africa and Lesotho, the latter a landlocked country encircled by the first, are LMICs
100 with weak to non-existent infrastructure, since in many rural areas wastewater usually returns
101 untreated to the environment. This highlights the urgent need for introducing robust and
102 versatile wastewater systems in rural and peri-urban areas. Considering that struvite recovery
103 systems are relatively simple to install and easy to operate, these can be installed in such
104 areas. South Africa is rich in cryptocrystalline magnesite, which, from the techno-economic
105 perspective, is promising for struvite precipitation (Masindi et al., 2016b, c; Mavhungu et al.,
106 2019). Nevertheless, to sustainably scale up the process its environmental sustainability,
107 which remains grossly unknown, should also be identified. This is the main goal of this LCA
108 study. Specifically, bench-scale data from a batch experimental procedure were used to
109 comprehensively examine four process parameters, i.e. contact time (i.e. mixing duration),

magnesite dosage, and initial wastewater pH and temperature. To the best of the authors' knowledge this is the first time that a comprehensive LCA study dealing with process optimisation, from the environmental perspective and when using South African thermally activated cryptocrystalline magnesite, is carried out. Furthermore, the results of this work provide insight into the environmental sustainability of struvite recovery systems in LMICs, which is largely under-reported (Sena and Hicks, 2018). As such, this manuscript makes available new LCA data for an area where the existing body of knowledge is limited (e.g. in a recent review paper by Lam et al. (2020) only 2 out of the 65 manuscripts that were reviewed were referring to Africa). It also provides insight to decision- and policy-makers to promote and sustainably scale up the process at village- or industrial-scale in South Africa, Lesotho and further afield.

2 Materials and methods

First, the materials and methods for struvite precipitation experimental setup are discussed and then the methodology and tools that were used for the environmental analysis are given.

2.1 Wastewater and magnesite rock collection

In the context of this work real municipal wastewater, rich in P and N, was collected from a treatment facility in Pretoria, South Africa and used for batch laboratory experiments. The collected wastewater was pre-treated to remove solids and debris using Macherey-Nagel filter papers (MN 615, Ø125mm). In scaled up systems preliminary treatment, such as solid waste screening, will be used. P and N concentrations, before and after treatment, were measured by means of inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7500 Series, model 7500ce), while the pH levels by a HANNA Multi-parameter probe (HI-9828 Multi-Parameter Water Quality Portable Meter).

Regarding the source of magnesium oxide (MgO), this was obtained from cryptocrystalline magnesite, a mineral abundant in South Africa (Mavhungu et al., 2019). Specifically, raw magnesite was hand-picked from an old (abandoned) mine in Folovhodwe, Limpopo Province, South Africa. In scaled up systems large amounts of cryptocrystalline magnesite will be required, and therefore the magnesite rock will be mined, rather than hand-picked. For this reason, here magnesite was assumed to be mined following the mining activities described elsewhere (Cherubini et al., 2008). The morphology of the raw cryptocrystalline

magnesite comprises of irregular sheets homogenously distributed across its surface (Magagane et al., 2019).

2.2 Thermal activation of the raw cryptocrystalline magnesite

MgO, which is required towards struvite formation, was produced by a fairly simple thermal activation process. Specifically, the raw cryptocrystalline magnesite was first milled into fine powder, by means of a vibratory ball miller (15 min at 500 rpm). The milled cryptocrystalline magnesite was then thermally activated (calcined at 1,000 °C for 30 min) as to convert its magnesium carbonate ($MgCO_3$) content to MgO (Mavhungu et al., 2019). The calcined magnesite was then remilled (15 min at 500 rpm) and passed through a 32 microns perforated sieve to obtain the desired particle sizes suitable for struvite precipitation. The thermally activated magnesite (feed thereafter) is rich in MgO, while after calcination new phases are formed in the feed, such as periclase, brucite, and calcite (Masindi et al., 2016a; Mavhungu et al., 2019). These will be dissolved in the wastewater, leading to an increase in the pH of the supernatant (Masindi, 2017), which is favourable for struvite precipitation (Mavhungu et al., 2019). After calcination the morphology of raw magnesite changes to nanosheets with octagonal structures, i.e. calcination reduces the particle size of the non-reactive magnesite and increases the surface area and reactive sites (Magagane et al., 2019). Due to its large surface area and reactive sites the feed has been also used for the remediation of acid mine drainage (AMD) as well (Masindi et al., 2016a). The mineralogical characteristics, along with the elemental and microstructural properties of the feed can be found elsewhere (Masindi et al., 2016b; Masindi et al., 2018b). The feed was then used, at bench scale and in a batch experimental procedure, for wastewater treatment and towards struvite precipitation.

2.3. Environmental Analysis

The main goal of this LCA study is to identify the environmental performance and main environmental hotspots of a struvite precipitation system. The optimal conditions, in terms of pollutants removal, have been identified elsewhere (Mavhungu et al., 2019), but this is not the case for its environmental sustainability under the South African setting. For this reason the one-factor-at-a-time (OFAT) method was used examining four process parameters (Table 1). Even though, the OFAT method requires a relatively large number of experiments to obtain exploitable results, and more importantly cannot effectively estimate the interactions between the examined parameters, as designed experiments do, it provides a good estimate of the importance of each parameter and hence was employed herein. OFAT results were used

for the environmental modelling, through scenario analyses. It should be noted that although for some of the values, such as the low pH, it is well established that the process efficiency is very low (Mavhungu et al., 2019), their effect on the environmental sustainability remains grossly unknown and thus they were included in the analysis.

For the environmental modelling the LCA methodology was followed, using the software programme SimaPro (version 9.1.0.11). LCA can quantify environmental impacts attributed to a product, process, or system, since it can trace and list material and energy flows throughout an examined life span. It has been widely used to evaluate the environmental impacts of different wastewater treatment systems (Sena and Hicks, 2018). Furthermore, the effective removal of P and N from municipal wastewater, particularly when considering the projected increase of nutrient discharge to surface water (van Puijenbroek et al., 2019), constitutes an environmental problem of emerging concern, both in the developed and particularly in the developing world. Therefore, the results of this work are of interest not only to researchers, but also to decision- and policy-makers and the wastewater and agricultural industry. All the above constitute the intended audience of this LCA study.

The attributional, over the consequential life cycle assessment approach was followed. The reason is that the first estimates the environmental impacts of a system attributed to the delivery of a specific amount of a functional unit (FU), while the latter the environmental consequences of a change in the production chain (Chatzisymeon et al., 2017). Finally, the time-related coverage of this work refers to present, i.e. 2020, and the geographical coverage includes South Africa, Lesotho, and other LMICs that are rich in magnesite mineral, as well as the developed world where circular economy in wastewater treatment is emerging as a promising wastewater management method.

2.3.1 Functional unit

The FU quantifies the performance of the system under study by normalising all input and output life cycle inventory (LCI) data using the same reference. In order to provide context with the literature, the FU of this LCA study was chosen as the treatment of 1 L of real wastewater. Volume is a commonly used FU for LCAs of wastewater treatment plants (WWTPs), since their main purpose is to treat influent wastewater (Sena and Hicks, 2018). Furthermore, by using this FU the influence of each of the four examined parameters on the system's environmental sustainability was identified, along with the main environmental hotspots.

