



Renewable energy powered membrane technology: Fluoride removal in a rural community in northern Tanzania



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ARTICLE INFO

Article history:

Received 10 April 2015

Received in revised form 20 May 2015

Accepted 22 May 2015

Available online 23 May 2015

Keywords:

Community case study

Fluoride removal

Fluorosis

Nanofiltration

Reverse osmosis

Water supply

Photovoltaics

ABSTRACT

Water samples from 13 sites in the Oldonyosambu Ward in Northern Tanzania were collected and comprehensively analyzed for fluoride and other components. Local water resources are typically of sodium carbonate (Na-HCO_3) type with low calcium (Ca^{2+}) concentrations. Water sources contain excessive fluoride concentrations and are used as the principal water supply for drinking and cooking by the rural communities. The occurrence of fluoride has severe health impacts in the population from babies (pre-natal) to the elderly, resulting in severe dental and skeletal fluorosis in this area.

Purification technologies suitable for such situations in very rough rural communities are scarce due to the absence of electricity supply, infrastructure and operational skills. The development of advanced, yet appropriate technologies is hence a novel separation and purification challenge in that somewhat established technologies need to be adapted to new environments. To this effect two types of experiments were conducted:

- (1) The water with the highest fluoride concentration (Site 4, 17.9 mg/L, WHO guideline 1.5 mg/L, Tanzanian guideline 4 mg/L) was treated with different nanofiltration (NF) and reverse osmosis (RO) membranes in batch-scale experiments. Results show that fluoride and other components including sodium, potassium, calcium, magnesium, carbonate, sulfate, silica, and organic matter were removed reliably from these natural waters by NF/RO.
- (2) A pilot-scale autonomous membrane system powered directly by solar energy was used to treat the water with the highest fluoride concentration and examine the impact of fluctuating energy on three different membranes (BW30, NF90, NF270). The results demonstrated that the BW30 and NF90 membranes achieved the WHO guideline, while the NF270 membrane bettered the Tanzanian fluoride guideline. The total permeate volume produced with these membranes was 1000, 2444 and 2656 L, respectively. The resilience of the system to solar fluctuations was good during the short term tests.

The results suggest that such advanced membrane technologies powered directly by renewable energy are an attractive option for addressing the water problems in rural areas. While the technology is well established, the adaptation to villages such as those in the Oldonyosambu Ward is far from achieved. This requires further work in terms of integration, technology adaptation and operation and maintenance schemes.

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1. Introduction

The location for this case study is the East African Rift, which is a naturally high-fluoride zone [1–3]. The fluoride (F^-) concentrations are up to 2800 mg/L in local soda lakes and as high as 330 mg/L in groundwater [2,4]. The abnormal fluoride contents originate from geothermal weathering of fluoride-rich volcanic rocks and are further concentrated by evaporative processes in arid climate [1]. Calcium concentrations in these lakes, which would typically limit fluoride concentrations through fluorite precipitation (CaF_2), are very low due to precipitation of carbonate phases in a highly alkaline environment [5,6]. In fact, the lakes and groundwaters are typically characterized as $Na-HCO_3$ waters as a result of the reaction of dissolved carbon dioxide with the rocks [6]. Therefore, fluoride concentrations of extremely high levels are found and limited only by the saturation of highly soluble villaumite (NaF).

The significance of fluoride in human health has always been a subject of debate. The World Health Organization (WHO) recommends a fluoride guideline value in drinking water of 1.5 mg/L [7]. Although small amounts of fluoride, less than 1 mg/L, are known to be beneficial for human health in preventing dental caries, high fluoride concentration in water causes dental fluorosis (1.5–4 mg/L), skeletal fluorosis (4–10 mg/L) and crippling fluorosis (>10 mg/L) [7]. Research has also indicated that fluoride can cause cancer, brain damage and genetic disorders [8]. In East Africa alone, over 80 million people show symptoms of fluorosis [9].

Current methods of fluoride removal from drinking water include membrane filtration, adsorption, co-precipitation, ion-exchange and electrodialysis [7]. Nanofiltration (NF) and reverse osmosis (RO) are very applicable membrane technologies due to their high fluoride retention [10]. This is especially so when concentrations are relatively high and adsorption media saturate quickly, or when other contaminants interfere with adsorption. NF and RO involve a number of separation mechanisms including size exclusion, charge repulsion and solution-diffusion [11]. Fluoride retention by NF/RO depends on solute and membrane characteristics as well as water quality and operating conditions (especially pH and pressure) [12]. For remote and developing areas, one great advantage of NF/RO is that it can be used in decentralized units, either at household or community level, as well as the simultaneous removal of bacteria and viruses [9].

