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Environmental sustainability of forward osmosis: The role of draw solute and its management

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ABSTRACT

Forward osmosis (FO) is a promising technology for the treatment of complex water and wastewater streams. Studies around FO are focusing on identifying potential applications and on overcoming its technological limitations. Another important aspect to be addressed is the environmental sustainability of FO. With the aim to partially fill this gap, this study presents a life cycle analysis (LCA) of a potential full-scale FO system. From a purely environmental standpoint, results suggest that significantly higher impacts would be associated with the deployment of thermolytic, organic, and fertilizer-based draw solutes, compared to more accessible inorganic compounds. The influent draw osmotic pressure in FO influences the design of the real-scale filtration system and in turn its environmental sustainability. In systems combining FO with a pressure-driven membrane process to recover the draw solute (reverse osmosis or nanofiltration), the environmental sustainability is governed by a trade-off between the energy required by the regeneration step and the draw solution management. With the deployment of environmentally sustainable draw solutes (e.g., NaCl, Na_2SO_4), the impacts of the FO-based coupled system are almost completely associated to the energy required to run the downstream recovery step. On the contrary, the management of the draw solution, i.e., its replacement and the required additions due to potential losses during the filtration cycles, plays a dominant role in the environmental burdens associated with FO-based systems exploiting less sustainable draw solute, such as $MgCl_2$.

1. Introduction

Membrane-based separation processes are cutting-edge technologies for the management of various liquid streams. To date, pressure-driven membrane processes (PDMPs) are widely deployed for the treatment of several water sources, ranging from wastewater treatment via micro- and ultra-filtration to seawater desalination or groundwater remediation through nanofiltration (NF) and reverse osmosis (RO) (Arevalo et al., 2012; Giagnorio et al., 2018a; Shaffer et al., 2012; Zeman and Zydney, 2017). On the other hand, osmotically-driven membrane processes (ODMPs) are still the subject of ongoing development and few real-scale systems have been designed and developed worldwide (Awad et al., 2019). Among the various ODMPs, forward osmosis (FO) is a promising technology for innovative applications (Awad et al., 2019; Linares et al., 2014; Shaffer et al., 2015). In FO, a highly selective membrane is deployed in combination with an engineered draw solution used to generate a concentration gradient across the membrane, thus extracting high-quality water from the contaminated feed solution, while diluting the draw solution (Cath et al., 2006; Shaffer et al., 2015; Zhao et al., 2012). While sustained work has been reported with the goal: (i) to optimize FO membrane performance; (ii) to study innovative draw solutions; and (iii) of fouling assessment and control (Achilli et al., 2010; Grinic et al., 2018; Islam et al., 2019; Ricceri et al., 2021; Shaffer et al., 2015; Siddiqui et al., 2018; Yip et al., 2010), few studies have been conducted to analyze large-scale FO applications and configurations. Full-scale FO systems have been discussed in literature reports, for example, presenting potential configurations deployable to achieve minimal or zero liquid discharge when FO is combined with seawater RO desalination systems through the exploitation of the brine osmotic potential (Kazner et al., 2014; Martinetti et al., 2009; Martinez et al., 2020; Subramani and Jacangelo, 2014). Other studies evaluated FO for direct fertigation through pilot-scale analysis and techno-economic assessments (Banchik et al., 2016; Kim et al., 2019; Lotfi et al., 2015; Phuntsho et al., 2016). Finally, results have been reported for indirect wastewater desalination, discussing pilot-scale experiments of FO integrated in a membrane bioreactor or as a polishing system for secondary

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and tertiary wastewater effluents (Cath et al., 2010; Corzo et al., 2018; Hancock et al., 2013).

Membrane-based separation processes have been further analyzed from an environmental perspective through life cycle assessment (LCA) studies. For example, RO was the subject of a number of LCA analyses, presenting: (i) the environmental benefits derived by the deployment of RO systems over thermal technologies for seawater desalination; (ii) possible scenarios for the end-of-life of membrane modules; and (iii) comparative studies to assess the environmental burdens related to the management of the concentrate (de Paula and Amaral, 2017; Lawler et al., 2015; Raluy et al., 2006; Zhou et al., 2011). In-depth analyses have also been performed to assess the environmental sustainability of membrane technologies deployed for potable water production and wastewater treatment, such as microfiltration and ultrafiltration systems implemented in membrane bioreactors (Chen et al., 2018; Giagnorio et al., 2018b; Holloway et al., 2016; Ioannou-Ttofa et al., 2016; Stokes and Horvath, 2006; Vince et al., 2008).

Literature reports of LCA studies on FO are instead limited, mainly due to the shortage of real-scale plants based on this technology. Linares *et al.* and Hancock *et al.* (Hancock et al., 2012; Linares et al., 2016) showed that the implementation of hybrid FO systems would potentially result in significant savings and environmental benefits when applied to seawater desalination. Case-studies were also discussed in the environmental evaluation of fertilizers-drawn FO systems or hybrid FO systems integrated within conventional wastewater treatment trains (Kim et al., 2017; Vinardell et al., 2020). Nevertheless, studies have been so far comparative and have not reported a specific analysis of the environmental sustainability of the FO technology itself.

This study attempts to partially close this gap by evaluating the parameters influencing the environmental sustainability of potential real-scale FO systems. At first, an LCA analysis of the most common FO draw solutions (DS) is presented. Inorganic, organic, thermolytic, and fertilizer-based DS are evaluated, assessing the environmental impacts related to their production and subsequent deployment in potential full-scale FO configurations. An LCA analysis is thus performed to evaluate potential FO configurations whereby different draw solutes may be deployed but for which the same systems can be applied for their regeneration, i.e., inorganic DS used in FO coupled with pressure-driven membrane processes. The most and the least environmentally friendly inorganic draw solutes are chosen for this environmental assessment. Design and optimization of potential full-scale FO-PDMP systems is presented, followed by analysis of their environmental sustainability and the influence of operating parameters on environmental burdens.

2. Methods

2.1. LCA methodology

Two LCA approaches were adopted with the aim to evaluate the parameters influencing the environmental sustainability of large-scale FO systems. Firstly, an LCA analysis was performed on the most common draw solutions currently exploitable in large-scale FO systems. To obtain comparable results, the bulk osmotic pressure was used as the functional unit for this LCA analysis. Based on the results obtained in this first step, a study was performed on potential FO-PDMP systems, where different inorganic draw solutes can be deployed. To compare the different FO-PDMP solutions, the functional unit was defined as 1 m^3 of water produced by the coupled system. A schematic overview of the methodology followed in this study is reported in Fig. 1. Details are discussed in the sections below, considering different boundary conditions while maintaining the same environmental indicators used for the analysis.

