Perspective

Making wastewater obsolete: Selective separations to enable circular water treatment

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A B S T R A C T

By 2050, the societal needs and innovation drivers of the 21st century will be in full swing: mitigating climate change, minimizing anthropogenic effects on natural ecosystems, navigating scarcity of natural resources, and ensuring equitable access to quality of life will have matured from future needs to exigent realities. Water is one such natural resource, and will need to be treated and transported to maximize resource efficiency. In particular, wastewater will be mined for the valuable product precursors it contains, which will require highly selective separation processes capable of capturing specific target compounds from complex solutions. As a case study, we focus on the nitrogen cycle because it plays a central role in both natural and engineered systems. Nitrogen occurs as several species, including ammonia, a fertilizer and precursor to many nitrogen products, and nitrate, a fertilizer and component of explosives. We describe two applications of selective separations: selective materials and electrochemical processes. Ultimately, this perspective outlines the next thirty years of modular, selective, resource-efficient separations that will play a major role in enabling element-specific circular economies and redefining wastewater as a resource.

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1. Introduction: imagine a future

Imagine a future where wastewater is obsolete. Waste and water are completely dissociated from one another, and younger generations wonder why they were ever associated. Every wastewater is mined for maximal value before environmental discharge. We hardly remember 2020, when tons of untapped revenue was discharged daily due to 80% of wastewater not being collected [1]. Pollution is primarily prevented because it wastes money; revenue has supplemented regulation as a primary driver for wastewater treatment. We also marvel at the bygone days of aging, centralized infrastructure that required miles of pipe, millions of dollars in maintenance, and still left the majority of people in Asia and Africa with inadequate treatment [1,2]. In 2050, wastewater treatment will be diverse in terms of scale, techniques, and value-added products. Rather than always bringing water to treatment, decentralized and hybrid on-site/centralized systems that bring treatment to water will represent appreciable fractions of installations.

We will deploy an arsenal of modular, scalable treatment techniques that can address any particular pollutant in wastewaters of any composition, ranging from industrial effluents to fertilizer runoff and municipal wastewater and desalination brine. The portfolio of products that can be generated from wastewaters is diverse, robust, and ever increasing. The building blocks of this vision exist today, and can be cultivated into reality over the next three decades.

Imagine future water resource recovery facilities with cascading selective recovery steps. Whereas removal can be achieved with general steps (e.g., oxidation of organics and ammonium), recovery of marketable products requires high product purity and highly selective separations. These selective separations will leverage selective materials (e.g., membranes and adsorbents) and selective processes (e.g., electrochemical reactors). These modular unit processes can be recombined in various configurations and benchmarked with ubiquitous performance metrics that facilitate practitioner selection. Engineers will aim to minimize the number of separation steps by maximizing the selectivity and product purity of each step. Water resource recovery facilities have become refineries, producing broad portfolios of products tailored to their

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surrounding regions and variable consumer demands.

Imagine a future where selective sensors enable tailored treatment based on upstream water composition. Especially when electricity is a major driving force, these operating parameters can be integrated with adaptive process control that facilitates point-of-generation treatment. Such capabilities will more effectively engage chemical manufacturing companies in wastewater treatment as they prioritize and pursue sustainability to enhance profit and access to sustainability-minded consumers. Sensors for process control can also facilitate water treatment that is not only decentralized, but deployable. Treatment is no longer restricted to waters that we can collect, and legacy pollution is rapidly being addressed. Aquatic ecosystems like the Gulf of Mexico are returning to 20th century nutrient levels, along with wildlife populations. Remote sensors are regularly deployed to monitor nitrogen pollution before harmful algal blooms and pinpoint areas in need of treatment. If sensed levels exceed a threshold, mobile treatment units are deployed to affected regions. Both sensors and treatment devices can be collected periodically to combine their ammonia products and clean their components before redeployment.

We can and must imagine these future scenarios now to achieve element-specific circular economies that convert water pollutants into valuable products. Over the past decade, wastewater treatment plants have been increasingly labeled water resource recovery facilities; this perspective will further accelerate in the near future until municipal wastewater becomes one of many feedstocks. The next several decades of water treatment require rapid innovation to meet the staggering challenges of climate change, water scarcity, equitable access to food and energy, and public health. The grand challenges of today are poised to become the grand achievements of 2050 that sustain humans for centuries to come. This future is not far off, and starts with designing next-generation materials and processes for element-specific wastewater treatment and valorization. We note that co-designing innovative materials and efficient processes can accelerate process toward this future; thus, we discuss both applications in this perspective. Lastly, we identify urgent research needs to realize our vision for selective separations and prepare for the future today.