However, a primary concern of applying NF or RO units is the requirement for electricity as this is either not available or unreliable. This problem was resolved by using a membrane system that is directly powered by photovoltaics. A further concern is the

integration of such advanced technology in a rural development context. The management and operation of such systems requires regular monitoring, maintenance procedures and accessibility to spare parts for repairs. The required infrastructure is typically not yet in place for supporting such technology in sub-Saharan African countries [13]. While this is not an unusual problem for new technology, the gap is wide and this case study is a humble beginning in raising awareness of possibilities and challenges.

A remote community in the Oldonyosambu Ward, northern Tanzania, was selected for the case study due to the predominance of fluorosis and the availability of information on water management in the area. The main objectives of the study are to;

- (1) evaluate the occurrence of different inorganic and organic compounds in local drinking water sources, with an emphasis on fluoride;
- (2) identify the prevalence and severity of fluorosis symptoms among populations of the community;
- (3) investigate the performance of different NF and RO membranes in removing fluoride and other compounds at bench scale;
- (4) determine the short term performance of a decentralized and directly solar powered membrane system in terms of water quantity, quality and energy efficiency, and
- (5) assess the potential of implementation and likely challenges to membrane systems in this rural area.

2. Case study overview

2.1. Location

The Oldonyosambu Ward is located in the Arumeru District of northern Tanzania (see Fig. 1A) and in the north-western sector of the Mount Meru natural system [13]. The Oldonyosambu Ward includes three villages which are Oldonyowas, Lemongo and Losinoni. Oldonyowas and Lemongo are about 30 km north of Arusha city on the road to Nairobi, while Losinoni is located circa 8 km further east from the main road, toward the Ngarenanyuki Ward.

The ecology of the area belongs to the Southern Acacia-Commiphora bushlands and thickets ecoregion [14]. The climate is tropical with a bimodal rainfall pattern. The long rains occur from March to May and the short rains from November to December, erratic rainfall patterns have occurred more frequently in recent years. Mean rainfall is 600–800 mm annually through most of the region [15]. Mean maximum temperatures are around

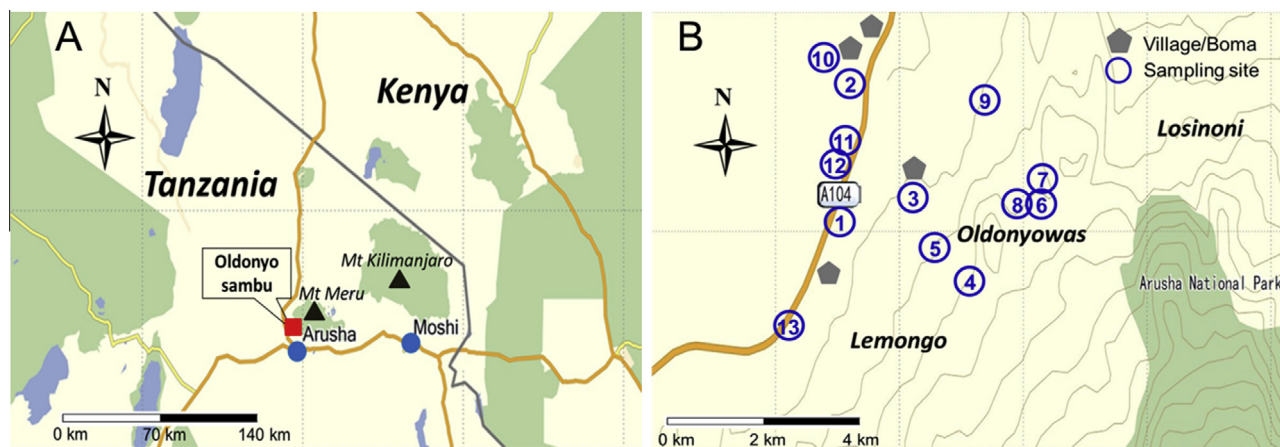


Fig. 1. Map showing the location of (A) the Oldonyosambu Ward and (B) the sampling sites (adapted from Tracks4Africa (T4A) GPS Maps).

