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INNOVATION, ADAPTATION AND ENGAGEMENT IN A CHANGING WORLD

**Gravity-driven membrane disinfection
for household drinking water treatment**

M. Peter-Varbanets, R. Johnston, R. Meierhofer, F. Kage & W. Pronk, Switzerland

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Ultrafiltration (UF) has been proven to be very effective in the treatment of water for the removal of particles, colloids and microorganisms. However, household application of UF is limited due to membrane fouling which results in complex and maintenance-intensive UF systems. In gravity-driven membrane disinfection (GDMD) technology, a stable membrane flux of 4-10 L.h⁻¹.m⁻² is observed during ultrafiltration without any backflushing, chemical cleaning or an external energy supply for over 24 months, while operated at relatively low pressures (40-65 cm of water column). This novel approach to operate UF systems at stable flux conditions can be considered an important breakthrough in membrane technology, as it allows development of a robust, maintenance-free, low-cost and user-friendly household water treatment system, which has a great potential for implementation.

Introduction

Household water treatment and safe storage (HWTS) has been associated with marked improvements in drinking water quality and reductions in disease (Fewtrell et al, 2005). However, time-consuming operation and maintenance, aesthetic concerns, limited effectiveness, high costs of existing technologies and lack of consideration of consumer preferences limit scale-up of HWTS. Recently some studies were published which evaluate efficiency as well as consumer preferences of existing HWTS systems. Albert et al., (2010) analysed user preferences for three household water treatment technologies i.e. diluted hypochlorite solution, a combined powdered flocculant-disinfectant mixture and porous ceramic filter in rural communities in Kenya. The study showed that households ranked filters most frequently as their most preferred product (Albert et al., 2010). The reasons of the choice were found to be potential durability of the filter and ease of use. Also USAID study of consumer preferences in Nepal showed that colloidal silver (CS) candle filters were most preferred option, although least affordable for the choices given (USAID, 2006). However, ceramic filtration was less effective in reduction of *Escherichia coli*, than other technologies tested (1-log (Albert, et al., 2010) to 2-log (Lantagne et al., 2009). Furthermore, ceramic filters are not expected to remove viruses as the pore size of ceramics is an order of magnitude higher than the size of a virus. Thus, some studies show that consumers prefer filters to other HWTS technologies, although the efficiency of this technology is of concern. Besides ceramic filtration, the choice of HWTS filters is limited. Some filters are available for middle-income population in intermediate countries (i.e. Tata-Swatch, PureIt from Hindustan Unilever, India). However, most of the filters are not widely available in other countries yet, and information is limited. Furthermore, the filters require regular replacement of cartridges, which lead to high operational costs. Thus, the potential of alternative filtration technologies should be considered and evaluated for HWTS applications.

Ultrafiltration (UF) provides an effective barrier for microorganisms, suspended particles and colloids and is increasingly implemented for the treatment of drinking water worldwide. For drinking water treatment, usually UF membranes are used with a pore size around 10 nm, which provide at least 7-log removal of bacteria and 3-log removal of viruses. Conventional large-scale ultrafiltration systems are operated at a transmembrane pressure of around 0.5 - 1.0 bar and require pumps for operation and backflushing. Decentralized systems designed in a similar manner are expensive due to the relatively high costs of

peripheral equipment (Peter-Varbanets et al., 2009). If operated by gravity, pump costs are avoided, and this can be an attractive option for decentralized, small-scale applications. Presently, only few gravity-driven ultrafiltration systems for decentralized application exist (SkyJuice for community water supply and LifeStraw Family for household water treatment (Peter-Varbanets et al., 2009, Clasen, et al., 2009)). These systems can be operated at ultra-low pressure (100-150 mbar) and require little maintenance compared to the conventionally operated UF (Peter-Varbanets et al., 2009). To control membrane fouling and prevent biofouling, irregular backflushing and disinfection with slow eluting chlorine tablets is used in LifeStraw (Clasen et al., 2009) and manual flushing and chemical cleaning is applied in SkyJuice (SkyJuice, 2009). However, no system is known so far, which are not dependent on cleaning or flushing.

