

MANUSCRIPT WATER RESEARCH

Title: Membrane stripping enables effective electrochemical ammonia recovery from urine while retaining microorganisms and micropollutants

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25 **ABSTRACT**

26 Ammonia recovery from urine avoids the need for nitrogen removal through
27 nitrification/denitrification and re-synthesis of ammonia (NH₃) *via* the Haber-Bosch
28 process. Previously, we coupled an alkalifying electrochemical cell to a stripping
29 column, and achieved competitive nitrogen removal and energy efficiencies using
30 only electricity as input, compared to other technologies such as conventional column
31 stripping with air. Direct liquid-liquid extraction with a hydrophobic gas membrane
32 could be an alternative to increase nitrogen recovery from urine into the absorbent
33 while minimizing energy requirements, as well as ensuring microbial and
34 micropollutant retention. Here we compared a column with a membrane stripping
35 reactor, each coupled to an electrochemical cell, fed with source-separated urine and
36 operated at 20 A m⁻². Both systems achieved similar nitrogen removal rates, 0.34 ±
37 0.21 and 0.35 ± 0.08 mol N L⁻¹ d⁻¹, and removal efficiencies, 45.1 ± 18.4 and 49.0 ±
38 9.3%, for the column and membrane reactor, respectively. The membrane reactor
39 improved nitrogen recovery to 0.27 ± 0.09 mol N L⁻¹ d⁻¹ (38.7 ± 13.5%) while lowering
40 the operational (electrochemical and pumping) energy to 6.5 kWh_e kg N⁻¹ recovered,
41 compared to the column reactor, which reached 0.15 ± 0.06 mol N L⁻¹ d⁻¹ (17.2 ±
42 8.1%) at 13.8 kWh_e kg N⁻¹.

43 Increased cell concentrations of an autofluorescent *E. coli* MG1655+prpsM spiked in
44 the urine influent were observed in the absorbent of the column stripping reactor after
45 24 h, but not for the membrane stripping reactor. None of six selected micropollutants
46 spiked in the urine were found in the absorbent of both technologies.

47 Overall, the membrane stripping reactor is preferred as it improved nitrogen recovery
48 with less energy input and generated an *E. coli*- and micropollutant-free product for

49 potential safe reuse. Nitrogen removal rate and efficiency can be further optimized by
50 increasing the NH_3 vapor pressure gradient and/or membrane surface area.

51

52 **KEYWORDS**

53 membrane; micropollutant; nutrient recovery; pathogen; stripping; urine

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55

56 1. INTRODUCTION

57 Nitrogen recovery from high-strength wastewater such as source-separated urine,
58 short-cuts the conventional removal *via* nitrification/denitrification and fixation *via* the
59 Haber-Bosch process. Multiple recovery technologies exist, ranging from adsorption
60 of NH_4^+ with zeolites or activated carbon (Ganrot et al. 2007, Tarpeh et al. 2017), to
61 precipitation as struvite for instance (Gagnon 2016, Ueno and Fujii 2001, Zamora et
62 al. 2017). Struvite precipitation is available on pilot scale (Desmidt et al. 2015), but
63 the lack of legislation (De Vrieze et al. 2016), social acceptance, and economic self-
64 sustainability (Etter et al. 2011) limits its application when recovered from human
65 waste sources.

66 Most recovery technologies are based on stripping *via* volatilization of ammonia
67 (NH_3) from the liquid waste stream through an increase in pH and/or temperature
68 (Arredondo et al. 2017, Böhler et al. 2015, Christiaens et al. 2017, Vanotti et al. 2017,
69 Xu et al. 2017)(SI Table A.1). In general, the higher the NH_3 concentration, pH, and
70 temperature, the more efficient the recovery technology will be. Column stripping is
71 an established process, even on full scale, requiring caustic, heating, and forced air
72 movement. The process has been developed for digester supernatant (Böhler et al.
73 2015) and urine (Antonini et al. 2011). A more recent stripping technology is
74 membrane distillation, requiring caustic and heat to create a vapor pressure gradient
75 across a hydrophobic membrane with gas or vacuum filled pores, separating the NH_3
76 liquid from the sweep gas, the applied vacuum (El-Bourawi et al. 2007), or the acid
77 (Ahn et al. 2011, Böhler et al. 2015, Lauterbock et al. 2012, Zarebska et al. 2014).
78 Membrane stripping, also called transmembrane chemisorption, or membrane
79 contactor, works similarly but only with caustic and no heat. An acid on the other side

80 of the membrane drives the NH_3 extraction by lowering the NH_3 concentration
81 (Amaral et al. 2016, Bernal et al. 2016, Dube et al. 2016, Ulbricht et al. 2013, Vanotti
82 et al. 2017). Hollow fiber configurations are commonly used because a large
83 membrane surface area improves ammonia extraction rates and efficiencies
84 (Darestani et al. 2017). A combination of (microbial) electrochemical or fuel cells with
85 column or membrane stripping units has the advantage to produce caustic *in situ*,
86 eliminating the need for chemicals, which are a concern, not only for their costs, but
87 also in terms of a safe and reliable supply (Arredondo et al. 2017, Christiaens et al.
88 2017).

89

90 In all these technologies, NH_3 is often recovered in strong acidic absorbents (e.g.
91 H_2SO_4 or HNO_3)(Arredondo et al. 2017, Bernal et al. 2016, Böhler et al. 2015,
92 Christiaens et al. 2017, Vanotti et al. 2017, Xu et al. 2017). This increases the pH
93 gradient and thus vapor pressure gradient between the alkaline NH_3 solution and the
94 absorbent. The products $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 can be applied as a fertilizer, since
95 the recovery process concentrates the nitrogen resulting in equal concentrations as
96 synthetic fertilizer. Recovery as NH_4HCO_3 precipitate is possible in a 3-compartment
97 microbial electrochemical cell in which the HCO_3^- is also extracted from urine
98 (Ledezma et al. 2017). If NH_3 is not recovered in acid but in a microbial growth
99 medium, this allows the recovered NH_3 to be used as a nitrogen source for microbial
100 conversions such as protein production. The protein can be used as a feed or even
101 food additive (Christiaens et al. 2017, Matassa 2016).

102

103 In any application, the quality of the recovered product needs to be safeguarded.
104 Human urine contains microbes, viruses (Bischel et al. 2015, Decrey and Kohn

105 2017), and antibiotic resistance genes (Bischel et al. 2015, Pruden 2014). These
106 mainly originate from fecal cross-contamination (Udert et al. 2015), and potentially
107 from the bladder (Anderson et al. 2004, Sianou et al. 2016, Wolfe and Brubaker
108 2015, 2016). The most abundant genera, covering 83.5% of classifiable sequences
109 found in male urine, had cell sizes with the smallest dimension ranging from 0.65-0.8
110 μm (*Lactobacillus* (Valik et al. 2008), *Veillonella* (Kraatz and Taras 2008),
111 *Streptococcus*, and *Enterococcus* (Kokkinosa et al. 1998)) down to 0.1 μm
112 (*Ureaplasma* (Shepard et al. 1974) and *Mycoplasma* (Waites and Talkington
113 2004))(Dong et al. 2011).