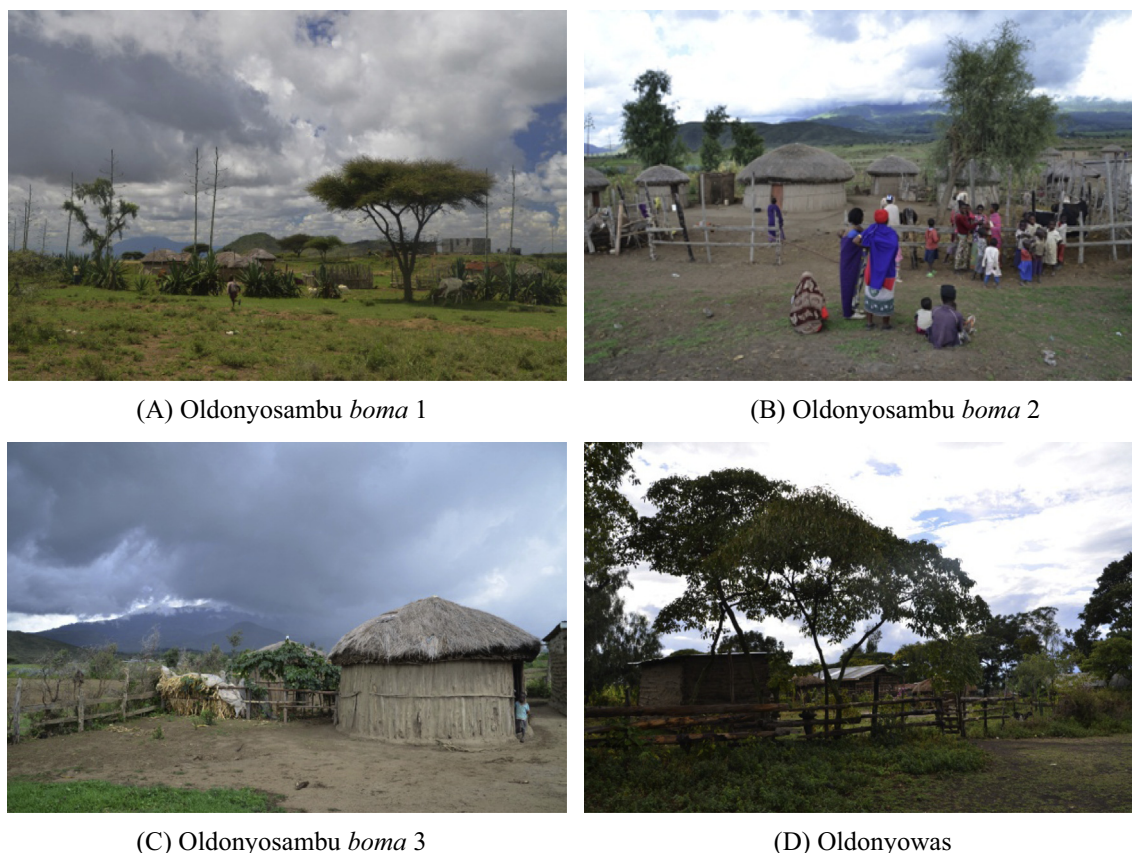


Fig. 2. Different *bomas* studied in Oldonyosambu Ward, villages of Lemongo and Oldonyowas with GPS coordinates (A) S03°07.692'E036°41.123', (B) S03°08.104'E036°40.844', (C) S03°08.379'E036°41.163', (D) S03°10.135'E036°41.779'.

30 °C at lower elevations. Mean minimum temperatures are normally between 13 °C and 16 °C [14]. Climate change predictions forecast an increase in both temperature and precipitation in this area [16]. The area is mainly covered with volcanic soils erupted from Mount Meru several thousand years ago.

2.2. Socio-demographic information

According to the 2012 national census, Oldonyosambu Ward had a total population of about 22,000 [17]. The dominant ethnic groups are people from the agro-pastoralists Warusha, pastoralists Maasai tribes and fewer Wameru, expert mountain farmers [18]. The Warusha separated from the Maasai approximately 200 years ago to settle on the fertile slopes of the Mount Meru [18]. The Warusha and Maasai live in polygamous communal settlements called *boma*. The number of homes in the *boma* reflects the size and wealth of the family. Typically, the Wameru farm banana and coffee, while the Warusha farm maize and beans. Both tribes farm some horticultural cash crops such as carrots, tomatoes, cabbages, onions, green leaves and potatoes. The livestock the Warusha and Maasai raise includes cattle, goats, sheep as well as donkeys for transportation. Diets of the Maasai and Warusha were traditionally based on milk and meat, but are lacking dairy products nowadays, whilst the Wameru, who have broadly adopted zero-grazing cattle keeping, have a higher and more regular access to milk [19]. The low consumption of milk is generally an unfavorable dietary transition for all children, but it is even more worrisome in such a fluoride-rich area given the potential of milk, with high calcium content, to reduce the bioavailability of fluoride in humans [20]. Pictures of some of the typical *bomas* in the area are shown in Fig. 2.

Table 1

Local population and estimated water demands in the Oldonyosambu Ward villages.

Village	Household number	Total population	Drinking water demand (L/d)	Non-drinking water demand (L/d)
Lemongo	2200	14,000	70,000	420,000
Oldonyowas	457	2650	13,250	79,500
Losinoni	600	6000	30,000	180,000

2.3. Water demand and alternative sources

Oldonyosambu Ward has a limited number of permanent water sources and those available are known to suffer from high levels of fluoride concentrations. The water demands in Oldonyosambu Ward were estimated (per person per day) at a minimum of 5 L for cooking and drinking water and 30 L of water that does not need to be free of fluoride for domestic use (Table 1).

