

## PAPER

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# Membrane technology for water reuse in decentralised non-sewered sanitation systems: comparison of pressure driven (reverse osmosis) and thermally driven processes (membrane distillation and pervaporation)<sup>†</sup>

E. Mercer,<sup>a</sup> C. Davey,<sup>a</sup> Y. Bajón Fernández,<sup>a</sup> S. Septien,<sup>b</sup> S. Tyrrel,<sup>a</sup> E. Cartmell,<sup>c</sup> M. Pidou<sup>a</sup> and E. J. McAdam<sup>a</sup>

Membrane processes are an established barrier technology for water reclamation from wastewater. Applied at a household scale to improve sanitation practice, membrane technology can disrupt the source–receptor pathway, alleviate water scarcity through eliminating flush water and recover clean water for reuse. However, blackwater comprises a distinct composition compared to municipal wastewater, and there is only limited understanding on whether membrane selectivity is sufficient to produce water of sufficient quality for reuse. In this study, pressure driven and thermally driven membranes are evaluated for their potential to treat blackwater, by relating selectivity to relevant water quality standards (ISO 30500) and the transmission of volatile organic compounds (VOCs) that are primarily associated with faecal odour, and thus constitute a critical challenge to water reuse. Both pressure driven (reverse osmosis) and thermally driven (membrane distillation and pervaporation) membranes were able to produce water that conformed to category B of the ISO 30500 standard for the majority of determinants. A critical limiting factor was in the selectivity for ammonia and odorous VOCs which were generally poorly removed by reverse osmosis and membrane distillation. The high ammonia transmission was accounted for by the elevated pH of blackwater which shifted the ammonium equilibria toward volatile ammonia which is poorly separated by RO polymers, and is free to diffuse through the gas-filled micropores of the membrane distillation membrane. In contrast, greater ammonia and VOC separation was evidenced for the pervaporation membrane due to advanced polymer–solute interactions. In a preliminary assessment, the hydrophilicity exhibited by the membrane was also advantageous to withstanding fouling. If complemented with a polishing step to target the residual COD and VOCs (that may be of similar origin), pervaporation could deliver to category A standard for non-potable reuse. This is particularly advantageous for water scarce regions where solar or liquified fuels may be applied in favour of electricity for off-grid sanitation.

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## Water impact

Thermally driven membrane processes have demonstrated water reuse potential from concentrated blackwater in terms of water quality, odour management and flux robustness. Facilitated by off grid heat sources such as waste heat or cooking gas, this presents an economically accessible alternative to conventional pressure driven membrane processes which rely on electricity.

## 1. Introduction

Two billion people lack access to adequate toilet facilities, resulting in over 673 million people having to defecate in the open, engendering transmission of diseases such as cholera, typhoid, polio, hepatitis A and trachoma.<sup>1</sup> The United Nations' Sustainable Development Goal 6 (SDG 6) recognises this substantive challenge to human health, setting an

<sup>a</sup> Cranfield Water Science Institute, Vincent Building, Cranfield University, Bedfordshire, UK. E-mail: e.mcadam@cranfield.ac.uk

<sup>b</sup> WASH R&D Centre, School of Engineering, University of KwaZulu-Natal, Durban, 4041, South Africa

<sup>c</sup> Scottish Water, Castle House, Carnegie Campus, Dunfermline, UK

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agenda to provide access to clean water and sanitation for all by 2030, whilst mandating water reuse as a critical tool to alleviate water scarcity.<sup>2</sup> However, implementing capital intensive centralised (sewered) wastewater infrastructure into low income countries (LICs) is economically challenging,<sup>3</sup> instead deferring to decentralised sanitation, primarily using pit latrines, which pose health risks through contamination of local water sources *via* poor installation, operation or through the unregulated environmental discharge of faecal sludge.<sup>4</sup> This challenge is not limited to LICs, where comparable inequity is identified in developed nations. For example, more than 2 million US citizens live without running water and basic indoor plumbing, whilst inadequate wastewater systems have been identified in up to 90% of households in discrete low-income regions of the US, resulting in high incidence of hookworm infection, transmitted *via* the faecal–oral route.<sup>1</sup>

Barrier technology that can be practically implemented within a decentralised context presents an opportunity to break the source–receptor pathway, and offers protection to 29% of the global population that depend on water supplies of unknown provenance, which are commonly contaminated from poor sanitation practice.<sup>2</sup> This is the key remit of the recently published ISO 30500 standard on non-sewered sanitation, which places emphasis on water reuse, to readdress water inequality introduced through water scarcity. This was exemplified by the approach of ‘day zero’ in Chennai and Cape Town, where communal water supply was no longer sustainable,<sup>5</sup> which emphasised how important recovering even small water volumes could be, and may help to revalue barrier technology for implementation into the decentralised context. Membrane processes are an established barrier technology capable of direct and indirect potable water reuse schemes from municipal wastewater.<sup>6</sup> The conventional two-stage ultrafiltration (UF) and reverse osmosis (RO) system used is commercially available for point-of-use potable applications (single household), demonstrating that economies of scale exist and may be amenable for decentralised sanitation. However, for point-of-use, power consumption is avoided through pre-pressurisation of the main supply, whereas a UF-RO sanitation system requires a feed pump to overcome osmotic pressures of ~30 bar, due to the high salt content of urine (~248 mEq L<sup>-1</sup>).<sup>7,8</sup> This increases system cost and introduces a high peak power demand (but a low net energy demand). Membrane distillation (MD) and pervaporation (PV) employ a vapour pressure gradient to deliver permeate quality comparable to RO for desalination applications, due to the selectivity that is achievable for non-volatile contaminants.<sup>9–11</sup> Whilst the source of heat restricts MD and PV, market penetration of liquefied fuels (propane and butane) has accelerated through government schemes (*e.g.* PMUY program) seeking to grow cleaner cooking fuels, and to ensure energy equity for women in below-poverty-line households.<sup>12</sup> This has resulted in market penetration of liquefied fuels into India of 95%, which exceeds grid

produced electricity coverage, and as the unitary cost (\$ kWh<sup>-1</sup>) is around 20% of power, thermally driven membrane processes may present a viable economic alternative for decentralised sanitation. This is complementary to other accessible heat sources including low grade waste heat and solar heat energy ranging from 4.5–7.5 kWh m<sup>-2</sup> in water scarce areas.<sup>13</sup>

Non-sewered sanitation (NSS) systems typically accept blackwater, often with limited or no flush water, as transport within the sewerage network is no longer demanded.<sup>14</sup> The resultant blackwater therefore comprises urine and faeces and is more concentrated than conventional blackwater, by over an order of magnitude.<sup>15</sup> This feed chemistry is considerably more complex, comprising higher concentrations of pathogens, a broader MW (molecular weight) range of organics, and a transient solution chemistry (*e.g.* pH), all of which risk reducing permeate quality. The ISO 30500 standard assesses the safety of the outputs of an NSS technology treating blackwater. Pressure driven UF-RO and thermally driven MD have shown to comply to most of the ISO 30500 standards treating CB,<sup>10,16–18</sup> while the robust MD operation achieving all ISO 30500 parameters has been demonstrated with upfront UF, which through size exclusion reduces the particulate and colloidal fraction contributing to membrane wetting, or utilising smaller pore sizes to increase the liquid entry pressure.<sup>18,19</sup>

