



OPEN LETTER

REVISED **Non-biological methods for phosphorus and nitrogen removal from wastewater: A gap analysis of reinvented-toilet technologies with respect to ISO 30500 [version 2; peer review: 2 approved, 1 not approved]**

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v2 **First published:** 24 Feb 2019, 3:559
<https://doi.org/10.12688/gatesopenres.12931.1>
Latest published: 15 May 2020, 3:559
<https://doi.org/10.12688/gatesopenres.12931.2>

Abstract

The aims of the Reinvent the Toilet Challenge (RTTC) include creation of an off-the-grid sanitation system with operating costs of less than US\$0.05 per user per day. Because of the small scale at which many reinvented toilets (RT) are intended to operate, non-biological treatment has been generally favored. The RTTC has already instigated notable technological advances in non-sewered sanitation systems (NSSS). However, increasingly stringent liquid effluent standards for N and P could limit the deployment of current RT in real-world scenarios, despite the urgent need for these systems. The newly adopted ISO 30500 standards for water reuse in NSSS dictate minimal use of chemical/biological additives, while at the same time requiring a 70% and 80% reduction in total nitrogen and phosphorus, respectively. This document provides a brief overview of the mature and emerging technologies for N and P (specifically ammonia/ammonium and orthophosphate) removal from wastewater. At present, the dearth of nutrient removal methods proven to be effective at small scales is a significant barrier to meeting ISO 30500 standards. Closing the gap between RTs and ISO 30500 will require significant investments in basic R&D of emerging technologies for non-biological N and P remediation and/or increased reliance on biological processes. Adaptation of existing nutrient-removal technologies to small-scale NSSS is a viable option that merits additional investigation.

Keywords

non-sewered sanitation systems (NSSS), reinvented toilet (RT), nutrient removal, nitrogen, phosphorus, ion exchange, membrane separation, hydrogels

Open Peer Review

Reviewer Status

	Invited Reviewers		
	1	2	3
version 2 (revision) 15 May 2020			 report
version 1 24 Feb 2019	 report	 report	 report

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Author roles: **Trotochaud L:** Conceptualization, Writing – Original Draft Preparation; **Hawkins BT:** Conceptualization, Writing – Review & Editing; **Stoner BR:** Conceptualization, Funding Acquisition, Writing – Review & Editing

Competing interests: No competing interests were disclosed.

Grant information: This work was funded by the Bill & Melinda Gates Foundation [OPP1174988].

The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

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How to cite this article: Trotochaud L, Hawkins BT and Stoner BR. **Non-biological methods for phosphorus and nitrogen removal from wastewater: A gap analysis of reinvented-toilet technologies with respect to ISO 30500 [version 2; peer review: 2 approved, 1 not approved]** Gates Open Research 2020, 3:559 <https://doi.org/10.12688/gatesopenres.12931.2>

First published: 24 Feb 2019, 3:559 <https://doi.org/10.12688/gatesopenres.12931.1>

REVISED Amendments from Version 1

This version has been updated in response to the valuable feedback provided by the Reviewers. Major changes include addition of text to clarify the scope of the Open Letter, expansion of the discussion of chemical precipitation methods and ion-exchange materials, and addition of a column to Table 1 to indicate which nutrients are removed by each method. Minor changes include rearrangement of some sections of text, changes to the formatting in Table 1, addition of several relevant references, correction of formatting errors and typos, and other small changes/additions in response to Reviewer feedback.

Any further responses from the reviewers can be found at the end of the article

Introduction and scope

Anthropogenic nutrient pollution of surface waters has severe negative impacts on the environment, human health, and the economy. In particular, algal blooms, caused by eutrophication due to high levels of nitrogen and phosphorus (N and P, primarily as ammonia/ammonium and orthophosphate), pose immediate risks to wildlife and human populations, and it can cost millions of dollars to treat a single body of water once a bloom occurs¹. Several effective, mature technologies for mitigating environmental release of N and P are widely employed at municipal wastewater treatment plants (WWTPs). However, a technological gap exists in providing similarly effective nutrient removal at smaller, decentralized treatment systems while maintaining affordability and sustainability². While the cost, efficacy, and practicality of nutrient removal technologies depends on many factors unique to each wastewater stream, including the influent quality, effluent requirements, climate, land and capital availability, local cultural and institutional attitudes, etc^{3,4}, a few general trends are observed^{1,2,5–8}:

- The smaller the scale of the water treatment system, the higher the per capita cost.
- More stringent effluent standards for N and P require increasingly complex systems with multiple remediation processes (both biological and physical/chemical) working in concert.
- Biological methods of N and P removal typically have lower capital and operational/maintenance costs than chemical/physical methods.

The Bill & Melinda Gates Foundation's Reinvent the Toilet Challenge (RTTC) has spurred significant advances in non-sewered sanitation systems (NSSS) designed to operate at less than US\$0.05 per user per day with minimal use of chemical/biological additives. Typically designed to work at the household or institutional scale, these NSSS fall into the category of small-scale, decentralized treatment systems. Smaller-scale treatment systems which rely on biological processes are more susceptible to system shocks and stresses, including non-continuous/inconsistent influent supply/quality^{3,9,10}. Thus, many NSSS in

the RTTC portfolio rely on non-biological[†] processes for water treatment and reuse (e.g. electrochemical oxidation; membrane separation)^{11–18}. Treatment of the liquid fraction of NSSS waste to enable non-potable water reuse (e.g. for toilet flushing) has been a focus of many reinvented toilet (RT) technologies and guides the scope of this Open Letter.

The technological approach for N and P removal from liquid effluent will depend to a great extent on whether, how, and when urine and feces are separated from each other. Also important is whether and to what extent urine is diluted by water used for toilet flushing (and personal washing, where applicable). Urine contains approximately 90% of the total N and 50–65% of the total P in human excreta, and the chemical components are largely inorganic compounds (after urea hydrolysis); in contrast, the contributions to N and P from feces are largely organic (proteins and bacterial biomass)¹⁹. For the purposes of this Open Letter, we focus on removal of ammonia/ammonium and orthophosphate, which are the majority water-soluble contributors to N and P in human excreta and the compounds of primary concern for anthropogenic nutrient pollution.

An additional layer of complexity has been introduced by the adoption of the new ISO 30500 standards for NSSS. The ISO 30500 standards provide guidance for safe onsite treatment of human excreta and non-potable water reuse, and includes threshold performance metrics for liquid effluent quality, including chemical oxygen demand (COD), total suspended solids (TSS), nutrients (N and P), and specific pathogens. Regardless of whether water is to be reused or discharged, ISO 30500 requires 70% and 80% reductions in total N and total P, respectively, in NSSS liquid effluent. Here, we briefly outline the current mature and emerging non-biological technologies for N and P removal. We discuss the advantages and limitations of these methods in the context of RTTC and their ability (or lack thereof) to reliably meet the ISO 30500 standards for nutrient removal. This discussion shows that, in general, the best conventional methods of N and P removal have not been shown to be compatible with small-scale RT systems. Throughout, we also provide perspective on current areas of research and development (R&D) opportunities that could spawn technological solutions which unify the goals of RTTC and ISO 30500. Finally, we briefly present potential opportunities for increasing the use of biological systems in the RTTC portfolio to meet more stringent effluent standards.

Mature and emerging technologies for non-biological nutrient removal

Several non-biological technologies are commonly applied at municipal- and industrial-scale WWTPs for N and P removal. However, very few of these technologies have been employed at the household/community scale where NSSS are most relevant.

[†]Because of the nature of blackwater, it is inevitable that biological processes occur at some point in many (if not all) of the RTTC systems. Here, we use “non-biological” to distinguish methods where biological processes are not being intentionally targeted or optimized to function as components of the wastewater treatment process.

Some methods (e.g. air stripping; breakpoint chlorination; chemical precipitation) are well-established but may be applicable only to one specific target nutrient, while others (e.g. hydrogel/polymer matrix encapsulation; ion-exchange materials; membrane-based separations) are in various stages of development and use depending on the nutrient targeted for removal/recovery. In this section, we give a brief overview of these mature and emerging technologies for N and P removal, while providing commentary on the relevant benefits and knowledge gaps as relates to their application in NSSS. A summary of the limitations and potential R&D opportunities for these technologies is presented in Table 1. It is important to note that the ISO 30500 standard is written with specific guidance (including liquid effluent reduction threshold values) for *removal* of N and P. While *recovery* and *reuse* of nutrients are critical topics for sustainability and implementation of NSSS technologies, the primary technological hurdle currently facing RT system deployment and water reuse is nutrient *removal*. We briefly mention situations where recovery/reuse is possible and indicate this also in Table 1, as we feel this is an important and exciting area for continued research. However, a comprehensive review of nutrient recovery/reuse is outside the scope of this Open Letter.