The obvious alternative water to fluoride contaminated sources is rainwater harvesting. In 2010, Instituto Oikos and Oikos East Africa installed, in the framework of a European Union and Intervita Onlus funded project, a total of 315,000 L water storage tanks fed by rainwater harvesting systems in primary schools of Oldonyosambu Ward [21]. In the Oldonyosambu Primary School, where more than 1100 students had no access to water whether safe or unsafe, a colonial water pipeline was restored to feed two of the 10,000 L rainwater tanks together with 185,000 L of rainwater storage. The rainwater harvesting supplies the school with a good back-up for personal hygiene and irrigating the school vegetable gardens. The major limiting factors of rainwater harvesting are (1) erratic and seasonal rainfall patterns [15] and (2) the costs

to install suitable roofs, gutters and storage systems. Despite those constraints rainwater harvesting ought to be considered to supplement supply and in some case may be the most viable solution.

In June 2012, Istituto Oikos facilitated the installation of two bone char defluoridation filters in the Oldonyosambu Primary School, and the Kimosonu Primary School in the neighboring Ngarenanyuki Ward. This intervention was driven by the lack of alternative fluoride-free water sources, which is always considered a preferential solution, even at when it is not cost-competitive. This was coupled with the distribution of approximately 60 domestic fluoride filters of 20 L capacity among households in Ngarenanyuki Ward. Monitoring data inform that both interventions had a relatively low success, mostly due to a lack of commitment and understanding of the importance of the replacement of the exhausted bone char. In both schools, in fact, 36 months after the installation of the filters and continue utilization, neither of the filters had the bone char replaced, in spite of a signed Memorandum of Understanding (MoU) between the donor and the beneficiaries where it was stated that the bone char had to be replaced on an annual basis. The same problem occurred at household level [22]. This highlights the importance of either being able to measure the fluoride concentration regularly on-site, or choosing a technology that guarantees a fluoride-free water supply as long as water is being treated. The community was not able to successfully maintain this relatively simple technology.

3. Health implications of fluoride

Oldonyowas and Lemongo are the most fluoride-affected villages. In early April 2013, a number of *bomas* were visited in Mareu and Lemanda sub-villages in order to ascertain the

prevalence and severity of fluorosis in this area. Observation of teeth and body impairments for the interviewed villagers was carried out informally by a team of visitors, village executives and Ministry of Water representatives. This was combined with an informal assessment of awareness and need. Such visits have been carried out for some decades, which have resulted in an increased awareness about fluoride issues. To reduce such fluoride exposure, many villagers who are financially able to send their children away for their education have done so.

Chronic ingestion of excess fluoride can cause fluorosis which affects teeth and bones [7]. Moderate amounts lead to dental fluorosis, and long-term ingestion of large amounts can lead to severe skeletal fluorosis. The clinical appearance of milder forms of dental fluorosis is characterized by narrow white lines following the perikymata and brown or yellowish stains in the opaque areas of the tooth. The more severe forms are subject to loss of outermost enamel and mechanical breakdown of the surface [23]. In skeletal fluorosis, fluoride accumulates in the bone progressively over a long time. The early symptoms of skeletal fluorosis include bone pains and stiffness in the joints. In severe cases, the bone structure may change and ligaments may calcify, resulting in progressive kyphosis and restricted movement [7,24].

Brown teeth are generally a very common sight in northern Tanzania. Many of the children aged between 5 and 15 years, as well as adults, were found to have serious dental fluorosis (Fig. 3A). Similarly, Kaseva [25] reported that in the Maji ya Chai Ward on the south-eastern side of Arusha National Park, all the 2217 children examined aged between 12 and 16 years had dental fluorosis while 74 percent of children in Ngurdoto Ward exhibited a severe degree. Dental fluorosis affects children more than adults because fluorosis is formed during the early stages of enamel development [23]. Dental fluorosis in an adult is likely to be the



(A) Adult with dental fluorosis



(B) Baby born with four toes and deformed leg



(C) Child with deformed and scarred legs



(D) Skeletal fluorosis of the aged

Fig. 3. Evidence of dental and skeletal fluorosis in the villagers within Oldonyosambu Ward.

result of high fluoride exposure when the adult was a child or adolescent [7].

Both adults and children in the area suffer visibly from skeletal fluorosis. It is believed that due to ingestion of contaminated local water, many children exhibit leg deformities and inhibited growth. It appears that symptoms often occur early, for example Fig. 3B shows a baby born with four toes and deformed legs. Some studies found that the human placenta does not prevent the passage of fluoride from a pregnant mother's bloodstream to the fetus and may in fact increase exposure [26,27]. As a result, a fetus can be harmed by fluoride ingested by the mother during pregnancy. Fig. 3C shows a five-year-old girl who underwent surgery to straighten her legs. Her legs are now less deformed but her condition is still not normal, she suffers like many others by inhibited growth and she is still drinking water from the local contaminated source. Adults above 40 years were observed to suffer from premature aging and were unable to walk straight due to serious backache. Severe kyphosis and mobility disability became apparent. Fig. 3D shows a woman who could no longer stand up straight and was using water containers as 'extended legs'. While families with sufficient means are sending children to school outside the ward to avoid this water exposure, others seemed somewhat oblivious to the problem or do not have the means and/or family connections to realize such an option.