2.2. Environmental indicators

ReCiPe, CED (Cumulative Energy Demand), and IPCC2013 methodologies were employed to conduct the environmental assessments, following the same protocols reported in our previous publication (Giagnorio et al., 2017). Both midpoint and endpoint indicators from ReCiPe calculations were considered: they convey the direct cause-effect linkage and the overall impacts of the products/systems, respectively (Bare et al., 2000). The normalization of the endpoint indicators was carried out following the guidelines reported in the literature (Huijbregts et al., 2017; Sleeswijk et al., 2008). CED approach was applied to assess the required energy expressed as the primary energy demand coming from renewable and non-renewable sources, while IPCC 2013 analysis with a timeframe of 20 years was performed to assess the global warming potential of the various processes. Open LCA 1.10 equipped with Ecoinvent database 3.5 was used to perform the environmental impact calculations. Attributional LCA with a hierarchist (H) perspective was adopted. An overview of the methodologies, categories, and environmental indicators used in this work is reported in the Supporting Information (Table S1).

2.3. LCA methodology to assess the environmental impacts of FO draw solutions

Life cycle assessment was first performed to evaluate the environmental impacts related to the production and deployment of the most common FO draw solutions (DS). Sodium chloride (NaCl), magnesium chloride ($MgCl_2$), sodium sulfate (Na_2SO_4), and magnesium sulfate

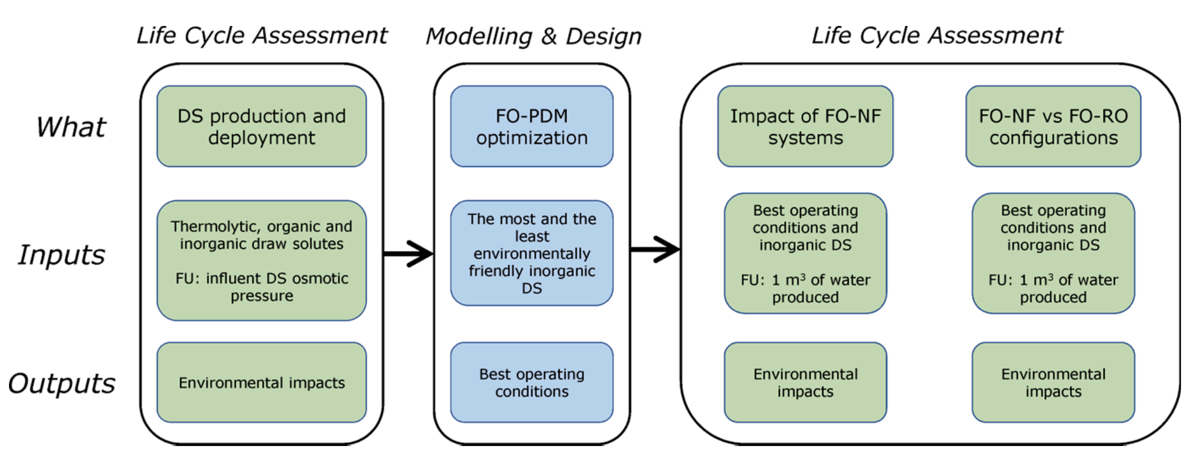

Fig. 1. Schematic representation of the scope of this study and the methodology followed to perform the environmental analyses.

($MgSO_4$) were chosen as representative of inorganic draw solutions composed by monovalent and multivalent ions, while ammonium bicarbonate was chosen as a representative thermolytic DS (Achilli et al., 2010; Giagnorio et al., 2019a; Giagnorio et al., 2019b; McCutcheon et al., 2005; McGinnis et al., 2013). A blended fertilizer was also investigated with the aim to include the main compounds usually employed for fertigation, namely, ammonium sulfate, sodium nitrate, potassium chloride, monopotassium phosphate (Phuntsho et al., 2013; Phuntsho et al., 2011). Finally, sodium propionate was considered as a promising organic draw solute, on the basis of recent reports of its use for the treatment of unconventional hypersaline waters (Islam et al., 2019). The draw solutions listed here are the only ones currently applied in full-scale forward osmosis units. A variety of alternative and innovative draw solutes have been proposed in the literature (Cai and Hu, 2016; Johnson et al., 2018), but with lack of substantial data for their implementation in real scale scenarios in the short-term. To obtain comparable results, the LCA inventory data were rationalized considering a common functional unit (FU), specifically, the amount of draw solute required to obtain a bulk osmotic pressure of 16 bar in an aqueous solution. This osmotic pressure was chosen in accordance with our previous research (Giagnorio et al., 2019a; Giagnorio et al., 2019b) and based on the system-scale analysis performed to evaluate the influence of operating parameters in large-scale FO systems (vide infra; next section). The LCA analysis was performed considering the material extraction and chemical synthesis followed by draw solution preparation and by its deployment, taking into account the potential post-recovery step for DS regeneration. An overview of the data analysis is presented in the SI (Table S2). Neither time frame nor specific regionalization was adopted to carry out the environmental assessment.

2.4. LCA methodology to assess the environmental impacts of FO-PDMP systems

Fertilizers do not require a post processing step, since their use as a draw solution in FO is performed with the objective of dilution and subsequent fertigation. Organic and thermolytic draw solutes require separate post-recovery thermal systems, i.e., thermal desalination and a distillation column respectively, to recover the draw solute and produce fresh water. On the other hand, all the various inorganic draw solutes are associated with the same post-recovery technologies, i.e., pressure-driven membrane processes. The LCA analysis was performed to evaluate the environmental sustainability of potential real-scale systems that include both the FO stage and an inorganic draw solution recovery step by pressure-driven membrane processes (FO-PDM). In this LCA, a specific FU of 1 m^3 of fresh water extracted from wastewater was adopted. In accordance with previous studies (Ansari et al., 2017; Linares et al., 2016), the systems were considered as having a lifetime of 15 years, accounting for the detrimental effects caused by the continuous circulation of a significant concentration of contaminants within the system. Consistent with previous studies (Binger and Achilli, 2020; Giagnorio et al., 2019a; Giagnorio et al., 2019b) and with the aim to limit the analysis to feasible operational conditions (i.e., to maximize the efficiency of the filtration system), the FO systems were modeled to achieve a high utilization of wastewater (i.e., an overall water recovery of 85%) and water fluxes greater than 5 L $m^{-2}h^{-1}$. The LCA was regionalized considering Milan (Italy) as installation site, more precisely San Rocco (Milan), the location of the wastewater treatment plant serving the Milan area. The following boundary conditions were applied to perform the LCA related to the FO-PDMP configurations:

1. An inlet flowrate of 100 m^3/h (~28 L/s) of wastewater was considered as feed solution entering the forward osmosis system, with the same characteristics of the real wastewater sample used in our previous study (Giagnorio et al., 2019a). The cleaning time and replacement of the FO membranes were estimated based on these characteristics. The bulk osmotic pressure of the feed solution was 0.5 bar on the basis of the total dissolved solid concentrations.
2. Both the concentrate produced by the forward osmosis system and the exhausted draw solution that needed replacement were modelled considering their disposal within the local wastewater treatment plant. This choice represents the worst-case scenario, since in future perspective, zero liquid discharge approaches may be pursued for the recovery of valuable products (e.g., water, minerals, organic material, nutrients).
3. Data related to the materials and building construction of the FO-PDMP systems were calculated based on previous research available in the literature, which reports the environmental impacts of real membrane desalination systems (Munoz and Fernandez-Alba, 2008; Zhou et al., 2011)
4. The FO-PDMP systems were designed to work with FO and NF/RO spiral wound membrane modules, namely, 4040 spiral wound membrane module for the FO unit (Kim and Park, 2011; Linares et al., 2016) and NF90-400/34i / SW30XHR-440i spiral wound membrane modules produced by DuPont for NF/RO. Data related to the materials used to fabricate the membrane modules were obtained from the literature and from membrane module autopsy (Bonton et al., 2012; Hancock et al., 2012; Kim et al., 2015; Yip et al., 2010).
5. The environmental burdens associated with transportation were calculated considering only the shipment of the (i) FO and (ii) NF/RO membrane modules from the production plant to the installation site, specifically: (i) from San Francisco, USA, to Milan, Italy, for FO membranes; and (ii) from Wilmington, USA, to Milan, Italy, for NF membranes. The shipping locations coincide with large membrane production plants. Given the availability of all the plant construction materials close to the installation site, their transportation was considered negligible compared to membrane module shipment, hence not included in the environmental assessment.
6. In accordance with what suggested by the membrane manufacturing producers and what discussed in literature reports (Hancock et al., 2012; Kim and Park, 2011), different chemical cleaning procedures were taken into account for the forward osmosis and for the post-recovery steps. For FO membranes, the typical cleaning procedure encompasses the employment of acid cleaning first (with the aim to remove scaling), followed by basic cleaning with surfactants (with the aim to remove organic matter and other colloids). For NF/RO membranes, only basic cleaning coupled with surfactants would be required, due to the better quality of the feed water to be processed in the DS recovery step. Low exergy sources were considered within the LCA analysis to account for the energy required by the clean-in-place system. Moreover, based on feed water characteristics, chemical cleanings for the forward osmosis membrane modules were modeled to be performed 4 times every year, twice as much as for the NF/RO membrane modules. This choice is a consequence of a fundamental consideration: in an FO-PDMP configuration, the FO membranes are those responsible for the filtration of the wastewater source, while the goal of the NF/RO membranes is to recover the diluted draw solution, which is virtually free of organics owing to the near complete removal performed in the first FO step. Moreover, the supposed lower fouling propensity of the forward osmosis technology is still under debate (Ricceri et al., 2021; Siddiqui et al., 2018), thus the choice of the cleaning cycles should account for worsts case scenarios where frequent chemical cleanings are needed to extend the lifetime of the membranes when treating wastewaters sources.
7. Membrane module replacement was considered as follows, based on the fouling considerations stated previously. For FO membrane modules: 3 years, coinciding with the worst case scenario, i.e., the shortest lifetime span of spiral wound modules. For NF/RO membrane modules: 5 years, that is, the average lifetime of spiral wound modules in real applications, when not prone to major fouling phenomena.

8. Based on research outputs presented in our previous publications (Giagnorio et al., 2019a; Giagnorio et al., 2019b), the recurring replacement of the entirety of the draw solution is strongly recommended, whether this operation occurs in batch or continuously. To assess the potential environmental burdens related to this operation, the average lifetime of the DS was set equal to 180 days of plant operation.
9. The energy needed for the flow of the feed water and draw solution within the FO membrane unit was considered negligible compared to the energy required by the pressure-driven membrane step. With the aim to guarantee a continuous operation, the energy supply was modeled following the Italian energy mix for electricity generation: 62% fossil fuels; 17% hydroelectric power; 14% solar power; 7% wind power.

A schematic overview of the FO-PDMP system scale modeling, together with a brief overview of the boundary conditions is presented in Fig. 2. A detailed description of the scenarios involved in this LCA analysis is reported in the SI (Figure S1) supplied with the inventory data (Table S3 and S4). The FO system was modeled to work with the feed solution facing the active layer of the membrane and in co-current configuration, that is, feed and draw solutions entering and exiting from the same side.

2.5. System-scale analysis of FO-PDMP configurations

As already discussed in our previous publications, FO system-scale analyses can be adequately performed by applying the mass transport equations governing the passage of water through the membrane (eq. (1)) and the reverse flux of the draw solute (eq. (2)) (Achilli et al., 2010; Tiraferri et al., 2013).

$$J_w = A\left\{\frac{\pi_D \exp\left(-\frac{J_w S}{D}\right) - \pi_F \exp\left(-\frac{J_w}{k}\right)}{1 - \frac{B}{J_w}\left[\exp\left(\frac{J_w}{k}\right) - \exp\left(-\frac{J_w S}{D}\right)\right]}\right\} \quad (1)$$

$$J_s = B\left\{\frac{C_D \exp\left(-\frac{J_w S}{D}\right) - C_F \exp\left(-\frac{J_w}{k}\right)}{1 - \frac{B}{J_w}\left[\exp\left(\frac{J_w}{k}\right) - \exp\left(-\frac{J_w S}{D}\right)\right]}\right\} \quad (2)$$