Air stripping

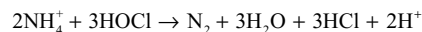
Air stripping is a process which relies on the aqueous equilibrium between the ammonium ion (NH_4^+) and free ammonia (NH_3). Under optimal conditions, this equilibrium is shifted to favor free ammonia which can leave the solution via evaporation. As a physical process, air stripping efficiency is affected by the ambient temperature and pressure. However, the solution pH is the predominant factor in determining availability of free NH_3 ; higher pH heavily favors NH_3 , with the optimal pH for air stripping being ≥ 10 . Optimized NH_3 removal via air stripping is in excess of 90%^{3,20,21}.

Chemical addition of large quantities of base is required to bring the wastewater pH into the optimal range for air stripping. It follows that acid addition is often required to bring the pH back down to the acceptable range for subsequent effluent discharge. Significant investments in infrastructure and/or land can be required for stripping towers, aeration ponds, etc²⁰. While it may be possible to effectively scale down the infrastructure, conventional ammonia air stripping is largely incompatible with RTTC guidelines due to the need for caustic chemical additives.

Electrochemical stripping may be one viable alternative to conventional air stripping for decentralized systems where intermittent electricity or photovoltaics are available²². In electrochemical stripping, base production is accomplished *in situ* using electricity and an appropriate anode material, and ammonia is separated from the solution via a gas-permeable membrane and without the need for a stripping tower. This emerging technology faces several challenges, including electrode poisoning/stability, membrane fouling, and generation of undesirable byproducts. These issues will be discussed in more detail in the subsections below.

Breakpoint chlorination

For ammonia-containing wastewater, breakpoint chlorination describes the chemical process whereby a sufficient amount of hypochlorous acid is present to completely oxidize ammonia to nitrogen gas:



The above description and equation are deceptively simplified, as multiple intermediate steps involving the formation and subsequent reaction of chloramines are not explicitly shown. Rather than adding chlorine directly to wastewater, hypochlorous acid can be generated *in situ* electrochemically by catalytic oxidation of chloride ions at an appropriate electrode surface. Breakpoint chlorination can reliably give ammonia-to-nitrogen conversions of $\geq 95\%$ with no measurable nitrous oxide formation under appropriate operating conditions^{21,23}.

Breakpoint chlorination initially appears to be an attractive method of N removal, as it typically requires no chemical additives (unless the chloride concentration in the wastewater influent is too low, in which case salt, e.g. sodium chloride, addition is warranted), generates harmless nitrogen gas, and uses technology already deployed in many NSSS. (Electrochemical oxidation of chloride for liquid disinfection is currently used in several RT systems^{14–16}.) However, there are some important limitations, including the possibility for formation of undesirable oxidation byproducts, many of which are toxic or carcinogenic^{24–26}. The formation of undesired byproducts has been well documented for electrochemical wastewater treatment processes, including chlorine disinfection^{20,27,28}. Breakpoint chlorination for ammonia removal uses the same active compounds as chlorine disinfection, but at far higher concentrations, which dramatically increases the chances for formation of harmful byproducts. In fact, the chloramine reaction intermediates themselves are considered undesired byproducts^{3,20,21}. This makes incomplete oxidation of ammonia to nitrogen problematic, which drives demand for an excess of reactive free chlorine, and necessitates a downstream dechlorination process. Control and remediation of byproducts downstream of the breakpoint chlorination process is thus required, which adds complexity and cost^{3,20,27}.

Additional issues with breakpoint chlorination include: (1) acidification of the treated effluent and subsequent creation of nitrogen trichloride; (2) need for chemical addition to neutralize pH, causing large increases in total dissolved solids in the treated effluent; (3) finite lifetimes of electrode materials (which contain expensive noble metals and metal oxides) due to surface inactivation/poisoning; (4) need for automated monitoring and control of pH, ammonia, and free chlorine levels; (5) practicality primarily as a polishing technique, rather than for removing high levels of N; (6) does not allow for recovery of N in a bioavailable form^{3,20,21}. Despite the reliability and efficiency of breakpoint chlorination, the associated downstream problems should exclude its use in the RTTC portfolio. There may be other electrochemical processes for N removal that are less problematic and more

Table 1. Non-biological methods of N and P removal. Values in *italic*, lowercase text indicate potential technological limitations for use in NSSS. Values in all capital letters indicate potential technological advantages for use in NSSS. Values in **bold text** indicate areas of existing R&D opportunity.

Method	Nutrient removed (N, P, or both)	Requirements/restrictions (YES/NO)						Notes
		Chemical additives?	Other consumables?	Power consumption?	Component fouling?	Regeneration possible?	Resource recovery possible?	
Air stripping	N (as NH ₃)	<i>yes*</i>	NO	<i>yes</i>	NO	n/a	YES #	*Large quantities of base required to raise pH. #Typically requires precipitation in sulfuric acid.
Breakpoint chlorination	N (as N ₂)	<i>yes*</i>	<i>yes</i> #	<i>yes</i>	<i>yes</i> #	<i>no</i> #	no	*Salt addition may be required depending on effluent quality. #Costly noble-metal electrodes are eventually poisoned.
Chemical precipitation	P (using Al, Fe, Ca) both (using Mg)	<i>yes</i>	<i>yes</i>	NO	NO	<i>no</i>	YES*	*Cost and processes not optimized.
Hydrogel/polymer matrix encapsulation	both	NO	yes*	NO	Unknown #	Unknown #	Unknown #	*Assumes hydrogel cannot be regenerated. #These technologies are not mature – requires additional R&D.
Ion-exchange materials	both	NO	yes*	NO	Unknown #	YES #	YES #	*Adsorbent must be replaced or regenerated. #R&D opportunities in materials discovery, surface chemistry modification, and resource recovery.
Membrane-based separations	both	NO	yes*	<i>yes</i>	yes #	Unknown #	YES	*Assumes membrane has a finite lifetime and cannot be regenerated. #Membrane technologies are not mature – requires additional R&D.

effective, e.g. electro dialysis, which will be discussed in more detail below.

Chemical precipitation

Chemical precipitation involves the addition of soluble salts to nutrient-rich wastewater to induce formation of nutrient-containing, insoluble compounds, which are removed by settling (gravity) or filtration. Chemical precipitation is currently the primary method of P remediation in wastewater treatment applications, and the most common compounds used for P removal contain Fe, Al, Ca, and/or Mg²⁹. Aside from the obvious requirement for chemical additives, a potential drawback of chemical precipitation is that P and/or N is merely sequestered rather than being removed from the wastewater treatment system. A consequence of this is the generation of large quantities of sludge that still require disposal and/or subsequent treatment. The sludge quantity is further increased by the need for chemical additives in excess of the theoretical stoichiometric requirements, presumably due to various side reactions^{2,20}. Small-scale NSSS may lack the space and/or infrastructure required to handle large amounts of sludge on site.

The particular chemical additive to be used in any treatment system needs to be chosen with care, as problems such as scaling and slow precipitation kinetics will be heavily influenced by the influent water chemistry. Often, pH adjustment by addition of hydroxide salts is required to favor formation of the desired precipitate²⁰. The choice of cation used for precipitation is critical and affects myriad factors including overall cost, P removal efficacy, optimal pH and temperature required for P removal, and reusability/bioavailability of the P-containing precipitate³⁰. Resulting trade-offs need to be carefully considered depending on the context and effluent requirements. For example, the Fe and Al salts used for P precipitation are typically much less expensive than Ca and Mg salts; however, Fe and Al phosphate precipitates are not suitable for direct use as fertilizers due to low P bioavailability³¹. On the other hand, using Mg for struvite precipitation can enable simultaneous removal of phosphate and ammonium in a 1:1 stoichiometric ratio and has been shown to be widely bioavailable;³⁰ however, struvite precipitation has a narrow window of optimal pH and cannot be relied upon as the sole method of N removal due to the much higher concentrations of ammonia/ammonium relative to phosphate³². Furthermore, more research is required to address concerns regarding the presence of contaminants in precipitates where fertilizer use is intended³⁰⁻³².

