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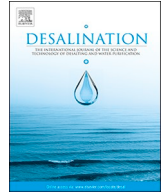To cite this publication, please use the final published version (if applicable).
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A five-stage treatment train for water recovery from urine and shower water for long-term human Space missions

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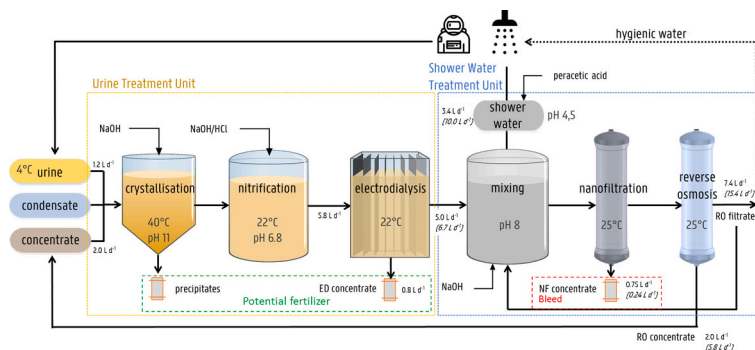[e] VITO, Boeretang 2, 2400 Mol, Belgium
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GRAPHICAL ABSTRACT

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ABSTRACT

Long-term human Space missions will rely on regenerative life support as resupply of water, oxygen and food comes with constraints. The International Space Station (ISS) relies on an evaporation/condensation system to recover 74–85% of the water in urine, yet suffers from repetitive scaling and biofouling while employing hazardous chemicals. In this study, an alternative non-sanitary five-stage treatment train for one "astronaut" was integrated through a sophisticated monitoring and control system. This so-called Water Treatment Unit Breadboard (WTUB) successfully treated urine (1.2-L-d^{-1}) with crystallisation, COD-removal, ammonification, nitrification and electrodialysis, before it was mixed with shower water (3.4-L-d^{-1}). Subsequently, ceramic nanofiltration and single-pass flat-sheet RO were used. A four-months proof-of-concept period yielded: (i) chemical water quality meeting the hygienic standards of the European Space Agency, (ii) a 87- ± -5% permeate

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recovery with an estimated theoretical primary energy requirement of $0.2\text{-kWh}_p\text{-L}^{-1}$, (iii) reduced scaling potential without anti-scalant addition and (iv) and a significant biological reduction in biofouling potential resulted in stable but biofouling-limited RO permeability of $0.5\ \text{L-m}^{-2}\text{-h}^{-1}\text{-bar}^{-1}$. Estimated mass breakeven dates and a comparison with the ISS Water Recovery System for a hypothetical Mars transit mission show that WTUB is a promising biological membrane-based alternative to heat-based systems for manned Space missions.

Abbreviations

BWRO	Brackish Water Reverse Osmosis		PAA	PerAcetic Acid
CCU	Crystallisation and Coagulation Unit		SI	Saturation index
COD	Chemical Oxygen Demand		SF	Supplemental Material
CED	Cumulative Energy Demand		SRT	Sludge Retention Time
ECLSS	Environmental Control and Life Support System		SWRO	Sea Water Reverse Osmosis
ED	ElectroDialysis		SWTU	shower water treatment unit
EDU	ElectroDialysis Unit		TAN	Total Ammonia Nitrogen
ESA	European Space Agency		TDS	Total Dissolved Solids
HRT	Hydraulic Retention Time		TOC	Total Organic Carbon
ID	Internal Diameter		UF	UltraFiltration
ISS	International Space Station		UTU	Urine Treatment Unit
LCD	Limiting Current Density		VCD	Vapor Compression Distillation
NF	NanoFiltration		VCF	Volumetric Concentration Factor
			WRS	Water Recovery System
			WTUB	Water Treatment Unit Breadboard

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1. Introduction

With the on-going developments in the commercialization of the space industry, human Space exploration has been attracting renewed attention. To sustain life in Space, fresh supply of water, oxygen and food is essential. With a metabolic minimum for humans of just over 3 L d^{-1} [1–3], and only 10 L d^{-1} of shower water for comfort on a 900 days mission to Mars, 4 crew members would already need almost 50 ton of fresh water supplies, which is larger than the mass of O_2, CO_2-capture and food combined [4,5].

Taking into account that the International Space Station (ISS) weighs approximately 400 ton, the current limitations in launcher capacity (~1 ton H_2O) and the high launching cost [6], recycling of water is essential [7]. Respiration and perspiration condensate, urine and flush water, and hygiene, wash and occasionally shower water are considered the most relevant sources for renewed freshwater supply [8]. The combination of urine (~1.5 L person^{-1} d^{-1}) and a low flow of shower water ($\leq$10 L person^{-1} d^{-1}) is challenging due to hardness (30–390 mg Ca^{2+} L^{-1}, 20–205 mg Mg^{2+} L^{-1}), salinity (0.3–23 mS cm^{-1}), bioavailable organics (up to 10 g COD L^{-1}) and ammoniacal nitrogen (up to 9 g N L^{-1}) present in urine and the surfactants and bioavailable organics present in shower water [9,10].

Although it can rely on occasional resupply, due to the vicinity to Earth, the environmental control and life support system (ECLSS) of the ISS is the state-of-the-art with regard to sustaining human presence in Space [11]. The Water Recycling System (WRS) at the ISS consists of a urine processing assembly (UPA) and a Water Processing Assembly (WPA). The core of the UPA is a Vapor Compression Distillation (VCD) unit to recover product water. It is particularly sensitive to organic fouling and scaling while consuming ~0.8 kWh L^{-1} for treating 9 L urine per day along with sulfuric/phosphoric acid and chromium(IV) trioxide. Only after recent upgrades (e.g., replacement of sulfuric acid by phosphoric acid), the recovery increased from 74 to 85% [11–13]. The product water is fed to the WPA, in which it is blended with condensate and water from the Sabatier process (formed with CH_4 from CO_2 and H_2). Together it undergoes, after a particle filter, chemical treatment (multifiltration beds, chemical oxidation and ion exchange) [12,14] to upgrade it to potable water and water suitable for

electrochemical oxygen production. The efficiency of the WPA is 97–99%, enabling a total water recovery of > 90% [15]. Extensive details, pictures and a flow scheme of the UPA and WPA were presented by Williamson et al. [11] and have been made publicly available on the NASA Technical Reports Server (https://ntrs.nasa.gov/).