We developed an alternative approach to UF systems. The pressure head is generated by gravity, and this gravity-driven membrane disinfection (GDMD) system is operated in a dead-end mode without any pre-treatment, flushing or cleaning. As a feed, natural water (river, spring, well or rainwater) can be used without pre- or post-treatment. According to generally accepted membrane filtration theory, operation of such a system on a long term leads to fouling and biofouling and results in flux decline and clogging of the membrane. In contrast to this common view, flux stabilization is observed at flux values of 4-10 L.h⁻¹.m² at hydrostatic pressure of 65 mbar or less for periods of at least 2 years. This phenomenon was not observed before and was first documented in our previous study (Peter-Varbanets, et al., 2010).

The objective of the paper is to evaluate the potential of this novel approach to operate UF systems for household water treatment. This paper summarizes and evaluates results of the previous studies and presents new data regarding membrane integrity, system design and cost estimation. In the outlook, the objectives of the new GDMD project started at Eawag (Switzerland) in collaboration with Kenya Water for Health Organization (Kenya) are summarized and the first results of this project will be presented at the conference.

The phenomenon of flux stabilization

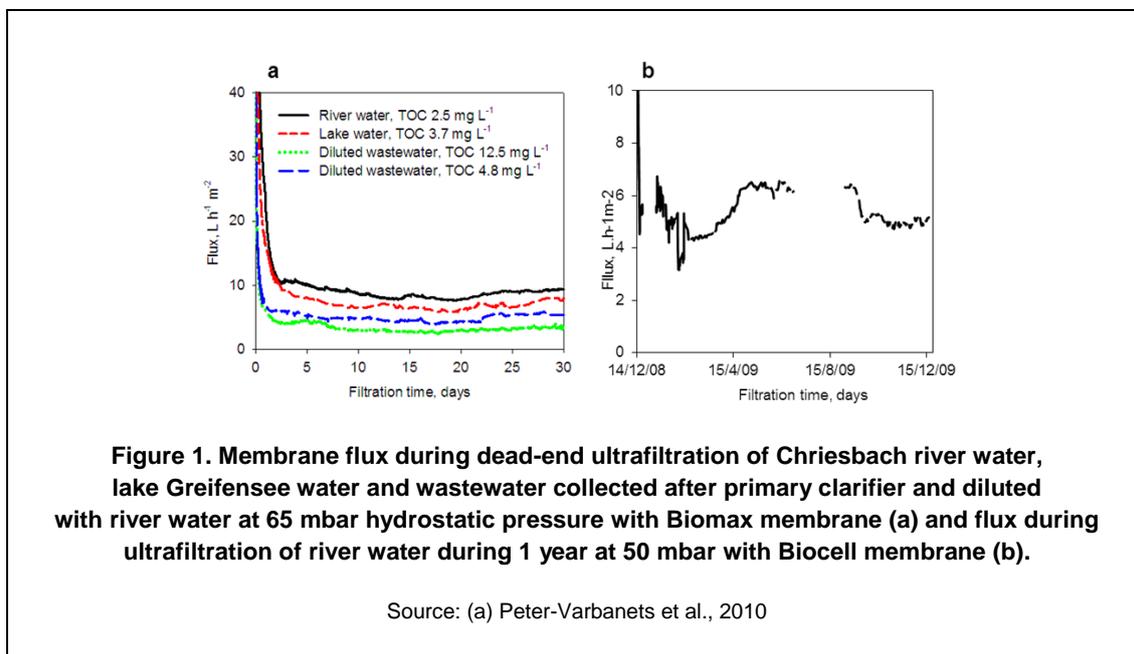
Figure 1 shows that flux stabilization was observed after 7-10 days of filtration through UF membrane (Biomax® Millipore, Material: polyethersulfon (PES), membrane cut-off: 100 kiloDaltons (kDa), which corresponds to a pore size of 10-15 nm, surface area 16.6 cm²) with different water types, including river water (Chriesbach, Switzerland), lake water (Greifensee, Switzerland) and diluted wastewater (10-30% Primary Effluent, Duebendorf, Switzerland). Depending on the water type, stable flux values were in the range of 4 - 10 L.h⁻¹.m² at a gravitational pressure of only 65 mbar. The flux remained stable during at least 120 days with all feed water types (the first 30 days are shown in Figure 1a) and was not affected by increase of turbidity of water up to 30 NTU during rain events. Figure 1b shows flux values measured during 1 year of continuous ultrafiltration of Chriesbach river water without flushing or cleaning at 50 mbar hydrostatic pressure with Biocell membrane from Microdyn-Nadir (Membrane material: PES, cut-off 150 kDa, surface area 285 cm²). Although fluctuations of flux occurred during filtration period, the flux values were dominantly in the range of 4-6 L.h⁻¹.m². This system is still in operation (about 2 years in total).