There still exists some room for improvement of chemical precipitation technologies, e.g. with the novel application of materials such as calcium silicate hydrate, which can initiate precipitation and simultaneously adjust pH³³, thus decreasing the quantity of added chemicals needed for phosphate removal. Electrochemically-induced coagulation or precipitation using sacrificial anodes can provide better control of critical process parameters, including pH and metal ion dosing, and may be easier to implement than direct salt addition in decentralized treatment settings³⁴⁻³⁶. If the nutrient-containing precipitate is a valuable commodity (e.g. fertilizer) and can be efficiently

separated from sludge, the use of a chemical additive may be justified by the benefits endowed from nutrient recovery. However, converting chemically-bonded nutrients in some precipitates to a bioavailable form can be difficult². New nanocomposite materials could be designed to increase capacity and regeneration capability³⁷. Depending on the influent characteristics, some industrial byproducts (e.g. gypsum, fly ash, slag) may provide a cost-effective solution for nutrient remediation²⁹. Ultimately, the overall cost, availability, sludge disposal, and potential downstream environmental impact of chemical precipitation processes will need to be evaluated on a case-by-case basis for each wastewater stream and its corresponding effluent requirements.

Hydrogel/polymer matrix encapsulation

Hydrogels and polymer matrices are a relatively new class of low-cost adsorbent materials being studied for P and N capture³⁸⁻⁴². Hydrogels have a high water content and porous structure that facilitates solute diffusion⁴³. As these materials are synthetic, they offer the opportunity to tune the absorptive selectivity and capacity by altering the polymer chemistry. Hybrid hydrogels can also be created by embedding inorganic particles into the polymer matrix, which can further increase the adsorption capacity⁴³. Hydrogels also have high potential for recycling and regeneration^{43,44}. To date, research for wastewater treatment with hydrogels has primarily focused on adsorption of organic compounds, radionuclides, and dyes^{43,44}. Recently, a bentonite-based hybrid hydrogel showed selectivity for phosphate adsorption in a mixed-anion waste stream with 99% phosphate removal under optimal conditions³⁸. A commercially available polymer hydrogel was demonstrated to have ammonia removal capacity up to 80% at pH 5.0–8.0, and showed minimal performance loss after regeneration with mild acid washing⁴². Development of new, tailored hydrogels for P and N removal is a promising area for future research.

Ion-exchange materials

Ion-exchange materials are highly porous structures loaded with ions which are selectively displaced by target ions (e.g. ammonium; phosphate) in the wastewater stream. Ion-exchange materials offer several benefits when compared to conventional chemical precipitation methods. For example, while chemical precipitation can handle higher nutrient concentrations, the kinetics of ion-exchange processes are much faster in comparison and thus the removal efficiency is not highly dependent on retention time. Additionally, nutrients captured by ion exchange are more easily recovered in comparison to e.g. chemical precipitation, as ion-exchange processes are reversible²⁹.

A fundamental limitation of ion-exchange materials is that their capacity is inherently limited by the accessible surface area and ion loading of the material; at some point, the adsorbent will need to be replaced or regenerated. Synthetic ion-exchange resins, while potentially engineered to have better selectivity, adsorption and regeneration capacities, and durability than natural zeolites, can be far more costly⁴⁵. On the other hand, certain natural zeolites may be geographically scarce. Conventional methods of material regeneration require large volumes of water and quantities of salts, including caustic acids and bases⁴⁶. There is also the issue

of the so-called “paradox of P sorption materials”, wherein the materials that are highly effective at P removal generally show poor performance for water transport²⁹.

Despite these issues, there is opportunity for using ion-exchange materials in a RTTC context. In particular, the option to regenerate the adsorbent material and recover a nutrient-rich solution is attractive when considering strategies for integrating NSSS into a circular N and/or P economy. For any particular ion-exchange material, testing with the real, intended wastewater stream is critical, as influent properties such as pH and organic loading can have dramatic effects on adsorption and regeneration capacities^{9,45}. Offsite zeolite regeneration using a spoke-hub model could create local employment opportunities, as the regeneration/recovery process need not be highly technical and the regeneration solution can be reused many times. However, access to and cost of salts required for regeneration would still need to be considered⁴⁵. Optimizing the zeolite regeneration process to operate on a small scale with limited additives would be a significant achievement that merits investigation⁴⁶.

Even without regeneration, the low cost of certain natural zeolites and other minerals may make them effective as consumable filtration media in small-scale systems. For example, clinoptilolite is one of the best natural ion exchange materials for ammonium ions (capacity = 2–30 mg NH₄⁺ g⁻¹), and has been demonstrated to have a lower material cost (\$ per g of N removed) than conventional biological nitrogen removal, even when treated as a one-time-use consumable⁴⁵. Spent natural zeolites may have additional value as soil conditioners and fertilizers^{46–48}. Polonite® is a particularly attractive material for P removal and recovery due to its high P capacity (up to 12% by weight)^{49–51}, and reasonable price (< US\$1 kg⁻¹). The exhausted filter media can be applied directly to soil for agricultural use as a [slow-P-releasing fertilizer](#) and soil conditioner⁵². If the two processes are used in series, the more basic Polonite®-filtered effluent could counteract the undesired pH drop induced by *in situ* chlorine disinfection. We are currently evaluating the use of Polonite® and clinoptilolite for ISO 30500 compliance in NSSS.

Ammonia removal from wastewater using natural⁵³ and synthetic^{9,48} ion-exchange materials has been studied for several decades; for the natural zeolite clinoptilolite, the average ammonia removal efficiency is ≥95%²¹. Despite the use of ion-exchange materials for decades in remediation of ammonia, ion-exchange materials can still be considered an “emerging technology” for P removal in wastewater treatment applications. Hybrid anion-exchange resins (HAIX) consist of iron (hydr)oxide particles embedded within anion-exchange polymers and have been tested with a variety of wastewater streams, including source-separated urine^{54,55}. The operational capacity of HAIX media depends on both the influent P concentration and the target effluent P concentration^{56,55}, and thus use of HAIX in NSSS will need to be assessed on a case-by-case basis. More work is also needed to demonstrate the economic feasibility of these materials (including the regeneration process) for use in small-scale NSSS; field testing and pilot-scale studies will be critical for evaluating long-term viability of HAIX. Overall,

there are ample opportunities for fundamental R&D in synthetic ion exchange resins, as well as modified and unmodified natural zeolites, for nutrient removal in NSSS^{48,57}.

Membrane-based separations

Membrane-based separation technologies are emerging as promising methods of nutrient removal for small-scale water treatment systems. Both active (energy required to drive separation; e.g. electrodialysis) and passive (separation driven by chemical equilibrium; e.g. osmosis, ion-selective membranes) membrane separation methods are dynamic areas of research for nutrient removal applications. For example, forward osmosis from source separated urine was recently shown to give at least 50% N and 93% P recovery into a diluted fertilizer draw solution. However, typical issues of membrane scaling and fouling were found to increase the operating costs of this system, due to the need for membrane replacement⁵⁸.

All membrane-based technologies suffer from a few key limitations in the membranes themselves, including membrane fouling and poor selectivity⁵⁹. However, important fundamental advances are continually being made in the development of tailored membrane materials⁶⁰. For example, highly selective membranes have recently been fabricated using covalent organic frameworks with tunable functional groups and nanoporous structures⁶⁰. However, cost and scalability of any new membrane materials must be considered from the earliest stages of development for application in NSSS, particularly for deployment in low- and middle-income countries. Opportunities also exist in modification of existing membranes and processes to tailor them for use in NSSS. Membrane surface modification can be used to increase hydrophilicity and membrane selectivity. Surface modification was recently used to demonstrate >89% ammonium removal in a non-optimized forward osmosis system⁶¹. Optimization of operating conditions and in depth techno-economic analyses of nutrient recovery may make membrane-separation-based technologies viable options under certain conditions. In particular, membrane fouling can be largely mitigated when membranes are used in “tertiary” nutrient removal processes (i.e. after COD removal).

Another promising method for mitigating membrane fouling is through electrodialysis reversal (EDR). In this process, the electrical polarity is switched periodically between the anode and cathode material to enable *in situ* self-cleaning of the electrode and membrane surfaces⁵⁹. EDR is already widely used in water desalination technologies, and one company (Saltworks Technologies Inc.) has developed a [modular EDR cell stack](#) demonstrating >95% ammonia removal in a municipal WWTP. Whether electrodialysis can be cost effective on a small scale and effectively handle more concentrated wastewater (blackwater) for use in NSSS will need to be demonstrated.