For advancing deeper into Space, longer mission duration will be required without regular resupply [8]. Therefore, Jones (2017) performed a mass payback analysis for the ECLSS in case it would be used for a hypothetical mission to Mars [4]. The study assumed a 450 day transit duration to Mars for 4 crewmembers, generating each 2.28, 2.00 and 1.29 kg per day of respectively perspiration and respiration water, urine, flush and wash water. Shower water was excluded. The study concluded that the ECLSS would be largely suitable, but advised on upgrading the urine treatment for longer mission durations [4].

Developments for terrestrial recovery of water from urine are relatively scarce, as only in extreme situations urine is considered a suitable source of potable water. Nonetheless, recovering water from urine can go hand in hand with nutrient recovery, as demonstrated in a membrane pervaporation approach for nitrogen and water recovery [16].

Shower water is at present not produced in the ISS, and was excluded from the study by Jones [4]. Only a limited number of studies have considered recycling this stream, although it is being considered for longer-term missions [17,18]. For terrestrial applications, recovery of shower water is often included in more extensive decentralized grey water recycling schemes that often use membrane-based systems, suitable for removing limited ions surfactants from the used detergents and potential fecal bacteria [19,20]. While most of the remote Antarctic research stations sufficiently treat grey water for safe discharge, the European Space Agency took the lead in developing an alternative membrane-based grey water recovery facility for the research station Concordia. This plant was designed to treat 2500 L d^{-1} of grey water based on a four-stage approach comprised of ultrafiltration (UF), nanofiltration (NF) and two reverse osmosis (RO) units, at a recovery efficiency of 75%. Black water (i.e. faeces, urine and rinse water) is fermented and the sludge is then transported to the mainland for incineration [21,22]. Afterwards, several other research stations, like Princess Elisabeth (Belgium) have adopted modified water recovery approaches [23].

Potable water production from saline streams often relies on either thermal or membrane-driven processes. Pressure-driven membrane systems are often preferred over thermal systems, due to associated energy costs [24]. For desalination of streams with a salinity similar to urine, i.e. brackish water, high water recoveries at reasonable energy cost are typically based on electrodialysis (ED) and/or pressure-driven membranes [25]. Yet, these systems are sensitive to biofouling and scaling, particularly when applied directly on wastewater [26,27], and therefore rely on anti-scalants and biocides [28]. For example, concentrate (with > 1000 mg Ca^{2+} L^{-1}) of brackish water RO (BWRO) plants has been treated using ED to increase the overall RO recovery to over 90%, requiring the addition of anti-scalant [29]. Hybrid brackish and seawater desalination schemes using BWRO and ED were also

proposed based on energetic calculations, but were not accommodated with long-term actual RO experiments [30]. However, neither seawater nor brackish groundwater has the same biofouling and scaling potential as urine. Furthermore, these streams are not high in ammoniacal nitrogen, with high salts (10–15 g TDS L^{-1}), nutrients (5–10 g TAN L^{-1} and 0.5–1.0 g P L^{-1}), and earth alkaline metals (50–200 mg Ca^{2+} L^{-1}) [31]. The low molecular weight of TAN-N combined with the speciation potential to uncharged NH_3, makes is a difficult compound to be removed by NF or RO [32].

A five-stage treatment train, the so-called Water Treatment Unit Breadboard (WTUB) was developed as a biological membrane-based water and nutrient recovery alternative for regenerative life support systems that could avoid the use of chromium(IV)trioxide. It consists of
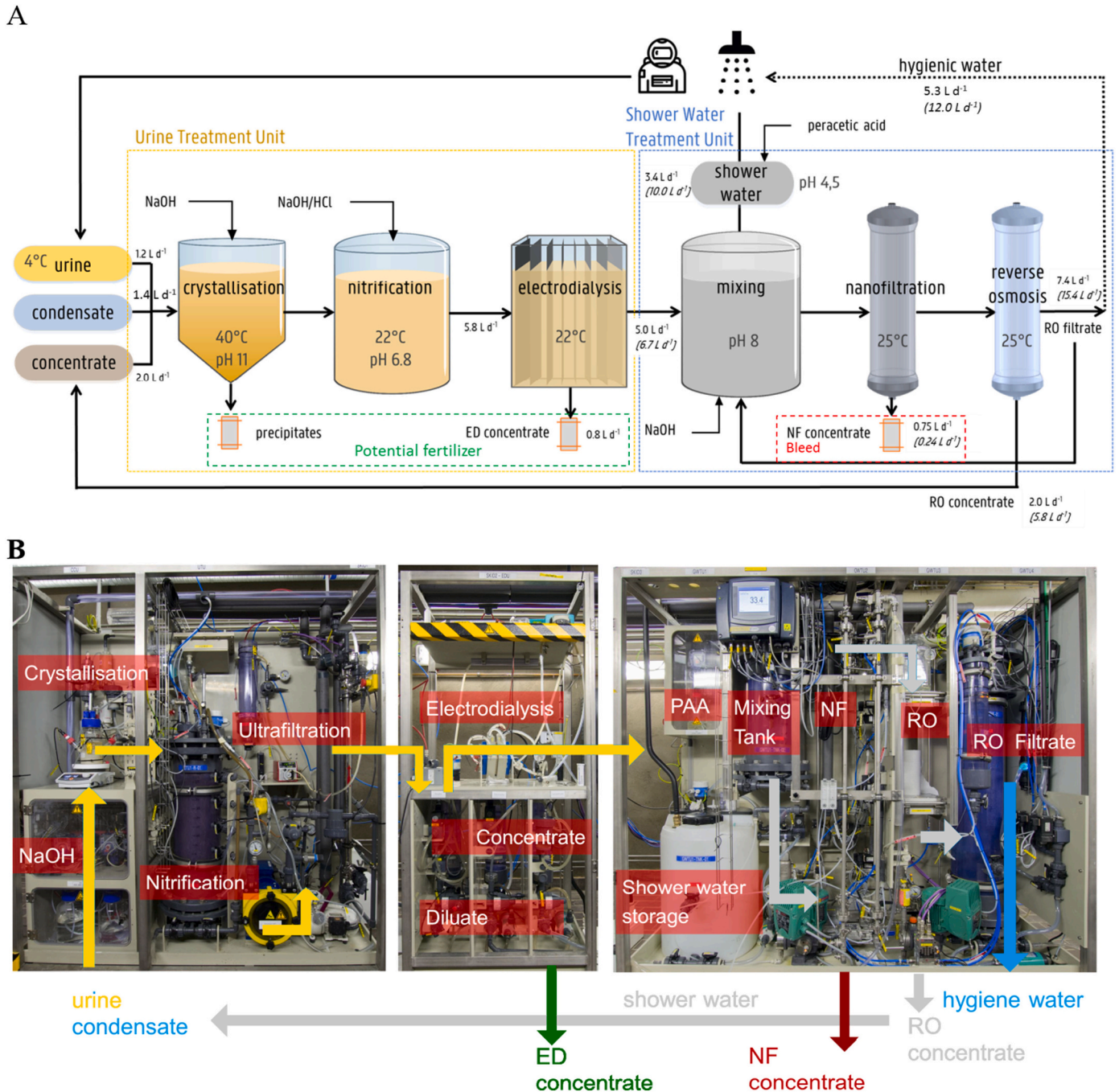