Although agreement exists regarding the endemic fluorosis in the area, fluoride may not be the only cause of the observed endemic diseases and it is very obvious that not all those exposed show similar symptoms. Enamel opacities similar to dental fluorosis are associated with other conditions, such as malnutrition with deficiency of calories, proteins or vitamins [28]. Lifestyle and diet will almost certainly play an important role and may offer important remediation strategies. Malnutrition is an important issue in the area. Oikos East Africa led a four-year biometric campaign (2010–2013) targeting 5–19 year old children in the Arusha Region. It was found that among the students of the Primary Schools in Oldonyosambu Ward, specifically Oldonyosambu, Leminyor, Losinoni and Engutukoit Primary Schools, the total prevalence of malnutrition measured was 32 percent of malnourished and 35 percent of severely malnourished ($n = 10,668$) [29]. Far greater studies are thus needed to relate life style, dietary pattern, and other possible contributing factors to the observed health effects. This will naturally lead to opportunities to correct the problem.

4. Water quality assessment

4.1. Water sampling

Water samples were collected on July 22, 2013, from water sources in 13 sites across the ward (see Fig. 1B). The location of field sites was recorded by a hand-held GPS device (Garmin GPSmap 60csx, USA). The waters stem mostly from artesian wells on the slopes of Mt Meru and are well protected by (i) overgrowth that prevents access to animals, (ii) fixtures such as taps or hand pumps, as well as (iii) guards. As such these are 'improved' water supplies. Samples were stored in closed containers at ambient temperature for further analysis or experiments. Physico-chemical parameters, including F^- , pH, electrical conductivity (EC), turbidity, total organic carbon (TOC), inorganic carbon (IC) were measured within days at Nelson Mandela African Institution of Science and Technology (NM-AIST) for bench-scale tests and Ngurdoto Defluoridation Research Station (NDRS) for pilot tests (using the same instruments, detailed below). Samples were shipped to Germany for further chemical analysis at Karlsruhe Institute of Technology (KIT). Some of the water sources were previously

examined in 1992 by the Ministry of Water and entered in the national water quality database.

4.2. Analytical methods

F^- content was determined using an ion-selective electrode (ISE) connected to a pH meter (826 pH Mobile Meter, Metrohm, UK). Samples were mixed at a 1:1 volume ratio with a total ionic strength adjustment buffer (TISAB) of pH 5.0–5.5. The pH and EC were measured in situ by a pH/conductivity meter (WTW Multi 340i, Germany). TOC and IC were measured using a TOC analyzer (Sievers 900 portable TOC analyzer with autosampler, GE Analytical Instruments, USA). Turbidity was measured by a turbidity meter (TN100, Eutech, The Netherlands).

After air shipment to KIT, cations (Al, Ca, Fe, K, Na, Mg, Sr, Zn) and B and Si were measured by an ICP-OES spectrometer (Vista-PRO CCD Simultaneous ICP-OES, Varian, The Netherlands). The aqueous samples were filtered prior to analysis by using 0.45 μm membrane filters (cellulose acetate, 25 mm syringe filters) and acidified with HNO_3 (65%, Suprapur, Merck, Germany). Samples were diluted with water (Milli-Q, Millipore, Merck, Germany) according to the calibration range set for the different elements. The calibration was verified using a certified reference material (ICP Multi element Standard Solution VI CertiPUR, Merck, Germany). Calibration was set from 10 to 2000 $\mu\text{g/L}$ for B, Fe and Sr, from 20 to 5000 $\mu\text{g/L}$ for Ca, K, Na, and Si, from 5 to 2000 $\mu\text{g/L}$ for Mg, and from 5 to 1000 $\mu\text{g/L}$ for Al and Zn. The relative standard deviation is $\pm 2\%$ for each element.

Major anions of chloride (Cl^-), sulfate (SO_4^{2-}), and nitrate (NO_3^-) were analyzed using an ion chromatograph equipped with a suppressor detection unit (IC 790, Metrohm, Germany). Calibration was set to 0.1–10 mg/L or to 10–100 mg/L according to the need of the sample. The relative standard deviation is $\pm 2\%$ for each anion.