In these equations, A represents the active layer water permeability coefficient; S, B, and D, represent the support layer structural parameter, the salt permeability coefficient, and the diffusion coefficient of the draw solute in water, respectively. The properties of typical polyamide FO membrane were considered for the simulations, namely, A equal to 2.75 ± 0.5 L m^{-2}h^{-1}bar^{-1} and S equal to 427 ± 19 μm (Giagnorio et al., 2019a; Giagnorio et al., 2019b). B and D are instead parameters related to the draw solutes. For this study, NaCl, $MgCl_2$, and Na_2SO_4 were considered to evaluate potential configurations of FO-NF/RO systems. Table 1 shows the B and D values for each of these draw solutes. Finally, k represents the mass transfer coefficient at the active layer-solution interface, function of the hydrodynamics in the membrane flow cell and maintained equal to 68 L m^{-2}h^{-1} in all the simulations. The design of the DS post-recovery system was performed through Wave software (DuPont), which includes the database of the nanofiltration and reverse osmosis spiral wound membrane modules available for real-scale application. An overview of the input data used in Wave software is reported in SI (Table S5).

3. Results and discussion

3.1. LCA assessment of draw solutions

The results of the environmental impact assessment associated with

Table 1
B and D of the draw solutes.

Draw Solute	B (LMH)*	D (m^2/s) (Achilli et al., 2010)
Na_2SO_4	0.06	7.6×10^{-10}
$MgCl_2$	0.07	1.1×10^{-9}
NaCl	0.94	1.5×10^{-9}

*Values obtained experimentally and reported in previous publications (Giagnorio et al., 2019a).

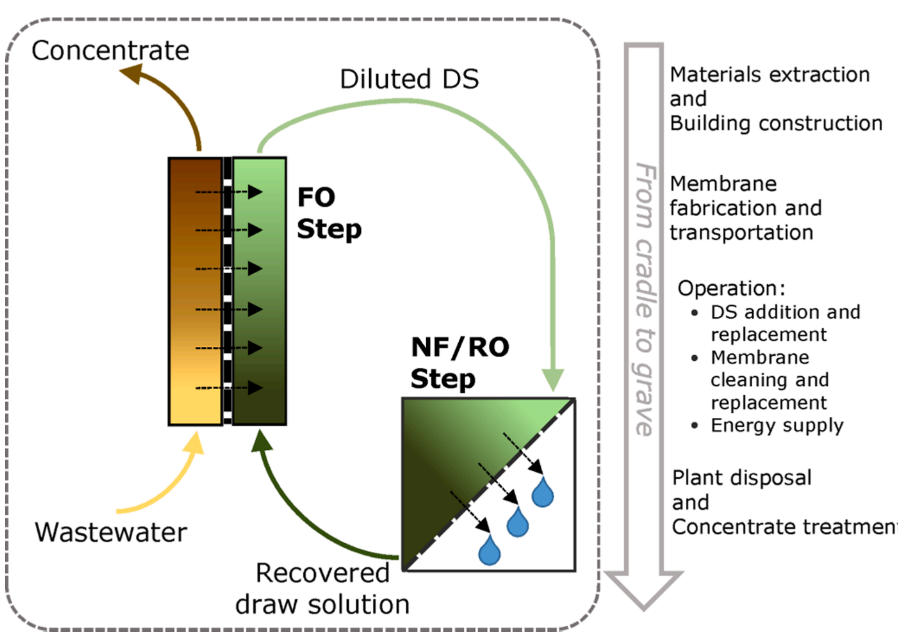

Fig. 2. Schematic diagram of the forward osmosis - pressure driven membrane system for wastewater treatment and schematic conceptualization of the “cradle-to-grave” LCA.

the production and deployment of the most common FO draw solutions are presented in Fig. 3. Among the various draw solutes, sodium chloride and sodium sulfate showed the lowest impacts. This observation can be ascribed to the widespread availability of these compounds in nature combined with their simple chemical processing (Grzmil and Kic, 2005). On the other hand, magnesium chloride is associated with the largest environmental impacts, mainly related to its production, which requires energy-intensive processes (e.g., electrolysis) (Choi et al., 2010; Sugasaka et al., 1970). While magnesium sulfate can be extracted from natural resources or from industrial effluents, thus beneficially exploiting waste streams, its production requires larger energy supply compared to the extraction of the sodium-based inorganic DS, resulting in higher overall impacts (Scheidema and Taskinen, 2011).

Note that the deployment of seawater as a widely available and virtually costless draw solution is not considered in this work. Various studies in the literature discussed the feasibility of large-scale FO-RO configurations in which contaminated sources are used to dilute the seawater draw solution, thus decreasing the energy input of the following reverse osmosis desalination step (Binger and Achilli, 2020; Hancock et al., 2012; Linares et al., 2016). However, to avoid irreversible fouling, seawater needs robust pre-treatment steps before entering the FO unit, which increases the overall environmental footprint of the plant. The direct utilization of seawater would not necessarily translate into a reduction of the environmental impacts of the system, especially if compared with the low environmental costs associated with the deployment of commercial NaCl.

In the case of ammonium bicarbonate, the environmental impacts shown in Fig. 3 are mainly associated with its deployment, due to the energy needed to dissociate and re-combine carbon dioxide and ammonia within the DS (McGinnis and Elimelech, 2007). In this case, the employment of low exergy grade sources (such as waste heat) would be beneficial, lowering the primary energy demand (Fig. 3b) and the required resources (Fig. 3a). However, the use of ammonium-based DS may result in losses of ammonia, potentially giving rise to toxic aqueous streams and environments (Braissant et al., 2013; Constable et al., 2003). Unexpected losses of CO_2 from the ammonium bicarbonate system would potentially contribute to an increase of the greenhouse gas emissions.