Fig. 1. Simplified process flow diagram with and actual measured flow rates and design values between (..) (A) and photograph of the WTUB testbed facility (B). Urine collection and composition.

a three-stage Urine Treatment Unit (UTU) [13].: 1) a crystallisation reactor for P recovery and decreasing scaling potential, 2) a membrane bioreactor for oxidation of organics and conversion of volatile ammoniacal nitrogen into NO_3^- and thereby reducing biofouling potential, 3) an ED to recover NO_3^- and remove accumulating monovalent ions to reduce the osmotic pressure for the RO. Subsequently, water is recovered from the ED diluate and Peracetic Acid (PAA)-stabilized shower water through a two-stage shower water treatment unit (SWTU), composed of 4) a ceramic NF as a selective sink for organic pollutants originating from the shower water and 5) RO as the final hygienic water recovery step, as inspired by the system in Concordia [33].

The goal of this study is to demonstrate the feasibility of this biological membrane-based alternative by (i) treating real urine and shower water in a highly automated pilot installation without relying on specialty chemicals and (ii) compare the chemical water composition, the water and energy balance to the state-of-the-art chemical and thermal-based system at the ISS. The UTU was operated for 4 months and the SWTU for 1 month. The proof-of-concept results presented in this study concern a representative period of combined operation on 0.3 L d^{-1} of condensate, 1.2 L d^{-1} of urine for 3.4 L d^{-1} of grey water.

2. Materials and methods

2.1. Wastewater collection and experimental setup

The integrated WTUB testbed facility is composed of a Urine Treatment Unit (UTU) and a shower water treatment unit (SWTU) that is depicted in Fig. 1A and B. More details on the design of the system can be found in Supplemental material (SM) Section D.

Urine was collected from healthy male donors not using any medication prior to the urine donation in 10–50 L batches using a water-less urinal (SF Fig. A.1), as described by De Paepe et al. [13]. The urine was diluted with demineralised water to simulate condensate and RO concentrate and the raw values are presented in Table 2.

2.1.1. Urine Treatment Unit (UTU)

Details on the crystallisation, nitrification bioreactor and ED unit can be found in our previous work [13] and SF Fig. A.2–4. For this specific experiment, urine was diluted approximately fivefold by condensate and RO concentrate at an average retention time of 3 h, 5 days and 6 h in the crystallizer, bioreactor and ED unit respectively (Table 1).

2.1.2. Shower water collection and composition

Shower water was collected in a shower installed next to the testbed facility (SF Fig. A.1) and was automatically pumped (Sani pump, SFA group, France) into the 60 L storage tank. Each volunteer received 2.5 g of fragrance-free soap (Neutrogena facial cleansing bar, Los Angeles, USA) and 10 L of Volvic water, consisting of 12, 8, 6, 9, 74, 32 and

15 mg L^{-1} of calcium, magnesium, sulphate, bicarbonate, silica and chloride according to supplier specifications (Danone, France). After each shower, 20 mL 100% Oxonia-solution (Ecolab, Leiden, The Netherlands) was added for biological stabilization to bring the peracetic acid (PAA) concentration to 100–200 mg L^{-1} at a pH ~4.5. PAA is a known "green oxidant" alternative to chlorination with eV ~1.8, which has been successfully used in MELiSSA hygienic designs before [21,34]. PAA is particularly useful below pH 8.2 where it slowly hydrolyses into H_2O_2 and water [35,36].

2.1.3. Shower water treatment unit (SWTU)

Photographs with labels have been used to describe SWTU in SF Fig. A.5. The shower water was pumped from the storage tank to the pH controlled mixing reactor, in which it could be mixed with RO permeate and ED diluate (for experiment 2). The NF unit itself consisted of two tubular TiO_2 membranes on Al_2O_3 support material (7 mm ID × 1200 m, Inopore, Germany) with a 750 Da cut-off, placed in series and providing 0.052 m^2 surface area [37]. A mass flow controller (mini-CORI FLOW, Bronckhorst, Ruurlo, The Netherlands) was used to meet the required permeate flow rate within a pre-specified pressure range of 2–10 bars at a crossflow velocity of 1.0–1.5 m s^{-1}.