4.3. Drinking water guidelines

According to WHO, fluoride concentration in drinking water should not exceed 1.5 mg/L [7]. The same is true for EU Drinking water guidelines [30,31]. The guideline value is based on an average per capita daily water consumption of 2 L. For countries with a hot and dry climate, the daily intake of water is much higher than the WHO proposed level. For this reason, Brouwer et al. [32] suggested that fluoride levels in Senegal should be below 0.6 mg/L in order to prevent the mildest dental fluorosis. In contrast, other countries, such as Ethiopia [33] and Tanzania [34], have other types of fluoride guidelines on basis of economical, practical and technical considerations. Tanzania temporarily allowed consumption of drinking water with fluoride concentration up to 8 mg/L in areas with no alternative water sources within acceptable walking distances [35]. This value has been reduced to 4 mg/L in 2008 [34], as a result of raising public awareness on fluoride issues and the development of defluoridation methods that promise alleviation of the problem such as bone char filters [36].

WHO recommends pH values of 6.5–8.5, based on aesthetic considerations such as taste and corrosion of materials (metals and cement based materials) [37]. There is no health based guideline value set for EC but it is an indicator parameter in the drinking water ordinance (EU: 2500 $\mu\text{S/cm}$ at 20 °C [30], Germany: 2790 $\mu\text{S/cm}$ at 25 °C [31]). Typically drinking water has a conductivity of 50–500 $\mu\text{S/cm}$, while seawater EC is about 50 mS/cm. TOC is used to quantify natural organic matter (NOM) which has been emphasized as the precursor to disinfection by-products including trihalomethanes (THMs) [38]. United States Environmental Protection Agency (USEPA) indicates that water with TOC above 2 mg/L requires treatment to avoid THMs formation [39] and in

consequence this is a target value for this case study as chlorination is often performed in an uncontrolled manner in Tanzania [40]. In the European guideline for drinking water, TOC is related to the indicator parameters, and it should not show abnormal change. The parameter oxidisability should not exceed 5 mg/L O_2 . This parameter need not be measured if the parameter TOC is analyzed [30].

4.4. Fluoride in water sources

All water sources, except the Site 9 (Amnai source, see Table S1) that was just installed by the village and has never been tested before this study, contain fluoride above the WHO guideline value (Table 2). Values measured in 2013 correspond well to those measured in 1992 in this case indicating that the waters have not changed significantly with time. This may be due to the fact that most waters are sources that flow naturally and are not pumped. The highest fluoride concentration (17.9 mg/L) is found in Site 4 (Mareu sub village). Site 5 (Nadung'oro spring) has a relatively low fluoride concentration (3.3 mg/L) but is supplied to this location only one day per week due to limited availability to Oldonyowas and Lemongo villages. The water from this source is called "Maji salama" (safe water) by the villagers and is used mainly for cooking and drinking. Another low-fluoride water source in Site 13 (Madukani sub village) stems from the nearby forest and supplies households in Lemongo. Results on the water analysis are summarized in Table 2. Out of the total 13 samples, 10 have a fluoride concentration of >10 mg/L.

4.5. Other water quality parameters

The EC is 200–700 $\mu S/cm$ and hence does not require desalination. The pH is slightly alkaline but not of concern, while IC is relatively high which is typical for waters in the region. The turbidity is very low, while the TOC is low for all samples except for Site 3 and 5. Seeing that Site 3 (field tap) is fed by Site 7 (Oldoyowas source), this TOC is likely originating from a leak or contamination in the pipe.

The results of major elements indicate that these water sources are typically of Na- HCO_3 type (at pH 7.9–8.3, the dominate carbonate species is HCO_3^- [41]). The results confirm previous findings that fluoride content in water is closely correlated with salinity,

Na^+ , and HCO_3^- [1]. Exceptions are Site 5 and Site 13 where the low F^- is associated with a high Ca^{2+} content (Table 2). Rango et al. [42] found water samples with similar compositions in the Main Ethiopian Rift which is also part of the East African Rift. High contents of Na, IC, F^- , SO_4^{2-} and Si are mostly from the dissolution of pyroclastic volcanic materials [42].

Another feature of these water samples is the relatively low contents of Ca^{2+} . Waters with low Ca^{2+} concentration are often found in volcanic regions which are dominated by alkaline volcanic rocks [1] and also in conditions where cation exchange occurs naturally [42]. Here, the uptake of Ca^{2+} by the aquifer matrix is balanced by the release of Na^+ into groundwater. In both cases, the waters become Na- HCO_3 type. The lack of calcium allows higher concentration of fluoride to be stable in solution as the solubility of CaF_2 (15 mg/L at 18 °C [43]) is no longer a limiting factor. In addition, the results reveal the occurrence of some trace elements in water, including Sr, B, Fe and Zn (Table 2). Other elements which were analyzed but not detected are shown in Table S2.