The impact of water quality on stable flux has been further evaluated. Increased levels of TOC in raw water led to the stabilization of flux at lower values (Peter-Varbanets et al., submitted (a)). However, flux stabilization was observed in all cases as long as water remained aerobic. Decline of dissolved oxygen values to <1mg/L resulted in a continuous decline of flux. A small community system which had a capacity of 4 m³/day has been operated in South Africa (Ogunjini, KwaZulu-Natal) for 6 months. During operation, raw water turbidity fluctuated and turbidity values up to 1000 NTU were measured. Turbidity values of up to 100 NTU did not influence the stable flux significantly while turbidity peaks of 600-1000 NTU resulted in a decline of flux (Boulestreau et al., 2010). Laboratory experiments with river water containing kaolin showed that the system can be operated at stable flux conditions when raw water turbidity does not exceed 300 NTU (Peter-Varbanets et al., submitted(a)).

Further investigations showed that although initial flux of the clean membrane is proportional to pressure, the stable flux values do not considerably depend on pressure (see Peter-Varbanets et al., 2010). Thus, increase of pressure for UF systems operated at stable flux conditions does not lead to a considerable increase of production capacity of the system. This explains why the phenomenon of flux stabilization has been overlooked so far: most previous fouling studies have been conducted at higher pressures and shorter periods of time. At higher pressures, the values of stable flux which might have been observed were less than 5% of the initial flux and therefore would have been disregarded.

The phenomenon of flux stabilization can be considered an important breakthrough in membrane technology, as in conventional UF processes regular backflushing and cleaning is required, which results

into complex systems and hinders decentralized application of UF. The mechanisms of flux stabilization and evaluation of the technology for household water treatment are discussed in the following sections.



The mechanisms of flux stabilization

The mechanisms of flux stabilization were studied by microbiological investigations combined with confocal laser scanning microscopy (CLSM) (Peter-Varbanets et al., 2010). When new membranes were first used to treat water, a fouling layer rapidly developed on the membrane surface, causing an initial decline of flux. However, after about 1 week of filtration, the development of cavities, channels and other heterogeneous structures was observed in the fouling layer first with confocal laser microscopy on microscale (Figure 2b) and later on macroscale by direct observations (Figure 2a). The development of heterogeneities was monitored over time with CLSM and it was concluded that the heterogeneity of the fouling layer increased over time. Due to preferential flow through the channels in the fouling layer, the resistance decreased. This process counteracted the increase of thickness of the fouling layer, leading to an increase of resistance. Thus, the stabilization of resistance and flux were observed.