2. Selective separations address sustainability goals and grand challenges

Selective separations can address the priority agendas set by several organizations for the 21st century. For the United Nations Sustainable Development Goals (SDGs), recovery of high-purity products from wastewater can contribute to efforts to achieve clean water and sanitation (SDG 6) and responsible consumption and production (SDG 12). More broadly, recovering marketable products from wastewaters (e.g., energy from salinity gradients \[3\–5\]) can accelerate affordable and clean energy (SDG 7); facilitate modular, scalable treatment that enhances sustainable cities and communities (SDG 11); and improve equitable access to fertilizer and commodity chemicals (SDG 10). Of the fourteen grand engineering challenges identified by the U.S. National Academy of Engineering, selective water treatment addresses five: 1) developing carbon sequestration methods, 2) managing the nitrogen cycle, 3) providing access to clean water, 4) restoring and improving urban infrastructure, and 5) engineering the tools of scientific discovery [8]. Of five major environmental engineering challenges developed by the same organization (U.S. NAE), selective separations primarily advance designing a future without pollution and waste. They also contribute to sustainably supplying food, water, and energy to meet the staggering levels of energy, costs, and transport. Adsorption and electrochemical approaches both exhibit the requisite modularity for selective recovery. Conventional ion-exchange columns can use conventional resins, sustainably sourced adsorbent (e.g., biochar), or novel adsorbent materials (see Section 4) to recover nutrients and metal ions (Fig. 2A). Regeneration plays a key role in recovery because of the elution of target compounds. Electrochemical techniques are well-suited for selective recovery separations because they use electricity as their major input, facilitate process control, and reduce transport-related chemical and energy inputs [22]. Additionally, unlike many conventional aqueous separations that separate large volumes of water from solutes, electro-driven processes directly remove ions, which favors ion-selective recovery. Although full electrochemical treatment trains that combine electrochemical unit processes have not been thoroughly realized, by 2050 we can expect to see many installations of modular, tailored electrochemical treatment. For example, electrochemical unit processes have been proposed for nutrient recovery [23–29], disinfection [30–33], desalination [34–36], transformation of dissolved organic compounds [37–40], and metal recovery [41–43]. Replacing chemical inputs with electricity, particularly from renewable energy sources, could offset the 75% of human health, environmental, and climate damages from water treatment processes that chemical inputs contribute [44,45]. Designing future selective processes is closely related to the advancement of novel selective materials, and integrating materials and processes can lead to reactive separations that enhance modularity and recovery by facilitating desired reactions and separations within one reactor. In the remainder of this section, we focus on the roles of next-
Electrochemical processes were first used for contaminant removal from wastewater and drinking water, such as removing salts via electrodialysis and removing organic compounds through electrochemical oxidation [39,46,47]. While removal applications continue to be vital, electrochemical processes can also accelerate recovery-oriented separations. The combination of electrochemical potential as a driving force and selective membranes can achieve highly selective separations and requisite product purity. More broadly, researchers are electrifying chemical manufacturing by investigating electrochemical potential as an additional driving force beyond more conventional temperature and pressure. For example, electrochemical water splitting can achieve pH gradients with ion exchange membranes between electrodes that perform oxidation (anodes) and reduction (cathodes). In some applications, this replacement of periodic acid or base addition with electrical current can be advantageous, particularly if the electricity is supplied by renewable energy sources. Here, we consider three general categories of electrochemical processes: electrodialysis, capacitive deionization/ion insertion, and electrocatalysis (Fig. 2).