The RO unit consisted of two custom-made stainless steel high pressure cells designed to mimic spiral-wound hydrodynamic conditions (Drukhuis 10-730 and 10-731, European Membrane Institute-Twente, The Netherlands) suitable for operation at 40 bar and 110 °C. Seawater reverse osmosis membranes (SWRO) and spacers were cut out of spiral wound Dow Filmtec elements (DOWSW30-2540, Dow, USA) at a total effective surface area of 0.04 m^2 for two membranes. In the latter stage of the experiment also brackish water reverse osmosis membranes (BWRO) were tested (DOWBW30-2540, Dow, USA). The system was operated at a crossflow velocity of 0.6–1.0 m s^{-1} and a mass flow controller (mini Coriflow, Bronkhorst, The Netherlands) combined with the PLC feedback loop in order to meet the required flow rate within a pre-specified pressure range of 5–15 bars.

2.1.4. Online measurements and control

The WTUB setup was equipped with instrumentation and a Siemens programmable logic controller (programmed by IEC NV, Riemst, Belgium) allowing a continuous operation of the pilot. In total, 5 pH (glass electrode, Hach Lange, Düsseldorf, Germany), 1 DO (LDO sc, Düsseldorf, Germany) and 6 EC (Inductive probe 3700 HC, Hach, Germany) electrodes were connected to a SC1000 (Hach, Düsseldorf, Germany) controller. The process parameters were automatically logged at a time interval of 20–60 s depending on the measured parameter. An online ammonium analyzer (AMTAX sc, Hach, Düsseldorf, Germany) was installed in between the bioreactor and the ED to continuously monitor the TAN (total ammonia nitrogen, sum of ammonia and ammonium) concentration.

Table 1
Summary of operational parameters for the WTUB units during representative operation.

	Crystallizer	Bioreactor	ED	Storage tank SWTU	NF	RO
Flow rate (L d^{-1})	5–6	5–6	5–6	3.0–4.5	9.0–13.5	6.0–10.0
Volume (L)	0.8–1.0	28	2	30	18	3
Hydraulic Retention Time	~6 h	~5d	8 h	6–10d	1.3–2d	0.3–0.5d
Surface area (m^2)		0.057	0.064		0.052	0.04
Membrane type		α-Al$_2$O$_3$	SCE/SAE		TiO$_2$ 750 Da	SW30/BW30
Crossflow velocity (m s^{-1})		0.5	N.D.		1.0–1.5	0.6–1.0
Pressure (bar)		< 0.5	N.D.		2–5	10–15
EC (mS cm^{-1})	8–10	10–12	4–5	< 1	1–3	5–10
pH	10.8–11.1	6.6–7.0	6.1–6.4	3.5–4.5	8.0–8.4	8.0
Temperature (°C)	40	19–22	18–20	18–20	25–28	25–26
Potential (V)			4.1 ± 2.1			
Current (A)			0.07 ± 0.03			

2.2. Design, start-up and experimental plan

The conceptual design was based on a steady-state water balance built in MSExcel and Visual Basic (Microsoft, Redmond, USA) and complemented with phreeqC simulations (version 3, USGS). After detailed design and procurement, the bioreactor was inoculated according to previous work [13] with a mixture of three mixed cultures that were shown to survive Low Earth Orbit exposure and contained genera that travelled to the ISS [38,39]. Subsequently, the chemical composition was measured online and offline using previously described analytical methods [13]. Finally, the flow rate, temperature, pressure and power supply were determined to complete the water and energy balance.

Prior to the integrated experiment several smaller and separate experiments were performed on the UTU and the SWTU to ensure proper functionality of the entire system, but fall beyond the scope of this work. Once these were performed, the mass balance based on which the system was engineered, was used to set the following operational parameters for each of the integrated process units.

In order to facilitate integration of the five treatment steps, flows had to be recirculated and mixed: i) a part of the produced RO permeate was recycled back to the mixing tank to ensure sufficient crossflow velocity over the NF and RO membrane at ~80% water recovery, b) The mixing tank was fed in a ratio of 1:1:1 with ED diluate, chemically stabilized shower water and RO permeate and c) RO concentrate was recycled back over the UTU to ensure the removal of hardness salts in the crystallisation unit, accumulating nutrients and carbon in the bioreactor and monovalent ions in the ED.

2.2.1. Data processing & evaluation

MSExcel was used for recording and processing offline measurements and RStudio (R version 3.2.2 (2015-08-14) on a $\times$86 64-w64-mingw32/$\times$64 (64-bit) platform running under Windows 64-bit was used to collect and process the more extensive online datalog files.

More extensive evaluation based on the permeability and underlying analysis of biofouling and scaling potential has been described in SF Section C.

The theoretical energy balance required assumption and measurements of power input into all hardware. These were then compiled in MSexcel in combination with Gross Energy Requirement estimations for used chemical elements (SF Section D).

3. Results & discussion

3.1. Chemical water composition

Table 2 summarizes the chemical water composition of the primary and recycled inputs (Urine, Shower Water and ED-diluate and RO-concentrate), hygienic water and the sinks (ED-concentrate and NF-concentrate). Underlying data is presented in SF Fig. C.1 The primary input condensate was substituted by demineralised water for the experiments and was therefore not included in the table.

The performance of the UTU, i.e. the Ca^{2+}, Mg^{2+} and $P-PO_4^{3-}$-removal in the crystallisation, the COD-removal, ammonification and nitrification in the bioreactor, and the nutrient recovery potential of the ED have been described previously [13].

The measured RO-permeate quality demonstrates that water recovered with the SWTU from the ED diluate and PAA stabilized shower water could meet ESA hygienic water requirements and the WHO drinking water guideline quality on most parameters [40]. However, both for TAN, NO_3^--N and COD/TOC, the RO permeate quality did not entirely meet requirements with < 2 mg TAN L^{-1}, 13 $\pm$ 3 mg NO_3^--N L^{-1} and 10–30 mg COD L^{-1}, respectively.

TAN concentrations peaks in the RO-permeate exceeding 0.4 mg N L^{-1} could be traced back to the performance of the bioreactor, due to the known but stable limited rejection of TAN on the RO. pH-upsets in the bioreactor increased outgoing TAN concentration to > 20 mg N L^{-1} from steady-state values of close to 0 mg N L^{-1} [13].