The environmental impacts of the blended fertilizer are strongly related to the needs of employing various chemical compounds to reach the desired nutrient ratio. The impact assessment reported in Fig. 3 was calculated based on the standard configuration for fertilizer-drawn FO systems, that is, one filtration cycle and no DS regeneration. However, in order to guarantee the adequate osmotic pressure and subsequent fertigation over the lifetime of the system, new chemicals must be constantly added within the FO filtration step, thus exploiting energy and resources (Shi et al., 2018). Among the fertilizing agents, sodium nitrate was found to be the most impactful, due to its well-known toxicity to human health and to aquatic ecosystems; see Fig. 3a and Figure S2 of the Supporting Information (SI) (Camargo et al., 2005; Umar and Iqbal, 2007). Another issue of fertilizer-based DS is that membranes are not completely or equally selective to the various ions composing the mixture or to the compounds contained in the feed waters, thus the loss of draw solute and the passage of feed components may induce a departure from the ideal nutrient ratio needed for the subsequent fertigation. This phenomenon was shown in previous studies (Kim et al., 2017; Phuntsho et al., 2013), which recommended the use of a downstream filtration system to process the diluted DS with the aim to obtain an adequate nutrient chemical composition. Clearly, this additional step would strongly increase the environmental impacts of the final FO-based configuration.

Finally, a promising organic draw solute, namely, sodium propionate, showed large environmental impacts, due to the exploitation of a significant amount of natural resources (Fig. 3a) and requiring the largest primary energy input among the various draw solutes analyzed (Fig. 3b). These results can be rationalized considering the NaPRO production process, which involves the combination of sodium hydroxide and propionic acid, the latter being by far the most impactful compound due to the involvement of energy-intensive oxidative reactions in processes adopted today (Ahmadi et al., 2017). The combination of FO with thermal systems for DS recovery represents the most promising technological solution to exploit the unique property of the NaPRO, that is, the possibility to treat complex hypersaline water sources through the achievement of extreme osmotic pressure in FO (Islam et al., 2019). However, the use of a thermal desalination process, if not supplied with low-exergy or waste heat, increases the primary energy demand of the overall system, as well as its carbon footprint.

3.2. *Evaluation of process parameters in hybrid FO-NF system configurations*

A detailed investigation was performed for FO-based systems working with Na_2SO_4 or $MgCl_2$ as DS. This analysis is particularly interesting because the two systems are analogous, both being based on FO followed by an NF step for DS recovery. That being said, the deployment of different DS entails significant changes in the characteristics of the systems, even for the analogous configuration. Also, the two DS were found to be at the opposite ends of the range of environmental burdens associated with their production and deployment (Fig. 3). Before performing the LCA analysis, the possible FO-based system configurations should be understood and described. Therefore, Fig. 4 presents a system-scale analysis carried out to assess the influence of process parameters in such hybrid FO-NF systems, and performed by applying the same boundary conditions used for the subsequent LCA analysis.

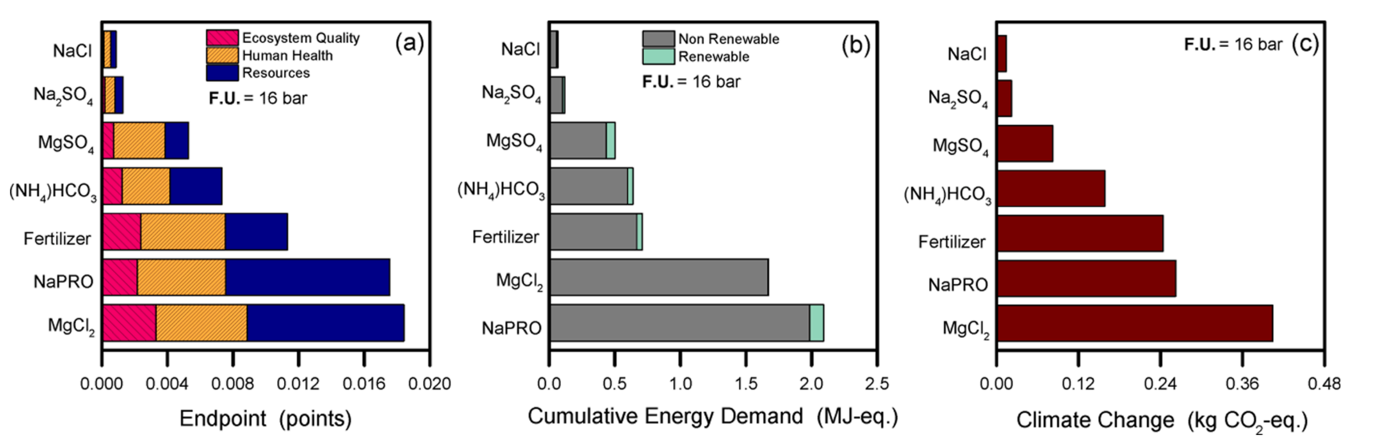

Fig. 3. Environmental impact assessment of potential forward osmosis draw solutions calculated based on the influent DS osmotic pressure as functional unit equal to 16 bar. Results are presented for (a) ReCiPe, (b) CED, (c) IPCC2013 methodologies.