It was also shown that the formation of channels and other heterogeneous structures was related to the biological activity in the fouling layer and was not observed when biological activity was suppressed by addition of disinfectants (e.g. sodium azide) or at low temperatures (4°C) (Figure 2c). In this case, flux declined steadily and no stabilization was observed. From these results, it was concluded that biofouling, always assumed to be a major limitation of ultrafiltration, actually causes stabilization of flux in case of gravity-driven membrane disinfection.

Impact of membrane type and cut-off on flux and removal of microorganisms

Membranes serve as absolute physical barriers and the retention of protozoa, bacteria and viruses depends on the pore size or cut-off of the membrane. In general, most microfiltration (MF) and UF membranes retain protozoa, tight MF membranes (pore size < 0.22 µm) provide complete removal of bacteria, and tight UF membranes (cut-off < 100 kDa) retain both bacteria and viruses. Nanofiltration (NF) membranes completely exclude viruses, colloids and some micropollutants.

Permeability also affects the selection of a membrane. In general, membranes with lower cut-offs have lower permeability which reduces capacity of the system. However, in GDMD technology stable flux does not depend on the membrane cut-off for MF and UF membranes tested (Figure 3). For tight UF membrane UP010 and NF membranes NF270 the initial flux at pressures used in the experiment is lower than the stable flux for other types of membranes which causes stabilization of flux at lower values. Thus, in general our results show that any type of membrane can be used, when the initial membrane flux is higher than the stable flux value.

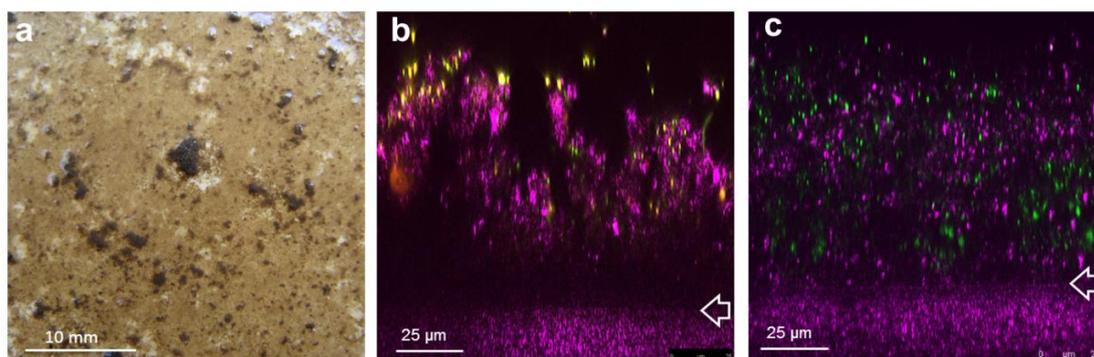


Figure 2. Figure 2. The fouling layer structure formed during ultrafiltration of river water for 27 (b, c) and 44 (a) days without (a, b) and with (c) suppression of biological activity
 The arrows show the separation plane between the membrane and the fouling layer. Green – SYBR® Gold stain, indicating presence of all bacterial cells; Red - Concanavalin A stain indicating presence of α -D-mannose and α -D-glucose groups in biopolymers; Purple - reflection of the solid surfaces

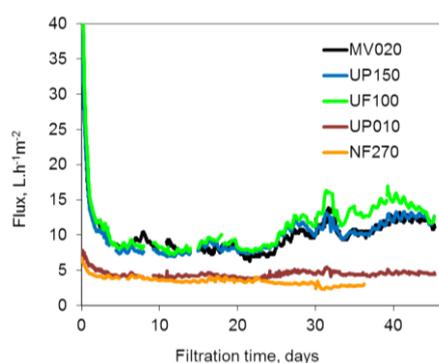


Figure 3. Impact of membrane cut-off on stable flux during filtration of Chriesbach river water. MF membrane MV020 had a pore size of 0.20 μ m, UF membrane UP150 had a cut-off of 150 kDa, UF100 membrane had a cut-off of 100 kDa and UP010 had a cut-off of 10 kDa