However, pollutants removal efficiency largely varies depending on the examined parameter, i.e. the final characteristics of the wastewater after treatment are not similar. For this reason, the total environmental footprint of each OFAT run was also normalised per mg L^{-1} of P and N removed. Expressing results per P removed or recovered is also commonly encountered in LCA studies dealing with struvite recovery systems (Sena and Hicks, 2018). This normalisation also enables the direct comparison of each scenario and the identification of the optimal conditions, in terms of environmental relevance. It should be noted that laboratory experiments were conducted in 500 mL volumetric flasks and results were extrapolated to the examined functional unit, i.e. 1 L of wastewater.

Table 1: The range of the examined parameters for struvite formation from real wastewater.

Parameter	Input	Examined values
Treatment duration	min	1; 5; 10; 30; 60; 90; 120; 150; 180; 240; and 300
Feed dosage	g L^{-1}	0.2; 1; 2; 4; 6; 8; 10; 16; and 20
pH of solution	N/A	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, and 12
Temperature	°C	35; 45; 55; 65; and 75

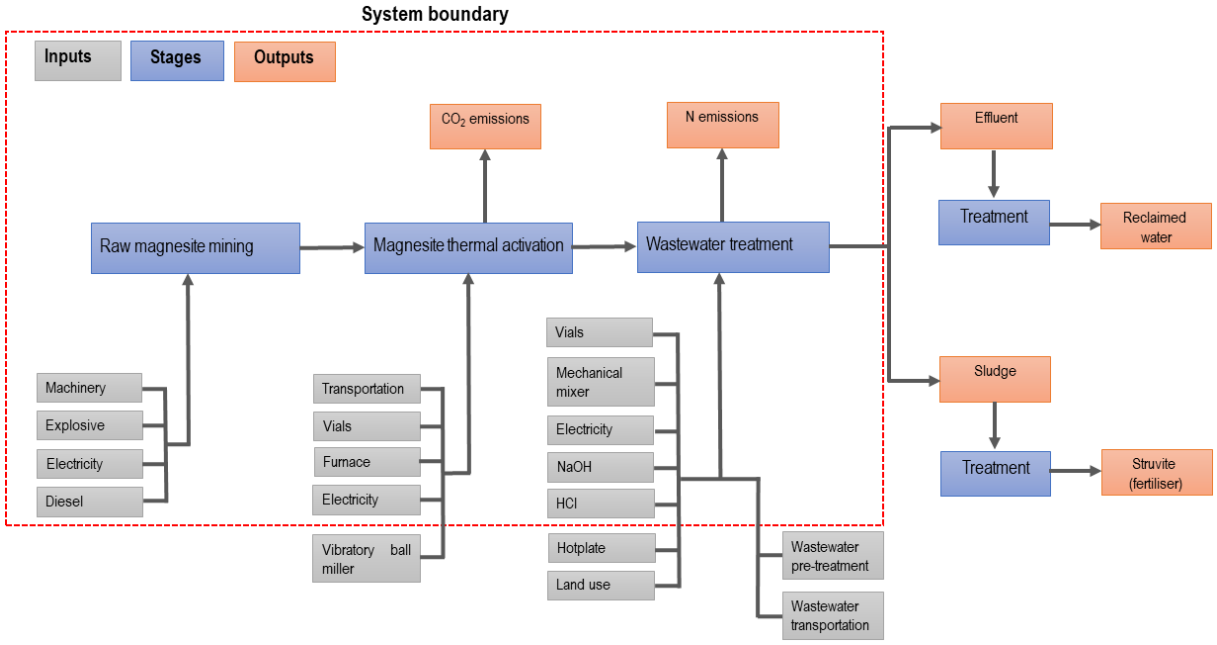
2.3.2 System boundary

The system boundary defines the smallest elements, i.e. unit processes, for which input and output data are quantified in the LCI analysis and thus are included in the LCA study. Here, all main inputs and outputs of the wastewater treatment process were considered (Schematic 1). Specifically, raw cryptocrystalline magnesite mining, as well as its transportation and processing (i.e. energy, equipment used and associated emissions from magnesite milling and calcination) are included in the analysis. This is also the case for the reagents used to regulate the pH (NaOH and HCl), as well as the energy required for stirring and wastewater temperature control. Ammonia airborne emissions, due to high temperature or pH values, are included in the analysis. Wastewater transportation to the laboratory, where the batch experiments were carried out, is external to the system boundary, since scaled up treatment systems are expected to be within existing or included in newly build wastewater treatment facilities.

The stirrer, as a material, is included in the analysis, while the hotplate and the vibratory ball miller, as materials, are external to the system boundary. The trivial laboratory equipment used, i.e. volumetric flasks, which will be replaced by tanks in scaled up systems is also

included in the analysis. In general, the operational phase of wastewater treatment processes is the main contributor to the environmental impacts, compared to the construction phase (Ioannou-Ttofa et al., 2016; Ioannou-Ttofa et al., 2017) and thus these material are not expected to largely influence the system's environmental sustainability.

The feed and the electricity input required for milling, mixing, and temperature control are also included in the analysis. Finally, a cradle to gate LCA was carried out and, as such, the final use of the treated effluent and of struvite are external to the system boundary (Schematic 1). Specifically, the effluent can be either released to the environment, if it meets the required standards, or further treated and reclaimed for various uses, such as for agriculture or even for drinking purposes. Each route has its own environmental impacts and benefits, and therefore including a specific route will make the LCA study case specific. This is also the case for the generated sludge (struvite), where further treatment is required to produce high quality fertiliser. For these reasons the treated effluent and the generated sludge (struvite) are external to the system boundary.



Schematic 1: The system boundary considered in this LCA study.

2.3.3 Life cycle inventory analysis

In the LCI analysis the resources, energy, and emissions entering and leaving the system under study are listed. In this work, primary LCI data were collected from batch laboratory experiments and used towards the environmental modelling. After the total environmental

footprint of each examined value was quantified per FU and the main environmental hotspots were identified, results were then used to identifying the optimal conditions, in terms of environmental relevance. This was achieved by normalising the results with the corresponding pollutant removal efficiency, i.e. expressing results per mg L^{-1} of P and N removed.

For the environmental modelling LCI data from SimaPro's proprietary databases were used, with the preferred option being ecoinvent v3.6 database (Table 2). In cases where relevant data were not identified in SimaPro's databases, proxy LCI data were collected from the literature. Specifically, for the stirrer LCI data were adapted from Griffiths et al. (2013), assuming a 10 year lifespan. Furthermore, the wastewater's specific heat capacity was considered to be 4.18 kJ per kilogram per degree Kelvin or Celsius, i.e. it takes 4,180 J to raise the temperature of 1 kg of wastewater by 1°C (McConnell and Tolley, 2015). The energy required to account for heat losses was assumed to be 50 % of the specific heat capacity per hour. Following (de Bakker, 2014) it was assumed that the specific energy required for the milling of 1 kg of magnesite is 60 Wh, when using a stirred media detritor. The rated power output of the overhead stirrer was 8.4 W, assuming it operated at 4 W, i.e. not at full power, when treating 1 L of wastewater. The electricity input considered throughout this work was assumed to originate from South Africa's fossil fuel-dependent energy mix, i.e. ~85 % hard coal according to ecoinvent database.

For the vials, ecoinvent's data for borosilicate glass tube were used, assuming a 5 year lifespan. After the end of their useful lifetime a 70 % recycling and a 30 % disposal, as inert waste, was considered. For magnesite mining, literature data were used (Cherubini et al., 2008), while the transportation distance ascribed to raw magnesite, i.e. from the place that is it mined to the place that is thermally activated and used for struvite precipitation, was 500 km (i.e. from Folovhodwe to Pretoria, South Africa). Transportation was assumed to be carried out by means of a diesel-powered truck with emissions standard EURO 3. Following (Krähenbühl et al., 2016) the energy consumption for raw magnesite calcination, i.e. at 1 000 ^{o}C for 30 min, was approximated at 0.47 kWh per kg of raw magnesite. For the furnace used for raw magnesite calcination LCI data were acquired from the literature corresponding to a high temperature furnace (Griffiths et al., 2013), assuming a 10 year lifespan. The efficiency of the furnace was assumed to be 70 %.