114 Micropollutants found in urine were mainly (metabolites of) pharmaceuticals (Bischel
115 et al. 2015, Jaatinen et al. 2016, Kovalova et al. 2012), but also alternative
116 plasticizers (Alves et al. 2017). Some of these micropollutants and metabolites (e.g.
117 hydrochlorothiazide, sulfamethoxazole) are mainly hydrophilic at pH 9, that of
118 hydrolysed urine, while others (e.g. irbesartan, clarithromycin, carbamazepine,
119 diclofenac) are rather hydrophobic, based on the octanol-water partition ratio (log
120 D_{ow})(<https://chemicalize.com>).

121

122 In our previous study, a strip and absorption column were coupled to the alkalifying
123 cathode compartment of an electrochemical cell for NH_3 removal and recovery
124 (Christiaens et al. 2017). Whereas the nitrogen removal percentage for real urine
125 was $87 \pm 6\%$ at 20 A m^{-2} with $3\text{M H}_2\text{SO}_4$ as absorbent, recovery was only $25 \pm 12\%$,
126 most likely due to nitrogen loss in the condensates throughout the lab set-up. In the
127 present study, membrane stripping was chosen as an alternative technology for the
128 strip and absorption column, since direct urine-absorbent contact eliminates the gas
129 phase compartment and prevents the potential adsorption and loss of NH_3 in

130 condensates, as already shown by Arredondo et al. (2017). Reactor set-ups with
131 column and membrane stripping were compared in terms of nitrogen removal,
132 recovery, and energy input. This study presents the first continuous test for
133 membrane stripping of NH_3 .

134 In addition to nitrogen transfer, the quality of the recovered product with respect to
135 microorganisms and micropollutants originating from urine, was quantified. Three
136 hypotheses were studied. First, the introduction of a membrane as a physical barrier
137 between urine and the absorbent, retains microorganisms based on their size. In
138 column stripping, microorganisms could move with the gas flow. Second, hydrophilic
139 micropollutants are more easily transferred to the absorbent *via* water droplets
140 moved with the gas flow, while hydrophobic compounds are not. Third, a hydrophobic
141 membrane prevents transfer of any micropollutant due to the gas filled pores.

142 2. MATERIALS AND METHODS

143 2.1 Urine

144 Hydrolysed urine originated from a central storage and hydrolysis tank that collects
145 men's undiluted urine *via* NoMix toilets and urinals at Eawag (Dübendorf,
146 Switzerland). The average hydraulic retention time (HRT) in the tank was about 14
147 days at room temperature. Female urine collected (33% diluted with flush water)
148 made up a limited part of the men's urine used in the present study, as a second
149 storage tank for female urine had an overflow in the men's storage tank. One batch
150 of urine was used for per test (Table 1).

151

152 **Table 1** Average (\pm standard deviation, SD) composition of hydrolysed urine. (n \geq 26)

pH	(-)	9.3 \pm 0.1
electrical conductivity	(mS cm ⁻¹)	28 \pm 1
Na ⁺	(mg L ⁻¹)	2041 \pm 536
Total Ammonium Nitrogen	(mg N L ⁻¹)	4622 \pm 719
K ⁺	(mg L ⁻¹)	1551 \pm 205
Cl ⁻	(mg L ⁻¹)	2604 \pm 297
NO ₂ ⁻	(mg N L ⁻¹)	<LOQ ^a
NO ₃ ⁻	(mg N L ⁻¹)	<LOQ ^b
PO ₄ ³⁻	(mg L ⁻¹)	472 \pm 184
SO ₄ ²⁻	(mg L ⁻¹)	471 \pm 56

153 ^aLOQ NO₂⁻ = 0.15 mg N L⁻¹

154 ^bLOQ NO₃⁻ = 0.18 mg N L⁻¹

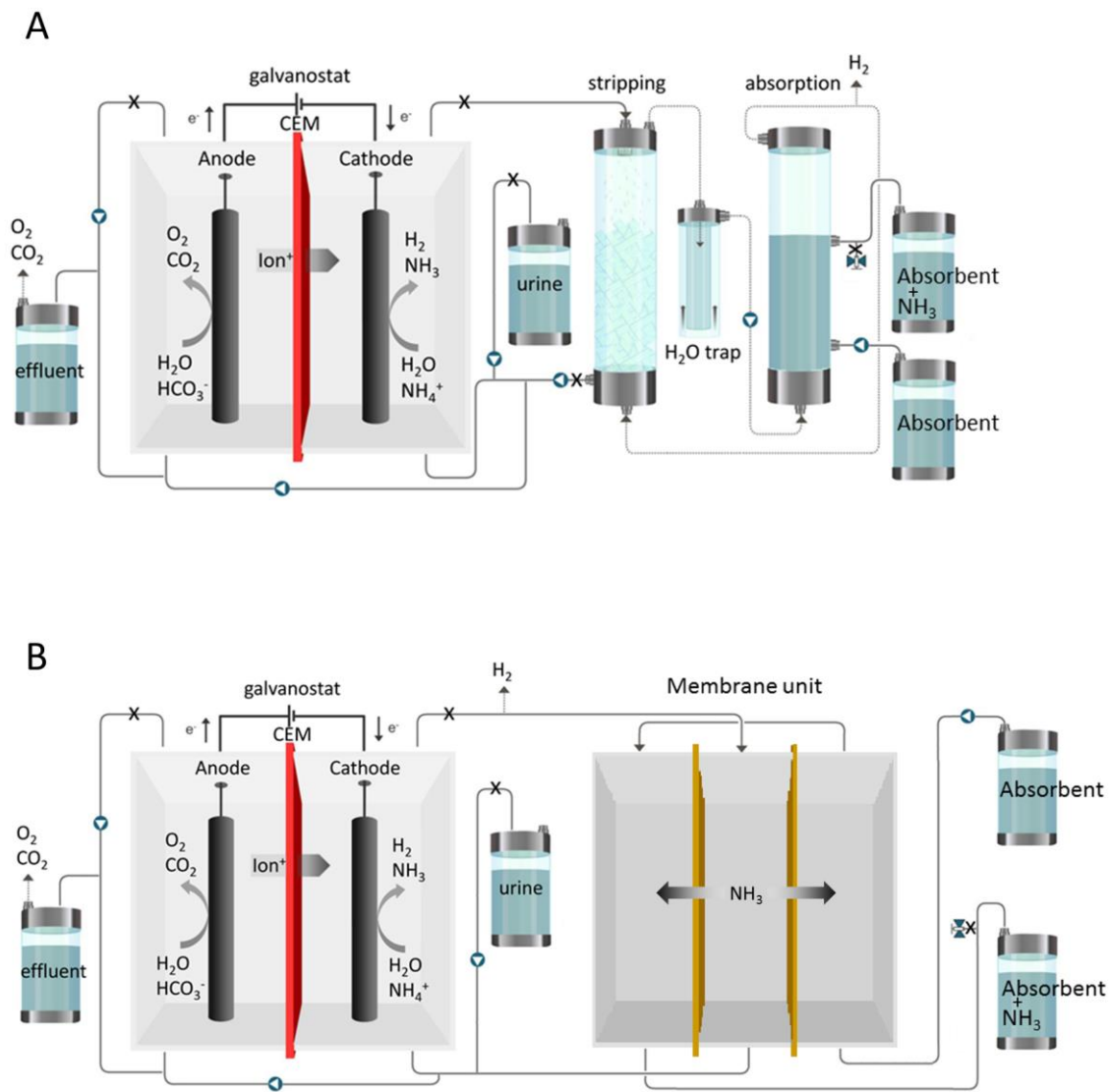
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157 2.2 Experimental set-up