The transport mechanisms in RO and MD are distinct and their role in fostering selectivity remains poorly understood for a wider variety of compound chemistries distinct to CB, particularly gaseous compounds or odour causing volatile organic compounds. While odour emissions are considered in the ISO 30500 standards, they are specific to the gas phase and not the liquid phase. However, taste and odour (T&O) is an aspect critical to water reuse as data from LIC surveys indicate that even if safe, malodorous water would likely be discarded, in preference for visually lower quality water that was odourless.<sup>20</sup> Due to the volatile nature of these contaminants, the selectivity of the thermally driven processes is also challenged. Mercer *et al.*<sup>21</sup> demonstrated the rejection and selective enrichment of faecal volatile T&O compounds by PV using hydrophilic and hydrophobic polymers respectively. This ultimately altered the compound concentration and mixture profile of VOCs, which are known to influence perception.<sup>21–24</sup> Pervaporation is a thermally driven membrane process like MD. Although, unlike MD which is microporous like UF, it also possesses similarities to RO through the use of a dense membrane, where mass transfer is governed by interactions between the compound and polymer through a solution-diffusion model, with additional selectivity though steric hindrance size exclusion.<sup>25</sup> Pervaporation may therefore provide an additional selective barrier for gas phase separation in thermal processes, with the polymer offering distinctive selectivity to the polymers employed in RO. While PV has been investigated for water



recovery from urine<sup>26–32</sup> and model faecal volatile organic compounds, it has not been examined with CB.

This study therefore aims to evaluate the suitability of pressure driven (dense RO) and thermally driven membrane processes (microporous MD and dense PV-hydrophilic and hydrophobic) for delivering safe sanitation and water reuse within a decentralised context. Specific objectives are to: (i) benchmark permeate quality against water reuse standards proposed for non-sewered sanitation (ISO 30500); (ii) evaluate the transport of the volatile fraction to determine how the driving force and material characteristics influence reuse quality; and (iii) undertake preliminary assessment of membrane permeability in response to concentrated blackwater treatment.

## 2. Materials and methods

### 2.1 Experimental setup

Pressure driven RO was operated in a HP4750 cell (Sterlitech, USA) at 12 bar (supplied by compressed air) in dead-end batch mode (Table 1). A magnetic stirrer (part of the HP4750 cell) was operated at 400 rpm on the feed side of the membrane to limit concentration polarisation. Flat sheet membranes from Sterlitech (TriSep X201, USA) were cut to an effective membrane area of 0.00146 m<sup>2</sup>. Permeate samples were collected within an airtight 50 mL glass conical flask. Flux was determined by measurement of the temporal permeate mass up to a standardised filtered volume of 10 L m<sup>−2</sup> (PR410 Symmetry, Cole-Parmer Ltd., London, UK). For thermally driven membranes (PV and MD, Fig. 1 and Table 1), the feed was circulated at 0.2 L min<sup>−1</sup> and vacuum was applied at 50 mbar on the permeate side using a

diaphragm vacuum pump (MD 4C NT, Vacuubrand, Brackley). Flat sheet hydrophilic polyvinyl alcohol (PVA) membranes (DeltaMem, Pervap 4101, Switzerland) provided a membrane area of 0.0153 m<sup>2</sup> (Model Products, UK). Hollow fibre (HF) polydimethylsiloxane (PDMS) (Permselect, PDMSXA-2500, USA) and hydrophobic microporous polypropylene (pore size 0.04 µm) (3M-Liqui-Cel, MM 1.7 × 5.5, USA) possessed membrane areas of 0.25 m<sup>2</sup> and 0.54 m<sup>2</sup> respectively. The permeate was collected through a condenser (2 °C) until a standardised filtered volume of 1 L m<sup>−2</sup> was reached (Fig. 1). A liquid nitrogen cold trap (−196 °C) was used for permeate collection during the determination of VOCs. All membranes were operated at 50 °C within a thermostatic bath (Grant TC120, Cambridge, UK) to provide parity between each process. Pressure driven membranes were also operated at ambient temperature (20 °C). A virgin membrane was utilised for each experiment. Relative water flux was calculated by:

$$\text{Relative flux} = \frac{J}{J_0} \quad (1)$$

where  $J$  is the faecally contaminated urine flux (kg m<sup>−2</sup> h<sup>−1</sup>) and  $J_0$  is the deionised water flux (kg m<sup>−2</sup> h<sup>−1</sup>).

### 2.2 Feed preparation and analysis

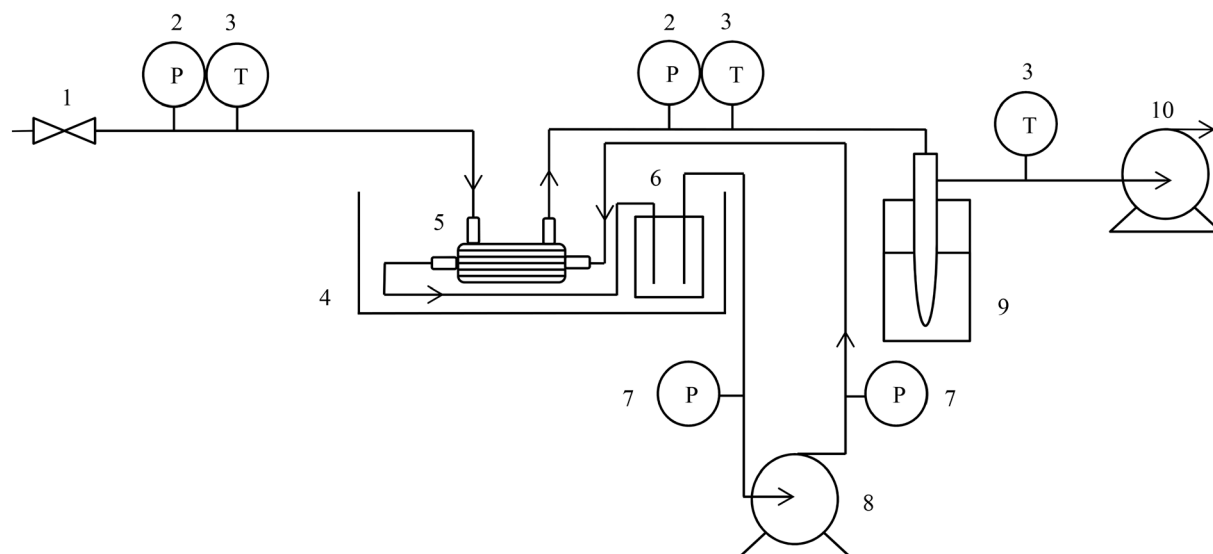
Concentrated blackwater was prepared at a 9:1 urine to faeces mass ratio, as this represents the typical proportion produced by an individual per day.<sup>37</sup> Samples were vortexed for 30 seconds to homogenise, to simulate conditions similar to a dynamic system where agitation occurs. This ratio can be considered representative of the maximum faecal