Electrodialysis, which uses electrochemical potential to split water molecules, has been a major technique used in water treatment for decades (Fig. 2B and C). The pH gradients supplied by anodic oxygen evolution and cathodic hydrogen evolution or oxygen reduction can be used to control the solubility of dissolved species. Recently, ammonia stripping has emerged as an ideal candidate for selective electrochemical separations. Early investigations started with performing electrodialysis and counter-current ammonia stripping in series [48,49], where the alkaline catholyte was passed to a conventional air stripping module. Membrane stripping with hydrophobic porous membranes (such as polypropylene) was then substituted for the conventional air stripping, thus reducing required energy [50]. These processes can be intensified with electrochemical stripping, a combined unit process in which ambient temperature membrane stripping is conducted in the same reactor as electrodialysis [24,51]. Nitrogen is separated first by charge across a cation exchange membrane, and then by volatility due to an in situ cathodic pH increase arising from water electrolysis. Ammonia is then recovered in an acidic gas trap stream producing ammonium sulfate, a common liquid fertilizer. Similar electrochemical stripping approaches can be used for gases that equilibrate with protonated aqueous salts, such as electrochemical carbon dioxide stripping to counter ocean acidification [52].

Parallel to water splitting approaches, capacitive techniques will play an important role in advancing selective electrochemical separations (Fig. 2D). Nitrogen as ammonium or nitrate can be inserted into ion-insertion electrodes without the need to split water, which dramatically decreases required energy [53,54]. Electrodes with pore sizes tuned for ammonium molecules can screen out other competing cations, with the exception of potassium, which has a similar hydrated ionic radius. For this separation, ligand-based approaches (e.g., Cu–NH₄) are often required to achieve high selectivity; alternatively, wastewaters with high NH₄⁺/K⁺ ratios may exhibit sufficient selectivity and subsequent product purity. Ammonium can also be migrated at potentials below that of water splitting to produce alkaline ammonia, a common disinfectant and chemical precursor for other nitrogenous products [25]. Recent efforts have also modified typical carbon electrodes with polymeric functional groups to perform selective ion exchange, such as nitrate-selective capacitive deionization [55]. Eventually, cascades of varying potentials, electrodes, and surface modifications could be used to selectively remove and recover multiples species of interest as valuable products.

Electrocatalytic approaches can also facilitate selective separations (Fig. 2E). For example, electrochemical struvite precipitation can be achieved by oxidizing elemental magnesium anodes into a phosphate-rich solution [28]. This electrocatalytic oxidation avoids magnesium inputs along with their associated impurities and environmental impacts, such as transport. For nitrogen, electrocatalytic nitrate reduction is a burgeoning field [56–58]. Originally focused on removal of NO₃ to N₂, the field has also recently focused on purposeful production and recovery of ammonia. Ammonia separation techniques will still need to be leveraged, and possibly combined from Faradaic and capacitive methods. In particular, an emphasis on designing reactive separations, rather than separately designing catalysis and separation unit processes, is needed to advance the science and engineering of selective nutrient recovery. Lastly, alternating current has been leveraged for insertion, such as for uranium and lithium [41,59]. These approaches have exhibited selectivity of several orders of magnitude for target compounds over competing ions, including in seawater.

Within all electrochemical systems, the interfacial interactions between the primary components, such as electrodes, membranes, reactors, and electrolyte (or choice of feed water) govern mass
transport and kinetics. Dimensionally stable anodes, such as titanium coated with mixed metal oxides, are most commonly used in electrodialysis processes [39]; carbon and metallic electrodes and metals are more common for capacitive and electrocatalytic approaches due to their molecular tunability [35,60]. Bimetallic electrodes are also being explored to simultaneously achieve selectivity, activity, and stability in aqueous electrolytes [61,62]. Cation exchange membranes, based on original Nafion proton exchange membranes, achieve acidic anode chambers and alkaline cathode chambers. Anion exchange membranes, often used in alkaline fuel cells, allow transport of hydroxide ions to balance pH. Bipolar membranes, which have both anion exchange and cation exchange layers along with a catalyst between them, have also been investigated for more energy-efficient water splitting [63,64]. Ongoing research at this water-energy nexus of fuel cells, water electrolyzers, and water treatment includes the effects of water composition (or electrolyte effects) on observed catalyst stability, selectivity, and activity. These investigations can improve the understanding of applicability to various wastewaters that may vary in composition, including due to shock loads or extreme weather events. Integrating component-level studies of electrodes, membranes, reactors, and electrolytes will help address a critical challenge for electrochemical process development: moving beyond proof-of-concept for novel unit processes towards proof-of-system that prioritizes integration of these unit processes and applications to specific wastewaters where ascribed benefits are maximized.