Reconsidering biological N and P remediation at small scales

There is likely not a “one-size-fits-all” solution to tackling the problem of nutrient removal at small scales². Above, we outlined some of the opportunities for R&D in physical/chemical methods

of nutrient removal in the context of NSSS. At present, there is also a significant knowledge gap as to whether biological systems can provide efficient nutrient removal in NSSS. With the exception of constructed wetlands⁶², there is very little data available at the household/community scale². Some of the major limitations for using biological systems in NSSS is their susceptibility to environmental stress, long start-up times, infrastructure requirements, and technical complexity. Performance of biological systems would be highly dependent on the local climate, seasonal variations, and ecosystem, and would require appropriate pilot tests in addition to laboratory efforts. Addressing these issues at small scales has not been adequately investigated, which presents opportunities for fundamental R&D that could support the goals of RTTC. Some opportunities for further investigation are briefly outlined below.

The lines between traditional “biological” treatment processes (i.e. systems intentionally inoculated with specific microbial strains) and truly “non-biological” treatment methods are likely already blurred in many RT systems due to the high microbial load of feces, which could yield formation of “native” biofilms on or within system components. The role of native biofilms in nutrient sequestration thus cannot be ignored³³, and the conditions which favor biological N and P sequestration in native biofilms could be optimized; more work is needed in this area, and would be specific to a given wastewater stream and treatment context². Gravity-driven membrane filtration systems have been reviewed recently and it is proposed that these systems can be economically viable at household- and community-scales⁶³. The formation of a stable biofilm on the membrane surface is a critical factor in their operation. The nutrient removal capabilities of these membrane systems have not been optimized for use in small-scale NSSS.

Hybrid chemical/biological systems are known to meet ISO 30500 standards for nutrient removal on large scales, but adapting these technologies to small scales will require extensive re-engineering and research efforts. For P removal in particular, it has been shown that it may not be possible to downscale existing technologies while maintaining performance². Nevertheless, some emerging technologies which combine chemical and biological processes show promise for efficient nutrient removal and recovery at small scales^{28,64,65}. Combining FeCl₃ dosing with a membrane bioreactor and cofermentation was recently demonstrated, yielding >98% P removal and subsequent recovery (>60%) from the bioreactor sludge⁶⁶. (The precipitate recovered in this case was vivianite, which may have value in electronic applications and as a fertilizer precursor.)^{67,68} An electrochemically assisted, small-footprint, constructed wetland

recently demonstrated >99% phosphate and >93% total nitrogen removal in tertiary wastewater treatment⁶⁹. Developing hybrid chemical/biological technologies specifically for small-scale blackwater treatment could yield breakthroughs in nutrient removal efficiencies.

The generation of biomass inherently consumes large quantities of N and P. The biomass product can serve as a valuable commodity, e.g., as an animal food supplement or via conversion to biofuel. One interesting area for future research would be in exploiting the eutrophication process by intentionally cultivating algae in a NSSS^{70,71}. Algae can remove N and P from wastewater at least as effectively as many chemical treatments and can be a high-value biomass commodity^{2,72}. As algae species are ubiquitous in aqueous environments, biological additives may not be necessary if the conditions for sustained algal growth are optimized, and nutrient removal efficiency may be independent of the particular algal community cultivated⁷³. Algae cultivated in a NSSS could be dried passively in the sun and used as a nutrient supplement for livestock in the local community. For example, adding small amount of dried algae to cows’ food can increase the omega-3 content of their milk⁷⁴ and decrease methane emissions⁷⁵. Engineering innovations are still needed to optimize small-scale algae cultivation, as well as algal biofilm stability, separation, and recovery^{2,76–78}. In particular, adapting algal-based systems to optimize light exposure in a minimal areal footprint would be critical for implementation in high population-density areas.

Conclusion

Rather than seeking a silver-bullet solution, installing sustainable methods for small-scale, decentralized nutrient removal will necessitate a case-by-case approach that takes into account the myriad technical, cultural, and economic constraints unique to each water-use scenario. The need for customized sanitation solutions should motivate investment in basic R&D for emerging technologies, including non-biological, biological, and hybrid solutions. The development of reliable non-biological nutrient removal methods may be critical especially for use in colder climates, and there is ample research space to explore options including ion-exchange processes, hydrogel materials, and membrane-driven separations. Furthermore, the potential of biological solutions for nutrient removal has been underappreciated in the RT portfolio to date. Incorporation of biological components may be indispensable in meeting the ISO 30500 effluent requirements in certain situations.

Data availability

No data is associated with this article.

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Current Peer Review Status:   

Version 2

Reviewer Report 15 May 2020

<https://doi.org/10.21956/gatesopenres.14324.r28852>

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Lauren F. Greenlee 

Ralph E. Martin Department of Chemical Engineering, University of Arkansas, Fayetteville, AR, USA

No additional comments.

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Water treatment; electrochemical engineering; electrocatalysis; nutrient recovery; nanotechnology

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Version 1

Reviewer Report 24 April 2019

<https://doi.org/10.21956/gatesopenres.14033.r27001>

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Lauren F. Greenlee 

Ralph E. Martin Department of Chemical Engineering, University of Arkansas, Fayetteville, AR, USA

The article presents a useful overview of N and P removal/recovery options from the perspective of small-scale RT systems.

Specific comments:

1. The introduction mentions several times “mature” or “conventional” technology solutions for removing N and P. I think it would be helpful to have a brief description early on (maybe just a few words or specific examples) of what these technology solutions are so that the reader knows up front what these technologies are – it could make the transition from the introduction to the core of the article better.
2. Is there a large energy demand to air stripping, and/or for the other processes mentioned? How does the energy demand compare across the technology space and how are the energy supply challenges addressed for the small-scale RT systems?
3. I would recommend electrochemical N and P recovery be added to Table 1. See for example: Hug and Udert (2013[ref-1]); Lin *et al.* (2018²); <https://www.igb.fraunhofer.de/en/research/competences/physical-process-technology/nutrient-mar>.
4. Is there any movement in the hydrogel/polymer matrix or ion exchange resin research toward not just N and P removal but recovery as a usable composite for fertilization? For example, designing systems where the polymer or resin used could actually be directly land applied as a soil amendment? You have Polonite as an example, but are there other efforts in this area?
5. The sentence on the “paradox of P sorption materials” describes the materials that are good at P removal and are poor water conductors. I would recommend changing the wording to say “poor performance for water transport”. The word “conductor” implies conduction, which would be transport of heat/electrons, but I think you mean that the movement of water molecules through these materials is poor, which would be related to the convective/diffusive transport of water.
6. The authors mention new developments in the membrane design/materials field. It might be important to also comment on the cost and scalability of these new membrane designs, as these two factors are often extremely limiting for new membrane materials. For researchers in this area reading this article, it’s important for people to think about cost and scalability from the beginning when designing new materials for this application.

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Is the rationale for the Open Letter provided in sufficient detail?

Yes

Does the article adequately reference differing views and opinions?

Partly

Are all factual statements correct, and are statements and arguments made adequately supported by citations?

Yes

Is the Open Letter written in accessible language?

Yes

Where applicable, are recommendations and next steps explained clearly for others to follow?

Partly

Competing Interests: No competing interests were disclosed.**Reviewer Expertise:** Water treatment; electrochemical engineering; electrocatalysis; nutrient recovery; nanotechnology**I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.**

Author Response 08 May 2020

Lena Trotochaud, Duke University, Durham, USA

We sincerely thank the Reviewer for their insightful comments and suggestions. We have addressed each of their comments below, and we feel that the manuscript is much improved due to their feedback. The Reviewer's comments are copied below in *italic text*, and our responses are in plain text. Changes to the manuscript are shown in **bold text**.

The article presents a useful overview of N and P removal/recovery options from the perspective of small-scale RT systems.

We sincerely thank the Reviewer for their comments and for the helpful suggestions for improvement.

Specific comments:

1. The introduction mentions several times “mature” or “conventional” technology solutions for removing N and P. I think it would be helpful to have a brief description early on (maybe just a few words or specific examples) of what these technology solutions are so that the reader knows up front what these technologies are – it could make the transition from the introduction to the core of the article better.

We have added more specific examples to the beginning of the section “Mature and emerging technologies for non-biological nutrient removal” to better integrate with and transition to the later subsections:

“Some methods (e.g. air stripping; **breakpoint chlorination; chemical precipitation**) are well-established **but may be** applicable only to one specific target nutrient, while others (e.g. **hydrogel/polymer matrix encapsulation**; ion-exchange materials; **membrane-based separations**) are in various stages of development...”