158 Two technologies were compared for nitrogen removal and recovery, and bacterial or
159 micropollutant contamination of the recovered nitrogen product: column and

160 membrane stripping (Fig. 1). Column stripping was established *via* the same set-up
161 as previously described (Christiaens et al. 2017) except for the gas counters which
162 were replaced by a gastight acid trap, no gas bags on the feed vessel, different liquid
163 pumps (Ismatec, Cole-Parmer GmbH, Germany), and a bubble column with sintered
164 glass frit (40-250 μm pore size) as absorption column (6x110 cm^2 , filled with 19 cm
165 absorbent). A second set-up was built with a membrane stripping unit instead of the
166 stripping and absorption columns. Two flat-sheet PTFE membranes (active surface
167 area of 10.5x12 cm^2 ; 0.45, 0.2, or 0.1 μm pore size)(SI Table B.1) were mounted on
168 both sides of a plexiglass holder, creating a middle compartment for the urine
169 (10.5x12x0.65 cm^3). Bolting this unit with teflon rubbers and metal frames between
170 two acrylic glass plates created two absorbent chambers (10.5x12x0.25 cm^3).
171 As with the column stripping, the catholyte of the electrochemical cell was
172 recirculated over the middle compartment of the membrane unit to allow nitrogen
173 removal and recovery before being fed in the anode compartment for additional
174 nitrogen extraction. Analog manometers (WIKA, Switzerland) on the urine input and
175 output of the membrane unit could indicate pressure loss and thus wetting of the
176 membrane, which was not observed. Prior to entering the membrane unit, degassing
177 of the catholyte was allowed. The exhaust gas was bubbled through 2M H_2SO_4 to
178 determine potential nitrogen losses. Measurements indicated these were limited. The
179 absorbent was fed in one of the two absorbent chambers of the membrane unit and
180 was recirculated over the other chamber.



181

182

183 **Fig. 1** Column (A) and membrane (B) stripping reactors. Sampling ports are indicated

184 by X.

185

186 2.3 Reactor operation and product quality tests

187 Some applications of recovered nitrogen, such as microbial protein (MP) production,

188 require a pH-neutral absorbent to benefit from the nitrogen alkalinity and to avoid

189 additional chemicals for pH control (Christiaens et al. 2017). Ammonia absorption

190 with a pH-neutral absorbent can be improved by increasing the buffer capacity and

191 by using more absorbent *via* HRT reduction. The required phosphate buffer
192 concentration of a neutral absorbent was calculated based on the desired maximum
193 nitrogen recovery (SI section C. Absorbent composition) and set at 0.21 M (11.15 g
194 $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$ L^{-1} , 23.05 g $\text{Na}_2\text{HPO}_4 \cdot 2\text{H}_2\text{O}$ L^{-1}). To avoid ion diffusion over the
195 hydrophobic membrane, a sodium buffer was chosen to which 12.82 g KHCO_3 L^{-1}
196 was added to set both Na^+ and K^+ concentrations similar to urine. The absorbent
197 HRT (HRT_{abs}) was determined by 24 h total ammonia nitrogen (TAN) breakthrough
198 curves with continuous urine flow and an applied current density (j) of 20 A m^{-2} (Table
199 2 and 4). The absorbent volume was set by the membrane unit at 0.12 L and also
200 applied in the column stripping reactor. Liquid and gas recirculation over the
201 membrane and column units, respectively, ensured mixing (Table 4). Samples for pH
202 and cation analysis were collected over a 24 h time period.

203 Using the optimal HRT_{abs} for both technologies, their nitrogen removal and recovery
204 was compared in continuous tests with applied current densities of 0.1 (control) and
205 20 A m^{-2} . Three samples were taken at steady state (*i.e.*, after 4 HRTs) and analysed
206 for pH, EC, cations, and anions.

207 For bacterial and micropollutant detection in the absorbent, the reactors were
208 operated for 24 h with continuous urine flow, as has been done for the TAN
209 breakthrough curves. Potential bacterial transfer from urine to the absorbent product
210 was tested by spiking the urine with an autofluorescent *E. coli* MG1655+prpsM
211 containing a GFP plasmid that is constitutively expressed (Eawag, Dübendorf,
212 Switzerland). In preparation for the reactor tests, the strain was grown for 24 h at
213 37°C and 220 rpm in LB Lennox medium (10 g L^{-1} tryptone, 5 g L^{-1} yeast extract, 5 g
214 L^{-1} NaCl) with addition of $50 \mu\text{g mL}^{-1}$ kanamycin to retain the autofluorescent plasmid.
215 Pellets obtained after centrifugation (10 min, 2000 rpm) were resuspended in urine.

216 Kanamycin was added to both urine and absorbent before filling the reactors.
 217 Samples were collected over 24 h tests for bacterial cell counts (autofluorescent
 218 FITC and live/dead staining SGPI, see 2.4 Chemical and microbial analyses), pH,
 219 and cations.

220 To study micropollutant transfer in both technologies, six wastewater micropollutants
 221 (Table 3) were selected (personal communication of Isabell Köpping, Eawag) ranging
 222 from hydrophilic to hydrophobic at pH 9. Concentrations spiked in urine were taken
 223 from the calculated reference urine (personal communication of Isabell Köpping,
 224 Eawag). Samples were taken over 24 h for micropollutant quantification, pH, and
 225 cations.