**Table 1** Summary of membrane characteristics and operation conditions used in this study

	Reverse osmosis	Pervaporation		Membrane distillation
Manufacturer (model)	TriSep	DeltaMem	Permselect	3M™ Liqui-Cel™
Model	(X201)	(Pervap™ 4101)	(PDMSXA-2500)	(MM1.7 × 5.5)
Material	Polyamide-urea	Polyvinyl alcohol	Polydimethylsiloxane	Polypropylene
Typical application	Desalination	Purification of organic mixtures	Removal of trace organic solvents from in industrial wastewaters	Desalination, process water treatment
Driving force	Pressure	Vapour pressure gradient	Vapour pressure gradient	Vapour pressure gradient
Membrane area (m <sup>2</sup> )	0.00146	0.0153	0.25	0.54
Membrane thickness (µm)	100–150	0.5	55	NAv
Pore size (µm)	<0.0005 <sup>b</sup>	<0.0005 <sup>b</sup>	<0.0005 <sup>b</sup>	0.04
Contact angle	28.5 <sup>c</sup>	43 <sup>d</sup>	116 <sup>d</sup>	104 <sup>e</sup>
Hansen solubility $\delta$ (MPa m <sup>−1/2</sup> )	NAv	25.78 <sup>f</sup>	15.59 <sup>f</sup>	NAp
Geometry	Flat sheet	Flat sheet	Hollow fibre	Hollow fibre
Feed volume (mL)	300	600	500	500
Permeate volume (mL)	20	20	5, 10, 20	5, 10, 20
Recommended pressure (bar)	7–21	≤1	≤1	0.04–0.2
Operating pressure (bar)	12	0.05 <sup>a</sup>	0.05 <sup>a</sup>	0.05 <sup>a</sup>
Recommended operating temperature (°C)	2–45	≤50	≤60	40
Operating temperature (°C)	20, 50	50	50	50

<sup>a</sup> Vacuum pressure; NAp: not applicable; NAv: not available. <sup>b</sup> Baker *et al.*<sup>25</sup> <sup>c</sup> Liu *et al.*<sup>33</sup> <sup>d</sup> Experimentally derived using the sessile drop method, Strobel *et al.*<sup>34</sup> <sup>e</sup> Mark.<sup>35</sup> <sup>f</sup> Bormashenko *et al.*<sup>36</sup>





**Fig. 1** Membrane distillation and pervaporation membrane rig schematic. 1: Needle valve; 2: absolute pressure transducer; 3: temperature probe; 4: thermostatic bath; 5: membrane; 6: feed solution; 7: pressure gauge; 8: peristaltic pump; 9: liquid nitrogen cold trap or condenser; 10: diaphragm vacuum pump.

contamination of the solution phase, expected within NSS systems using a waterless flush (*i.e.* no flush water). The sample was pretreated by a passive filtration step through sand and cotton wool to limit the coarse material (*e.g.* unmasticated food) which could lead to blockage during pumping. All experiments were performed in accordance with guidelines set out on the anonymous collection of fresh urine and faeces from consenting volunteers and disposed of as biohazardous waste or through the normal sewerage system. This protocol was approved by the Cranfield University Research Ethics System (CURES, project ID 3022). Informed consents were obtained from human participants volunteering for this study.

The discharge and reuse standards contained within the recently published ISO 30500 standard on 'Non Sewered Sanitation Systems' sets out specified guideline values for ammonium ( $\text{NH}_4^+\text{-N}$ ), total phosphorus (TP), chemical oxygen demand (COD), coliform forming units (CFU), total suspended solids (TSS) and pH as key parameters for evaluation.<sup>16</sup> The purpose of this standard is for the certification of technologies explicitly developed to deliver safe sanitation at a single household scale, in order to reduce consumer risk at procurement. Consequently, membrane selectivity was characterised using these parameters together with conductivity. Data was triplicated however insufficient to conform to a 20% variance of at least five trials (as stated in the standard) and therefore the mean is only used as a quick reference to the guidelines, to benchmark the membrane technologies. For the determination of TP,  $\text{NH}_4^+\text{-N}$  and COD, proprietary wet chemistry methods were used coupled with quantitation by spectrophotometry (NOVA60 photometer, VWR, UK). The TSS, electrical conductivity and pH were measured using standard methods<sup>38</sup> and a Jenway 4330 meter respectively (Cole Parmer, Staffordshire, UK). Total coliforms

and *E. coli* coliform colony counts were based on methods 9215C, 9215D, 9922B and 9922D.<sup>38</sup> Pathogen reduction was characterised using the log removal value (LRV):

$$\text{LRV} = \log_{10} \left[ \frac{C_0}{C} \right] \quad (2)$$

where  $C_0$  is the initial pathogen concentration ( $\text{CFU mL}^{-1}$ ) and  $C$  is the pathogen concentration in the permeate ( $\text{CFU mL}^{-1}$ ). The permeate limit of detection (LD) was determined by enumerating 10 mL permeate for RO, and 100 mL for PV and MD using EZ-Fit filtration units (Merck Millipore, Watford, UK) which yielded LDs of  $1 \text{ CFU } 10 \text{ mL}^{-1}$  and  $1 \text{ CFU } 100 \text{ mL}^{-1}$  respectively. Non detected (ND) was classified as sample concentrations below the LD which did not contain coliforms within the filtered volumes.

### 2.3 Volatile organic compound sampling and analysis

Membrane odour analysis was carried out using both a synthetic and real CB matrix. Odourant production within the feed occurred during the duration of the trial altering the feed concentration (Fig. S1†), as T&O causing compounds (VOCs) are bacterial by-products.<sup>39</sup> Therefore, in order to accurately investigate polymer and process selectivity towards VOCs, the quantitative assessment of membrane separation was conducted using the synthetic matrix where the feed concentration was stable, and qualitative odour characterisation was carried out using the real matrix.

Nine VOCs were identified that represent commonly occurring compounds with differing chemical structures found in urine and faeces:<sup>24</sup> sulfides (dimethyl disulfide), aromatic heterocycles (indole, skatole), phenols (*p*-cresol), alcohols (1-butanol), aldehydes (benzaldehyde), ketones (2-butanone) and esters (ethyl propionate, ethyl butyrate)



**Table 2** Removal efficiencies of conductivity, total phosphorus (TP) and chemical oxygen demand (COD) on treating concentrated blackwater with membranes

	Conductivity	TP	COD
	Reduction (%)	Removal efficiency (%)	Removal efficiency (%)
RO (PA-UREA) 20 °C	88.3 ± 3.3	99.4 ± 0.2	98.6 ± 0.5
RO (PA-UREA)	86.4 ± 1.4	97.4 ± 1.2	98.8 ± 0.1
PV (PVA)	93.8 ± 5.5	100 ± 0	99.1 ± 0.4
PV (PDMS)	76.5 ± 4.8	99.9 ± 0	98.8 ± 0.7
MD (PP)	60.6 ± 8.1	99.7 ± 0.2	99.1 ± 0.3

Feed water temperature 50 °C, unless stated otherwise. Average feed conductivity, TP and COD is 13.07 mS cm<sup>-1</sup>, 203.39 mg L<sup>-1</sup> and 15.36 g L<sup>-1</sup> respectively. RO (reverse osmosis); PA-UREA (polyamide-urea); PV (pervaporation); PVA (polyvinyl alcohol); PDMS (polydimethylsiloxane); MD (membrane distillation); PP (polypropylene). Error represented as standard deviation of a triplicated experiment.