2. Is there a large energy demand to air stripping, and/or for the other processes mentioned? How does the energy demand compare across the technology space and how are the energy supply challenges addressed for the small-scale RT systems?

We specifically included a column in Table 1 to acknowledge which technologies will require electricity from some source, however, it is difficult to put a number on energy demand for any given technology for use in small-scale NSSS when the only available data are for conventional systems at large wastewater treatment plants (e.g. air stripping). Furthermore, the energy demand of some technologies can fluctuate over time and during periods of intermittent use, which will be more noticeable in a small-scale system than for a large WWTP. In our experience, it is extremely difficult to estimate energy demand for small-scale NSSS without context-specific field/pilot testing and/or an in-depth techno-economic analysis.

The original aim of the Reinvent the Toilet Challenge was to create a toilet that “operates ‘off the grid without connections to water, sewer, or electrical lines”. Strictly speaking, any RT system would ideally generate its own electricity (e.g. with thermoelectrics) or use solely renewable sources (e.g. solar) for all power needs. However in practice, some basic electrical connectivity is assumed in many RT prototype systems. Nevertheless, energy requirements are a critical consideration for RT systems, due to lack of reliability of grid power in many rural areas and some low- and middle-income countries, as well as the high cost of electricity in some areas, even when it is reliably available.

3. I would recommend electrochemical N and P recovery be added to Table 1. See for example: Hug and Udert (2013[ref-1]); Lin et al. (2018²);

<https://www.igb.fraunhofer.de/en/research/competences/physical-process-technology/nutrient-manage>

Electrochemically-induced coagulation or struvite precipitation is indeed an interesting area of research, with several benefits over salt addition, including better control of pH and metal ion dosing. However, the chemistry involved in formation of precipitates is identical, so we believe adding it as a separate item in the table is not necessary. However, we have added the following text and citations to acknowledge the benefits of electrochemically-induced coagulation/precipitation:

“Electrochemically-induced coagulation or precipitation using sacrificial anodes can provide better control of critical process parameters, including pH and metal ion dosing, and may be easier to implement than direct salt addition in decentralized treatment settings.[Lacasa 2011, Hug 2013, Garcia-Segura 2017]

Lacasa E, Cañizares P, Sáez C, Fernández FJ, Rodrigo MA: Electrochemical phosphates removal using iron and aluminium electrodes. *Chem. Eng. J.* 2011;172(1):137-143

Hug A, Udert KM: Struvite precipitation from urine with electrochemical magnesium dosage. *Water Res.* 2013;47(1):289-99. 10.1016/j.watres.2012.09.036

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4. *Is there any movement in the hydrogel/polymer matrix or ion exchange resin research toward not just N and P removal but recovery as a usable composite for fertilization? For example, designing systems where the polymer or resin used could actually be directly land applied as a soil amendment? You have Polonite as an example, but are there other efforts in this area?*

We are not aware of any examples of hydrogel/polymer or ion exchange resin development for direct application as fertilizer or soil amendment, although this would be an interesting area for future research. There are several examples of mineral/zeolite based materials, including Polonite and clinoptilolite which we mentioned. However, the general strategy and research focus with ion exchange and polymer-based materials seems to be to regenerate the material and use it repeatedly, and recover nutrients from the regeneration solution. Nevertheless, it seems that many of these materials cannot be regenerated indefinitely, which raises the question as to what to do with them when they have reached full exhaustion. While considering this comment, we realized that we neglected to mention the hybrid-anion exchange resins (HAIX) that have been tested recently for phosphate removal and recovery. We have added the following text and references to include these important materials:

“Hybrid anion-exchange resins (HAIX) consist of iron (hydr)oxide particles embedded within anion-exchange polymers and have been tested with a variety of wastewater streams, including source-separated urine.[Sengupta 2011, O’Neal 2013] The operational capacity of HAIX media depends on both the influent P concentration and the target effluent P concentration,[Martin 2017, O’Neal 2013] and thus use of HAIX in NSSS will need to be assessed on a case-by-case basis. More work is also needed to demonstrate the economic feasibility of these materials (including the regeneration process) for use in small-scale NSSS; field testing and pilot-scale studies will be critical for evaluating long-term viability of HAIX.”

Sengupta S, Pandit A: Selective removal of phosphorus from wastewater combined with its recovery as a solid-phase fertilizer. *Water Res.* 2011;45(11):3318-30.

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Martin BD, De Kock L, Gallot M, Guery E, Stanowski S, MacAdam J, McAdam EJ, Parsons SA, Jefferson B: Quantifying the performance of a hybrid anion exchanger/adsorbent for phosphorus removal using mass spectrometry coupled with batch kinetic trials. *Environ. Technol.*

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O’Neal JA, Boyer TH: Phosphate recovery using hybrid anion exchange: Applications to source-separated urine and combined wastewater streams. *Water Res.* 2013;47(14):5003-17.

10.1016/j.watres.2013.05.037

5. *The sentence on the “paradox of P sorption materials” describes the materials that are good at P removal and are poor water conductors. I would recommend changing the wording to say “poor performance for water transport”. The word “conductor” implies conduction, which would be transport of heat/electrons, but I think you mean that the movement of water molecules through these materials is poor, which would be related to the convective/diffusive transport of water.*

We have changed the text in this section to clarify this point:

“...wherein the materials that are highly effective at P removal generally **show poor performance for water transport.**”

6. The authors mention new developments in the membrane design/materials field. It might be important to also comment on the cost and scalability of these new membrane designs, as these two factors are often extremely limiting for new membrane materials. For researchers in this area reading this article, it's important for people to think about cost and scalability from the beginning when designing new materials for this application.

We agree with the Reviewer that cost and scalability of new membrane materials are very important to consider from the earliest stages of research. We also think that there are some membrane technologies using off-the-shelf membranes (e.g. electrodialysis) that are more well-established and could be adapted for use with NSSS potentially without the need for development of new membranes. We have added the following to this section to clarify this point:

“However, cost and scalability of any new membrane materials must be considered from the earliest stages of development for application in NSSS, particularly for deployment in low- and middle-income countries. Opportunities also exist in modification of existing membranes and processes to tailor them for use in NSSS.”

Competing Interests: No competing interests were disclosed.

Reviewer Report 11 April 2019

<https://doi.org/10.21956/gatesopenres.14033.r27000>

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Kara L. Nelson 

Department of Civil and Environmental Engineering, University of California, Berkeley, Berkeley, CA, USA

The authors address an important challenge for emerging sanitation technologies, which is the removal of nitrogen and phosphorus. A new international standard requires certain removals, and yet mature technologies that work at small scales do not yet exist. Overall the review is insightful and should help to inspire further innovation. However, more attention is needed to some topics:

1. The authors start with the premise that the N and P removals required by ISO 30500 make sense. Do the authors agree that this requirement makes sense in all scenarios?
2. The authors don't discuss that NSSS can produce different types of waste streams, depending on whether water is used for flushing, and whether urine and feces are separated. Also, some NSSS

treatment technologies produce concentrated waste streams that need further treatment (N and P removal). It would be helpful if the authors can briefly summarize these different streams that can be targeted for N and P removal.

3. Table 1: Caption states that the table provides a summary of N and P removal methods, but some of the methods only work for N (air stripping and breakpoint chlorination), and chemical precipitation is primarily developed for P (via struvite, which only removes about 10% of N). These are crucial distinctions. The Table should be revised to indicate which approaches work for N, P, or both.
4. Breakpoint chlorination section: Although stated in Table 1, it is worth mentioning in the text that this method does not allow recovery of N in a usable form (only removal as N₂).
5. Chemical precipitation section: This section makes it seem as if all precipitation strategies are similar in terms of their removal levels of N and P, but in fact the removal potential completely depends on the stoichiometry of the precipitate that is produced. These distinctions should be made.
6. Ion exchange section: Is there any literature that can be cited for Polonite?
7. Algal-based systems: These will require large outdoor areal footprints to provide algae with sufficient sunlight. This limitation should be mentioned.

Is the rationale for the Open Letter provided in sufficient detail?

Yes

Does the article adequately reference differing views and opinions?

Partly

Are all factual statements correct, and are statements and arguments made adequately supported by citations?

Partly

Is the Open Letter written in accessible language?

Yes

Where applicable, are recommendations and next steps explained clearly for others to follow?

Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Treatment technologies for wastewater, urine, fecal sludge, drinking water, water reuse.

I confirm that I have read this submission and believe that I have an appropriate level of expertise to state that I do not consider it to be of an acceptable scientific standard, for reasons outlined above.