226 All tests were conducted at room temperature ($20.8 \pm 0.6^\circ\text{C}$) in duplicate with
 227 continuous urine feeding. Thorough cleaning with 1% HOCl and water occurred in
 228 between all (replicate) tests. Electrode potentials versus a Ag/AgCl reference
 229 electrode (± 0.247 V vs standard hydrogen electrode, 3 M NaCl, Bio-Logic, France)
 230 and cell voltages were logged with a multimeter.

231

232 **Table 2** Overview of experiments, conducted in duplicate. Urine was always fed
 233 continuously.

Test	j (A m ⁻²)	Urine	Absorbent	Membrane pore size (μm)	Test duration (h)
TAN breakthrough curves	20	plain	Batch	0.1	24
Continuous TAN tests	0.1, 20	plain	Continuous	0.45	48
Pathogen tests	20	<i>E. coli</i> MG1655prpsM	Batch; kanamycin dosed	0.1	24
Micropollutant tests	20	6 micropollutants	Batch	0.2	24

234

235 **Table 3** Selected micropollutants present in wastewater ranging from strongly
 236 hydrophobic to hydrophilic at pH 9. The log D_{ow} represents the octanol-water

237 distribution ratio of ionized and neutral forms of a compound and indicates
 238 hydrophobicity.

Micropollutant	Medical use	Log D _{ow} at pH 9 ^a (-)	Aqueous solubility at pH 9 ^a (g L ⁻¹)	Concentration tested ^b (µg L ⁻¹)
Irbesartan	antihypertensive	4	0.02	6.05
Clarithromycin	antibacterial	3.1	3.51	8.26
Carbamazepine	antiepileptic	2.8	0.04	4.49
Diclofenac	analgesic	0.7	296	97.2
Sulfamethoxazole	antibacterial	-0.1	253	23.5
Hydrochlorothiazide	diuretic	-0.8	9.32	102.2

239 ^aSource: <https://chemicalize.com/>

240 ^bBased on reference urine as determined by Isabell Köpping – personal
 241 communication)

242

243

244 **Table 4** Hydraulic retention times (HRT, h), volumes (V, L), and average (± SD) rates
 245 (L d⁻¹) for inflow (Q) and recirculation (Q_{recirculation}) for all compartments in both column
 246 and membrane stripping reactors. (n = 3)

Reactor	Compartment	HRT (h)	V (L)	Q (L d ⁻¹)	Q _{recirculation} (L d ⁻¹)
Column	Cathode & stripping column	7.7 ^a	0.41	1.27 ± 0.01	326 ± 17
	Anode	3.9	0.21	1.24 ± 0.05	274 ± 25
	Absorbent	2.0 ^{b,d}	0.55 ^d	6.7 ± 0.2 ^d	7 ± 0 ^e
Membrane	Cathode & middle compartment membrane unit	8.3 ^a	0.44	1.24 ± 0.03	288 ± 0
	Anode	3.4	0.18	1.26 ± 0.03	301 ± 1
	Absorbent	1.1 ^{c,d}	0.12	2.8 ± 0.1 ^d	285 ± 3

247 ^aSimilar as Christiaens et al. (2017)

248 ^bQuantified in the nitrogen breakthrough curves for the neutral pH absorbent

249 ^cIntended to be the same as the HRT of the absorbent in the column stripping set-up

250 ^dOnly in tests in which the absorbent was continuously refreshed. In batch, the
 251 absorbent volume in the column reactor was 0.12 L with an HRT of 24 h.

252 ^eGas recirculation

253

254 **2.4 Chemical and microbial analyses**

255 Liquid samples were filtered (0.22 μm) before analyzing pH, electrical conductivity,
256 and ion concentrations. Cations (Na^+ , TAN, K^+) were quantified with a 761 Compact
257 IC with a Metrosep C4/4.0 guard and a C4-150/4.0 main column (Metrohm,
258 Switzerland)(LOQ of 1-20, 0.78-30, and 1-20 mg L^{-1} for Na^+ , TAN, K^+ , respectively).
259 Anions (Cl^- , NO_2^- , NO_3^- , PO_4^{3-} , SO_4^{2-}) were quantified by a 881 compact IC Pro with a
260 Metrosep A Supp 5/4.0 guard and a Metrosep A Supp 7 250/4.0 main column
261 (Metrohm, Switzerland)(LOQ of 0.8-40, 0.15-7.6, 0.18-9, 0.13-6.5, 0.8-40 mg L^{-1} for
262 Cl^- , NO_2^- -N, NO_3^- -N, PO_4^{3-} -P, SO_4^{2-} , respectively). Both devices were equipped with
263 conductivity detectors (Metrohm, Switzerland).

264 Intact, membrane-ruptured, and autofluorescent bacterial cell populations were
265 quantified by flow cytometry (Beckman Coulter Cytoflex, IN, US) after dilution with
266 0.22 μm filtered evian water (limit of detection, LOD; 10^3 events mL^{-1}). To assess the
267 viability of the bacteria, samples were stained with a mixture of SYBR Green I
268 (SG)(10 000 times diluted from stock, Invitrogen, US), binding all DNA, and
269 Propidium Iodide (PI)(3 μM final concentration, Invitrogen), only staining permeable
270 cells. After incubation at 37°C for 13 min., samples were excited by a 488 nm laser
271 and quantified by two fluorescent detectors (525/40 and 585/42 for SG and PI,
272 respectively). Unstained samples allowed quantifying the autofluorescent cell
273 population by excitation at 488 nm and detection by a 525/40 fluorescent detector.

274 Heat-killed and 0.22 μm filtered controls allowed the identification and quantification
275 of non-viable populations and noise, respectively, *via* manual gating.

276 Target micropollutants were extracted from the urine matrix, and separated by an
277 UHPLC system (Dionex, Amsterdam, The Netherlands). Chromatographic separation
278 was achieved using reversed phase chromatography with gradient elution. The

279 detection of target compounds was carried out using a Q-Exactive™ Benchtop
280 HRMS (Thermo Fisher Scientific, San-Francisco, USA) fitted with a Heated
281 Electrospray Ionization (HESI-II) source. More detailed information can be found in SI
282 (section D. Micropollutant analysis).

283 All calculations performed are described in SI (section E. Calculations).

284

285

286

3. RESULTS & DISCUSSION

287

3.1 ABSORBENT HRT OPTIMIZATION FOR NH₃ RECOVERY

288

Ammonia recovery technologies often include absorption in (strong) acid for the

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production of (NH₄)₂SO₄ or NH₄NO₃, applicable as fertilizer. The conditional

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dissociation constant (pKa') of NH₃ for hydrolysed urine at 20.8°C is 9.5, as

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calculated after Stumm and Morgan (1996). An acid absorbent guarantees strong

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dissociation and thus absorption capacity. A pH-neutral solution still allows

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dissociation but results in a lower absorption capacity. This can be improved by

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reducing the HRT in the absorption unit, *i.e.*, using a higher absorbent volume

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compared to the urine volume, and by applying a strong phosphate (P) buffer. In this

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way, a pH-neutral absorbent could be an alternative to produce a slightly alkaline

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nitrogen solution that can be used as a nitrogen source and pH correction agent, *e.g.*

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in microbial protein production (Christiaens et al. 2017). This is a more safe

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alternative for the chemicals required as pH control in microbial production

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processes.