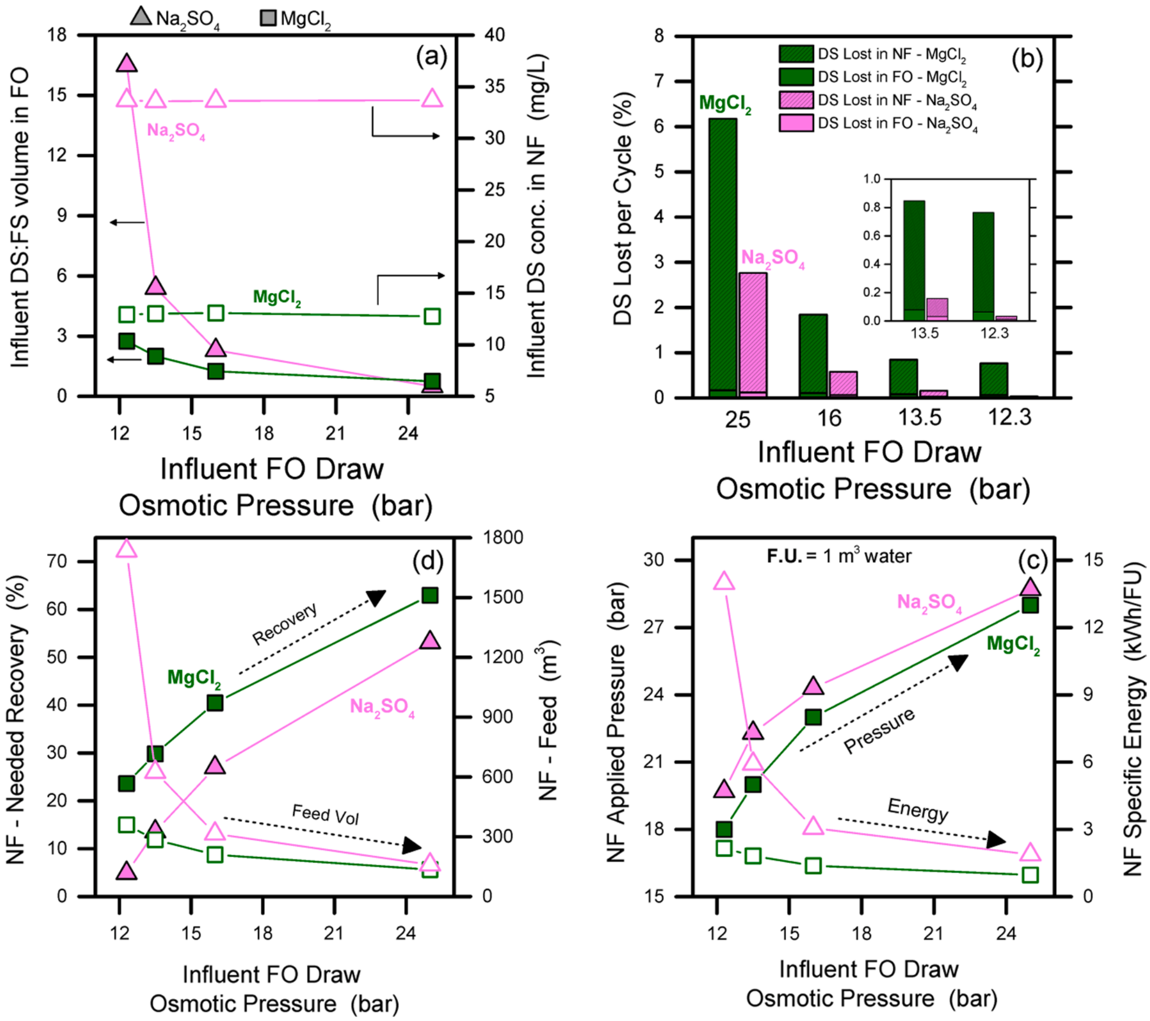

Fig. 4. Variation of the process parameters in real-scale FO-NF applications as a function of the osmotic pressure of the draw solution entering the FO step, with wither sodium sulfate or magnesium chloride as draw solute. The FO recovery rate was set at 85% from simulated wastewater. (a) Required DS:FS flow rate in FO and the resulting concentration of diluted DS entering into the recovery NF step as feed solution. (b) Percentage of draw solute mass lost at the end of each filtration cycle. (c) Correlation between the external hydraulic pressure needed in NF to recover the original DS volume and the energy required to carry out the NF separation. (d) Correlation between the feed volume entering the NF recovery step and the resulting required recovery rate, rationalized considering the functional unit of 1 m^3 of water produced. The lines in (a), (c), and (d) are only intended as guide for the eyes.

When setting a high water recovery rate (85%) and imposing that FO water flux should never fall below 5 L $m^{-2}h^{-1}$, either large influent draw osmotic pressures or influent DS flow rates are required in the FO step. In particular, a higher influent DS osmotic pressure would allow the use of a lower DS:FS ratio, and vice versa; see Fig. 4a, left axis. This effect results in a larger dilution factor for draw solutions that are initially more concentrated, which can be observed looking at the trend of the FO water flux as a function of the membrane area (Figure S3 of the Supporting Information). Interestingly, these different operating conditions have negligible effect on the draw solute concentration of the diluted stream exiting the FO system (Fig. 4a, right axis). The need of higher DS flow rates for lower draw osmotic pressures is much more pronounced for sodium sulfate compared with magnesium chloride. This result is due to the intrinsic lower osmotic pressure achievable per unit mass with the sodium-based DS, which is also characterized by a lower diffusion coefficient (Table 1). Specifically, deploying Na_2SO_4 with an influent DS osmotic pressure lower than ~ 15 bar, results in an extremely large DS flow rate needed to achieve the desired recovery. The subsequent need of pressurizing larger feed flows of diluted draw solution in NF requires excessive energy supply with respect to the correspondingly low recovery ratio (Fig. 4c, d), making the overall FO-NF configuration unfeasible. In summary, at fixed recovery rate, lower DS osmotic pressures in the FO step ultimately translate into larger energy needs for the NF system. For this reason, the use of magnesium chloride in real-scale FO-NF systems for wastewater treatment may be advantageous compared to the deployment of sodium sulfate, especially when working within a low-range of influent draw osmotic pressures (i.e., below 20 bar). Note that similar total membrane area would be needed in FO units running with $MgCl_2$ or Na_2SO_4 (Figure S3).

The results presented in Fig. 4b present the losses of draw solutes calculated per filtration cycle within the FO and the NF filtration steps. Estimated DS losses are low in FO, thanks to the intrinsically high selectivity of the membrane toward high multivalent ions, while losses were estimated to be much larger through the looser NF membrane. In accordance with previous studies, sodium sulfate is more easily retained by NF membranes with respect to magnesium chloride, for which significant losses may be achieved when increasing the influent osmotic pressure of the draw solution, reaching almost 10% DS loss per filtration cycle when working with an influent draw osmotic pressure above 25 bar (Giagnorio et al., 2019a; Giagnorio et al., 2019b; Giagnorio et al., 2018a; Giagnorio et al., 2018b).

3.3. Environmental sustainability of FO-NF systems

LCA analyses were performed to assess the environmental sustainability of the FO-NF systems described in the previous section and in Fig. 4, which may be potentially applied for wastewater treatment. The endpoint environmental impacts (ReCiPe methodology) are presented in Fig. 5a for a draw solution of magnesium chloride and in Fig. 5b for a draw solution of sodium sulfate. The related midpoint outcome can be found in Figure S5 of the SI. The results suggest that the energy supply and the management of the draw solution play the most significant roles. On the other hand, membrane fabrication and transportation, material extraction and building construction, as well as plant disposal, do not contribute significantly to the environmental burdens within the total plant lifetime. The environmental contribution related to the discharge of the concentrate stream and of the exhausted draw solution, and that related to their treatment within a wastewater treatment plant may be observed in specific midpoints (e.g., marine and freshwater eutrophication), but they do not play a significant role at the endpoint. However, it should be noted that depending on the DS, the discharge of the exhausted DS may impact the biological processes in wastewater treatment, if present. Notably, the environmental sustainability of FO-NF units would change greatly depending on the draw solute.