Author Response 08 May 2020

Lena Trotochaud, Duke University, Durham, USA

We sincerely thank the Reviewer for their insightful comments and suggestions. We have addressed each of their comments below, and we feel that the manuscript is much improved due to their feedback. The Reviewer's comments are copied below in *italic text*, and our responses are in plain text. Changes to the Manuscript are shown in **bold text**.

The authors address an important challenge for emerging sanitation technologies, which is the removal of nitrogen and phosphorus. A new international standard requires certain removals, and yet mature technologies that work at small scales do not yet exist. Overall the review is insightful and should help to inspire further innovation. However, more attention is needed to some topics:

We sincerely thank the Reviewer for their positive comments regarding the manuscript and for the helpful suggestions for improvement.

1. The authors start with the premise that the N and P removals required by ISO 30500 make sense. Do the authors agree that this requirement makes sense in all scenarios?

The Reviewer raises an excellent point. However, we point out that we make no claims regarding the suitability nor the sensibility of the ISO 30500 standards; the fact is that these standards exist and are likely to be applied to at least some of the RTTC systems. We think that the appropriateness of the ISO 30500 standards is an important conversation that researchers and policy makers in this field should continue to have. However, we believe that it is not within the scope of the current work to opine on this issue.

To add a bit of commentary: One overarching conclusion from our Open Letter is that a one-size-fits-all approach to NSSS is not likely to be successful. One could extrapolate then also that a one-size-fits-all approach to regulation of NSSS may be inappropriate. While one could argue that certain metrics (e.g. pathogen reduction) are non-negotiable due to the immediate implications for human health, blanket requirements for N and P removal may be inappropriate or restrictive in certain cases. For example, what if a certain NSSS produces pathogen-free, liquid effluent that contains ammonia and phosphate in concentrations appropriate for direct application as a fertigation solution? Does it make sense to require that the N and P be removed, for example by precipitation, only for that solid product to be re-dissolved later in the same reclaimed water at a later time point for fertigation purposes? Ultimately, the ISO 30500 standards have already been adopted in several countries (including Senegal, the USA, and Canada) and adoption is expected to continue, so the sensibility of the standards may already be a moot point (unless there are future opportunities for revision of the standards).

2. The authors don't discuss that NSSS can produce different types of waste streams, depending on whether water is used for flushing, and whether urine and feces are separated. Also, some NSSS treatment technologies produce concentrated waste streams that need further treatment (N and P removal). It would be helpful if the authors can briefly summarize these different streams that can be targeted for N and P removal.

We thank the reviewer for this comment, which is related to concerns raised also by Reviewer 1. The ISO 30500 standards specifically dictate removal from N and P from *liquid* effluent; we have made changes at several points in the manuscript to clarify this limit in scope. (Please see the response to Reviewer 1 for details.)

We have also added text in the introduction to address the point of different types of waste streams:

“The technological approach for N and P removal from liquid effluent will depend to a great extent on whether, how, and when urine and feces are separated from each other. Also important is whether and to what extent urine is diluted by water used for toilet flushing (and personal washing, where applicable).”

3. Table 1: Caption states that the table provides a summary of N and P removal methods, but some of the methods only work for N (air stripping and breakpoint chlorination), and chemical precipitation is primarily developed for P (via struvite, which only removes about 10% of N). These are crucial distinctions. The Table should be revised to indicate which approaches work for N, P, or both.

We have added a column to the table indicating which approaches work for N, P, or both.

4. Breakpoint chlorination section: Although stated in Table 1, it is worth mentioning in the text that this method does not allow recovery of N in a usable form (only removal as N₂).

We have added item 6 to the list of “additional issues with breakpoint chlorination”:

“...(6) does not allow for recovery of N in a bioavailable form.”

5. Chemical precipitation section: This section makes it seem as if all precipitation strategies are similar in terms of their removal levels of N and P, but in fact the removal potential completely depends on the stoichiometry of the precipitate that is produced. These distinctions should be made.

We thank the Reviewer for this comment, which is also similar to concerns raised by Reviewer 1. We have added a discussion and several references related to the nuances of chemical precipitation. We have also clarified in Table 1 which cations are appropriate for only P removal vs both N and P removal together.

“The choice of cation used for precipitation is critical and affects myriad factors including overall cost, P removal efficacy, optimal pH and temperature required for P removal, and reusability/bioavailability of the P-containing precipitate.[Melia 2017] Resulting trade-offs need to be carefully considered depending on the context and effluent requirements. For example, the Fe and Al salts used for P precipitation are typically much less expensive than Ca and Mg salts; however, Fe and Al phosphate precipitates are not suitable for direct use as fertilizers due to low P bioavailability.[Desmidt, 2014] On the other hand, using Mg for struvite precipitation can enable simultaneous removal of phosphate and

ammonium in a 1:1 stoichiometric ratio and has been shown to be widely bioavailable;[Melia 2017] however, struvite precipitation has a narrow window of optimal pH and cannot be relied upon as the sole method of N removal due to the much higher concentrations of ammonia/ammonium relative to phosphate.[Mehta, 2015] Furthermore, more research is required to address concerns regarding the presence of contaminants in precipitates where fertilizer use is intended. [Desmidt 2014, Mehta 2015, Melia 2017]”

6. *Ion exchange section: Is there any literature that can be cited for Polonite?*

We thank the reviewer for pointing out this oversight. We have added the following references which describe testing of Polonite:

Brogowski A, Renman G: Characterization of opoka as a basis for its use in wastewater treatment. *Pol. J. Environ. Stud.* 2004;13(1):15-20.

Gustafsson JP, Renman A, Renman G, Poll K: Phosphate removal by mineral-based sorbents used in filters for small-scale wastewater treatment. *Water Res.* 2008;42(1-2):189-97.
10.1016/j.watres.2007.06.058

Renman A, Renman G: Long-term phosphate removal by the calcium-silicate material Polonite in wastewater filtration systems. *Chemosphere.* 2010;79(6):659-64.
10.1016/j.chemosphere.2010.02.035

Cucarella V, Zaleski T, Mazurek R, Renman G: Effect of reactive substrates used for the removal of phosphorus from wastewater on the fertility of acid soils. *Bioresour. Technol.* 2008;99(10):4308-14.
10.1016/j.biortech.2007.08.037

We have also re-arranged parts of this section to improve the flow of the narrative, and added a section describing work on hybrid anion-exchange (HAIX) materials for phosphate removal, in part as a response to a query raised by Reviewer 3:

“Hybrid anion-exchange resins (HAIX) consist of iron (hydr)oxide particles embedded within anion-exchange polymers and have been tested with a variety of wastewater streams, including source-separated urine.[Sengupta 2011, O’Neal 2013] The operational capacity of HAIX media depends on both the influent P concentration and the target effluent P concentration,[Martin 2017, O’Neal 2013] and thus use of HAIX in NSSS will need to be assessed on a case-by-case basis. More work is also needed to demonstrate the economic feasibility of these materials (including the regeneration process) for use in small-scale NSSS.”

Sengupta S, Pandit A: Selective removal of phosphorus from wastewater combined with its recovery as a solid-phase fertilizer. *Water Res.* 2011;45(11):3318-30.
10.1016/j.watres.2011.03.044

O’Neal JA, Boyer TH: Phosphate recovery using hybrid anion exchange: Applications to source-separated urine and combined wastewater streams. *Water Res.* 2013;47(14):5003-17.
10.1016/j.watres.2013.05.037

Martin BD, De Kock L, Gallot M, Guery E, Stanowski S, MacAdam J, McAdam EJ, Parsons SA, Jefferson B: Quantifying the performance of a hybrid anion exhcnager/adsorbent for phosphorus removal using mass spectrometry coupled with batch kinetic trials. *Environ. Technol.*

2018;39(18):2304-14. 10.1080/09593330.2017.1354076

7. *Algal-based systems: These will require large outdoor areal footprints to provide algae with sufficient sunlight. This limitation should be mentioned.*

We have added the following text to the section on algal-based systems:

“In particular, adapting algal-based systems to optimize light exposure in a minimal areal footprint would be critical for implementation in high population-density areas.”

Competing Interests: No competing interests were disclosed.