281 Furthermore, during magnesite calcination mass loss takes place, attributed to the conversion
282 of $MgCO_3$ to MgO and leading to CO_2 volatilization. Here, 50 % of the raw cryptocrystalline
283 magnesite was considered to be volatilized, which is in line with existing literature
284 (Krähenbühl et al., 2016). The corresponding CO_2 emissions are included in the analysis,
285 which is also the case for the ammonia emissions attributed to high pH and temperature
286 values. Specifically, N evaporates to air at temperatures higher than 30 °C and at pH values
287 higher than 10 (Sutiyono et al., 2016). In order to account for these losses, 50 % of the
288 additional N removal, compared to N removal for pH 10 and for ambient temperature (i.e. 35
289 ^{o}C) respectively, were considered as ammonia airborne emissions. Land use and land use
290 change were not taken into account, since when the process will be scaled up it would require
291 a limited space to operate and is also not expected to cause land use change. The chemicals
292 used for pH adjustment, i.e. 0.1 mol L^{-1} NaOH and 0.1 mol L^{-1} HCl, were taken directly from
293 ecoinvent database, however insignificant amounts, of the order of mL L^{-1} per pH range,
294 were used. The LCI data and the corresponding sources are summarised in Table 2.

295 Table 2: The collected LCI for the system under study.

Name	**Input**	**Unit**	**Source**
Electricity	South African mix	kWh	Ecoinvent 3.6
Mechanical stirrer	1 - 300	min	(Griffiths et al., 2013)
Electricity required for stirring	0 - 20	Wh	Ecoinvent 3.6
Energy required for wastewater temperature increase	4,180 J	J °C kg^{-1}	(McConnell and Tolley, 2015)
Electricity required for temperature increase*	0 – 69.72	Wh	Ecoinvent 3.6
Energy required for magnesite milling	60	Wh kg^{-1}	(de Bakker, 2014)
Electricity required for milling*	0.024 – 2.4	Wh	Ecoinvent 3.6
Vials (material) use	1 - 300	min	Ecoinvent 3.6 (borosilicate glass)
Raw magnesite	0.4 - 40	g	(Cherubini et al., 2008)
Raw magnesite transportation	500	km	Ecoinvent 3.6
Furnace	30	min	(Griffiths et al., 2013)

Energy required for raw magnesite calcination	0.47	kWh kg^{-1}	(Krähenbühl et al., 2016)
Electricity required for calcination*	0.671	kWh kg^{-1}	Ecoinvent 3.6
CO_2 emissions from calcination	50	% (w/w)	(Krähenbühl et al., 2016)
CO2 emissions to atmosphere	0.2 - 20	g L^{-1}	Ecoinvent 3.6
Feed dosage	0.2 - 20	g L-1	Calculated from the above data
NaOH	1 - 10	mL L^{-1}	Ecoinvent 3.6
HCl	1 - 7	mL L^{-1}	Ecoinvent 3.6
Ammonia loss during treatment (> 35 ^{o}C)	50	% (w/w)**	(Sutiyono et al., 2016)
Ammonia loss during treatment (pH > 10)	50	% (w/w)**	(Sutiyono et al., 2016)
Ammonia airborne emissions (> 35 ^{o}C)	0 – 44	mg L^{-1}	Ecoinvent 3.6
Ammonia airborne emissions (pH > 10)	0 – 8.5	mg L^{-1}	Ecoinvent 3.6
Vials recycling	30	% (w/w)	Ecoinvent 3.6
Vials landfilling	70	% (w/w)	Ecoinvent 3.6

296 * Values include electricity losses.

297 ** 50 % of the additional N that is removed compared to N that is removed in pH 10 or in 35 ^{o}C.

298 **2.3.4. Life cycle impact assessment**

299 In the life cycle impact assessment (LCIA) stage the collected LCI data are associated with
300 specific environmental impacts/damages and an understanding of those impacts/damages is
301 attempted. In this work, ReCiPe 2016 (version 1.04), a robust and widely used multi-issue
302 LCIA method, was employed. The ReCiPe model expresses results both at midpoint level,
303 where environmental impacts are examined earlier in the cause-effect chain, and endpoint
304 level, where environmental impacts are translated into issues of concern (e.g. damage on
305 human health) and thus are examined at the end of the cause-effect chain (Ioannou-Ttofa et
306 al., 2017). The endpoint or damage-oriented approach is associated with higher levels of
307 statistical uncertainty, compared to the midpoint or problem-oriented approach, due to data
308 gaps and assumptions stacking up along the cause-effect chain. However, results are easier to

communicate to decision- and policy-makers and the general public, while it also provides a common reference for comparison between different processes/systems (Chatzisymeon et al., 2017).

In order to reach endpoint, ReCiPe converts and aggregates midpoint impact categories into three damage categories, namely, damage to human health, damage to ecosystem diversity, and damage to resource availability, which can be further aggregated into a single score. The single score approach is used here to identify the total environmental footprint of each examined parameter and main environmental hotspots. In addition, the net impacts approach was also preliminary examined, focusing on the benefits from reducing the eutrophication potential of receiving water bodies, which is directly related to P and N removal. To this end, ReCiPe was applied at midpoint level examining two midpoint impact categories, i.e. freshwater eutrophication (P enrichment of freshwater) and marine eutrophication (N enrichment of seawater). Finally, average technology was assumed and the Hierarchist perspective (H) was employed, using the normalisation values of the world and average weighting (i.e. world ReCiPe H/A).