NO_3^--N fluctuation in the RO permeate were attributed to the performance of the ED or elevated nitrogen concentrations in the urine, because consistent removal efficiencies of NO_3^- were observed of 70–80% in the ED, 3% on the NF, and 92% on the RO.

The COD in the RO permeate contained acetic acid and seemed to

Table 2
Average chemical composition of the urine, ED diluate, chemically stabilized shower water, RO concentrate (Conc.) and final RO permeate over representative operation.

		Primary and recycled inputs				Hygienic water		Sinks	
		Urine	ED diluate[e]	Shower water[d,e]	RO Conc.[e]	RO permeate[f]	ESA standard	ED Conc.	NF Conc.[e]
N-total	mg-N L^{-1}	5365 $\pm$ 909	312 $\pm$ 63	8 $\pm$ 1	186 $\pm$ 94	16 $\pm$ 1		3936* $\pm$ 746	98 $\pm$ 33
TAN	mg-N L^{-1}	771 $\pm$ 325	9 $\pm$ 12	2 $\pm$ 1	0–50	< 2	**0.4**	131 $\pm$ 187	3 $\pm$ 5
N-NO_3^-	mg-N L^{-1}	2 $\pm$ 3	303 $\pm$ 51	5 $\pm$ 0	161 $\pm$ 65	13 $\pm$ 3	**11.3**	3798 $\pm$ 556	89 $\pm$ 19
N-NO_2^-	mg-N L^{-1}	< 2	< 1	< 1	19 $\pm$ 18	< 2		6 $\pm$ 4	5 $\pm$ 9
COD	mg L^{-1}	6098 $\pm$ 11	68 $\pm$ 11	1517 $\pm$ 73[c]	447 $\pm$ 224	< 30		151 $\pm$ 50	463 $\pm$ 130
TOC	mg L^{-1}	ND	ND	ND	ND	< 10[a]	**10**	ND	ND
Ca^{2+}	mg L^{-1}	135 $\pm$ 77	0.2 $\pm$ 0	13 $\pm$ 7	25 $\pm$ 36	1 $\pm$ 7		0	8 $\pm$ 5
K^+	mg L^{-1}	1971 $\pm$ 326	117 $\pm$ 23	26 $\pm$ 5	127 $\pm$ 49	8 $\pm$ 5	**120**	1662 $\pm$ 343	44 $\pm$ 13
Na^+	mg L^{-1}	2209 $\pm$ 309	741 $\pm$ 103	45 $\pm$ 7	1533 $\pm$ 623	66 $\pm$ 7	**750**	8975 $\pm$ 1352	560 $\pm$ 158
Mg^{2+}	mg L^{-1}	39 $\pm$ 31	< 1	6 $\pm$ 1	5 $\pm$ 6	< 1		ND	2 $\pm$ 2
$P-PO_4^{3-}$	mg L^{-1}	649 $\pm$ 30	51 $\pm$ 5	18 $\pm$ 0	17 $\pm$ 7	2 $\pm$ 0	**16.3**	62 $\pm$ 22	11 $\pm$ 1
HCO_3^-/CO_3^{2-}	mg L^{-1}	5000[b]	52[b]	80[b]	217[b]	63[b]		ND	ND
Cl^-	mg L^{-1}	3449 $\pm$ 527	283 $\pm$ 41	25 $\pm$ 15	532 $\pm$ 285	37 $\pm$ 15	**1000**	3159 $\pm$ 535	384 $\pm$ 17
$S-SO_4^{2-}$	mg L^{-1}	153 $\pm$ 85	54 $\pm$ 4	7 $\pm$ 2	13 $\pm$ 7	2 $\pm$ 2	**TBD**	267 $\pm$ 47	8 $\pm$ 1
Turbidity	FNU	ND	ND	ND	ND	ND	**10**	**ND**	**ND**
pH		6–8	5.9–6.5	6.4–7.0	7.3–9.0	7.5–8.5	**5.0–8.5**	**5.9–6.5**	8.5
TDS	g L^{-1}	17.3 $\pm$ 2.6	2.8 $\pm$ 0.4	1.7 $\pm$ 0.1	5.9 $\pm$ 2.6	< 0.2	1.92	32	ND
Average flowrate	L d^{-1}	1.2 $\pm$ 0.4	5.0 $\pm$ 1.0	3.4 $\pm$ 1.7	2.0 $\pm$ 1.3	7.4 $\pm$ 2.5		0.8 $\pm$ 0.8	0.8 $\pm$ 0.4
						5.3 $\pm$ 1.8			

[a] Estimated (TOC = COD/3).
[b] Calculated based on ionic balance.
[c] ~250 mg COD L^{-1} originated from the soap, the remainder from PAA.
[d] Chemically stabilized shower water.
[e] ED diluate, chemically stabilized shower water and RO permeate make up NF-feed.
[f] Gross and net RO permeate production due to automatic feedback loop and required flow rates.

originate from the shower water. The use of PAA for disinfection purposes increased COD values in the shower water from 0.25 g_{soap} L^{-1} to > 1.5 g COD L^{-1} by its decay product acetic acid. Although limited retention by NF was reported earlier for acetic acid/acetate [41], acetic acid was not detected in the modest 68 ± 11 mg COD L^{-1} remaining in the ED-diluate.

3.2. Effect of UTU on SWTU operation

Based on an environmental biotechnological approach, a ratio of COD:N:P of 100:5:1 was used to determine the growth limiting compound and the potential growth (mg biomass L^{-1}) in the SWTU as compared to a situation without urine treatment. A heterotrophic growth yield of 0.67 g COD per g COD under anoxic conditions and 1.8862 g COD g biomass^{-1} were assumed [42]. Urine has a high C:N:P concentrations compared to typical drinking water values and can thus be considered biologically unstable [43]. Due to controlled carbon and nutrient removal and recovery in the UTU, 96, 78 and 69% of COD:N:P removal from the mass balance perspective, a significant improvement in biofouling potential was achieved in the ED diluate (Fig. 2A). Nevertheless, the addition of PAA and the mixing of ED diluate and shower water increased biofouling potential compared to shower water treatment only particularly in the NF permeate due to addition of N in a nitrogen limited systems. This is in line with biofouling visually observed in the RO spacers and likely causing the recorded limitations in RO permeability. A more detailed analysis has been included in SF Section C.