Reviewer Report 13 March 2019

<https://doi.org/10.21956/gatesopenres.14033.r26951>

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Susan K. De Long 

Department of Civil and Environmental Engineering, Colorado State University, Fort Collins, CO, USA

General comments:

This article presents a very useful and timely review of non-biological methods for removing N&P from wastewater that are suitable for small-scale systems. Generally I found the review well organized and well-written, and the presented content is highly practical. My overarching suggestions are that I would have liked to see a larger focus on opportunities for nutrient reuse throughout the manuscript text, and additionally the authors should do a better job of delineating which technologies are for N vs P in Table 1 and in the text. Additionally, the various forms of N and P should be more clearly indicated (e.g. inorganic vs. organic forms). My only other suggestions relate to adding more detail and clarification on some points, which I have listed below.

Specific comments:

1. I think it's worth briefly explaining the ISO 30500 standards in the introduction, at their first mention. Not all readers will be familiar, and this document is actually quite broadly useful beyond NSSS.
2. The design of Table 1 is not optimal. The use of italic text is very subtle. I would suggest a different and more obvious notation for indicating the limitations for use in NSSS. Also, by the title and text it is really not clear which methods are for N and with are for P. I suggest the applicability for each nutrient be separately indicated.
3. I would have liked to see text specifically discussing the column in Table 1 dealing with nutrient recovery. Ability to reuse nutrients should be considered as a critical criterion for technology development and selection. Although reuse possibilities are summarized in the table, mentioning

them in the text would bring more attention to the importance of this issue.

4. Breakpoint chlorination is said to release “almost no nitrous oxide”. It would be highly preferred to be more quantitative in this statement given the potency of N₂O as a GHG.
5. For breakpoint chlorination, a reference should be added related to byproducts which are toxic and carcinogenic to direct the reader to detailed information.
6. Please explain why breakpoint chlorination leads to high TDS in the effluent.
7. The chemical precipitation section should be expanded to individually discuss the different metals added. Please add text explaining how the choice of Fe vs. Al vs. Ca vs. Mg affects efficiency and whether the end product is reusable. Struvite is mentioned, but nothing is said about the other precipitates.
8. For the ion exchange section, change “rate” to “efficiency”. 95% is not a rate.
9. For “Reconsidering biological N and P remediation at small scales”, the meaning of “native” for biofilms is not specified. Some of the transitions in this section are rough as well.
10. Please clarify that for inorganic P capture with FeCl₃, reuse is not viable. Please also more clearly discuss inorganic vs organic forms of P.

Is the rationale for the Open Letter provided in sufficient detail?

Yes

Does the article adequately reference differing views and opinions?

Yes

Are all factual statements correct, and are statements and arguments made adequately supported by citations?

Partly

Is the Open Letter written in accessible language?

Yes

Where applicable, are recommendations and next steps explained clearly for others to follow?

Partly

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Environmental engineering, biological treatment, applied microbiology, contaminant biodegradation, nutrient recovery

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Author Response 08 May 2020

Lena Trotochaud, Duke University, Durham, USA

We sincerely thank the Reviewer for their insightful comments and suggestions. We have addressed each of their comments below, and we feel that the manuscript is much improved due to their feedback. The Reviewer's comments are copied below in *italic text*, and our responses are in plain text. Changes to the Manuscript are shown in **bold text**.

This article presents a very useful and timely review of non-biological methods for removing N&P from wastewater that are suitable for small-scale systems. Generally I found the review well organized and well-written, and the presented content is highly practical. My overarching suggestions are that I would have liked to see a larger focus on opportunities for nutrient reuse throughout the manuscript text, and additionally the authors should do a better job of delineating which technologies are for N vs P in Table 1 and in the text. Additionally, the various forms of N and P should be more clearly indicated (e.g. inorganic vs. organic forms). My only other suggestions relate to adding more detail and clarification on some points, which I have listed below.

We sincerely thank the Reviewer for their positive comments regarding the manuscript and for the helpful suggestions for improvement.

On the point of the Reviewer's desire to "see a larger focus on opportunities for nutrient reuse throughout", we agree that nutrient reuse is indeed a critically important topic in addition to nutrient removal. However, the scope of this article was specifically limited to analysis of nutrient *removal*, as this is the metric specified in the ISO 30500 standard for water reuse.

There are many questions that surround nutrient reuse that are extremely case specific (e.g. Is the reuse for landscaping, agriculture, or livestock nutritional supplement?; For agricultural use, what are the specific crops to be cultivated and does the method of nutrient recovery support this?; What additional costs/added chemicals/processes are involved for nutrient reuse and are these affordable in a low- and/or middle-income country context?; Is the reuse product safe in terms of pathogen/heavy metals/trace pharmaceuticals content?) We believe that an adequate treatment of nutrient *reuse* is therefore outside the scope of the current article and would best be reviewed separately. We have added the following text (emphasis included) to the main text to indicate this limitation in scope while acknowledging the importance of nutrient reuse:

"It is important to note that the ISO 30500 standard is written with specific guidance (including liquid effluent reduction threshold values) for *removal* of N and P. While *recovery* and *reuse* of nutrients are critical topics for sustainability and implementation of NSSS technologies, the primary technological hurdle currently facing RT system deployment and water reuse is nutrient *removal*. We briefly mention situations where *recovery/reuse* is possible and indicate this also in Table 1, as we feel this is an important and exciting area for continued research. However, a comprehensive review of nutrient *recovery/reuse* is outside the scope of this Open Letter."

On the point of "delineating which technologies are for N and P in Table 1 and in the text", we received comments from all Reviewers regarding the formatting of Table 1 and have made several changes in the revised version. A new column has been added which indicates whether the technologies mentioned are appropriate for removal of N, P, or both. Additionally, we have

changed the formatting of some values, including shading of certain cells, to more clearly delineate differences. (Note – our original version of this table included different colors of text, but we were informed by the editorial staff that the use of colored text is not allowed.)

On the point of more clearly indicating the organic and inorganic forms of N and P, we thank the Reviewer for pointing out this ambiguity. The term “nutrients” or “N and P” are used frequently in the literature (including in the original version of our manuscript), but are not sufficiently precise when discussing the chemistry required for removal of organic vs inorganic N and P components. The discussion of organic vs inorganic fractions of N and P in human excreta has been covered in an extensive survey of the literature by Rose et al. (2015); specifically, 90% of total N and 50-65% of total P is contained in the urine fraction. More specifically, most of the organic fractions of N and P are excreted in feces, and thus the inorganic N and P (primarily ammonia from hydrolyzed urine and orthophosphate) is the fraction with the highest contribution to the liquid effluent. Ammonia and orthophosphate are also the nutrients of highest concern with regard to anthropogenic nutrient pollution. We have added the following text to the abstract and introduction to clarify this point:

In the abstract: “...emerging technologies for N and P (**specifically ammonia/ammonium and orthophosphate**) removal...”

In the introduction, paragraph 1: “In particular, algal blooms, caused by eutrophication due to high levels of nitrogen and phosphorus (N and P, **primarily as ammonia/ammonium and orthophosphate**),...”

In the introduction, after paragraph 2: “**The technological approach for N and P removal from liquid effluent will depend to a great extent on whether, how, and when urine and feces are separated from each other. Also important is whether and to what extent urine is diluted by water used for toilet flushing (and personal washing, where applicable). Urine contains approximately 90% of the total N and 50-65% of the total P in human excreta, and the chemical components are largely inorganic compounds (assuming urea hydrolysis); in contrast, the contributions to N and P from feces are largely organic (proteins and bacterial biomass). [Rose et al., 2015] For the purposes of this Open Letter, we focus on removal of ammonia/ammonium and orthophosphate, which are the majority water-soluble contributors to N and P in human excreta and the compounds of primary concern for anthropogenic nutrient pollution.**”

The introduction of the above text requires an additional citation to the manuscript:
Rose C, Parker A, Jefferson B, et al.: The characterization of feces and urine: A review of the literature to inform advanced treatment technology. *Crit Rev Environ Sci Tec.* 2015;45(17):1827-79. 10.1080/10643389.2014.1000761

The Reviewer’s suggestion to clarify that ISO 30500 refers specifically to removal of N and P from *liquid* effluent also clarifies that the water soluble fractions of N and P are most relevant for this discussion. We have further clarified that that we are focused on liquid effluent:

In the abstract: “ However, increasingly stringent **liquid** effluent standards for N and P...”

In the introduction, paragraph 2: “**Treatment of the liquid fraction of NSSS waste to enable non-potable water reuse (e.g. for toilet flushing) has been a focus of many reinvented toilet (RT) technologies and guides the scope of this Open Letter.**”

and paragraph 3: "...ISO 30500 requires 70% and 80% reductions in total N and total P, respectively, in NSSS **liquid** effluent."