3 Results and discussion

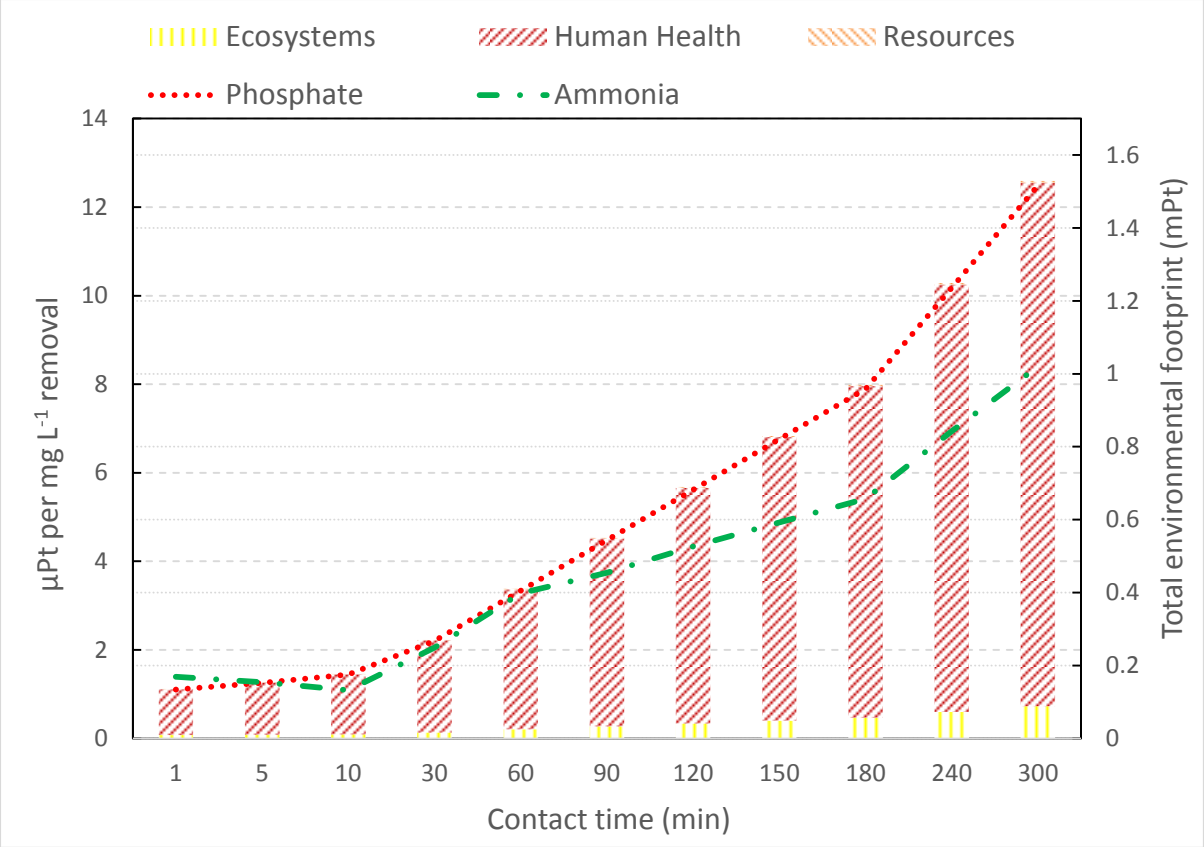
This is a comparative LCA study with the main goals being to identify the effect of operating parameters and optimal conditions from the environmental perspective. For this reason the total environmental footprint of each examined parameter was first estimated per FU, as to identify the effect of operating parameters, while the optimal treatment conditions were estimated by normalising results per mg L^{-1} of P and N removed.

3.1 Effect of operating parameters

First the effect of contact time was examined. Eleven mixing durations, ranging from as low as 1 min to as high as 300 min (Table 1), were considered, ceteris paribus pH (~7.5), temperature (~35 ^{o}C), and feed dosage (2 g L^{-1}). Results are shown in Figure 1, where it is observed that the higher the contact time the higher the environmental footprint. Specifically, when treatment time increases so does the electricity input required for wastewater mixing (or for wastewater pumping in scaled up systems) leading to increased footprints. However, in the first three examined contact times, i.e. 1, 5, and 10 min, the mixing duration has a relatively small effect on the system's environmental sustainability, i.e. the total environmental of 10 min is around 27 % higher than that of the 1 min (Figure 1). This implies

that for mixing duration of up to 10 min the remaining parameters play an important role. Nevertheless, for longer contact times a strong influence is observed (e.g. the total environmental footprint for 300 min is ~11-fold higher than the one of the 1 min) suggesting that contact time is a process environmental hotspot.

The normalised results, i.e. per mg L^{-1} P and N removed, reveal a similar pattern with the total environmental footprint, however the influence of contact time depends on the targeted pollutant (Figure 1 and Table 3). Specifically, for N a decrease in the normalised environmental footprint was observed in treatment time 10 min, compared to the 1 min value, however thereafter largely increases with increasing contact time. On the other hand, for P an almost linear relationship between contact time and the normalised environmental footprint was identified with increasing contact time and particularly after 10 min. For practical applications the 1 min contact time might be too short and therefore the 5 or 10 min contact time might be selected (Figure 1 and Table 3). The underlying reason for this pattern is that P is practically removed starting from 1 min contact time and afterwards it remains relatively constant. Therefore, the longer the mixing the higher the electricity consumption and by extension the higher the environmental footprint per mg L^{-1} of P removed. Overall, the optimal contact time for both P and N removal is 10 min, however higher durations (e.g. 180 min) increase process efficiency (Table 3), which is desirable for real-world applications.



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359 Figure 1: The total environmental footprint per FU (treatment of 1 L of wastewater) and per
360 mg L^{-1} of P and N removed with varied contact time. Fixed parameters: pH ~7.5, temperature
361 ~35 ^{o}C, and feed dosage 2 g L^{-1}.

362 Feed dosage, i.e. the amount of the thermally activated cryptocrystalline magnesite added
363 into the wastewater, was then examined, ceteris paribus contact time (30 min), pH (~7.5) and
364 temperature (~35 ^{o}C). Similarly to contact time, the higher the feed dosage the higher the
365 total environmental footprint of the process (Figure 2), with the environmental footprint of
366 the last examined value first (20 g L^{-1}) being around 9 times higher than the one of the first
367 examined value (0.2 g L^{-1}). Therefore, feed dosage has a similar influence on the system's
368 environmental sustainability with the one observed in contact time. The feed influence can be
369 traced back to: i) the mining activities, where explosives, machinery, and fossil-fuel energy
370 are used to mine the raw magnesite, ii) the fossil-fuel dependent energy consumed for raw
371 magnesite processing, mainly for calcination and to a lesser extent milling, and iii) the CO_2
372 emissions originating from the calcination process. Even though raw magnesite transportation
373 distance was 500 km, it was identified that it plays only a small role in the feed's total
374 environmental footprint.

Furthermore, as shown in Figure 2 it was found that the system's environmental performance is optimised at the lowest used dosage, i.e. 0.2 g L^{-1}, while for higher values, and particularly for values higher than 2 g L^{-1} the system's environmental performance drastically declines. This is attributed to the facts that: i) the feed dosage is an environmental hotspot and as such increasing the dosage largely increases the total environmental footprint, and ii) pollutants, and particularly P, are grossly attenuated even at very low feed dosages (i.e. when increasing the dosage from 0.2 to 1 g L^{-1} only a ~ 0.3% increase in P removal is achieved and then P removal stabilises (Table 3)). A similar pattern is observed in the case of N removal, i.e. the higher the feed dosage the higher the normalised environmental footprint, however in this case the optimal conditions are achieved at 1 g L^{-1}. Nonetheless, N removal efficiency is optimised at the higher end of the examined range (Table 3), with the 16 g L^{-1} dosage having a slightly lower N removal efficiency but being more environmentally friendly than the 20 g L^{-1} dosage.