301

The absorption breakthrough profiles for the column and membrane stripping

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reactors enabled selecting the absorbent HRT at which the buffer capacity (here:

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0.21 mol H⁺ L⁻¹) was not exceeded (Fig. 2). As a safety margin, a buffer capacity of

304

0.14 mol H⁺ L⁻¹ was considered allowing a concentration of 0.14 mol N L⁻¹ or 2 g TAN

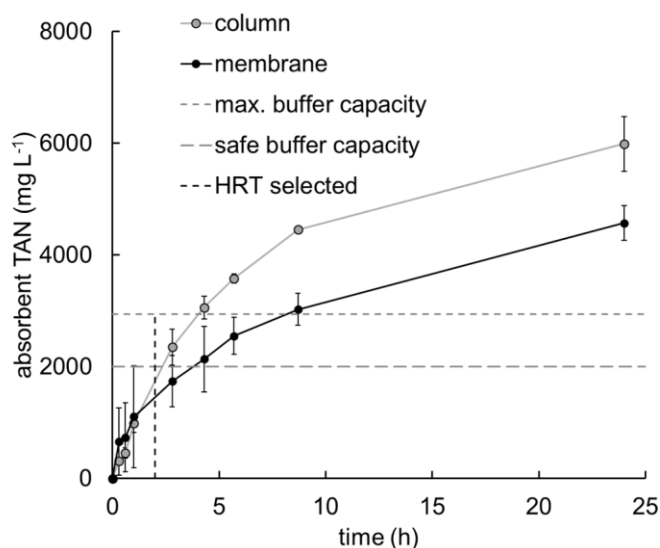
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L⁻¹ to build up in the absorbent. The corresponding HRT is thus 2 h and was applied

306

in the continuous tests further on.

307



308
 309 **Fig. 2** Average (\pm SD) 24 h total ammonia nitrogen (TAN, mg N L⁻¹) concentration
 310 profiles in a phosphate buffer absorbent for the continuously operated column and
 311 membrane stripping reactors at 20 A m⁻². The maximum (0.21 mol P L⁻¹) and safe
 312 (0.14 mol P L⁻¹) buffer capacity and the selected HRT_{abs} are indicated by dashed
 313 lines. (n = 2)

314

315 3.2 COLUMN AND MEMBRANE STRIPPING EQUALLY EFFECTIVE FOR NITROGEN REMOVAL

316 To quantify nitrogen removal, recovery, and the required energy input, both the
 317 column and membrane stripping reactors were continuously operated at 0.1 (control)
 318 and 20 A m⁻² with real urine as ammonia source and the phosphate buffer as
 319 absorbent. Both systems performed similarly in terms of nitrogen removal rates,
 320 efficiencies, and electrochemical energy efficiency under the low and high applied
 321 current density (Table 5).

322 Nitrogen removal rates at 20 A m⁻² in the column stripping reactor (0.34 ± 0.21 mol N
 323 L⁻¹ d⁻¹) were, however, lower compared to values reported earlier (0.58 ± 0.07 mol N
 324 L⁻¹ d⁻¹ in Christiaens et al. (2017)), resulting in 50% lower efficiencies and a three
 325 times higher energy demand (Table 5). Reactor set-ups and operation were the
 326 same and the hydrolysed urine had a similar composition, except for 1 g N L⁻¹ less

327 nitrogen relative to the previous study. The lower nitrogen removal rate was
 328 accompanied by a lower catholyte pH (8.7 ± 0.1 compared to 9.4 ± 0.1 for
 329 Christiaens et al. (2017)), which is usually only observed in control experiments with
 330 NH_3 stripping and limited OH^- production (SI Table F.1). As the applied current
 331 density and cathode potentials were similar in both studies (SI Table F.1), not
 332 electrocatalytic OH^- production but its consumption might be different. A hypothesis
 333 could be *in situ* Fe^{3+} precipitation as $\text{Fe}(\text{OH})_3$ (lowest solubility at pH 8) since the
 334 cathode (*i.e.*, stainless steel) might have been partly oxidized as it was exposed to
 335 bleach during a cleaning step before starting the experiments and some precipitation
 336 was observed during the experiments. To date, no decision could be taken about this
 337 observation which does not modify our comparison between the technologies.

338 The membrane reactor had equally low nitrogen removal rates as the column reactor
 339 in this study, although the catholyte pH was as expected (9.5 ± 0.1). The hydrophobic
 340 membrane was limiting the nitrogen flux ($3.9 \pm 1.5 \text{ g N m}^{-2} \text{ h}^{-1}$) as the removal rate
 341 almost equaled the recovery rate. However, this flux was found to be similar to other
 342 membrane stripping units (Arredondo et al. 2017, Dube et al. 2016), except for
 343 Tarpeh et al. (2018), who reported $42 \text{ g N m}^{-2} \text{ h}^{-1}$ for undiluted urine stripping.

344
 345 **Table 5** Average (\pm SD) nitrogen removal and recovery rates (mol N d^{-1} and mol N L^{-1}
 346 d^{-1}), efficiencies (%), and electrochemical energy requirements ($\text{kWh}_e \text{ kg}^{-1} \text{ N}$) in
 347 steady state for both column and membrane stripping reactors in this study (col and
 348 mem, respectively) with phosphate buffer as absorbent, and the column stripping
 349 reactor (ColSCP) operated with acid absorbent as described in Christiaens et al.
 350 (2017). ($n = 3$)

j	A m^{-2}		0.1		20	
			removal	recovery	removal	recovery
Rate	mol N d^{-1}	ColSCP	-0.06 ± 0.07	0.07 ± 0.06	0.32 ± 0.04	0.10 ± 0.05
		Col	0.05 ± 0.05	0.03 ± 0.01	0.21 ± 0.13	0.09 ± 0.04