These membranes were operated at hydrostatic pressure of 65 mbar. NF membrane NF270 was operated at 900 mbar

The structural integrity of membranes and membrane modules was examined using flow cytometry - a novel highly reliable and fast method to measure total cell count in water samples (Berney et al., 2008; Hammes et al., 2008). The procedure was as follows: *Brevundimonas diminuta* (ATCC 19146), a standard filter testing organism of the American Society for Testing and Materials (ASTM, 2005), was cultivated in Luria-Bertani medium (30°C, 48 h). The bacteria were stained with SYBR® Green I, separated by centrifugation (3000 rpm), re-suspended in 1 mL of tap water and diluted with river water to a final cell concentration of $3 \cdot 10^5$ cells/mL. This solution was passed through several membrane modules which had been treating river water prior to the experiment for 30 days. The fluorescent bacterial count was measured in the feed as well as in the permeate vessel at 0, 20 and 50 minutes after start of the permeation by flow cytometry according to (Berney et al., 2008). As shown in Figure 4, the amount of organisms detected in the permeate was below the detection limit, which confirms that the membrane and membrane module were intact and did not have any leakage.

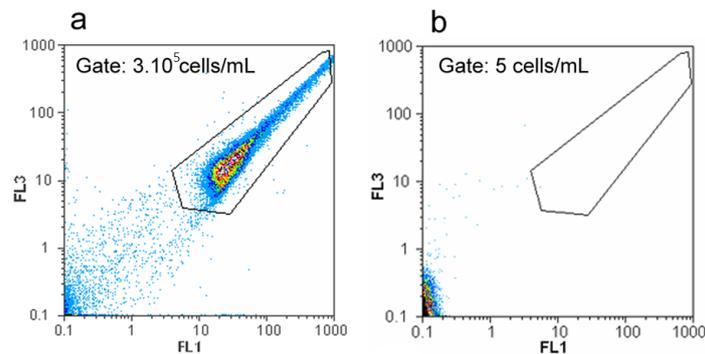


Figure 4. Flow cytometry dot plots of the feed (left) spiked with fluorescently labeled *B. diminuta*, and the permeate (right) collected after 50 min filtration through an UF membrane with a cut-off of 100 kDa. FL1 denotes green fluorescence intensity (520 nm) and FL3 denotes red fluorescence intensity (615 nm) in arbitrary units

The impact of long term operation on the retention properties of the fouled membranes was studied. MS2 phage (ATCC 15597-B1, DSMZ) was used in this study. The procedure of this experiment was as follows: 50 ml of a phosphate buffer solution containing MS2 phage (2×10^7 phages/ml) was filtered through an UF and a MF membrane placed in standard filter holders (48 mm inner diameter, Whatman). Permeate was collected during approx. 3 hours of filtration. Counts of MS2 phages in the cumulative feed, permeate and room temperature control samples were determined for each membrane in triplicate using a standard double agar layer assay (Adams, 1959; methodology provided by DSMZ). The detection limit of this experiment was 6.1-log reduction of MS2 phages. An UF membrane with a cut-off of 150 kDa, (approx. $0.04 \mu\text{m}$ according to producer) and a MF membrane with a pore size of $0.2 \mu\text{m}$ were selected for this experiment. The pore size of the selected membranes exceeded the radius of MS2 phage (about 24–25 nm), and thus little retention of MS2 phage was expected for new membranes. Indeed our results showed 1-log reduction of MS2 in the permeate for both membranes. However, after 5 weeks of operation, a biological layer of approx. 40–50 μm had formed on the surface of both membranes. Filtration of MS2 through the fouled membranes showed 4-log reduction for UP150 membrane, while for MV020 membrane MS2 removal remained the same. These results indicate that the development of the fouling layer on the membrane surface does not affect or even improves removal of MS2 bacteriophage. However, in order to assure complete disinfection of the water from the start, membranes with a pore size of max. 20 nm should be used and their integrity should be tested experimentally.