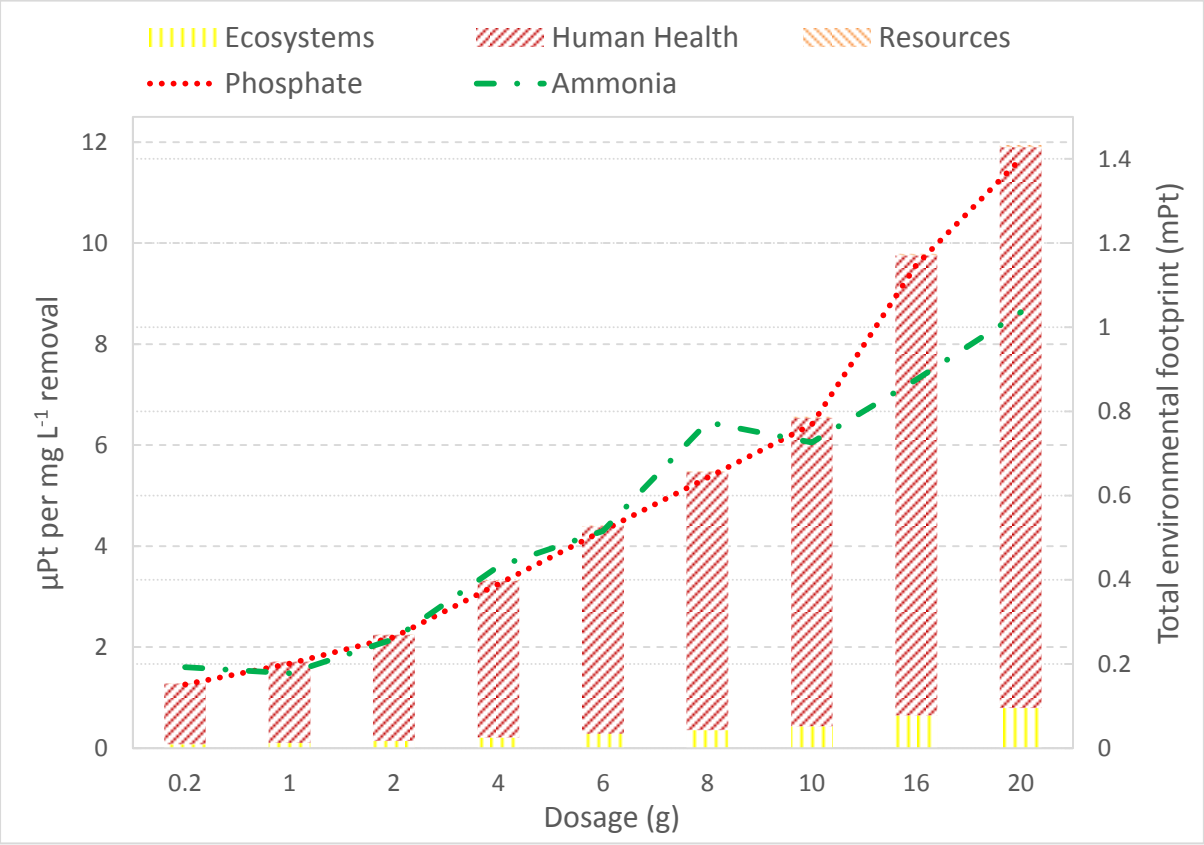

Figure 2: The total environmental footprint for the treatment of 1 L of wastewater and per mg L^{-1} of P and N removed from real wastewater with varied feed dosage. Fixed parameters: pH ~7.5, temperature ~35 ^{o}C, and contact time 30 min.

The effect of increasing/decreasing the initial pH of the wastewater was studied by taking into account a wide range of pH values, from as low as 1 to as high as 12 (Table 1). As feed dosage 16 g L^{-1} was used, which optimizes N removal efficiency (Table 3), the contact time was 60 min and ambient water temperature (~35 ^{o}C) was considered. It was observed that pH diversification only slightly affects the process environmental sustainability, and therefore cannot be considered as an environmental hotspot (Figure 3). Specifically, it was found that chemical addition (i.e. NaOH and HCl) has a very small influence on the system's environmental sustainability, compared to the above-mentioned hotspots (contact time and feed dosage). The main reasons for this low influence can be attributed to the facts that: i) only small amounts of these chemicals are used, and ii) these chemicals are not suspected to be carcinogenic, mutagenic or toxic to reproduction (European Commission, 2006). As a result, the total environmental footprint of pH 1, where the largest amount of chemicals is used, is around 21 % higher than the ambient pH scenario (here taken as pH 8). The underlying reason is that contact time and feed dosage remain constant, and therefore chemicals addition for pH control only slightly affects the system's environmental performance. This is also reflected in the normalised environmental footprints shown in Figure 3. Specifically, for P, apart from the pH 12 value where the removal efficiency (and by extension the environmental sustainability) strongly deteriorates, the normalised environmental footprint remains practically constant with varying pH. On the other hand, for N the worst value is observed at pH 1, then the normalised environmental footprint remains relatively stable, while for pH values higher than 10 it reduces (Figure 3 and Table 3). It should be noted that pH values higher than 10 promote ammonia evaporation to air (Sutiyono et al., 2016), which is reflected in the normalised environmental footprint.

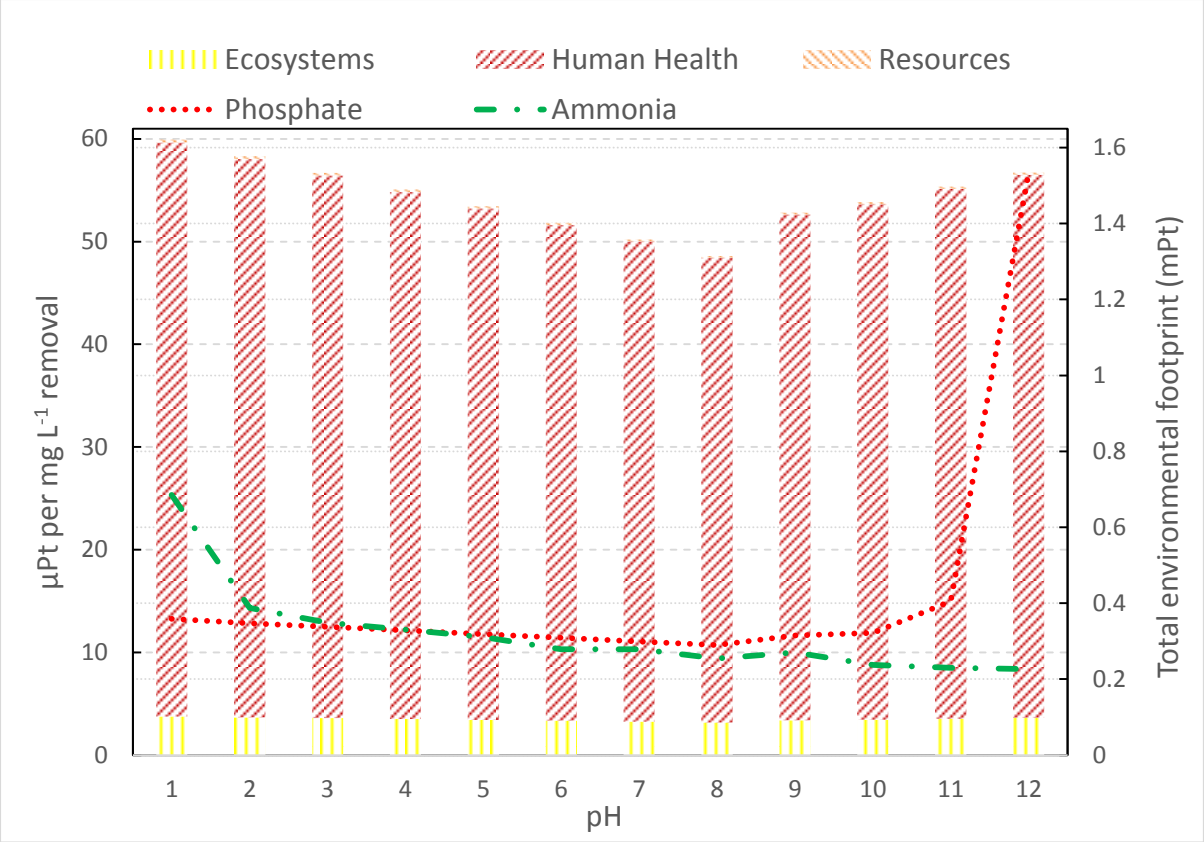

Figure 3: Total environmental footprint for the treatment of 1 L of wastewater and for the removal of 1 mg L^{-1} of P and N from real wastewater with varied pH values. Fixed parameters: temperature ~35 ^{o}C, feed dosage 16 g L^{-1}, and contact time 60 min.