(Table S1†), to provide an understanding of membrane selectivity. For synthetic solutions, a 1000 mg L<sup>-1</sup> stock solution containing pure VOCs was first prepared in propylene glycol to dissolve all compounds. An aliquot was subsequently added to a pH 6.5 (to mimic fresh urine)<sup>37</sup> buffered solution according to Robinson and Stokes,<sup>40</sup> for the preparation of a synthetic feed standardised at 10 mg L<sup>-1</sup> for all VOCs. All the samples were stored within gas tight 10 ml glass centrifugal vials (Cole Parmer, UK) and analysed on the same day as the trial. Analysis of VOCs involved a solid phase pre-concentration step (Oasis HLB SPE cartridge, 1 g, Waters, USA) followed by quantification using gas chromatography mass spectrometry (GC-MS) (TQ 8040, Shimadzu, UK). Full details of quantification and method validation can be found in the ESI† and Mercer *et al.*<sup>21</sup> Membrane processes were compared by determining the separation of odorous VOCs using the ratio between the water flux and VOC flux, defined as:

$$\frac{J_{\text{VOC}}}{J_{\text{w}}} = \frac{\left(\frac{Q_{\text{VOC}}}{A_{\text{m}}}\right)}{\left(\frac{Q_{\text{w}}}{A_{\text{m}}}\right)} \quad (3)$$

where  $J_{\text{VOC}}$  is the VOC flux (kg m<sup>-2</sup> h<sup>-1</sup>),  $Q_{\text{VOC}}$  is the VOC flow rate (kg h<sup>-1</sup>),  $Q_{\text{w}}$  is the water flow rate (kg h<sup>-1</sup>) and  $A_{\text{m}}$  is the effective membrane area (m<sup>2</sup>).

### 3. Results and discussion

#### 3.1 Pervaporation offers competitive separation for ISO 30500 contaminants

The selective capabilities of each membrane were evaluated against key wastewater determinants set out in ISO 30500 (ref. 16) with the respective average loadings of 15 ± 3 gCOD L<sup>-1</sup>, 203 ± 142 mgTP L<sup>-1</sup>, 8.11 × 10<sup>6</sup> ± 9.43 × 10<sup>6</sup> CFU TC mL<sup>-1</sup> and 998 ± 740 mgNH<sub>4</sub>-N L<sup>-1</sup>. Organic (as COD) removal efficiencies were >98.6% for RO (20 °C, 50 °C), MD and PV (Table 2). Liu *et al.*<sup>41</sup> recorded a similar organic removal efficiency of 97% (as total organic carbon) for a RO membrane operated in forward osmosis (FO) mode at 25 °C, using a urine feed matrix, suggesting that RO can maintain high rejection capabilities with faecal organic loading (>3 times higher COD concentration). Kamranvand *et al.*<sup>10,42</sup> demonstrated >95% COD rejection using MD for CB, which was improved to 98.9% with a reduced pore size of 0.1 µm. In this study, the MD pore size is 0.04 µm which could explain the enhanced organics separation efficiency of >99% due to the additional barrier of gas phase size exclusion. With respect to the ISO 30500 standard, COD category B (≤150 mg L<sup>-1</sup>) was achieved by the thermal processes hydrophilic PV and MD, which represents restricted urban reuse or safe discharge (Table 3). Non-compliance for RO could be attributed to the poor rejection of small and

**Table 3** ISO 30500 (2018) membrane (mean) filtrate compliance (✓) and failure (✗) from concentrated blackwater

	RO (PA-UREA)		PV (PVA)	PV (PDMS)	MD (PP)
	20 °C	50 °C	50 °C	50 °C	50 °C
COD Category A (≤50 mg L <sup>-1</sup> )	✗ (202)	✗ (159)	✗ (134)	✗ (231)	✗ (149)
COD Category B (≤150 mg L <sup>-1</sup> )	✗ (202)	✗ (159)	✓ (134)	✗ (231)	✓ (149)
TP (≤2 mg L <sup>-1</sup> )	✓ (0.9)	✗ (4)	✓ (ND)	✓ (0.05)	✓ (1.19)
TP (≥80 % reduction)	✓ (99)	✓ (98)	✓ (>99)	✓ (>99)	✓ (>99)
NH <sub>4</sub> <sup>+</sup> -N (TN ≤ 15 mg L <sup>-1</sup> )	✗ (462)	✗ (382)	✗ (36)	✗ (854)	✗ (2375)
NH <sub>4</sub> <sup>+</sup> -N (TN ≥ 70 % reduction)	✓ (74)	✓ (78)	✓ (87)	✗ (-162)	✗ (-736)
CFU mL <sup>-1</sup> (≤0.1 CFU mL <sup>-1</sup> )	✓ (ND)	✓ (ND)	✓ (ND)	✓ (ND)	✓ (ND)
pH (6–9)	✗ (9.5)	✗ (10.1)	✗ (9.3)	✗ (9.86)	✗ (10.5)

RO (reverse osmosis); PA-UREA (polyamide-urea); PV (pervaporation); PVA (polyvinyl alcohol); PDMS (polydimethylsiloxane); MD (membrane distillation); PP (polypropylene); COD (chemical oxygen demand); TP (total phosphorus); NH<sub>4</sub><sup>+</sup>-N (ammoniacal nitrogen); TN (total nitrogen) CFU (colony forming units). Shaded rows represent urban wastewater discharge limits according to the European Commission (91/271/EEC) as a reference.



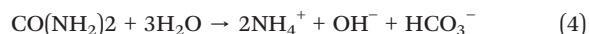


uncharged molecules such as urea (solute radius: 0.18 nm urea compared to 0.26 nm of water), where removal efficiencies have been reported to be as low as 22%,<sup>43</sup> and the selective enrichment of the volatile organic compounds for hydrophobic PV.<sup>21</sup>

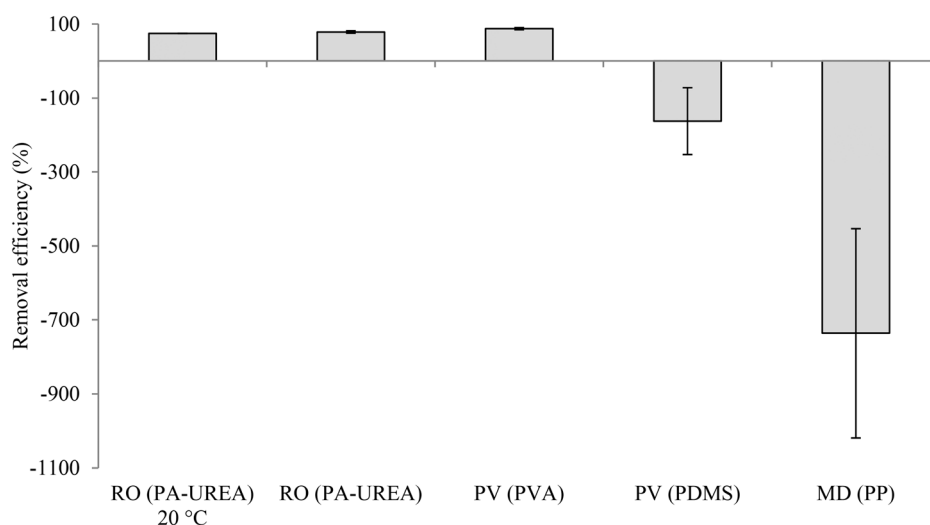
ISO 30500 compliance was met by all membranes for the non-volatile contaminants. Rejections of >99% for TP were encountered for all processes other than RO operated at 50 °C where rejection was reduced to 97% (Table 2). The high RO rejection in this study with CB was similarly observed by Davey *et al.*<sup>17</sup> who also observed >99% rejection for TP after UF-RO, demonstrating the robustness of RO when directly fed with CB. This consistently high rejection could be attributed to the fact that the majority of the TP content is present as the phosphate ion which is preferentially rejected due to electrostatic interaction.<sup>44,45</sup> Furthermore, TP is predominantly non-volatile and therefore its transport was restricted for all thermally driven processes. The TP limit of 2 mg L<sup>-1</sup> set out in the EU directive for urban waste-water treatment (91/271/EEC)<sup>46</sup> was met by RO at 20 °C and all thermal processes (Table 3). Decreased rejection of solutes by RO at elevated temperatures can be ascribed to changes in the polymer structure, combined with reduced solvent viscosity, and increased solute diffusivity.<sup>47</sup> However, the less stringent ISO 30500 removal efficiency target could also be achieved by RO at 50 °C (Table 3). The permeate of all membranes also presented no coliforms above the detection limit when challenged with concentrated blackwater *E. coli* concentrations up to 10<sup>7</sup> CFU mL<sup>-1</sup> (Fig. 3), which achieved the ISO 30500 specifications (≤0.1 CFU mL<sup>-1</sup>, Table 3). The LRVs obtained were influenced by feed concentration and available permeate volume which determined detection limits. Reverse osmosis is dense and provides an absolute barrier to pathogens, hence its wide scale adoption for