The utilization of magnesium chloride would result in more environmentally friendly configurations when operating at mid-low influent draw osmotic pressures (<16 bar) in FO. This observation can be ascribed to the lower energy required by the downstream NF system, which is strongly dependent on the influent DS volume needed to reach the desired FO osmotic pressures, as discussed above. However, above a certain threshold of osmotic pressure (> ~16 bar), the loss of draw solutes (Fig. 4b) would generally translate in higher impacts, especially for compounds like magnesium chloride, associated with large burdens relative to its production(Fig. 3). In fact, when the osmotic pressure of the influent solution of the FO step is high, the use of Na_2SO_4 results in more sustainable large-scale systems. The vast majority of the environmental impacts associated with this DS are in fact related to the energy supply of the downstream NF step. Therefore, as this energy is smaller for high initial osmotic pressure (Fig. 4c), sodium sulfate becomes more sustainable than magnesium chloride. Indeed, while higher concentrations are needed to obtain high osmotic pressures, the production of sodium sulfate is related to low environmental burdens (Fig. 3). Analogous results of those obtained with the ReCiPe approach were obtained carrying out the LCA analysis using CED and IPCC2013 methodologies (Figure S4 of the SI)

3.4. Environmental sustainability of FO-NF vs. FO-RO hybrid systems

The discussion has so far focused on FO-NF systems. However, the results suggest that it is useful to also evaluate the possible deployment of more selective membranes in the downstream regeneration step, i.e., the use of RO instead of NF to recover the draw solution, which would be associated with lower DS losses. Given the intrinsically lower productivity (Fujioka et al., 2013; Vrijenhoek et al., 2001; Yang et al., 2019), reverse osmosis membranes would require a larger number of modules to produce the same permeate flow. However, from a purely environmental standpoint, the plant installation and disposal have been found to be less impactful than draw solution losses and energy supply. The use of RO may specifically reduce the significant environmental burdens associated with the loss of $MgCl_2$ and observed at high concentrations of this compound as DS. Also, RO units would allow the deployment of NaCl, which was found to be associated with lower environmental impacts related to its production and deployment, with respect to both $MgCl_2$ and Na_2SO_4. The LCA analysis was thus also performed to compare FO-NF systems with FO-RO systems, with the main results summarized in Fig. 6 for an influent draw osmotic pressure in FO of 16 bar.

Fig. 6a presents an overview of the results from the LCA analysis performed through ReCiPe methodology. For three out of the four systems, specifically when RO is used as a DS recovery step and for the FO-NF system based on Na_2SO_4, more than 90% of the total environmental impacts can be ascribed to the energy supply. This result can be rationalized with the exploitation of mostly non-renewable energy sources (Fig. 6c), which are required to ensure the non-intermittent operation of the plant. In perspective, improvement of the renewable energy harvesting and storage would significantly reduce the environmental burdens of real-scale FO-PDMP systems. The loss of magnesium chloride in the FO-NF system would instead increase the relative importance of draw solute management in the overall environmental analysis of this system. Even working with a mid-range osmotic pressure in FO (16 bar), the losses of $MgCl_2$ within the NF process and the subsequent replenishment would offset the benefits of a lower energy supply and translate into nearly 60% of the total environmental impacts of the technology.

In this respect, further interesting conclusions may be drawn from the results presented in Fig. 6b, which reports the fate of the various inorganic draw solutes within the potential FO-PDMP systems. The largest losses of draw solutes among the four configurations would be achieved in the FO-NF systems, with most of the Na_2SO_4 or $MgCl_2$ lost in the nanofiltration step. The deployment of a more selective process (i.e., RO) as downstream recovery step would reduce the loss of multivalent

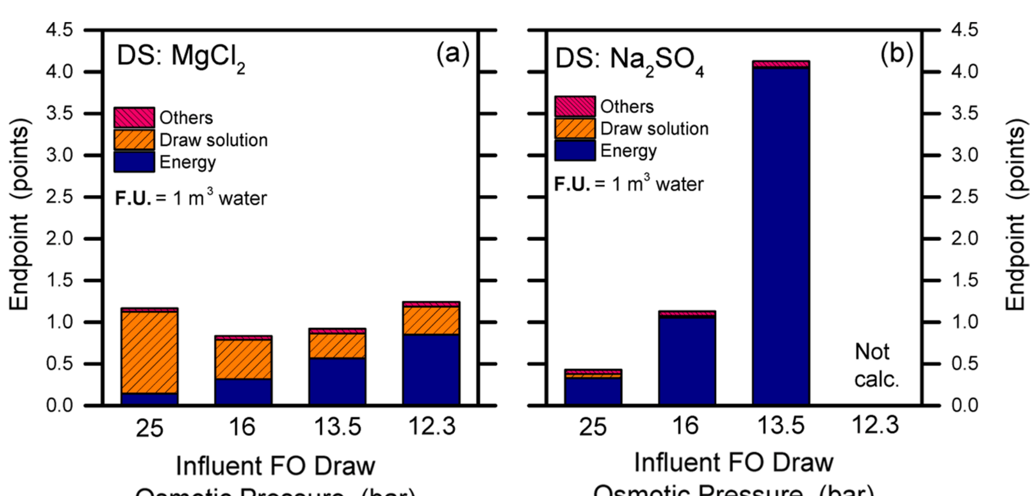

Fig. 5. Environmental impact assessment of different FO-NF system configurations calculated based on the functional unit of 1 m^3 of water produced, using the ReCiPe endpoint methodology, for draw solution consisting of (a) $MgCl_2$ or (b) Na_2SO_4.