Finally, to examine the effect of the wastewater initial temperature, five different values, ranging from ambient temperature under South Africa's climatic conditions, i.e. 35 ^{o}C, to values as high as 75 ^{o}C, were examined (Table 1). The fixed parameters were 8 g L^{-1} feed dosage, 60 min contact time, and ambient pH (~7.5). As shown in Figure 4, raising the wastewater temperature appears to be environmentally unsound and should be avoided. The reason is that a very large electricity input, compared to the remaining process (Table 2), is required to raise and maintain wastewater's temperature during treatment. This is the reason behind the very large total environmental footprints for the values where the wastewater temperature was raised (i.e. 45 ^{o}C and upward). Furthermore, the improvement in treatment efficiency, when wastewater temperature is raised, is relatively small (Table 3), which also leads to very high normalised environmental footprints (Figure 4). More specifically, the improved N removal efficiency for pH values higher than 10 is not large, while ammonia emissions, attributed to elevated temperature, negatively influence the system's environmental sustainability, but to a small extent compare to the electricity input required

for temperature increase and maintenance. Therefore, the optimal conditions are achieved at ambient temperature (35 °C).

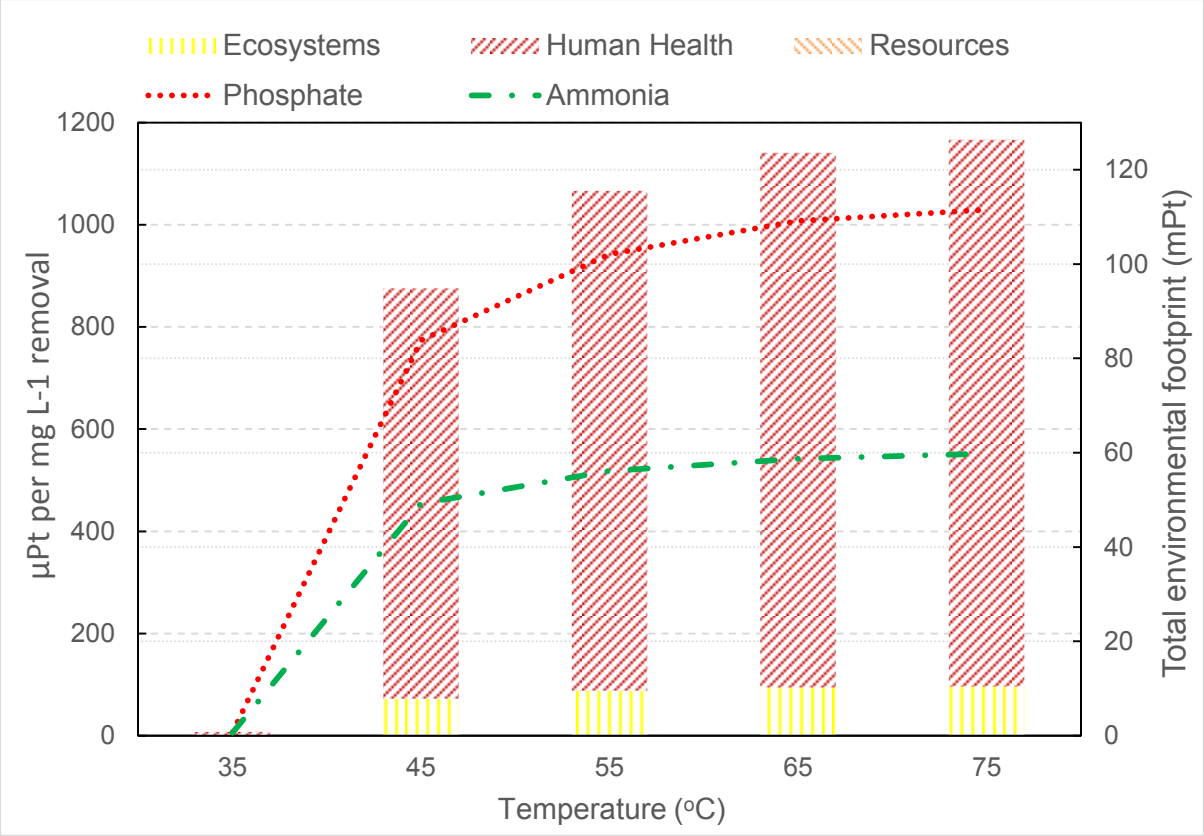

Figure 4: Total environmental footprint for the treatment of 1 L of wastewater and for removal of 1 mg L^{-1} of P and N with varying wastewater temperature. Fixed parameters: pH ~7.5, feed dosage 8 g L^{-1}, and contact time 60 min.

Regarding the contribution of the construction and operational phase, the latter was the main contributor. For example, in the 10 min, 0.2 g L^{-1}, and ambient temperature and pH scenario the contribution of the construction phase on the total environmental footprint is slightly over 6 %. However, this contribution in the scenario that optimises N removal (180 min, 16 g L^{-1} and ambient temperature and pH) reduces to under 2.5 %, since a higher amount of feed and of electricity (for stirring) is consumed, which drives up the operational phase contribution on the total environmental footprint. Even though these results are suggestive, since the examined system operates at bench scale, they are indicative of the much larger contribution of the operational phase, attributed to feed and electricity consumption (their contribution varies with their corresponding input). The results concerning the contribution of the construction and operational phase are also comparable with those of other wastewater treatment processes (Ioannou-Ttofa et al., 2016; Ioannou-Ttofa et al., 2017).

Furthermore, in all examined parameters the endpoint damage category that was affected, by and large, was human health, followed, to a lesser extent, by resources, while ecosystems was only slightly affected (Figure 1 to 4). Electricity input is required for magnesite milling and calcination and for stirring the magnesite-wastewater matrix. As such, the operational phase can be power-hungry, while the South African energy mix, as well as the global energy mix, are grossly dependent on fossil fuels (Masindi et al., 2018a). Emissions originating from fossil fuel extraction, and particularly from fossil fuel combustion, directly affect the damage category human health, while the burning of fossil fuels leads to their depletion, thus affecting the resource availability category. However, when using the global weighting factors to reach endpoint in ReCiPe, the damage category ecosystems is only slightly affected. Therefore, using renewable energy to drive the process could substantially reduce environmental impacts.

Table 3: Initial and final (after treatment) pollutants concentration and their normalised, i.e. per mg L^{-1} of P and N removed, environmental footprint.

	Initial P (mg L^{-1})	Initial N (mg L^{-1})	Final P (mg L^{-1})	Final N (mg L^{-1})	P removal (%)	N removal (%)	µPt per mg L^{-1} P removed	µPt per mg L^{-1} N removed
Contact time (min)								
1	123	239	0.56	143	99.55	40.59	1.10	1.39
5	123	239	<0,20	100	>99.84	50.63	1.25	1.27
10	123	239	<0,20	114	>99.84	68.20	1.44	1.09
30	123	239	<0,20	128	>99.84	55.23	2.20	2.05
60	123	239	<0,20	116	>99.84	52.30	3.34	3.28
90	123	239	<0,20	137	>99.84	61.51	4.48	3.74
120	123	239	<0,20	109	>99.84	66.53	5.62	4.34
150	123	239	<0,20	78	>99.84	71.13	6.76	4.88
180	123	239	<0,20	73	>99.84	74.90	7.89	5.42
240	123	239	0.56	143	>99.79	75.31	10.18	6.94
300	123	239	<0,20	100	>99.54	76.15	12.49	8.40
Feed dosage (g)								
0.2	123	239	0.55	142	99.54	40.17	1.26	1.61
1	123	239	<0,20	118	>99.84	58.16	1.68	1.48
2	123	239	<0,20	76	>99.84	52.30	2.20	2.16
4	123	239	<0,20	107	>99.84	46.44	3.25	3.60
6	123	239	<0,20	114	>99.84	51.46	4.31	4.30
8	123	239	<0,20	92	>99.84	42.68	5.36	6.45
10	123	239	<0,20	80	>99.84	54.39	6.41	6.05
16	123	239	<0,20	69	>99.84	67.36	9.56	7.30
20	123	239	<0,20	60	>99.84	69.46	11.67	8.63
Initial wastewater pH								
1	123	239	0.92	175	99.25	26.78	13.28	25.32