potable reuse from wastewater.<sup>48</sup> Similarly, PV and MD (despite being microporous) also act as absolute barriers since the separation constitutes both size exclusion and selectivity toward volatile constituents.

The volatile ISO 30500 contaminant, ammonia, challenged the selectivity of the membrane processes which demonstrated NH<sub>4</sub><sup>+</sup>-N separations of 74 ± 0.1, 78 ± 3, 87 ± 3, -162 ± 91 and -736 ± 283% for RO (20 °C), RO (50 °C), PV (PVA), PV (PDMS) and MD respectively. This is because ammonium rejection was impacted by urea hydrolysis, a naturally occurring process during urine storage which converts urea into ammonium and increases pH:<sup>49,50</sup>

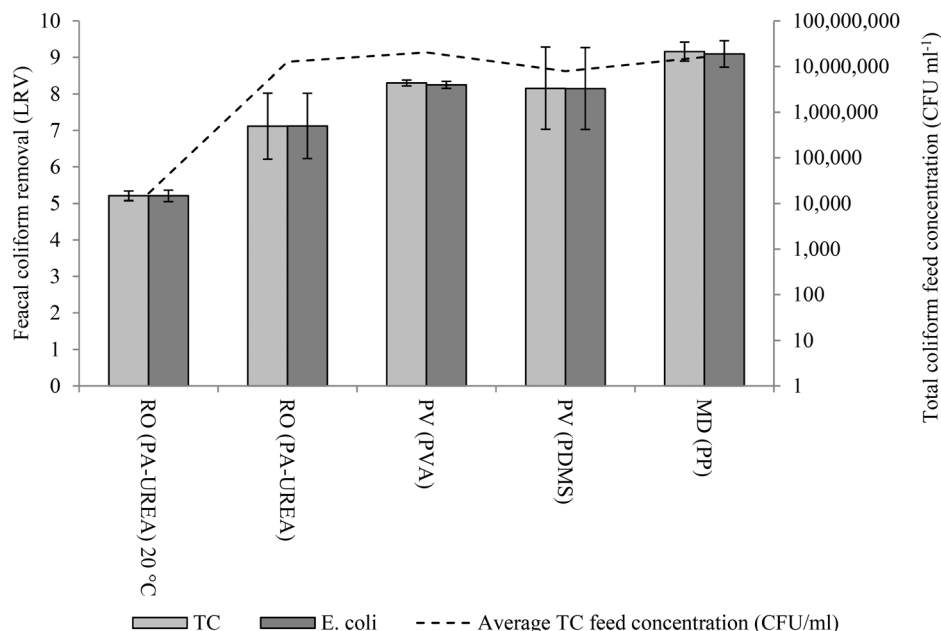


Initial pH values (before faecal contamination) were below pH 7 and increased to a mean of pH 8.64 ± 0.61 within two hours of faecal contamination (Table S2 and Fig. S2†). As the pK<sub>a</sub> for ammonium is 8.54 at 50 °C, the equilibrium is shifted towards ammonia, a non-ionic, polar and volatile compound which negatively impacts the respective separation mechanisms of RO, PV and hydrophobic thermally driven membrane processes. As a result, reported rejections for these processes were not comparable to literature values in which ammonium was maintained in the ionic form by pH adjustment (Fig. 2). For RO, electrostatic interaction is the principal mechanism for the removal of nitrogenous compounds, compared to molecular weight and chemical structure.<sup>44</sup> To illustrate, at a similar ammonium feed concentration (1300 mg L<sup>-1</sup>) maintained at pH 6, Bódalo *et al.*<sup>51</sup> reported 98.9% NH<sub>4</sub><sup>+</sup>-N removal, whereas in this study the final permeate pH was 9.5–10.1 with a ~20% lower removal efficiency. Ray *et al.*<sup>30</sup> demonstrated that once ammonium is fully hydrolysed to unionised ammonia at pH



**Fig. 2** Removal efficiencies of NH<sub>4</sub><sup>+</sup>-N on treating concentrated blackwater with membranes. Feed water temperature 50 °C, unless stated otherwise. Average feed concentration of NH<sub>4</sub><sup>+</sup>-N is 998.24 mg L<sup>-1</sup>. RO (reverse osmosis); PA-UREA (polyamide-urea); PV (pervaporation); PVA (polyvinyl alcohol); PDMS (polydimethylsiloxane); MD (membrane distillation); PP (polypropylene). Error bars represent standard deviation of a triplicated experiment.





**Fig. 3** Coliform log removal values (LRV) for *E. coli* and total coliforms (TC) using RO (PA-UREA), PV (PVA), PV (PDMS) and MD (PP) processes to treat concentrated blackwater. Feed water temperature 50 °C, unless stated otherwise. Average feed *E. coli* and total coliforms are 7.63 and 8.11 CFU mL<sup>-1</sup> respectively. RO (reverse osmosis); PA-UREA (polyamide-urea); PV (pervaporation); PVA (polyvinyl alcohol); PDMS (polydimethylsiloxane); MD (membrane distillation); PP (polypropylene). Error bars represent standard deviation of a triplicated experiment.