4. Next-generation selective materials

We focus here on two classes of separating agents: adsorbents and membranes. Both facilitate modular, selective unit processes capable of separating target ions from complex wastewaters. For adsorbents, three key fundamental metrics are capacity (number of sites available to target ion), selectivity (preferential sorption of target species), and regenerability (desorption of target species and multiple use of adsorbent). Selectivity has historically connoted selective adsorption, but will likely also refer to selective regeneration in the future. For membranes, several metrics exist for two key physical characteristics: selectivity and target species flux. Consensus is still developing on which metrics should be used to benchmark selective aqueous separations for resource recovery, but commonly reported metrics include permselectivity, separation factors, recovery efficiency, and time to replacement.

4.1. Solute-selective adsorbents

General principles of adsorbent selectivity are illustrated in Fig. 3A. During adsorption, observed selectivity is a function of both the wastewater being treated and the adsorbents used. Several adsorbents, such as weak acid cation exchangers, exhibit a majority of sites occupied by ammonia in nitrogen-rich wastewaters, such as urine [65,66]; however, the high ratio of ammonium to competitors like sodium and potassium in urine contributes significantly to this observed selectivity. Experiments in equimolar, binary solutions establish intrinsic selectivity without the influence of wastewater composition. For example, Cu- and Zn-loaded weak acid cation exchangers exhibit 7–10 times intrinsic selectivity toward ammonium over potassium [67]. Because NH$_4^+$ and K$^+$ have similar hydrated ionic radii, physical separations by pore size are insufficient; chemical specificity is required. This specificity can be achieved by ligand complexation [67–69], crown ethers [70,71], and molecular imprinting [72]. Many studies to date describe novel materials in ideal wastewaters or established adsorbents in real wastewaters. Tandem research that both identifies new materials and systematically evaluates the effects of aquatic chemistry in simulated solutions to real wastewaters will accelerate implementation and mechanistic understanding of material and solution effects on performance. Beyond nitrogen, ligand exchange and molecular imprinting are general strategies that can be tailored to various adsorbates (e.g., nitrate [73], arsenic [74–76], phosphate [77,78]). Emerging materials such as metal-organic frameworks and covalent-organic frameworks may also play a role in selective separations, especially if an increasing number of studies consider realistic wastewater compositions. Adsorbents can also be operated in series, such as a hybrid anion exchanger followed by a hybrid cation exchanger that removes phosphorus and then nitrogen. Careful understanding of each adsorbent and its unit process facilitates full treatment trains with cascades of selective separation steps.

4.2. Regenerating adsorbents

Regenerating adsorbents will be a major area of research for recovery-oriented selective separations. Removal processes have not historically emphasized regeneration because it primarily increases resin lifetime, which was less important than selective adsorption for pollutant removal. Regeneration also produces brine waste from adsorption processes that often requires additional treatment or disposal. Many columns are constructed to be regenerated infrequently by overestimating the column dimensions, and some adsorbents are intended to be disposed rather than regenerated. In contrast, resin regeneration during recovery processes doubles as the product generation step, which is one of the most vital considerations. Selective regeneration could further enhance product purity by replacing or augmenting selective adsorption; both are critical to the future of resource-efficient wastewater treatment (Fig. 3B).

Regeneration also contributes to embedded energy and emissions for adsorption. During nitrogen recovery via ion exchange, acid regeneration consumed more than 70% of the energy and greenhouse gas emissions [79]. Alternative regenerants, including other strong acids, exhibit similar environmental impacts; brine solutions exhibit lower environmental impacts, along with lower recovery efficiencies and product purities [66]. Regeneration also occupies the majority of costs associated with ion exchange for recovery [80]. Efforts to decrease cost and environmental impacts focus on electrifying water treatment through several strategies, such as on-site electrolysis to produce strong acids and bases [81]. More recently, electro-assisted mild regeneration has been used to regenerate phosphorus and nitrogen on HAIX and WAC resins [82]. Avoiding transport and conventional manufacturing can reduce the environmental impacts of adsorption and make it a more competitive option for selective separations. These regeneration strategies may be tailored to various new and existing adsorbent materials.