		Mem	0.08 ± 0.06	0.06 ± 0.03	0.22 ± 0.05	0.17 ± 0.06
	mol N L ⁻¹ d ⁻¹	ColSCP	-0.11 ± 0.13	0.13 ± 0.11	0.58 ± 0.07	0.18 ± 0.09
		Col	0.08 ± 0.08	0.05 ± 0.02	0.34 ± 0.21	0.15 ± 0.06
		Mem	0.13 ± 0.09	0.09 ± 0.05	0.35 ± 0.08	0.27 ± 0.09
Membrane flux ^a	g N m ⁻² h ⁻¹	Mem	n.a.	1.5 ± 0.7	n.a.	3.9 ± 1.5
Efficiency	%	ColSCP	4.7 ± 1.4	25.3 ± 23.1	87.1 ± 6	25.0 ± 12.1
		Col	11.4 ± 12.8	7.5 ± 3.4	45.1 ± 18.4	17.2 ± 8.1
		Mem	19.7 ± 16.8	14.7 ± 9.8	49.0 ± 9.3	38.7 ± 13.5
Energy	kWh _e kg ⁻¹ N	ColSCP	0.05	0.04	1.9	5.8
		Col	0.01 ± 0.06	0.1 ± 0.03	5.9 ± 3.1	12.2 ± 4.8
		Mem	0.04 ± 0.01	0.1 ± 0.03	4.6 ± 1.1	6.3 ± 2.3

351 n.a. means not applicable

352 ^aParameter only applicable to membrane set-up

353

354 **3.3 MEMBRANE STRIPPING IMPROVES NITROGEN RECOVERY, REDUCING ENERGY COST**

355 Nitrogen recovery at 20 A m⁻² was almost two fold higher for membrane stripping
356 (0.27 ± 0.09 mol N L⁻¹ d⁻¹) compared to the column stripping reactor (0.15 ± 0.06 mol
357 N L⁻¹ d⁻¹). As a result, the nitrogen recovery efficiency doubled with only half the
358 electrochemical energy requirement of the column stripping reactor (Table 5). The
359 direct liquid-liquid contact likely prevented evaporative losses that might have
360 occurred in the column stripping reactor. The unintended lower absorbent HRT in the
361 membrane reactor could have contributed to this improved nitrogen recovery (Table
362 4). The extra energy for this lower absorbent HRT did hardly make a difference in the
363 overall operational energy requirements on lab scale, since the absorbent volume
364 and thus flow rate for the column reactor was higher (Table 4). The total
365 (electrochemical and pumping) operational energy required for the membrane reactor
366 was calculated at 6.5 kWh_e kg N⁻¹ recovered while for the column reactor this was
367 13.8 kWh_e kg N⁻¹ recovered (SI Eq. E.10-13; Table G.1). These values include power
368 consumption of the electrochemical cell, feed and recirculation pumps, and the gas

369 recirculation pump for the column reactor. Differences in HRT and thus flow rate
370 made up only minor differences in energy requirements between both reactor set-
371 ups. Even the gas recirculation pump was not a major contribution in this lab scale
372 set-up. The liquid recirculation over the stripping column made the difference as a
373 height of at least 1 m needed to be overcome. Additionally, the membrane unit is
374 considered to be safer because the H₂ gas, *in situ* produced at the cathode that
375 enhances NH₃ stripping, is not recirculated in a separate gas loop.

376

377 Overall, the membrane reactor consumed less energy, was safer to operate, and
378 improved nitrogen recovery compared to the column reactor, but nitrogen removal
379 should be optimized. Increasing the NH₃ vapor pressure gradient could already be
380 sufficient since the average absorbent pH in steady state at 20 A m⁻² (9 ± 1 for the
381 membrane and 7.9 ± 0.2 for the column reactor) was close to the feed pH (9.3 ± 0.1
382 and 9.2 ± 0.1 , respectively). This could be implemented by further decreasing the
383 absorbent HRT (Ahn et al. 2011). In addition, heat could be applied, shifting to
384 membrane distillation. However, a temperature difference of 30-40°C is required to
385 drastically improve the nitrogen removal and recovery efficiency, increasing the
386 operational energy costs (Ahn et al. 2011, Derese 2018). Enlarging the membrane
387 surface area could have an impact although the surface area was calculated based
388 on the nitrogen flux expected in membrane distillation (Derese 2018).

389 Coupling a membrane stripping unit to an electrochemical cell comes with
390 advantages if conceptually compared to other nitrogen recovery technologies, such
391 as conventional column stripping, membrane distillation, or membrane contactors.
392 First, the *in situ* production of H₂ gas and caustic, with only sustainably produced
393 electricity, is considered safer, eliminates the need for chemicals, and thus their cost

394 and transport. Second, the absorbent can be a microbial medium instead of a strong
395 acid, which can be used to make a high-value product, such as microbial protein
396 (Matassa 2016).

397

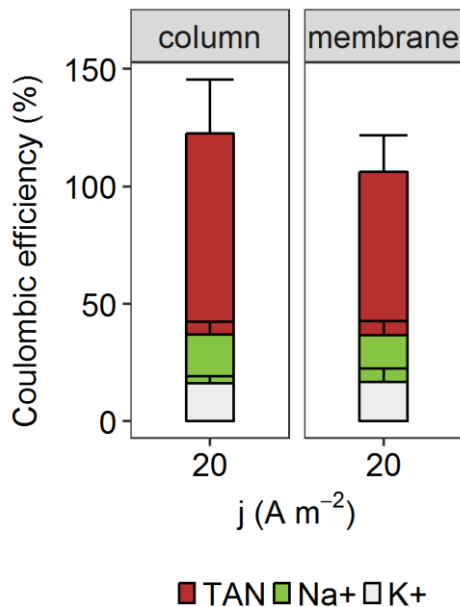
398 **3.4 TAN ELECTROMIGRATION INSTEAD OF DIFFUSION**

399 The coulombic efficiency at 20 A m^{-2} , defined as the amount of cations extracted from
400 the anolyte over the cation exchange membrane (CEM) compared to the amount of
401 applied electrons, indicated that the nitrogen flux over the CEM, or the
402 transmembrane flux, was mainly driven by electromigration in both the membrane
403 and column stripping reactors (Fig. 3). The TAN transmembrane flux towards the
404 cathode compartment was three times higher compared to the one reported in
405 Christiaens et al. (2017)(SI Table F.1), where diffusion was pointed as the main
406 driver for the nitrogen flux.

407 The anolyte ion concentration, but also electrical charge and diffusion coefficients,
408 determine which ions will account for the charge balance *via* electromigration
409 (Christiaens et al. 2017, Cord-Ruwisch et al. 2011, Desloover et al. 2012). The lower
410 nitrogen removal *via* stripping in this study resulted in a TAN accumulation in the
411 catholyte and thus also in the anolyte. The TAN anolyte concentrations were three
412 times higher (3126 ± 403 and $3070 \pm 841 \text{ mg N L}^{-1}$ for column and membrane
413 stripping, respectively) compared to Christiaens et al. (2017)($1155 \pm 692 \text{ mg N L}^{-1}$).
414 As the anolyte pH was 7.7 ± 0.2 and 8.0 ± 0.6 for column and membrane stripping,
415 respectively, anolyte TAN was mainly present as $\text{NH}_4^+\text{-N}$ (98% and 96%,
416 respectively). Conversely, the anolyte concentrations of other main ions (Na^+ and K^+)
417 were similar in both studies and two times lower compared to the tripled TAN
418 concentrations. Consequently, electromigration of TAN was favored. These results

419 confirm earlier work where an EC fed with real hydrolysed urine in the anode
420 compartment was coupled to a stripping column (Luther et al. 2015) and coupled to a
421 membrane contactor (Arredondo et al. 2017).