Fig. 2B visualizes the results of simulations with stepwise water-removal to mimic the effect of an increasing theoretical Volumetric Concentration Factor (VCF) on the estimated precipitation of tricalcium phosphate (TCP or $Ca_3(PO_4)_2$) and struvite ($MgNH_4PO_4$) in the RO [28,44]. This figure clearly presents the significantly reduced precipitation of TCP from the addition of ED diluate instead of urine or compared to shower water alone. A similar trend can be observed for struvite. It was therefore concluded that the crystallisation reactor, operated at pH 11 with alkalinity anyhow required for the bioreactor, and subsequent addition of ED diluate to shower water successfully decreased the scaling potential in the NF and RO. A more detailed description of the underlying methodology as well as the phreeqC code has been included in SF Section C and SF Tables C.3 and C.4.

From Table 2 it can also be derived that the ED-concentrate

functioned as a sink for monovalent ions (Na^+, Cl^-, K^+, NH_4^+ and NO_3^-). From a mass balance perspective the ED removed an equivalent of 0.9 mol ions d^{-1}, which when not removed would have contributed an additional osmotic pressure of ~9 bar for the RO, when assuming 9.4 L of NF permeate under a VCF of 4 at 293 K.

3.3. Water balance

The water balance was made for a 10-day representative operational period of the integrated water treatment system, in which chemical water quality exceeded the hygienic water quality. The actual water flows were however lower than the original design values (Fig. 1A) as the both the SWRO and BWRO permeate flux was limited by a stable but limited RO permeability of 0.5 $L\text{-}m^{-2}\text{-}h^{-1}\text{-}bar^{-1}$. Further details of the measured water balance have been described in SF Table C.2. Actual daily input was 6.1 L day^{-1} and consisted of urine (19%), condensate (22%) and shower water (59%), corresponding to daily averages of 1.2, 1.4 and 3.4 L d^{-1}. In average 2.0 L d^{-1} of RO concentrate was sent back to dilute the urine. After inclusion of the BWRO membrane on day 7, this was increased to 4.6 L d^{-1}. However, as during the initial days of combined operation only 3.0 L d^{-1} of shower water was fed, insufficient RO concentrate was produced to maintain stable bioreactor operation. To stabilize operation, urine and condensate were complemented with tap water when needed to produce a stable flow of 5.8 L d^{-1} of bioreactor effluent.

Overall, 5.8 L d^{-1} entered the ED of which 0.5 L d^{-1} was used for online TAN analysis and 0.8 L d^{-1} was collected in the ED concentrate forming a potential nutrient solution [13]. The missing flow of 0.3 L day^{-1} (5%) is likely caused by sampling (more than 100 mL day^{-1}), water losses during maintenance and evaporation in the bioreactor. The waste liquid for online ammonia measurements was also lost, but contained a citric acid solution and ammonium, that could theoretically be recycled back into the bioreactor. In average, 3.4 ± 1.8 L d^{-1} of shower water was introduced into the system. This was mixed with 4.5–5.3 L d^{-1} of ED diluate. Excess ED diluate was discarded. From this flow, 0.8 L d^{-1} was bled through the NF, which was higher than the design bleed of 0.24 L d^{-1} because of draining due to overpressure. The recovery of the NF therefore decreased from the planned 95% to 92%. The remainder NF permeate was filtered over the RO, in which a 78–80% recovery was maintained. The RO concentrate produced here was sent back to dilute the urine, and was thus not lost.
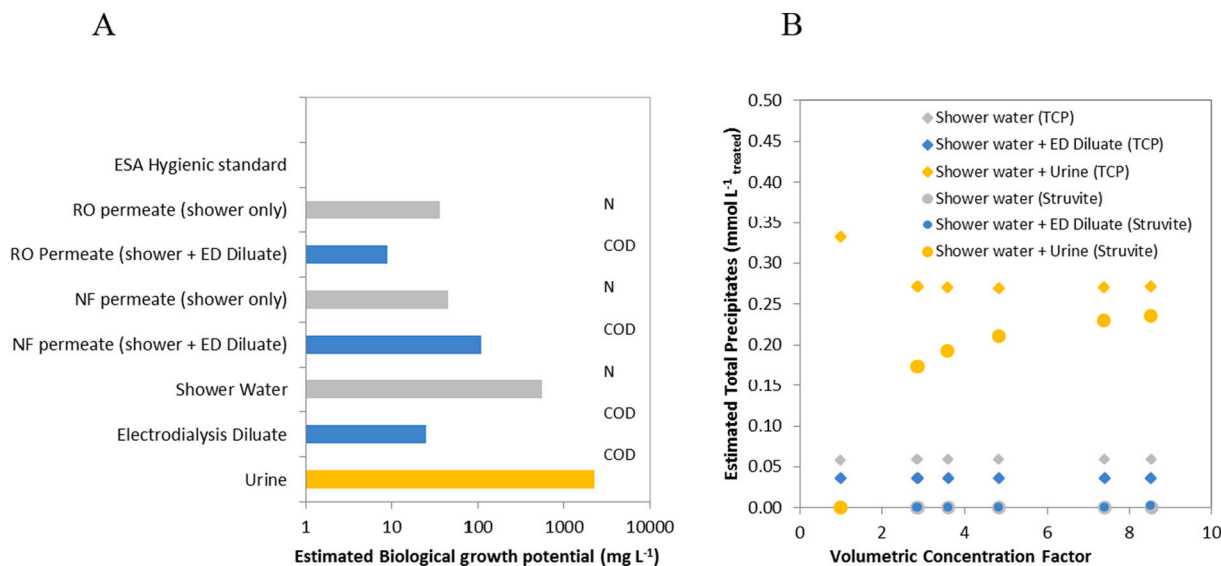
A

B

Fig. 2. A) Estimated Biological Growth potential and B) phreeqC simulations results showing the estimated total precipitates of $Ca_3(PO_4)_2$ and $MgNH_4PO_4$ in shower water, shower water + ED diluate and shower water + urine against an increase in volumetric concentration factor. TCP = tricalcium phosphate ($Ca_3(PO_4)_2$), and Struvite ($MgNH_4PO_4$).