Evaluation of GDMD technology for applications in household systems

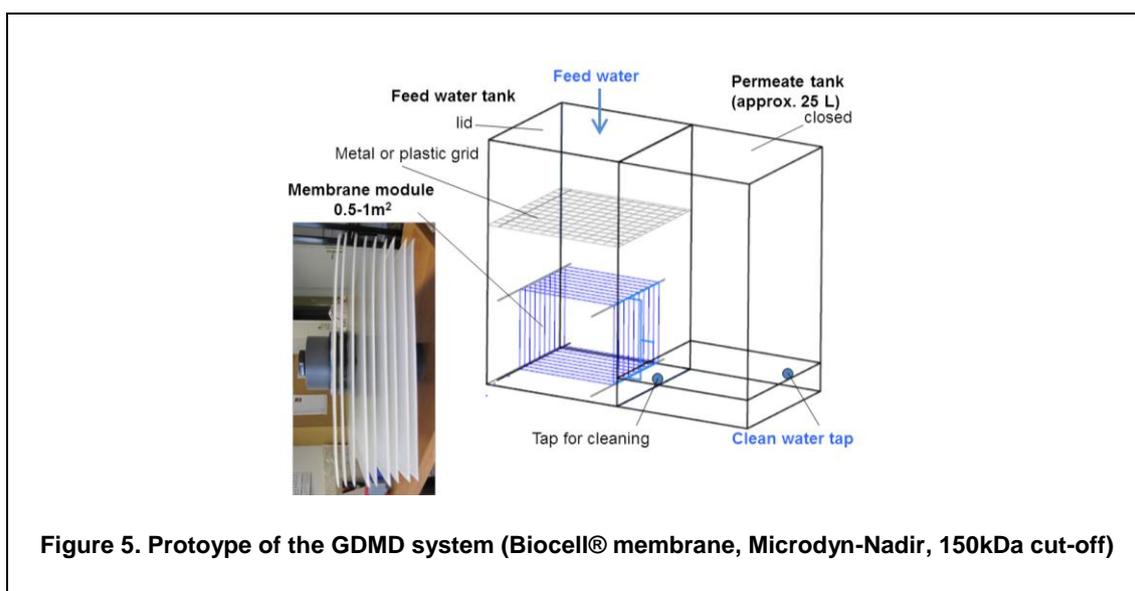
Summarizing, our results show that the phenomenon of flux stabilization is observed during dead-end ultrafiltration of natural waters without back-flushing, cross-flow, chemical cleaning or disinfection at hydrostatic pressure of 50–65 mbar. UF systems, based on the described phenomenon of flux stabilization would not require any back-flushing, cross-flow, chemical cleaning or disinfection, which would greatly simplify the maintenance and operation of ultrafiltration systems and reduce operational costs of the system almost to null. The pressure required for ultrafiltration can easily be obtained by gravity in most situations. This makes GDMD attractive for drinking water treatment on a household scale.

System configuration and design

Figure 5 shows one of the possible configuration of a household system. In this system, feed and treated water (permeate) tanks are assembled next to each other, with a membrane immersed in the feed water tank and protected from user by a plastic or metal grid. This configuration prevents overflow of the permeate tank as well as protects membrane from complete drying. Complete drying of the membranes can damage membrane integrity and should be prevented. However, drying does not occur when only part of the membrane remains submerged in water due to capillary flow of water through the entire membrane. In the

system shown on Figure 5, the permeate collection pipe is located in the middle of the membrane module, which keeps part of the membrane always submerged. The tap for cleaning shown on figure 6 can be used to drain the system in order to remove accumulated sediments as needed.