11.5, rejection reduces to 36%, which highlights the inhibiting impact of retention time. Ammonia is highly volatile (vapour pressure 7500 mmHg compared to 24 mmHg of water at 25 °C),<sup>52</sup> and therefore the reduced permeate vapour pressures coupled with high feed temperatures used for thermally driven processes encourage its transport over water.<sup>49,53,54</sup> The enhanced ammonia separation garnered with hydrophilic PV (PVA) can be accounted for by the material selectivity of PVA for water over ammonia due its higher polarity (electronegativity difference:  $\Delta E_{N-O-H} = 1.24$ ,  $\Delta E_{N-H} = 0.84$ ). Hydrophobic PV (PDMS) and MD exhibited NH<sub>4</sub><sup>+</sup>-N permeate production, due to the continuous urea hydrolysis during the experimental period. The moderate permeability of ammonia to water for PV(PDMS) is attributed to its lower polarity.<sup>55</sup> However, membrane interaction by size exclusion also plays a role in the control of ammonia (molecular size: ammonia 0.326 nm, water 0.26 nm)<sup>56</sup> when compared to MD where ammonia can freely diffuse through vapour filled micropores. As a result, PV (PDMS) and MD have been applied for permeate side ammonia recovery for high pH feed waters.<sup>54,55</sup> A negative ammonium rejection was similarly observed by Davey *et al.*<sup>18</sup> treating CB using direct contact membrane distillation operated at 30 °C at pH 8.5. The final permeates were at pH 9.9 and 10.5 for PV(PDMS) and MD respectively, illustrating the extent of urea hydrolysis which took place in this study. The permeate ammonia concentration correlated to an increase in permeate conductivity (Table 2) which resulted in poor salt rejection in the permeate at  $76.5 \pm 4.8$  (PV, PDMS) and  $60.6 \pm 8.1$  (MD), as similarly observed by Zhao *et al.*<sup>49</sup> who reported urea hydrolysis. In this study, the ISO 30500 standard of 70%

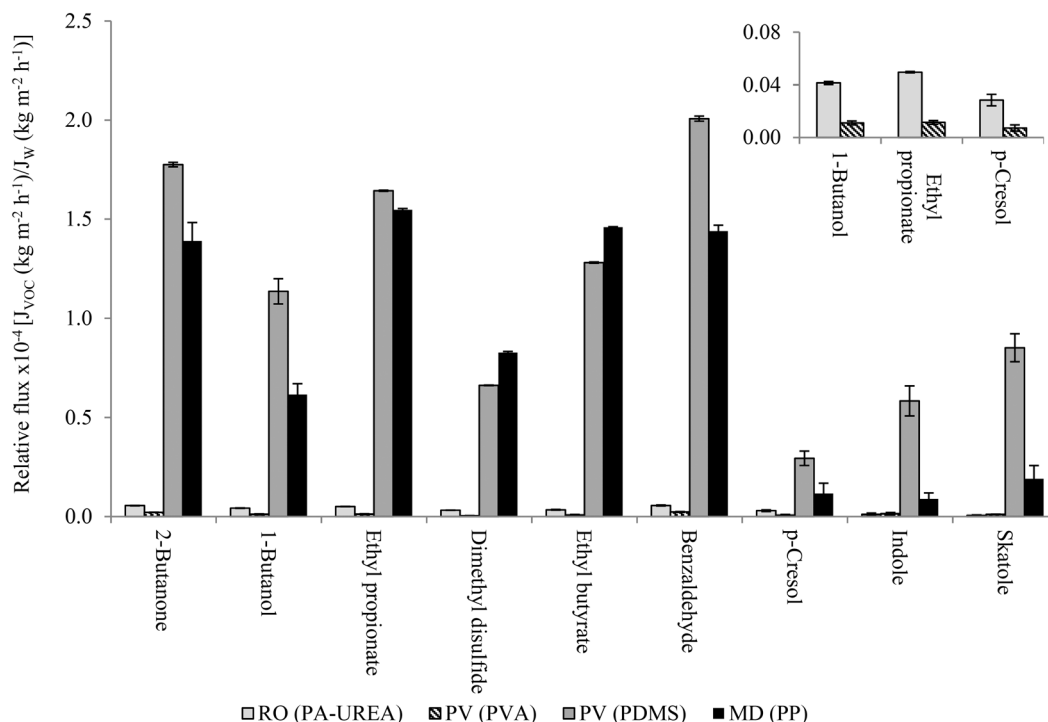
removal of NH<sub>4</sub><sup>+</sup>-N was achieved by the dense hydrophilic membranes RO and PV(PVA), with the final permeate concentration >10 times lower for PV(PVA) than RO (Table 3). The permeate pH produced from each membrane was alkaline, exceeding the pH range (pH 6–9) specified in the ISO 30500 standard (Table 3), which can be attributed to the basicity of low MW carried over (including NH<sub>4</sub><sup>+</sup>-N), the removal of which with a secondary unit process subsequently adjusting pH to within the relevant range without the requirement for acid dosing (Table S3).†

### 3.2 T&O is influenced by membrane selectivity, microbial activity and detection thresholds

The selectivity of taste and odour compounds was assessed with synthetic and real CB using a discrete range of VOCs including the key malodorous faecal compounds: indole, skatole, *p*-cresol and dimethyl disulfide; encompassing seven functional groups and a wide range of physico-chemical properties (Table S1†).

The relative VOC to water flux for all membrane processes was first benchmarked using a synthetic VOC solution at 10 ppm (Fig. 4). Reverse osmosis demonstrated a VOC removal of 20–85% with selectivity (Fig. 5) strongly correlated to molecular weight ( $r = -0.85$ ,  $n = 9$ ,  $p = 0.003$ ) and octanol water coefficient ( $r = -0.911$ ,  $n = 9$ ,  $p = 0.001$ ) as reported by Altayan *et al.*<sup>57</sup> Rejection for RO was lower than PV(PVA) which removed VOCs by 65–80%. While transport phenomena are described by a solution-diffusion mechanism for both membranes, which comprise of glassy hydrophilic polymer characteristics, the difference can be ascribed to





**Fig. 4** A comparison of membrane odour separation (RO, PV and MD) expressed as the ratio of volatile organic compound (VOC) flux to water flux. Synthetic solution feed concentration  $10\ mg\ L^{-1}$ . RO (reverse osmosis); PA-UREA (polyamide-urea); PV (pervaporation); PVA (polyvinyl alcohol); PDMS (polydimethylsiloxane); MD (membrane distillation); PP (polypropylene). All membranes operated at  $50\ ^\circ C$ . Error bars represent standard deviation of a triplicated experiment.

differences in the driving force applied, and distinctions in affinity of the polymeric substrate. For RO, pressure induces an osmotic gradient and the subsequent concentration polarisation causes solute flux to be higher than the water flux, thereby reducing selectivity. Concentration polarisation is similarly considered in pervaporation, but the role of the vapour pressure gradient is distinctive and thus selectivity is less sensitive to rejection. In contrast, the hydrophobic polydimethylsiloxane membrane concentrated all VOCs due to the material's selective preference to non-polar compounds.<sup>25</sup> With the suite of compounds used in this study encompassing a large range of physico-chemical properties, selectivity in the PDMS was governed by vapour pressure, volatility and hydrophobicity as reported by Mercer *et al.*<sup>21</sup> Membrane distillation illustrated a similar profile to PV(PDMS), however separation had a general dependency on boiling point ( $r = -0.862$ ,  $n = 9$ ,  $p = 0.003$ ) or volatility ( $r = -0.701$ ,  $n = 9$ ,  $p = 0.036$ ) due to the fact that the VOC transport is reliant on the vapour pressure gradient, rather than material interactions specifically.<sup>25</sup>