The system produced 7.4 ± 2.5 $L_{RO\ permeate}$ d^{-1} and based on system water losses a maximum total recovery of 87% was determined. Due to the smaller flow rate than the system was designed for, a larger part than anticipated of the RO permeate was sent back to the mixing tank to maintain a minimal operational water level. Without this, permeate production would have been 5.3 L d^{-1}.

3.4. Energy balance

The designed reactor setup was not designed to test minimal energy requirements. An initial estimation of the potential energy and mass saving that could be reached by using the proposed system has been performed. A cumulative energy demand of 0.2 kWh_p L^{-1} recovered water was calculated with an energy balance constructed on assumptions regarding energy use of basic equipment on pilot and industrial scale and gross energy requirements for chemical production (SF Tables D.1 and D.2). The mayor relative contributor, with 71% of energy requirement was the nitrifying bioreactor, followed by NF (14%), RO (8%), Crystallizer (4%) and ED (3%) (Fig. 3B). A high air flow was employed in the bioreactor, higher than the necessary to supply the biological oxygen demand, in order to keep under agitation the bulk volume of the bioreactor. Only 25 g O_2 day^{-1} was stoichiometrically required for the biological conversions. In comparison, an adult astronaut is considered to require 830 g of O_2 day^{-1}. The power demand for O_2-production was nevertheless estimated at 0.1 kWh L^{-1}, 50% of the bioreactor demand (SF Table D.1).

Future energy savings could be obtained by opting for a mechanical agitation and pressurized O_2 control loop with DO feedback or by using air slug flow to reduce the required crossflow velocity, similar to what was proposed in previous work [45]. In the case of the NF and RO, the power consumption per m^3 of permeate, 13.2 and 9.7 kWh m^{-3} permeate respectively, are one order of magnitude higher than the power requirement of industrial systems designed for large flow rates. The power consumption of the RO is smaller than for the NF, because the small dimensions of a one person permeate production required a relatively high optimal surface velocity in the membranes compared to

the power needed to maintain the operational pressure. Ultimately, the osmotic pressure in the RO was reduced by ~9 bar due to the ion removal in the ED. Since additional RO energy savings and ED power supply consumption were in a similar order of magnitude, this potential mass saving opportunity appears near energy neutral.

3.5. Comparison to state-of-the-art

Given the high costs associated with mass payload significant mass saving is often described as a preliminary indication of feasibility for judging the merit of alternative water recovery systems [4]. Therefore a mass payback analysis will be performed for the WTUB and compared with the results of the study by Jones [4] on the WRS-ISS feasibility for a Mars transit mission of 450 days. A qualitative comparison of both systems is presented in Table 3.

Although this study concerned a terrestrial proof-of-concept, with 870 kg and 5.3 kg d^{-1} of measured output for one crewmember a 167 days mass breakeven date is reached for the WTUB. Allocation of mass to either urine treatment or shower water treatment is not as straightforward due to integration of the system but when basing it on input distribution 2.2 kg d^{-1} for the UTU leads to 188 days and 3.1 kg d^{-1} results in 144 days for the SWTU, excluding the spare parts. The WRS-ISS system with 3 sets of spare parts reaches a mass breakeven of 190 days but it should be noted the spare parts represent 72% of the mass in the basic WRS scenario. Although over the operational period of 4 months, no spare parts were replaced in the UTU, and a few minor parts in the SWTU, from a reliability perspective at least 3 sets of spares will need to be considered. Given the current status of the WTUB it is therefore unlikely it would lead to a shorter mass breakeven, than for the WRS-ISS. It should however be noted that the design capacity of 11.3 kg d^{-1} could likely be reached if the RO permeability limitation would be resolved by either increasing the membrane surface area or improving the management of the biofouling potential. Both appear feasible without an associated system mass increase and would thus decrease the mass breakeven date to below 100 days. Fig. 3 compares the mass payload per crewmember, assuming an input of 13 L CM-d^{-1}
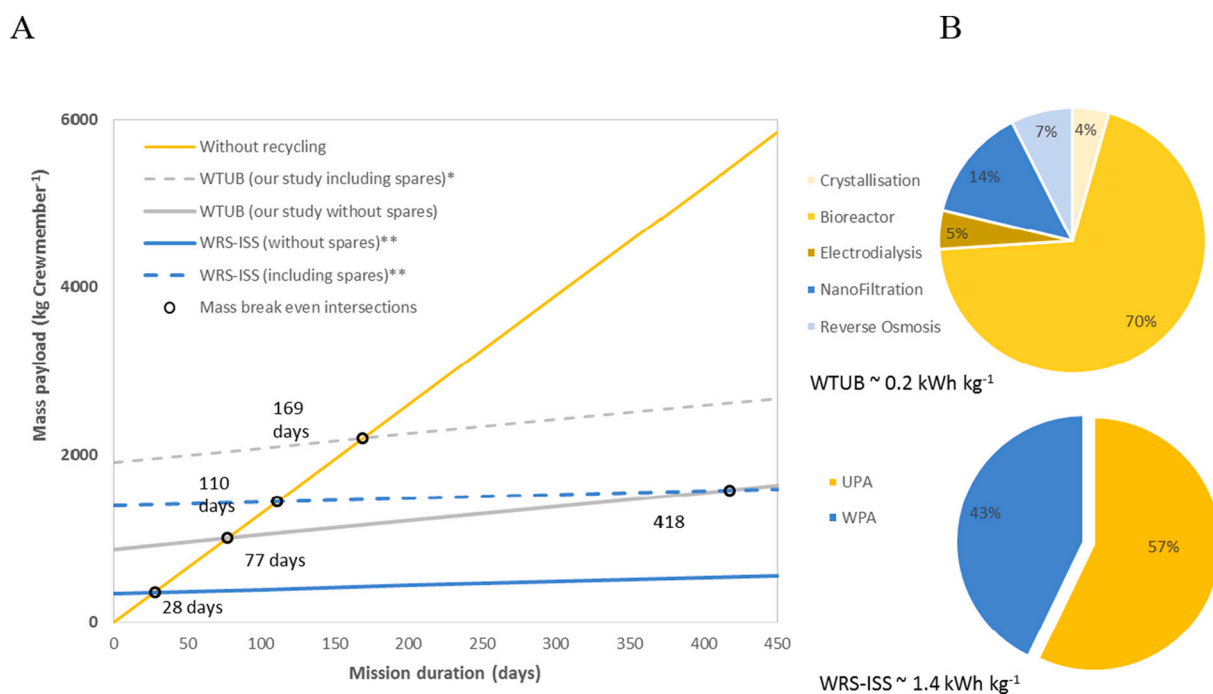
A   **B**