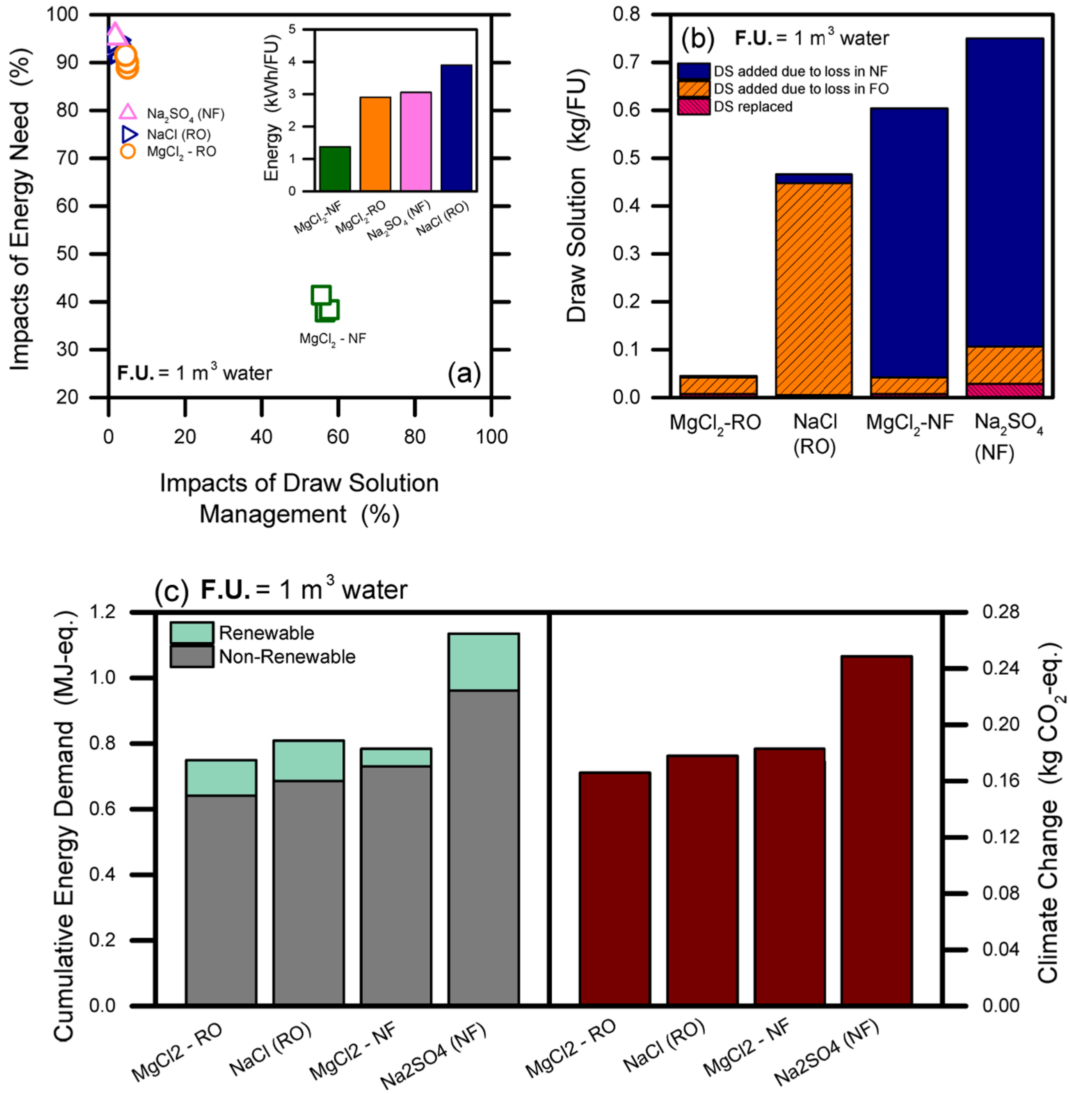

Fig. 6. Environmental impact contributions related to large-scale FO-NF and FO-RO configurations calculated based on the functional unit of 1 m^3 of water produced. (a) Results of LCA using ReCiPe methodology, extrapolating the relative environmental burdens associated to the draw solution and the energy requirement of the DS recovery step. (b) Amount of draw solution that needs to be supplemented per FU in the various configurations. (c) Impacts calculated using CED, IPCC2013 methodologies.

ions-based DS but at the expense of an increase in the energy supply.

There seem to be a clear a trade-off between the environmental impacts associated with the loss and replenishment of draw solute (i.e., use of NF as DS recovery step) and those associated with the energy supply (i.e., use of RO). This result is apparent when confronting the endpoint indicators for the two configurations (FO-NF vs. FO-RO) encompassing the deployment of $MgCl_2$ (Supporting Information, Figure S6). Overall, the two systems comprising an RO downstream step would be associated with the lowest and similar environmental burdens, while the Na_2SO_4-based system would be characterized by the largest impacts. Please note that for this draw solute, the use of RO as a recovery step would not be advantageous because it would translate in larger energy needs without significant gains in terms of DS losses, which are already low in the FO-NF configuration.

4. Concluding remarks

In this work, the environmental sustainability of forward osmosis was evaluated looking at the influence of the process parameters involved in potential real-scale FO-based systems, also comprising the downstream draw solute reconcentration step. At first, the most suitable draw solutes were analyzed through life cycle assessments. Significantly higher environmental impacts would be associated with the deployment of thermolytic, organic, and fertilizer-based draw solutes in FO, compared to simpler and more accessible inorganic DS.

When focusing on the use of inorganic draw solutes in the complete FO-based systems, the influent draw osmotic pressure in FO strongly influences the design of the real-scale filtration systems. The results suggested that the most important contributions to environmental impacts of potential large-scale FO-based plants with the objective of fresh water production, are associated with energy supply and draw solute management, while plant installation, disposal, and other factors are negligible when considering the lifetime of the systems.

Even if the production and management of sodium-based DS (NaCl and Na_2SO_4) are associated with the lowest intrinsic environmental impacts, the deployment of these compounds in FO-PDMP systems was found to translate into higher impacts related to the energy required by the downstream PDMP system needed for draw solution recovery. On the other hand, for draw solutes characterized by high intrinsic environmental impacts, such as $MgCl_2$, a trade-off regarding environmental burdens was observed between the effect of DS losses and that of energy supply: when attempting to minimize losses by the use of more selective membranes in the downstream recovery step, the environmental gains are offset by the larger associated energy needs due to the lower RO membrane productivity. It should be noted that this study only analyzed the environmental impacts through LCA of an FO-based system to extract high-quality water from wastewater. Economic considerations are not taken into account, nor are systems with different treatment goals, such as, for example, desalination.

CRediT authorship contribution statement

Mattia Giagnorio: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Visualization, Writing - original draft, Writing - review & editing. **Alessandro Casasso:** Resources, Data curation, Writing - review & editing. **Alberto Tiraferri:** Funding acquisition, Project administration, Supervision, Visualization, Writing - review & editing.

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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