4.3. Solute-selective membranes

Solute-selective membranes will also play a critical role in advancing selective separations from now until 2050. Membrane processes are increasingly investigated and widely adopted in desalination, water purification, and reuse. Reverse osmosis (RO) has rapidly become the most common desalination method due to the modularity and scalability that membrane modules exhibit [83–86]. The same benefits apply for selective separations, and a new frontier for resource recovery is ion-selective membranes that can facilitate the transport of a target ion while rejecting other undesired compounds of complex wastewaters (Fig. 3C). Nitrogen-selective membranes, and in particular ammonia- and nitrate-
selective membranes, will play a critical role in preventing and remediating pollution that contributes to harmful algal blooms and threatens drinking water sources. Phosphate-selective membranes could similarly be used to extract phosphorus from wastewater, and potentially combined with struvite precipitation. Brine mining using metal ion-selective membranes will allow efficient recovery of lithium from battery leachate and rare earth metals from produced water, thus contributing to the circular economy of the energy sector.

Unlike gas separation membranes that discriminate among single gas solute molecules, conventional aqueous separation membranes are predominantly focused on controlling solvend-solute selectivity, i.e., improving water permeability while minimizing solute passage [87–90]. Current membrane materials exhibit limited solute-solute selectivity. Osmotic-driven nanofiltration (NF) membranes with relatively large pores are more permeable to small monovalent ions than large multivalent ions [87,91]. Ion-exchange membranes with fixed charged functional groups facilitate the transport of counter-ions while rejecting cations based on electrostatic interactions, and can distinguish ions with different valency (monovalent ion permselective membrane) [87,92,93]. However, precise separation of solutes with similar physicochemical properties, e.g., molecular size and charging condition is still challenging with current state-of-the-art membranes [94,95].

Ion-selective membranes require precisely tailored membrane structures and properties to control the solute transport process. While permeant selectivity for salt-rejecting dense membranes (RO and NF) is dominated by size-based mechanisms [87,93,96,97], synthesis of membrane materials with highly homogenous sub-nanometer pores can achieve precise selection of specific electrolyte solutes [98]. Molecular sieves (e.g., metal-organic frameworks, zeolitic imidazolate frameworks, carbon nanotubes) have also been recently investigated for solute-solute separation [99–102]. Solute-membrane interactions are critical to selective solute transport across the membrane [87,89,103]. Inspired by the coordination mechanisms of selective adsorbents, introducing ion-specific ligands into the membrane matrix has been identified as a promising strategy for favorable transport of target solutes [94,95,104].

4.4. Integrating materials and processes into intensified treatment trains

Conventionally, adsorbents and membranes are designed, tested in ideal solutions, and eventually integrated into water treatment processes. For both adsorbents and membranes, materials design and process engineering should be conducted in tandem. Novel materials enable record-setting process performance, and novel processes establish unique design criteria for existing and future materials. There is also a need for iterative, multiscale investigations that connect molecular mechanisms to process innovations and systems-level evaluations, as exhibited by the influence of regeneration on overall ion exchange process economics and environmental impacts [105]. In this application, electrochemical techniques may be able to address the major implementation challenge of adsorption: bed regeneration. Ion exchange resins can also be integrated into electrochemical processes, such as in electro-deionization, to overcome insufficient conductivity and reduce energy consumption [106]. Such integration of electrochemistry and ion exchange could lead to novel selective separations that exhibit unprecedented recovery efficiency, purity, and energy efficiency.

5. Preparing for the future now

The next several decades will experience rapid expansion of both the product and pollutant portfolios addressed by selective separations. Wastewater-derived products will expand from predominantly energy and fertilizer to commodity chemicals such as disinfectants, acids, bases, and precursors for conventional chemical manufacturing processes. Conversations and actions that circularize the economy will take place at several scales, from households and individual chemical manufacturing plants to municipalities, regions, and multinational organizations aiming to better manage the entire global nitrogen cycle. The pollutants that can be removed and recovered will also expand. Novel processes and materials will become platform technologies that can be tailored to specific nitrogen solutes, e.g., ammonia, nitrate, nitrous oxide (e.g., as well as a wider set of elements, including rare earth metals, transition metals, and micronutrients. This innovation will be accelerated by increasing the cross-sector conversations between different applications. For example, applications for sensors, sewered setting, non-sewered settings, and remote operations will help categorize this diverse portfolio of applications and identify synergies between them. Operation in extreme environments, such as selective separations in space, can also accelerate selective separations that improve biogeochemical cycles.