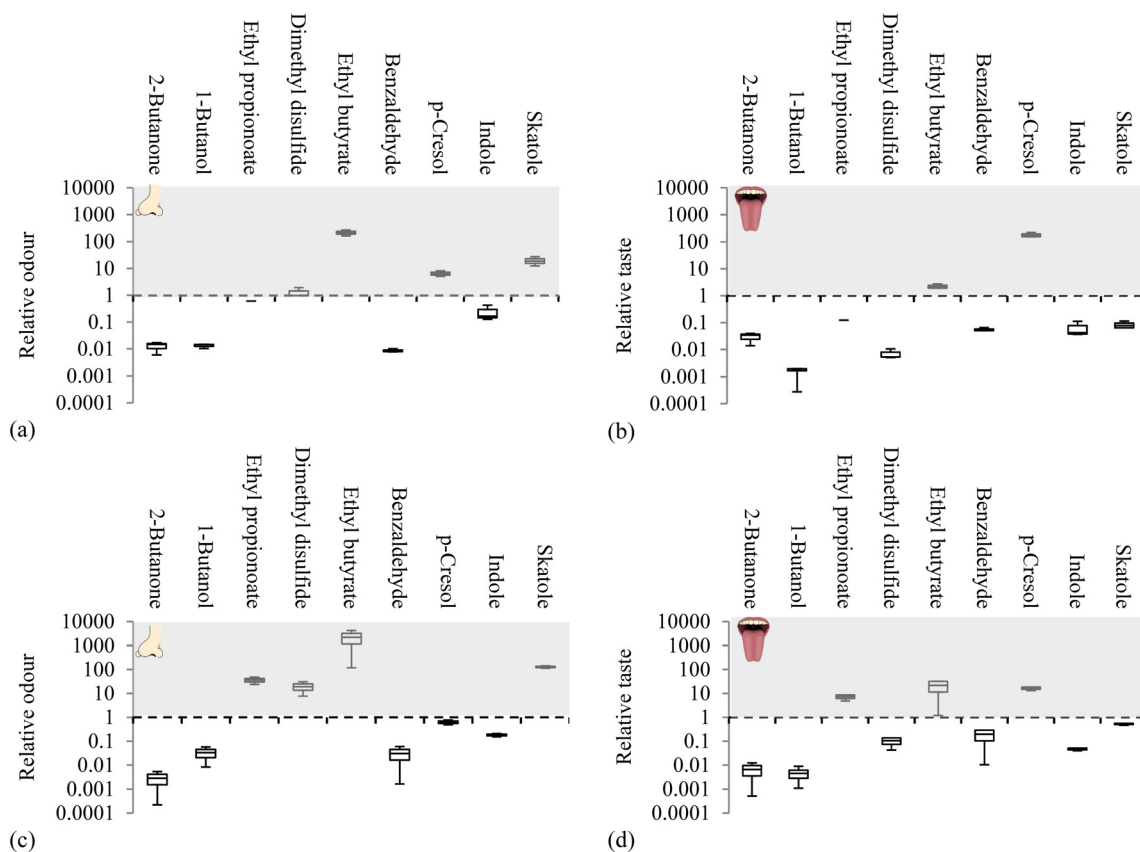
When assessing selectivity with a real CB matrix, it was evident that VOCs were being produced during the experiment due to feed side microbial activity.<sup>39</sup> For example, in the PV(PVA) evaluation, after almost 5 hours, the malodorous faecal compound skatole increased by 11-fold within the feed (Fig. S1†). Coupled with the extremely low odour threshold of skatole in water ( $0.0002\ mg\ kg^{-1}$ ),<sup>58</sup> its

odour would be detected in the permeates of RO and PV(PVA) (Fig. 5). However the taste threshold is higher ( $0.05\ mg\ kg^{-1}$ )<sup>58</sup> and therefore skatole would not be tasted using RO and PV(PVA) (Fig. 5). The breaches in T&O are therefore dependent on the respective thresholds of the compounds as also observed for ethyl butyrate, ethyl propionate, *p*-cresol and dimethyl disulfide which can be smelt or tasted at concentrations at least three orders of magnitude lower than the other compounds examined.<sup>58</sup>

Using the method of wastewater odour wheel classification,<sup>59,60</sup> odour was qualitatively characterised for the thermally driven processes which selectively enriched or rejected VOCs. Three different odour outcomes were encountered, with little resemblance of concentrated blackwater due to alterations in the respective odour profiles (Table 4). Hydrophobic pervaporation (PDMS) produced the most hedonistically pleasant odour which was characteristic of a cleaning product due to the selective enrichment of alcohols, aldehydes, esters and ketones over the key malodorous 'faecal like' odours of indole, skatole and *p*-cresol, from having a higher vapour pressure of over an order of magnitude (Table S1†).<sup>21</sup> The reduction of indole and skatole within the odour profile changed their overpowering faecal notes to floral notes, and as such these compounds are key constituents of jasmine perfume.<sup>22</sup> The permeate could be associated with 'sanitised' water and therefore widely accepted. Hydrophilic (PVA) pervaporation, which reduced the VOC concentration but did not change the







**Fig. 5** Concentrated blackwater relative odour and taste of reverse osmosis permeate (a and b) and hydrophilic pervaporation permeate (c and d) according to the lowest reported odour and taste thresholds in water.<sup>58</sup> Note: dotted line represents the threshold in which the compound can be detected with grey above white below the threshold respectively.

T&O profile, represented a chemical odour with similarities to body odour, unpleasant however not repulsive. Membrane distillation permeate was overpowered by ammonia causing a repulsive odour, even with a VOC profile similar to PDMS (Fig. 4). Distinctive T&O profiles are therefore facilitated by thermal and pressure driven processes, and can be strongly modified with polymeric solution-diffusion interactions. Importantly, this study has demonstrated how membranes can mitigate T&O detection for water reuse by reduction in concentration, and how material characterisation alter perception as practiced in the perfume industry.<sup>22</sup> Due to the concentration of CB, it may still be necessary for complementary technology for T&O polishing to increase

willingness to adopt water reuse from this specific water source.

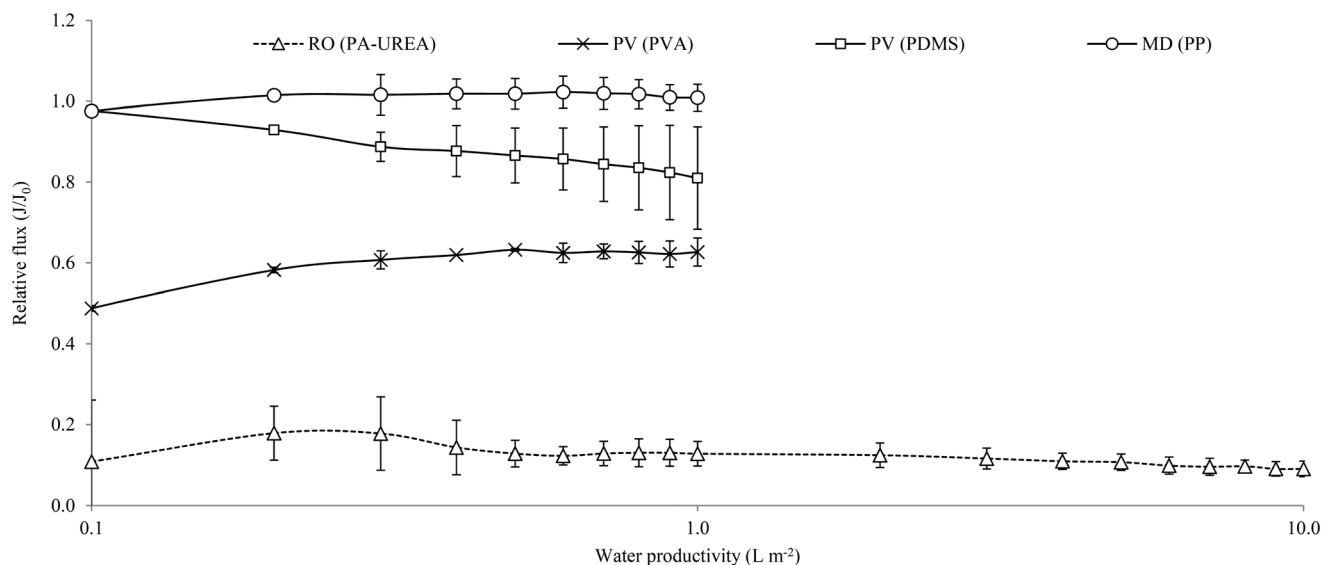
### 3.3 Water productivity may be more restricted from pressure driven membrane processes