422



423

424 **Fig. 3** Average (\pm SD) coulombic efficiency (%) at 20 A m⁻² for monovalent ions in
425 steady state for both column (left) and membrane (right) stripping reactors. The
426 coulombic efficiency is defined as the amount of cations extracted from the anolyte
427 over the cation exchange membrane compared to the amount of applied electrons.
428 The sum of all ions makes up for a 100% efficiency indicating electromigration. (n =
429 3)

430

431 3.5 A HYDROPHOBIC MEMBRANE PREVENTS TRANSFER OF CONTAMINATING BACTERIA

432 If the recovered nitrogen will be used to produce proteins for feed or food
433 applications, contamination by microorganisms present in urine needs to be avoided.

434 If microorganisms are still present, a post-treatment step might be required (e.g.
435 filtration or sterilization by ozone, UV, or chlorination)(Jones et al. 2018, Macauley et
436 al. 2006), which adds energy and chemical costs.

437 During 24 h batch NH₃ stripping tests, the feed and absorbent of both column and

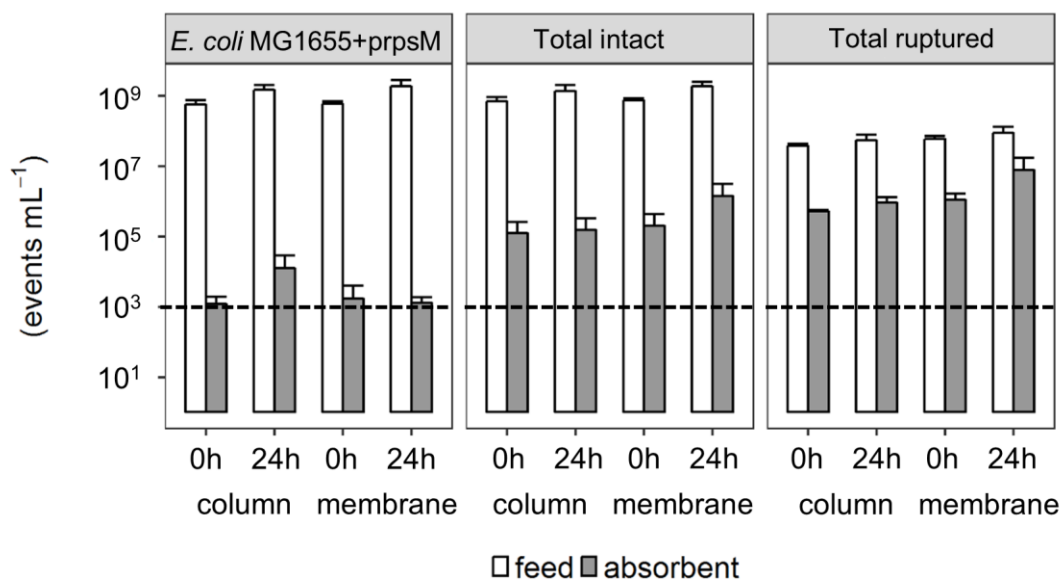
438 membrane stripping reactors were monitored for autofluorescent *E. coli*
439 MG1655+prpsM cells that had been spiked into the feed bottle as a tracer organism,
440 but also for total intact and ruptured cells. The initial concentration of *E. coli*
441 MG1655+prpsM in the feed bottles of the column and membrane reactors was
442 $5.8 \cdot 10^8 \pm 1.9 \cdot 10^8$ and $6.1 \cdot 10^8 \pm 9.9 \cdot 10^7$ events mL⁻¹, respectively, while the
443 concentration in the absorbents was below the LOD (10^3 events mL⁻¹)(Fig. 4). After
444 24 h there was a slight increase in spiked *E. coli*, intact, and ruptured cells of the
445 influent as the intake tubing reached the settled biomass.

446 The spiked *E. coli* only increased in the absorbent of the column reactor, from $1.2 \cdot 10^3$
447 $\pm 7.5 \cdot 10^2$ to $1.3 \cdot 10^4 \pm 1.6 \cdot 10^4$ events mL⁻¹ after 24 h. The absorbent of the membrane
448 reactor increased for intact and ruptured cell counts after 24 h from $2.1 \cdot 10^5 \pm 2.2 \cdot 10^5$
449 to $1.4 \cdot 10^6 \pm 1.7 \cdot 10^6$ events mL⁻¹, and from $1.1 \cdot 10^6 \pm 5.4 \cdot 10^5$ to $7.9 \cdot 10^6 \pm 9.4 \cdot 10^6$
450 events mL⁻¹, respectively. Growth was rather unlikely as the absorbent accumulated
451 free ammonia (FA) to 4759 ± 550 and 3895 ± 408 mg N L⁻¹ after 24 h at a pH of $9.9 \pm$
452 0.1 and 10.2 ± 0.1 for the column and membrane reactors, respectively (Fig. H.1).
453 Vinneras et al. (2008) reported ammonia concentrations of 2100 mg N L⁻¹ at pH 8.9
454 to rapidly inactivate enteric pathogens. Allievi et al. (1994) found that mainly FA
455 inactivated bacterial cells if 10°C is reached, enhanced by an alkaline pH and/or
456 increased salinity, while other studies showed a pH increase to be sufficient (Diez-
457 Gonzalez et al. 2000, Ogunyoku et al. 2016). In microbial cells, FA probably
458 disintegrates the cell membrane (Jenkins et al. 1999) by membrane and protein
459 denaturation (Bujoczek 2001) and alkalinisation of the cytoplasm (Diez-Gonzalez et
460 al. 2000).

461 More plausible is the transfer of the tracer organism *E. coli* to the absorbent *via* urine
462 aerosols (Benami et al. 2016, Heinonen-Tanski et al. 2009). Transfer of the smallest

463 microorganisms reported in urine (0.1 μm)(Dong et al. 2011, Shepard et al. 1974,
 464 Waites and Talkington 2004, Wang et al. 2007) could have happened through the
 465 hydrophobic membrane which had an average measured pore size of 0.274 ± 0.003
 466 μm , whereas the manufacturer reported 0.1 μm (SI Table B.1). Only *via* wetting of the
 467 gas-filled pores microorganisms could have transferred. However, these small cells
 468 apparently could not transfer *via* the gas phase in the column stripping reactor,
 469 although *E. coli*, a large microorganism with minimum diameter of 1 μm (Gagnon
 470 2016), could. Innovative hydrophobic gas permeable membranes with track-etched
 471 pores could provide a tailor-made and extremely narrow pore size to prevent future
 472 transfer of microorganisms (Apel 2001).