2	123	239	<0,20	129	>99.84	46.03	12.84	14.34
3	123	239	<0,20	120	>99.84	49.79	12.49	12.88
4	123	239	<0,20	117	>99.84	51.05	12.13	12.21
5	123	239	<0,20	113	>99.84	52.72	11.77	11.47
6	123	239	<0,20	103	>99.84	56.90	11.42	10.31
7	123	239	<0,20	107	>99.84	55.23	11.06	10.29
8	123	239	<0,20	99	>99.84	58.58	10.70	9.39
9	123	239	<0,20	96	>99.84	59.83	11.64	9.99
10	123	239	0.55	73	99.55	69.46	11.89	8.77
11	123	239	23	63	81.30	73.64	14.97	8.51
12	123	239	96	56	21.95	76.57	56.82	8.38
Initial wastewater temperature (°C)								
35	123	239	0.47	97	99.62	59.41	6.51	5.62
45	123	239	0.28	30	99.77	87.45	773.46	454.16
55	123	239	0.45	16	99.63	93.31	942.48	517.94
65	123	239	0.29	11	99.76	95.40	1007.20	542.08
75	123	239	0.21	9.9	99.83	95.86	1029.51	551.78

3.2 Optimal conditions

As mentioned above, even though temperatures higher than 30 ^{o}C promote ammonia stripping, increasing the wastewater temperature is environmentally unsound. From the remaining three examined parameters, contact time and feed dosage were identified as environmental hotspots, which was not the case for pH control. Therefore, ambient pH (~7.5) and temperature (~35 ^{o}C) should be used. Regarding the other two examined parameters, the feed's influence is reflected in the fact that the system's environmental performance is optimized at the lower end of the examined range (0.2 to 1 g L^{-1}), while the worst performance is observed at the higher end (20 g L^{-1}). This was also the case for contact time, where the optimal conditions were achieved between 1 to 10 min. With these conditions the system has a very low environmental footprint (e.g. 10 min, 0.2 g L^{-1} environmental footprint is just 60.9 μPt L^{-1}, while the10 min, 1 g L^{-1} is 113 μPt L^{-1}). However, these conditions are not similar to the ones that optimise pollutants removal efficiency (Mavhungu et al., 2019).

Specifically, the total environmental footprint of the conditions that optimise the removal of both pollutants (30 min and 6 g L^{-1}) will be 529 μPt L^{-1}, while the conditions that optimise N removal (6 g L^{-1} and 180 min) are responsible for 1.87 mPt L^{-1}. Therefore, in terms of environmental relevance, it appears that the 0.2 to 1 g L^{-1} is the optimal feed dosage, when using contact times as low as 10 min. However, for practical applications, e.g. at village- or industrial-scale where pollutants removal is the main concern, higher dosages and contact times are required to optimise the process efficiency; albeit at the expense of the

485 environmental performance. This suggests that when using a low dosage (e.g. 0.2 to 1 g L^{-1})
486 struvite recovery systems can act as a very fast (even at 10 min contact time), efficient, and
487 environmentally friendly pre-treatment step to grossly remove P and also reduce N from
488 wastewater. This very low dosage and contact time is also favourable in cases of overloaded
489 wastewater treatment facilities, where this technology can be added to assist the treatment
490 process or at least used to pre-treat the wastewater before being released to the environment,
491 such as in rural and semi-urban South Africa. However, for real-world applications higher
492 contact times (up to 180 min) and feed dosages (up to 16 g L^{-1}) will probably be employed. If
493 required, other processes could be also coupled with this technology on a wastewater
494 treatment train system.

495 **3.3 Net impacts approach and contributions to the body of knowledge**

496 Regarding the avoided environmental impacts from treating wastewater, rather than releasing
497 it untreated to the environment, these were approximated using the net impacts approach.
498 Focus was placed on ReCiPe's midpoint impact categories freshwater eutrophication (FE - P
499 enrichment of freshwater) and marine eutrophication (ME - N enrichment of seawater). Three
500 scenarios were examined, one that focuses on P removal (i.e. 123 mg L^{-1} P and 121 mg L^{-1} N
501 removed at 5 min mixing and 1 g L^{-1} dosage), one that focuses on N removal (i.e. 123 mg L^{-1}
502 P and 179 mg L^{-1} N removed at 180 min and 16 g L^{-1}) and one that focuses on the removal of
503 both pollutants (i.e. 123 mg L^{-1} P and 132 mg L^{-1} N removed at 30 min and 6 g L^{-1})
504 (Mavhungu et al., 2019). As shown in Figure 5, in terms of eutrophication potential, the
505 avoided environmental impacts grossly outweigh the corresponding environmental impacts of
506 the system. Furthermore FE enjoys a larger reduction compared to ME (Figure 5). This was
507 expected, since marine ecosystems are less sensitive to eutrophication pressures, compared to
508 freshwater ecosystems (Ioannou-Ttofa et al., 2017).

509 The net impacts approach also revealed that the total environmental footprint of the scenario
510 that optimises the removal of both pollutants will be reduced by around □9 %, when taking
511 into account the avoided environmental impacts of the FE and ME categories. For the
512 scenario that optimises P removal the reduction is much higher (□52 %), whereas for the
513 scenario that optimises N removal the reduction is much lower (□2.5 %). The reason is that
514 the total environmental footprint of the first is much lower than the one of the scenario that
515 optimises the removal of both pollutants and much higher in the latter case. More
516 importantly, the reduction was solely on the damage category ecosystems, thus in all

examined scenarios this category had a negative score. This is of great significance for areas where raw wastewater is directly released to freshwater ecosystems and also implies the importance and the pressing need for treating wastewater before releasing it to the environment. Overall, results suggest the possible huge environmental benefits of struvite recovery systems in the eutrophication impact categories. These benefits are attributed to the fact that releasing untreated municipal wastewater negatively affects the receiving environment and particularly affecting the nutrient balance (eutrophication). It should be noted that this is an ongoing problem, mainly affecting the developing world, since globally more than 80% of all wastewater is released untreated to the environment (UNESCO, 2020) and struvite recovery systems could address, at least partly, this problem.