Preliminary analysis of water flux from each membrane was undertaken to provide an initial indication of how fouling mechanisms may differ based on driving force (thermal or pressure driven) and material characteristics when treating this challenging CB feed (Fig. 6). From an initial flux of 50 L m<sup>-2</sup> h<sup>-1</sup>, a flux decline (80%) was demonstrated for the pressure driven RO, which we suggest is due to the convective mass transport of organics toward the membrane surface during filtration. Organic fouling can be mitigated through the use of a UF pretreatment which has shown to enhance the resilience of both RO and MD configurations,<sup>8,19</sup> with sustainable UF operation evidenced for CB through hydrodynamic optimisation. Kamranvand *et al.*<sup>19</sup> observed how UF retained the high molecular weight (MW) fraction of CB comprising colloidal organics greater than 500 kDa which represents 30% of organics in CB. The initial increase in flux for the PVA membrane can be attributed to polymer swelling which raises the internal polymer volume, subsequently increasing flux. A more stable flux (relative flux of 0.6) was

**Table 4** Odour descriptors for permeate produced from pervaporation and membrane distillation membranes following treatment of concentrated blackwater

Membrane process (material)		Permeate odour descriptor
PV (PVA)		Sweaty, chemical, sweet, onion
PV (PDMS)		Sweet, chemical, earthy, floral
MD (PP)		Pungent, ammonia, fishy, citrus
PV (pervaporation); (polydimethylsiloxane); MD (polypropylene).	PVA (polyvinyl alcohol); MD (membrane distillation); PP (polypropylene).	





**Fig. 6** Normalised membrane flux with deionised water ( $J_0$ ) and concentrated blackwater ( $J$ ). Water productivities for RO standardised to  $10 \text{ L m}^{-2}$  membrane area and PV and MD standardised to  $1 \text{ L m}^{-2}$  membrane area. RO (reverse osmosis); PA-UREA (polyamide-urea); PV (pervaporation); PVA (polyvinyl alcohol); PDMS (polydimethylsiloxane); MD (membrane distillation); PP (polypropylene). All membranes were operated at  $50^\circ\text{C}$ . Average initial total suspended solids concentration  $6 \text{ g L}^{-1}$ . Error bars represent standard deviation of a triplicated experiment.

observed for the hydrophilic PV(PVA) membrane. We propose this to be analogous to forward osmosis, where the limited applied convective force mitigates fouling.<sup>61</sup> The hydrophobic microporous and dense thermally driven membranes facilitated similar recoveries to RO but only negligible permeability loss was identified. This can be ascribed to the material's resistance to mass transfer (as evidenced by the low water vapour flux of  $0.06$  and  $0.08 \text{ L m}^{-2} \text{ h}^{-1}$  for PDMS and MD respectively, Table S4†), which was in excess of the resistance of heat and mass transfer provided by the fouling layer. Consequently, fouling did not inhibit water vapour mass transport as the rate of separation was determined by the material permeability. Fouling was visually evidenced as clogging within the interstitial spacing of the hollow fibre configuration (HF) by coarse faecal solids. This could be resolved using UF for pretreatment and further optimisation of thermal membrane module geometry for wastewater applications.

## 4. Conclusions

The study assessed pressure and thermally driven membranes for their potential to realise water reuse as a household scale solution for sanitation:

- Pressure driven and hydrophobic thermally driven membranes mostly achieved category B of the ISO 30500 standards within a single stage. The critical weakness in selectivity lies with volatile contaminants such as ammonia and organics (residual COD as VOCs). Thermally driven hydrophilic PV complied closely to the category A ISO 30500 standards and demonstrated the highest rejection of VOCs by providing the greatest selectivity for water over all the

membrane processes, due to its affinity to water based on polarity.

- Improved ammonia separation could be practically implemented by limiting faecal bacterial enzymic activity to mitigate urea hydrolysis through reducing faecal contamination, shortening residence time and increasing operating temperature. Such modifications will also assist in the mitigation of feed side bacterial VOC production.

- As direct reuse from blackwater is considered high risk, ISO 30500 (ref. 16) advises that category A effluent should be reused for flushing or toilet cleaning purposes which saves up to 25% of household water.<sup>62</sup> However, this risk can be reduced if a multibarrier approach is implemented and the effluent is frequently monitored (*i.e.* through sudden changes in electrical conductivity as an indicator), enabling a further saving of 51% of household water if extended to laundry and all other non-food related reuse applications.<sup>62</sup>

Thermally driven PV membranes may offer a more robust solution to water reclamation by offering greater mitigation of volatile components ( $\text{NH}_3$ , VOCs) than RO, a more robust barrier to pathogens than MD, and less sensitivity to fouling due to the limited convection experienced in permeation. By complementing PV with UF pretreatment, as would be standard for the household application of RO, a lower organic load to PV could improve resilience and achieve category A water reuse compliance. Importantly, the use of thermal membrane separation can broaden the applicability and affordability of high-performance off-grid sanitation through exploiting widely accessible thermal energy. Increased crosslinking of PVA, or use of ceramic zeolites may further enhance robustness in the long-term by reducing swelling which can further enhance selectivity above that illustrated within the present study. Common to both



technologies is the challenge of concentrate production, which must be managed locally either through collection or treatment, but also offers significant opportunities for localised resource recovery enabling synergistic valuation to maintenance business models.

## Abbreviations

CB	Concentrated blackwater
CFU	Coliform forming units
COD	Chemical oxygen demand
HF	Hollow fibre
ISO	International Standards Organisation
LD	Limit of detection
LIC	Low-income country
LRV	Log removal value
MD	Membrane distillation
MW	Molecular weight
NH <sub>3</sub>	Ammonia
NH <sub>4</sub> <sup>+</sup>	Ammonium
NH <sub>4</sub> -N	Ammoniacal nitrogen
NSS	Non-sewered sanitation
PA-UREA	Polyamide-urea
PDMS	Polydimethylsiloxane
PES	Polyethersulphone
PP	Polypropylene
PV	Pervaporation
PVA	Polyvinyl alcohol
RO	Reverse osmosis
SDG	Sustainable Development Goal
T&O	Taste and odour
TP	Total phosphorus
TSS	Total suspended solids
UF	Ultrafiltration
VOC	Volatile organic compound

## Data availability

All relevant data are available from an online repository or repositories. Data underlying this paper can be accessed from: <https://dspace.lib.cranfield.ac.uk/handle/1826/20712>.

## Conflicts of interest

There are no conflicts to declare.

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