473



474

475 **Fig. 4** Average (\pm SD) cell counts (events mL⁻¹) at 0 and 24 hours for autofluorescent
 476 *E. coli* MG1655+prpsM, total intact, and total ruptured microorganisms, in the feed
 477 and absorbent compartments of the column and membrane reactors. The LOD for
 478 flow cytometry is at 10³ cells mL⁻¹, as indicated by the dashed line. (n = 2)

479

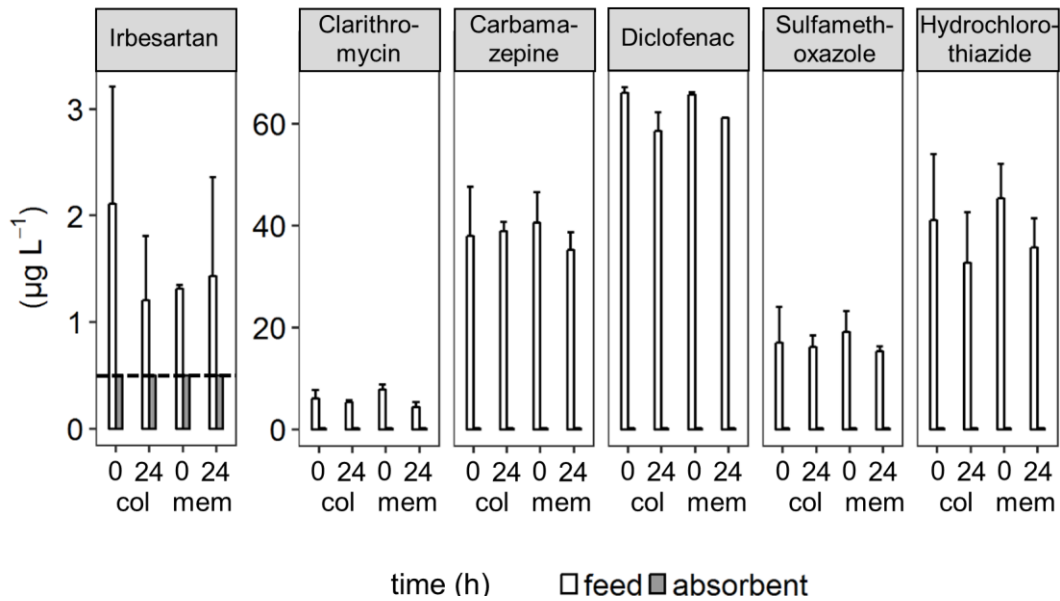
480 3.6 NO MEASURED MICROPOLLUTANTS TRANSFERRED TO THE RECOVERED PRODUCT

481

482 The absence of urine-derived micropollutants in the absorbent with recovered
483 nitrogen is a prerequisite for reuse applications. Six micropollutants, ranging from
484 hydrophobic to hydrophilic at pH 9 (Table 3), were spiked in the urine fed to both
485 column and membrane stripping reactors and monitored in both the feed and
486 absorbent during 24 h batch NH₃ stripping tests. Measured carbamazepine
487 concentrations in urine at the start of the tests exceeded the spiked concentration
488 (4.49 µg L⁻¹) as it was already present at 37.54 ± 7.83 µg L⁻¹ (replicate 1) and 35.38 ±
489 0.44 µg L⁻¹ (replicate 2).

490 Despite the hypothesis that hydrophilic compounds could transfer more easily in the
491 column reactor and hydrophobic compounds might transfer better through the
492 hydrophobic membrane, no difference could be observed between the column and
493 membrane stripping reactor (Fig. 5). Moreover, while micropollutant concentrations in
494 the feed urine reflected spiked concentrations (except for carbamazepine), no
495 micropollutants were detected in the absorbents (below LOD 0.25 and 0.50 µg L⁻¹).
496 These results are in line with recent work by Tarpeh et al. (2018) who reported the
497 absence of urine-derived trace organics (<0.1 µg L⁻¹) in the acid trap after a
498 hydrophobic NH₃ extraction membrane. However, Böhler et al. (2015) reported the
499 transfer of micropollutants *via* column stripping to the absorption column.
500 Carbamazepine, for instance, reached 0.1 µg L⁻¹ in the (NH₄)₂SO₄ recovered product.
501 With respect to the LODs reported here, we can conclude that both column and
502 membrane stripping techniques can be safely used for nitrogen recovery in view of
503 detectable micropollutant concentrations. However, additional concentration
504 quantification with devices where the LOD reaches to 0.1 µg L⁻¹ would be
505 recommended.

506



508

time (h) □ feed ■ absorbent

509 **Fig. 5** Average (\pm SD) micropollutant concentrations ($\mu\text{g L}^{-1}$) at 0 and 24 hours in the
 510 feed and absorbent compartments of the column (col) and membrane (mem)
 511 reactors. All concentrations for the absorbent were below the LOD ($0.5 \mu\text{g L}^{-1}$ for
 512 irbesartan, indicated by the dashed line, and $0.25 \mu\text{g L}^{-1}$ for the other
 513 micropollutants). (n = 2)

514 4. CONCLUSIONS

515

516 Nitrogen removal and recovery from source-separated urine and the safety of the
517 recovered product were evaluated for two technologies: column and membrane
518 stripping, both coupled to an electrochemical cell.

519

520 - Both technologies performed similarly for nitrogen removal. Nitrogen
521 recovery was clearly improved in a membrane stripping reactor by
522 reducing nitrogen losses, that potentially occurred *via* condense water in
523 the gas phase of a stripping column.

524 - Membrane stripping reduced operational energy requirements
525 (electrochemical and pumping) with 50% compared to the column stripping
526 reactor.

527 - Lower overall nitrogen removal rates for both stripping technologies
528 compared to earlier work resulted in nitrogen accumulation in the anolyte
529 compared to Na⁺ and K⁺, which caused TAN electromigration towards the
530 catholyte.

531 - Membrane stripping prevented the transfer of the autofluorescent *E. coli*
532 MG1655+prpsM spiked in the urine towards the absorbent, whereas
533 increased cell concentrations were observed in the absorbent of the
534 column stripping reactor.

535 - Urine-derived micropollutants were below the LOD in the absorbent of both
536 stripping technologies.

537

538 Overall, membrane stripping should be optimized for nitrogen removal by increasing

539 the NH_3 vapor pressure gradient and/or membrane surface area. However, it is
540 preferred over column stripping since it improved nitrogen recovery, consumed less
541 energy, and prevented transfer of spiked microbial cells and micropollutants into the
542 recovered nitrogen product.
543

544 The authors declare no competing financial interest.

545 The Supporting Information is available free of charge on the Elsevier website at

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547

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560

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