Another possible configuration of the GDMD system is a two bucket type of system similar to ceramic candle units. Disadvantages of this configuration are that overflow of water in the permeate tank could occur, and special adaptations would be needed to prevent the membrane from drying. Advantages include higher capacity of the system due to higher transmembrane pressure at the end of the cycle, and possibly a smaller footprint. Both configurations should be further evaluated and optimized.

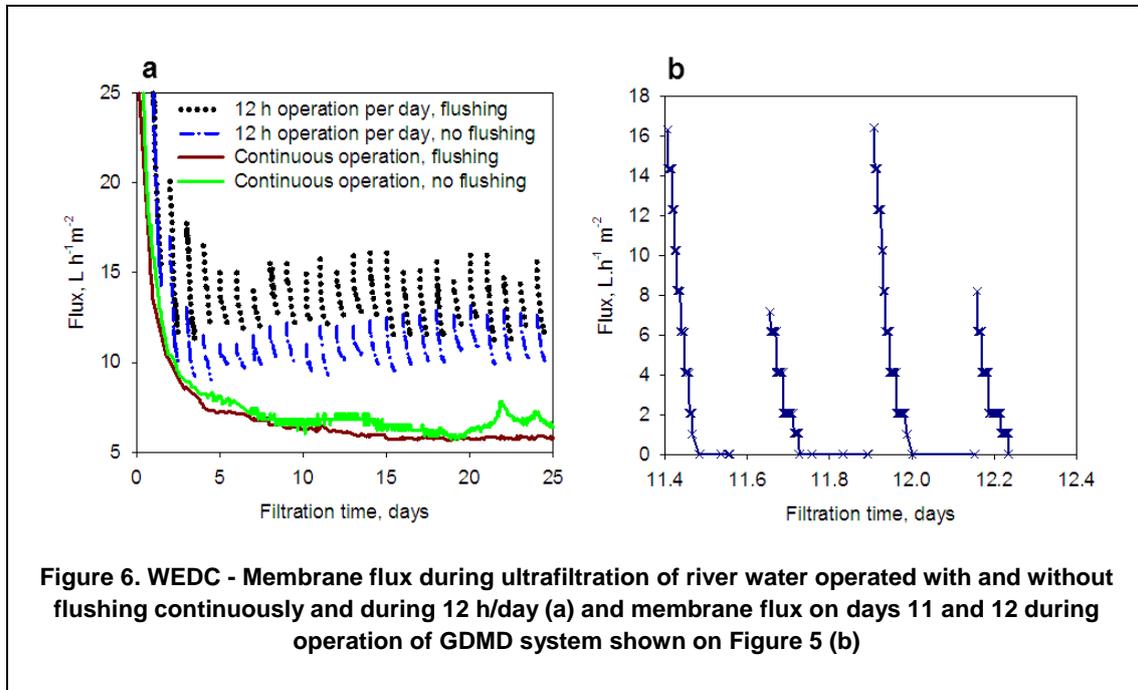


The impact of intermittent operation and changing pressure conditions on flux in GDMD system was studied and the results showed that intermittent operation results in an increase of flux at the beginning of the filtration cycle as well as average flux of the system (Figure 6a). This process can be further intensified when the water level in the feed water tank decreases and the fouled membrane is exposed to air. Partial draining of the system leads to formation of a more open biological layer and causes its sloughing from the membrane surface (Peter-Varbanets et al., submitted (b)). Furthermore, regular exposure of the biological layer to oxygen prevents risk of anaerobic conditions which can decrease flux or even lead to clogging (Peter-Varbanets et al., submitted (a)). Figure 6b shows flux measured during operation of a system shown on Figure 5. The first and third filtration cycles were measured after the system was filled with water but the clean water tap closed. After the end of these cycles (the level of water in feed and permeate tanks was equal), the clean water tap was opened, water discharged and filtration continued (second and fourth filtration cycles on figure 6b). The flux of zero observed between filtration cycles reflects hydrostatic equilibrium between the raw and permeate tanks, not biofouling. Figure 6b shows that initial flux measured after the system is filled with water is about 16 L/hm², which is approximately double the stable flux usually observed during continuous operation under constant pressure (about 6-8 L/hm²). Thus, intermittent operation and draining leads to an increase of peak flows of the system.