5. Bench-scale nanofiltration/reverse osmosis (NF/RO)

The aim of bench-scale experiments was to investigate the transfer and fate of fluoride and other solutes in NF/RO. Site 4 water (Mareu) with the highest fluoride concentration (17.9 mg/L) was selected as the worst-case water for this area to be treated. Water samples from Site 1 (A104 roadside tap) and Site 2 (church tank) were also treated and results are shown in the Supplementary Information.

5.1. Experimental set-up

A dead-end stirred cell with flat sheet membranes was used for the bench-scale experiments (Fig. 4). A brackish water RO membrane (BW30) and a relatively loose NF membrane (NF270) were selected as the representatives (Dow Chemicals, USA). Details of the system and the membranes are published elsewhere [44]. In brief, the cell was filled with 400 mL solution and pressurized by compressed medical air (TOL, Tanzania) with applied pressure up to 10 bar. Permeate solution was collected on an electronic balance (Adventurer Pro 2102, Ohaus, Switzerland) until recovery (the ratio of permeate to feed volume) reached 50%. Retention was calculated as the percentage of solute removed from the feed solution.

Table 2
Water quality at different field sites in Oldonyosambu Ward.

Site No.	1	2	3	4	5	6	7	8	9	10	11	12	13
pH (20 °C)	–	8.33	8.01	7.53	8.18	8.55	7.68	7.8	8.64	8.31	8.68	8.46	8.57
EC (20 °C)	$\mu S/cm$	548	504	443	441	540	433	430	498	202	544	527	502
TOC	mg/L	0.9	2.4	10.1	2.8	4.5	1.1	1.2	1.3	2.1	1.9	2.4	1.9
IC	mg/L	52.5	47.0	35.6	42.9	67.6	42.9	42.2	41.6	21.3	48.4	51.7	47.8
Turbidity	NTU	0.12	0.91	0.45	0.46	0.78	0	0	0.34	0.55	0.77	0.66	0.4
F^- (1992) ^a	mg/L	–	–	–	18.0	3.5	13.0	13.0	13.0	–	13.2	–	–
F^- (2013)	mg/L	10.9	12.8	13.4	17.9	3.3	13.4	13.9	13.9	0.9	14.5	15.1	15.0
Cl^-	mg/L	5.6	6.4	7.2	4.9	5.0	5.0	5.0	5.1	1.8	4.6	5.5	5.3
SO_4^{2-}	mg/L	11.4	10.0	12.6	11.8	7.5	9.7	9.8	9.7	2.2	9.5	11.5	11.5
Si	mg/L	20.8	20.6	20.3	23.8	24.7	21.9	21.9	22	11.2	20.9	23.3	21.1
Na	mg/L	112.3	94.1	86.1	89.2	96.1	86.1	86.8	87.9	24.4	95.8	109.7	101.4
K	mg/L	23.2	30.2	22.8	18.7	36.4	23.4	23.4	24	14.2	23.9	26.5	25.8
Mg	mg/L	0.6	0.7	0.5	0.3	2.0	0.2	0.2	0.2	0.4	0.5	0.4	0.5
Ca	mg/L	4.4	4.0	2.6	1.7	7.6	1.5	1.4	1.3	4.5	2.8	2.2	2.8
Hardness ^b	mg/L	13.5	12.9	8.6	5.5	27.2	4.6	4.3	4.1	12.9	9.1	7.1	9.1
Sr	$\mu g/L$	144.4	121.2	62.3	41.0	426.0	50.0	47.0	45.0	173.0	96.0	46.0	96.0
Al	$\mu g/L$	<10	28.8	37.0	22.9	34.3	22.9	17.7	55.1	28.1	23.9	15.6	23.9
B	$\mu g/L$	21.8	16.8	23.5	19.0	16.0	15.0	15.0	15.0	<10	18.0	19.0	19.0
Fe	$\mu g/L$	<10	39.7	48.1	12.0	38.0	<10	<10	20.0	<10	21.0	42.0	28.0
Zn	$\mu g/L$	15.0	5.0	133.0	<5	136.0	<5	<5	<5	48.0	<5	<5	29.0

^a Data from NDRS database.

^b Hardness is the concentration of Ca^{2+} and Mg^{2+} expressed as equivalent of $CaCO_3$.



Fig. 4. Stainless steel stirred cell experimental apparatus at the University of Dar es Salaam.

5.2. Membrane performance

Fluoride and other solutes including trace elements and organic matter were removed to a different extent by the NF and RO membranes (Fig. 5). The results demonstrate the typical difference between RO and NF. The BW30 membrane retained all ions and organic matter to more than 90%, while the relatively open NF270 membrane was more effective in removing multivalent ions (Ca^{2+} , Mg^{2+} , SO_4^{2-}) than monovalent ions (K^+ , Na^+ , F^- , HCO_3^-). The permeate concentration of trace elements (Sr, Fe, B, Zn) was very close or below the detection limits of the instrument, therefore the retention was not quantified due to a naturally very large analytical error. Both the NF and RO membranes achieved very high TOC retention as a result of size exclusion effect for large NOM molecules [45].