Specific comments:

1. *I think it's worth briefly explaining the ISO 30500 standards in the introduction, at their first mention. Not all readers will be familiar, and this document is actually quite broadly useful beyond NSSS.*

We thank the Reviewer for this suggestion. We have found it particularly helpful for clarifying some of the other points brought up by all three reviewers, in that the ISO 30500 specifically dictates nutrient removal requirements from *liquid effluent*, and this was not explicitly clear in the original version of our manuscript and indeed may not have been clear to readers who are unfamiliar with the standard. We have added the following to the introduction, paragraph 3, after the first mention of ISO 30500:

"The ISO 30500 standards provide guidance for safe onsite treatment of human excreta and non-potable water reuse, and includes threshold performance metrics for liquid effluent quality, including chemical oxygen demand (COD), total suspended solids (TSS), nutrients (N and P), and specific pathogens."

2. *The design of Table 1 is not optimal. The use of italic text is very subtle. I would suggest a different and more obvious notation for indicating the limitations for use in NSSS. Also, by the title and text it is really not clear which methods are for N and with are for P. I suggest the applicability for each nutrient be separately indicated.*

We thank the reviewer for this comment; indeed, all three Reviewers suggested improvements to the formatting and content of Table 1. We have updated the table in the new version and feel that the changes address the concerns of all Reviewers.

3. *I would have liked to see text specifically discussing the column in Table 1 dealing with nutrient recovery. Ability to reuse nutrients should be considered as a critical criterion for technology development and selection. Although reuse possibilities are summarized in the table, mentioning them in the text would bring more attention to the importance of this issue.*

We agree with the Reviewer that nutrient recovery should be considered as critical. However, the ISO 30500 specifically provides guidance for nutrient *removal* requirements and only briefly mentions nutrient *recovery*. We believe that the complex and multifaceted issues of nutrient recovery/reuse are outside the scope of the current work, and we have updated the language in the main text to clearly delineate the scope of this letter.

4. *Breakpoint chlorination is said to release "almost no nitrous oxide". It would be highly preferred to be more quantitative in this statement given the potency of N₂O as a GHG.*

It is possible in principle to form N₂O from decomposition of chloramines without an excess of

chlorine, however in practice, N_2O is not observed as a byproduct of breakpoint chlorination due to the large excess of chlorine that is present. The text has been edited and a citation added to clarify this point:

“with no **measurable** nitrous oxide formation **under appropriate operating conditions**²⁰.
[Pressley, 1972]”

Pressley TA, Bishop DF, Roan SG: Ammonia-nitrogen removal by breakpoint chlorination. *Environ. Sci. Technol.* 1972;6(7):622-8. 10.1021/es60066a006

5. *For breakpoint chlorination, a reference should be added related to byproducts which are toxic and carcinogenic to direct the reader to detailed information.*

Byproducts of breakpoint chlorination are typically the same as for other chlorine disinfection methods, e.g. chloramines, as described later in the paragraph in question. We have also added three references which specifically discuss breakpoint chlorination byproducts:

“...undesirable oxidation byproducts, many of which are toxic or carcinogenic.[Yang 2005; Shah 2012; How 2017]”

Yang X, Shang C, Huang J-C: DBP formation in breakpoint chlorination of wastewater. *Water Res.* 2005;39(19):4755-67. 10.1016/j.watres.2005.08.033

Shah AD, Mitch WA: Halonitroalkanes, halonitriles, haloamides, and N-nitrosamines: A critical review of nitrogenous disinfection byproduct formation pathways. *Environ. Sci. Technol.* 2012;46(1):119-31. 10.1021/es203312s

How ZT, Kristiana I, Buseti F, Linge KL, Joll CA: Organic chloramines in chlorine-based disinfected water systems: A critical review. *J. Environ. Sci.* 2017;58:2-18. 10.1016/j.jes.2017.05.025

6. *Please explain why breakpoint chlorination leads to high TDS in the effluent.*

We have edited the text to clarify this point:

“...(2) **need for chemical addition to neutralize pH, causing** large increases in total dissolved solids in the treated effluent...”

7. *The chemical precipitation section should be expanded to individually discuss the different metals added. Please add text explaining how the choice of Fe vs. Al vs. Ca vs. Mg affects efficiency and whether the end product is reusable. Struvite is mentioned, but nothing is said about the other precipitates.*

We have added to the text in this section to briefly describe the use of different metals for P removal and the trade-offs associated with each; the topic has been extensively reviewed elsewhere, and we have also added some relevant references.

“The choice of cation used for precipitation is critical and affects myriad factors including overall cost, P removal efficacy, optimal pH and temperature required for P removal, and

reusability/bioavailability of the P-containing precipitate.[Melia 2017] Resulting trade-offs need to be carefully considered depending on the context and effluent requirements. For example, the Fe and Al salts used for P precipitation are typically much less expensive than Ca and Mg salts; however, Fe and Al phosphate precipitates are not suitable for direct use as fertilizers due to low P bioavailability.[Desmidt, 2014] On the other hand, using Mg for struvite precipitation can enable simultaneous removal of phosphate and ammonium in a 1:1 stoichiometric ratio and has been shown to be widely bioavailable;[Melia 2017] however, struvite precipitation has a narrow window of optimal pH and cannot be relied upon as the sole method of N removal due to the much higher concentrations of ammonia/ammonium relative to phosphate.[Mehta, 2015] Furthermore, more research is required to address concerns regarding the presence of contaminants in precipitates where fertilizer use is intended.[Desmidt 2014, Mehta, 2015, Melia 2017]”

Melia PM, Cundy AB, Sohi SP, Hooda PS, Busquets R: Trends in the recovery of phosphorus in bioavailable forms from wastewater. *Chemosphere* 2017;186:381-95. 10.1016/j.chemosphere.2017.07.089

Desmidt E, Ghyselbrecht K, Zhang Y, Pinoy L, Van der Bruggen B, Verstraete W, Rabaey K, Meesschaert B: Global phosphorus scarcity and full-scale P-recovery techniques: A review. *Crit. Rev. Environ. Sci. Tec.* 2015;45(4):336-84. 10.1080/10643389.2013.866531

Mehta CM, Khunjar WO, Nguyen V, Tait S, Batstone DJ: Technologies to recover nutrients from waste streams: A critical review. *Crit. Rev. Environ. Sci. Tec.* 2015;45(4):385-427. 10.1080/10643389.2013.866621

8. For the ion exchange section, change “rate” to “efficiency”. 95% is not a rate.

This change has been made.

9. For “Reconsidering biological N and P remediation at small scales”, the meaning of “native” for biofilms is not specified. Some of the transitions in this section are rough as well.

We have added a sentence to more clearly define what we mean by “native” biofilms. Additional small changes in this paragraph have been made to help with transitions.

“The lines between traditional “biological” treatment processes (i.e. systems intentionally inoculated with specific microbial strains) and truly “non-biological” treatment methods are likely already blurred in many RT systems due to the high microbial load of feces, which could yield formation of “native” biofilms on or within system components. The role of native biofilms in nutrient sequestration **thus** cannot be ignored, and the conditions which favor biological N and P sequestration in **native** biofilms could be optimized; more work is needed in this area, and would be specific to a given wastewater stream **and treatment context.**”

10. Please clarify that for inorganic P capture with FeCl₃, reuse is not viable. Please also more clearly discuss inorganic vs organic forms of P.

We have already described clarifications to the introduction to limit the scope of this work to orthophosphate. We therefore do not feel it is necessary to clarify the discussion here with regards to inorganic vs organic P.

The precipitate recovered in the specific case referenced here is vivianite. While the direct bioavailability and use of vivianite is still somewhat controversial, there is promise for its use as a fertilizer precursor or in electronic applications, which was the subject of a review published shortly after submission of our manuscript. We have added the following text and citations:

“(The precipitate recovered in this case was vivianite, which may have value in electronic applications and as a fertilizer precursor.)[Wilfert 2018, Wu 2019]”

Wilfert P, Dugulan AI, Goubitz K, Korving L, Witkamp GJ, Van Loosdrecht MCM: Vivianite as the main phosphate mineral in digested sewage sludge and its role for phosphate recovery. *Water Res.* 2018;144:312-21. 10.1016/j.watres.2018.07.020

Wu Y, Luo J, Zhang Q, Aleem M, Fang F, Xue Z, Cao J: Potentials and challenges of phosphorus recovery as vivianite from wastewater: A review. *Chemosphere.* 2019;226:246-58. 10.1016/j.chemosphere.2019.03.138

Competing Interests: No competing interests were disclosed.