Fig. 3. Comparison of A) estimated mass payload per crewmember and B) mass specific energy demand (kWh kg^{-1}) against mission duration without and without spare parts for the WRS-ISS, WTUB and a no recycling scenario. *an identical spare sets mass per CM is assumed as for the WRS-ISS; **data from Jones [4] was normalized against the design capacity of the WTUB with an input of 13 kg CM-d^{-1}.

Table 3

Main characteristics of WTUB (this study) and the Water Recovery System of the International Space Station for a 4 crewmember 450 day transit mission to Mars (adapted from [4,11,46].

	WTUB	WRS-ISS
System units	UTU and SWTU	UPA and WPA
Core mechanism	UTU: Chemically induced crystallisation, biological nitrification and organic carbon oxidation, electrodialysis	UPA: Phosphoric acid dosage, Cr(IV)O3 based oxidation, VCD
	SWTU: PAA, NF and RO membrane	WPA: Particle filter, multifiltration beds, chemical oxidation and IEX
Water sources	Condensate to UTU; Shower water included	Condensate to WPA; Shower water excluded
Technology readiness level	6	9
Current design capacity (CM)	1	4
System mass (kg)	870 (Total); 420[a] (UTU); 450[a] (SWTU)	1383 (Total); 742 (UPA); 641 (WPA)
Required spare parts mass (kg)	Unknown	3540[b]; 4878[c]
Total water input/output (kg)	2745[d]; 2362[d]	10,026; 9213
Water recovery (%)	87% (Total); 86% (UTU); 91% (SWTU)	92% (Total); 80% (UPA); 99% (WPA)
Power consumption (kWh L^{-1})	0.2 (Total); 0.1–0.2 (UTU); 0.0–0.1 (SWTU)	1.4 (Total); 0.8 (UPA); 0.6 (WPA) [46]
Main operational issue	Biofouling –limited RO permeability	Scaling and fouling of UPA
Mass breakeven date (days)	167[e]	190(WRS)[a]; 319 (UPA)[a]; 132(WPA)[a]

[a] Mass equally allocated due to system integration and feedback loops.
[b] Scenario with 3 spare sets.
[c] Scenario with 0.001 failure rate based on real WRS-ISS performance.
[d] Based on one CrewMember (CM).
[e] No spares included.

against mission duration for a scenario without recycling, the WRS-ISS and the WTUB. It demonstrates the potential for significant mass savings against the no recycling scenario of the WTUB. It also identifies the gap with the WRS-ISS after the design capacity is reached and shows that the focus for future studies should be on upscaling to 4 crewmembers while minimizing the mass payload per crewmember and identifying the exact spare set requirement for sufficient reliability.

Also from an energy perspective the WTUB system (Fig. 3B) shows energy savings potential with an estimated cumulative energy demand (CED) of ~0.2 kWh kg^{-1} compared to the WRS-ISS ~1.4 kWh kg^{-1}. Particularly the UTU with required oxygen generation included, 0.1–0.2 kWh L^{-1} appears efficient compared to the 0.8 kWh L^{-1} of the VCD based UPA. Moreover it does not use toxic chromium(IV).

A last point to be taken into consideration is that the water quality assessment focused on basic chemical parameters so far, and did not yet include determination of bacterial or viral pathogens or micropollutants like pharmaceutical and personal care products (PPCP). For terrestrial potable water reuse systems, typically a multi-barrier approach and a monitoring-based risk mitigation strategy is in place as described elsewhere [47]. Although technically, a combination of UF, NF and RO should provide sufficient redundancy to ensure risk mitigation by such constituents, a follow-up study in the development of this treatment train could focus on detection potential membrane ruptures, along with for instance testing an additional barrier like UV treatment.

4. Conclusions

This study provided a proof-of-concept that the automated five stage biological membrane based treatment train (WTUB), can achieve a 87% ± 5% water recovery efficiency from urine and shower water without using anti-scalant and chromium(IV)with an estimated cumulative energy demand of 0.2 kWh L^{-1}, while meeting ESA's chemical hygienic water quality requirements.

Lowering RO recovery to around 80% and using the concentrate for urine dilution instead of flush water was key to reducing total system losses. The application of the ED reduced the osmotic pressure encountered in the RO by 9 bar and thus saved mass while recovering nutrients in a concentrated form. Geochemical speciation modelling showed that the scaling potential was kept under control through the crystallisation reactor and a 95% reduction in biofouling potential in the NF permeate could be attributed to the addition of the bioreactor.

Further optimization in TAN and nitrate removal, respectively by

the nitrification reactor and ED unit, is needed to maintain hygienic water quality during long-term operation. In order to reach the design input capacity of 13 L d^{-1}, the output limitation resulting from the stable but low RO permeability of 0.5 L m^{-2} h^{-1} bar^{-1} should be resolved.

Despite some anticipated shortcomings, already in this early stage of development WTUB shows mass break even dates for a hypothetical transit mission to Mars of in the same order of magnitude as the WRS-ISS. Overall, the biological membrane-based water recovery therefore showed to be a promising alternative to current energy-intensive heat-based systems for water recovery in future regenerative life support systems.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.desal.2020.114634.

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