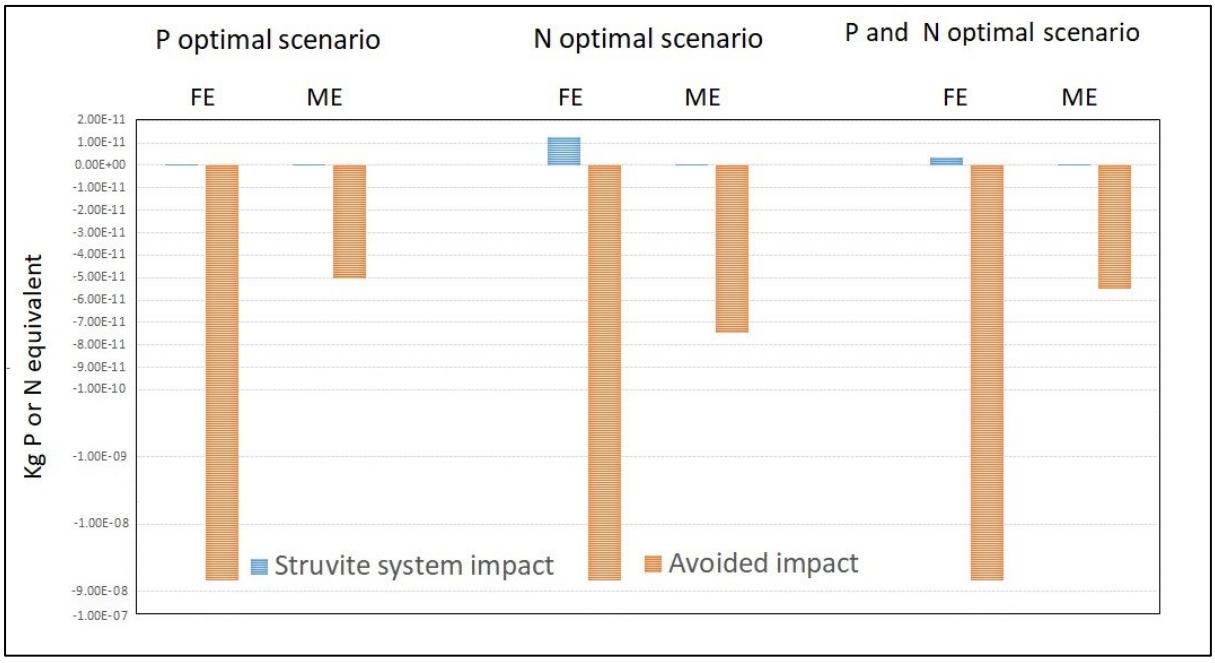

Figure 5: Comparison between the impacts of the examined system and the avoided impacts on ReCiPe's midpoint impact categories freshwater eutrophication (FE) and marine eutrophication (ME) for three examined scenarios, i.e. optimal P removal, optimal N removal, and optimal P and N removal.

Furthermore, the abovementioned P and N removal rates suggest that the treated wastewater needs further polishing to remove the excess N content, before being discharged to the environment or used for irrigation, among other possible uses. Specifically, according to the guidelines of the South African Department of Water and Sanitation (DWS), previously known as the Department of Water Affairs and Forestry (DWAF), the nitrogen content

(ammonia, ammonium, nitrite, and nitrate in their inorganic forms) should be equal or less than 5 mg L^{-1} for effluents intended for irrigation or for releasing to the environment (DWAF, 1996). To reduce the N content a biological nutrients removal (BNR) process (nitrification-denitrification process) or a polishing step or even dilution using clean water could be employed. The latter will require large volumes of clean water and hence it might not be feasible. Each chosen treatment process will have its own environmental footprint, while depending on the final use of the reclaimed water (e.g. irrigation) avoided environmental impacts will arise. This is also the case for the generated sludge (struvite). However, as mentioned above, including the recovered struvite and treated effluent in the system boundary depends on many different parameters and local conditions, such as treatment method, intended use, type and maturity of the technology, and therefore they are external to the system boundary. The effect of the recovered struvite, of the reclaimed water, as well as further insight on the contribution of the construction and operational phase a will be examined in future works of our group dealing with scaled up systems.

Overall, the abovementioned results suggest the large environmental benefits, particularly in the eutrophication potential, of struvite precipitation. They also provide context and add new information to the body of knowledge. Specifically, it has been identified that P recovered from large-scale struvite precipitation systems, as suggested in this work, is more environmentally friendly than P recovered from the ash of incinerated sludge (Linderholm et al., 2012). Furthermore, through struvire crystallisation the environmental impacts of WWTPs can be reduced, and particularly the eutrophication potential (Rodriguez-Garcia et al., 2014). In general, the reduction of the eutrophication potential is a well-documented argument for phosphorus recovery, however, P recovery might not necessarily present a net environmental benefit (Bradford-Hartke et al., 2015). For example, in a case study in Metro Manila, Phillipines it was identified that integrating nutrient recovery systems in WWTPs can decrease the impact on aquatic eutrophication and acidification by around 14 % and 4 % respectively, however, at the expense (5 % increase) of global warming potential (GWP) (Pausta et al., 2018). On the other hand, Zhou et al. (2019) examined the AirPrex® process for P recovery and reported better environmental credits in the GWP, along with the eutrophication impact categories.

Through the net impact approach the environmental benefits in eutrophication impact categories were also estimated and highlighted here. More importantly, the results of this work provide new insight on the environmental sustainability of struvite precipitation in a

typical LMIC in Africa. Specifically, Sena and Hicks (2018) reviewed the existing body of knowledge on LCA studies for struvite precipitation and highlighted the lack of studies outside Europe and the need for further studying the process environmental impacts. In a more recent literature review, from the 65 reviewed manuscripts only 2 were referring to Africa, with most of the works focusing in Europe, while the need for optimising, from the environmental perspective, the recovery processes, as is the case here, was also highlighted (Lam et al., 2020). The results of this work provide new insight on the process and on its main environmental hotspots under the African setting. They also provide a preliminary understanding of the avoided environmental impacts in the eutrophication impact categories, through the net impacts approach, highlight the potential of struvite recovery systems and the importance of wastewater treatment in LMICs, where currently wastewater is typically released untreated or poorly treated to the environment and water is not reclaimed.

5. Conclusions

The environmental sustainability and main environmental hotspots of wastewater treatment, through struvite precipitation, were examined in the South African setting. Four process parameters were examined, i.e. contact time, magnesite dosage, and initial wastewater pH and temperature, using the LCA methodology. The main environmental hotspot was, by and large, temperature increase and therefore raising wastewater temperature during treatment was identified to be an environmentally unsound practise. Magnesite dosage and contact time were identified as environmental hotspots, while pH only slightly affected the system's environmental performance. As such, the optimal conditions, from the environmental perspective, were identified as 0.2 - 1 g L^{-1} feed dosage, 5 - 10 min mixing time, ambient temperature and pH. Through this configuration, the system is able to practically remove the P content and grossly reduce the N content from the wastewater with a small environmental footprint, i.e. 60.9 μPt L^{-1} for the 0.2 g L^{-1} and 10 min contact time. However, in terms of process efficiency a higher dosage (6 g L^{-1}) and contact time (30 min) optimises pollutants removal efficiency, with the environmental footprint rising to 529 μPt L^{-1}. When N removal is the main focus, then magnesite dosage should be increased to 16 g L^{-1} and the contact time to 180 min, however at the expense of the environmental sustainability (total environmental footprint 1.87 mPt L^{-1}).

Therefore, the result of this work suggest that when using the low dosage, struvite precipitation can act as a fast, efficient, and environmentally friendly pre-treatment method,

particularly in cases where raw wastewater is directly released to the environment. In these cases, which include rural and peri-urban South Africa and Lesotho, struvire recovery systems could be a very efficient and fast alternative to effectively treat wastewater, instead of releasing it untreated to the environment. However, for village- or industrial-scale applications higher feed dosages and contact times might be require. The damage category that is affected the most by this technology is human health, followed by ecosystem diversity, while resource availability is only affected to a much lesser extent. Through the net impact approach is was identified that P and N removal from wastewater, before its release to the environment, would lead to large environmental benefits. Overall, the results of this work provide context and extent the body of knowledge on struvite precipitation in the South African and the African setting in general, which is grossly missing from the literature.

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