The future of wastewater may favor decentralized treatment, and must include technologies that can operate at multiple scales. In the U.S., 25% of the population operates on non-networked sanitation infrastructure [107]; in Africa and Asia, where the majority of population growth will occur over the next several decades, wastewater treatment must be proliferated quickly to match growing urban demand [1]. More broadly, decentralization has been observed to reduce energy consumption, cost, and environmental impacts for treating and valorizing wastewater [108,109]. Thus, selective separations must be flexible for various scales and tailored for suitable locations and configurations to enable efficient recovery of each element in Fig. 1. For example, recovering nitrogen from agricultural runoff or manure on-site can avoid diluting the nitrogen concentration, thus saving consumption of separation energy, and benefiting the circular agriculture. Similarly, recovery of nutrients from urine could occur at the household or building level, and can be directed to urban gardens or peri-urban farms (depending on regional fertilizer needs) [110]. The practical considerations of scale and location influence separation design due to the primary characteristics of the waste streams, including salinity, conductivity, temperature, and ionic compositions. While previous studies demonstrated that real urine is suitable for electrochemical treatment due to its high conductivity and nitrogen content [24,111], there is a need to establish a comprehensive framework that evaluates the feasibility of different waste streams for electrochemical treatment. In addition, economic aspects of electrochemically-driven selective separations require further

![Fig. 3. Next-generation materials for selective separations. a. adsorbent, b. regenerative adsorbent, and c. membrane. Circles in green denote the target ion for separation, whereas those in red represent undesirable ions. The light green color of materials represents affinity to target ions.](image)
study. Compared with conventional centralized treatment, electrochemical treatment processes avoid the large amount of chemical inputs and complicated operating procedures, showing great potential in saving capital costs and reducing carbon emissions. Operating expenses (OPEX) and capital expenses (CAPEX) should be optimized for implementing electrochemical selective processes based on data collected at various scales and with real effluent streams.

The future of wastewater may also include distributed treatment taken to the extreme of deployable treatment. Electrochemical wastewater separations could be applied as deployable sensors to monitor water quality. In the case of nutrients, there is an urgent need to track non-point sources (e.g., agricultural runoff) to prevent eutrophication that damages aquatic ecosystems and human health. In this context, reactive separations could play a critical role in electifying water treatment in another manner by facilitating an Internet of Things for water. Fit-for-purpose water and point-of-generation treatment require dynamic, high-resolution measurements of water composition that could be achieved by electrochemical sensors. These high-resolution data could also inform process control for treatment processes based on influent water composition. In the more distant future, markets might develop for environmental data, especially under cap-and-trade or other credits for waterborne emissions, which have already been discussed for nutrients at wastewater treatment plants [112]. Nutrient sensors could also serve as platforms for integration with existing and future sensors for wastewater treatment and environmental monitoring. Adding nutrient sensors would improve resolution of nutrient cycling in aquatic ecosystems, and anthropogenic effects on nutrient cycles in various settings.

The future of wastewater treatment may also be more participatory. As the median scale of installation drops, so do the barriers to citizens engaging with wastewater treatment infrastructure. For example, sensors can be handled by community members and monitored as parts of school projects, further engaging communities and recruiting the future global workforce of water treatment experts. Particularly if installations of sensors and on-site treatment are networked and enmeshed, visitors can easily learn about larger networks while leveraging the internet of things towards water treatment. In addition to the general public, the future of wastewater will require engaged input from industries that would purchase wastewater-derived products. Networked, smaller installations can double as research sites and demonstration facilities for orthogonal industries (e.g., fertilizer and disinfectant producers) that can develop for environmental data, especially under cap-and-trade or other credits for waterborne emissions, which have already been discussed for nutrients at wastewater treatment plants [112]. Nutrient sensors could also serve as platforms for integration with existing and future sensors for wastewater treatment and environmental monitoring. Adding nutrient sensors would improve resolution of nutrient cycling in aquatic ecosystems, and anthropogenic effects on nutrient cycles in various settings.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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