System costs

Evidently, for GDMD, the required membrane area is higher than for conventional UF. However, assuming that the required capacity for drinking and cooking water is about 20-40 L/day per family, and stable flux of 4-10 L·m⁻²·h⁻¹, the membrane area needed is less than 0.5 m². Increase of flux observed during intermittent operation leads to an increase of flux at the beginning of the filtration, and thus peak flow rate significantly higher than 2 L/h for waters with high TOC and turbidity is expected in GDMD system with 0.5m² of the membrane. Membrane costs have decreased significantly in the past decade and continue to decline. At present, high quality membranes for a market price of < 40 US\$/m² are available. The membrane service life expectancy is estimated to be approx. 7-8 years in large-scale applications. For GDMD, maximum duration of our laboratory experiments has been 2 years so far, and thus the laboratory data are not sufficient to

predict service life of the membranes in GDMD system. However, in GDMD no chemical cleaning is used and higher service life could be expected comparing to large scale applications. Assuming the service life of 8 years and membrane costs of 20 US\$, the average annualized cost will be about 2.5 US\$ per household per year for the membranes. If the housing of the system is produced locally it is expected to add at least 5-10 US\$ to the system costs, depending on the type of production and materials. Thus, the total costs are expected to be about 30 US\$ or 3.75 US\$ per household per year.



Outlook

Summarizing, our results show that GDMD technology has a great potential for implementation in simple and durable household systems which do not require external energy, addition of chemicals, replacement of membrane, or any other type of maintenance. However, further investigations and field evaluation are needed in order to evaluate limitations of the system with regard to water quality, system design and operation requirements, as well as user perception of the filter. Although annual water treatment costs are estimated to be significantly lower than affordability limit for the poorest part of the world population (10 US\$ per household and year (Sobsey, 2002)), high investment costs could represent a significant barrier to adoption of GDMD filter and solutions are needed to overcome this barrier.

The goal of the Gravity-Driven Membrane Disinfection (GDMD) project, started at Eawag in July 2010, is to develop, design, produce and implement GDMD system for low- and middle-income population in Kenya considering economic, technical and social constraints. In this project, the performance of GDMD systems will be evaluated in Nairobi and Kisumu, Kenya in cooperation with an NGO: Kenya Water for Health Organization (KWAHO) and involvement of the private sector.

The focus of the project is on technical evaluation of the technology as well as demand and supply side issues of the product development. Membrane modules and housing units will be further adapted and optimized depending on the end-user requirements and preferences. The potential for local production of housing and assembly of the systems will be assessed in close collaboration with the private sector. Potential distribution mechanisms, including supply chains and long term support will be assessed and marketing strategies developed in order to allow sustainable implementation of the system. Interventions to create demand including commercial marketing, behavioural change through household promotion and on-going interventions to maintain new behaviours will be developed and evaluated in case studies. Novel financial models to overcome high investment costs for end-users and distributor will be assessed and further developed. First results and outcomes of this project will be presented at the conference.

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Keywords

Household water treatment, ultrafiltration, membrane disinfection.

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Contact details

Maryna Peter-Varbanets

Eawag – Swiss Federal Institute of Aquatic Science and Technology, P.O.611 CH-8600 Dübendorf, Switzerland.

Tel: +41 44 823 5074 Fax: + 41 44 823 5399

Email: maryna.peter@eawag.ch www.eawag.ch

Rick Johnston

Eawag – Swiss Federal Institute of Aquatic Science and Technology, P.O.611 CH-8600 Dübendorf, Switzerland.

Tel: +41 44 823 5011 Fax: + 41 44 823 5399

Email: richard.johnston@eawag.ch www.eawag.ch