Permeate flux was monitored as an indicator of membrane performance and plotted as a function of recovery in this dead end batch filtration experiment, as shown in Fig. 6A. As expected, the flux of the NF270 membrane was significantly higher than that of the BW30 membrane because the NF270 membrane is more open and more hydrophilic [46]. The flux of NF270 declined gradually as a function of recovery. Due to the high flux the boundary layer concentration is higher and hence this decline is expected to occur predominantly due to osmotic pressure both in the boundary layer and in the cell as the concentration increases. Some solute penetration into the pores may be possible but this

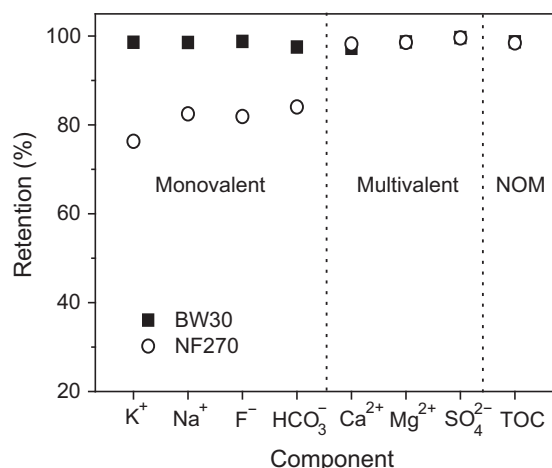


Fig. 5. Retention of major compounds in Site 4 water by BW30 and NF270 membranes at 50% recovery.

was not investigated further [47]. Gradual flux decline also occurred in the BW30 membrane but within a very narrow range.

5.3. Ion transport in NF/RO

With initial pH 8.18, Site 4 water is an alkaline solution where hydroxyl ion (OH^-) rather than hydrogen ion (H_3O^+) predominate. During filtration the permeate pH of each membrane was always lower than the concentrate pH, indicating that OH^- was rejected to some extent (Fig. 6B). For the tighter BW30 this OH^- rejection is higher than that for the NF270 membrane. Zhu et al. [48] reported the similar phenomenon at acidic and neutral solutions, and explained it with the negative rejection of positive H^+ under the influence of the Donnan equilibrium.

The two membranes showed distinct differences in removing fluoride. The BW30 membrane was able to remove fluoride below the guideline value of 1.5 mg/L for all recovery values (Fig. 6C). The NF270 membrane was only reliable at low recovery (<20%) and exceeded the WHO guideline value at higher recovery, although the Tanzanian guideline (4 mg/L) was met at all recovery values. The hydrated radius of fluoride (0.35 nm) falls in between the pore radii of the two membranes [12,49]. Therefore fluoride ion can penetrate into the NF270 membrane but be rejected by the BW30 due to size exclusion. Increasing recovery results in an elevated concentration gradient in the filtration system and across the membrane, which promotes the diffusive transport of fluoride [50]. It should be noted that the recommended element recovery rates for surface water are between 15% and 20% [51]. While in this recovery range, the defluoridation performance of the NF270 membrane was still satisfying, the water in this case study is very low turbidity spring water and hence this limitation may not be necessary. Further, the recovery changes along an element and further varies with fluctuating energy. Hence, it is important to understand the membrane behavior over a broader recovery range.

The trend in EC permeate concentration is similar to that of fluoride (Fig. 6D). The increased permeate EC with recovery can be attributed to diffusion. As a typical RO membrane, BW30 has a high salt retention. In consequence, the treated water may lack the essential composition of minerals and result in an unpleasant taste. Some health experts believe that mineral-free water may be harmful to human health by leading to demineralization of the body [52] and in the case of malnourishment this may further aggravate the situation. Hence the RO treated water may require remineralisation post-treatment. This problem should however be considered as secondary compared to the health effects of fluoride and considered together with nutritional strategies. The NF270 membrane, on the other hand, allows more ions to pass through due to its openness. This reveals a major NF advantage over RO that NF removes salts selectively without requiring remineralisation. Clearly, choices exist regarding specific membranes with regard to the desired degree of purification and should be selected on a case by case basis.

6. Renewable energy powered NF/RO system

Bench-scale experiments have confirmed that NF/RO membranes are able to reduce fluoride and other contaminants reliably and simultaneously from the Oldonyosambu water. A key challenge to apply such NF/RO technologies in the Oldonyosambu Ward is the lack of electricity and water infrastructure. The entire area is not connected to an electrical grid and no such connection is foreseeable in the future. Decentralized membrane systems powered by renewable energy are attractive options for such scenarios [13,53–55]. Specifically in this area, the high year-round availability of sunshine provides ideal conditions for the implementation of

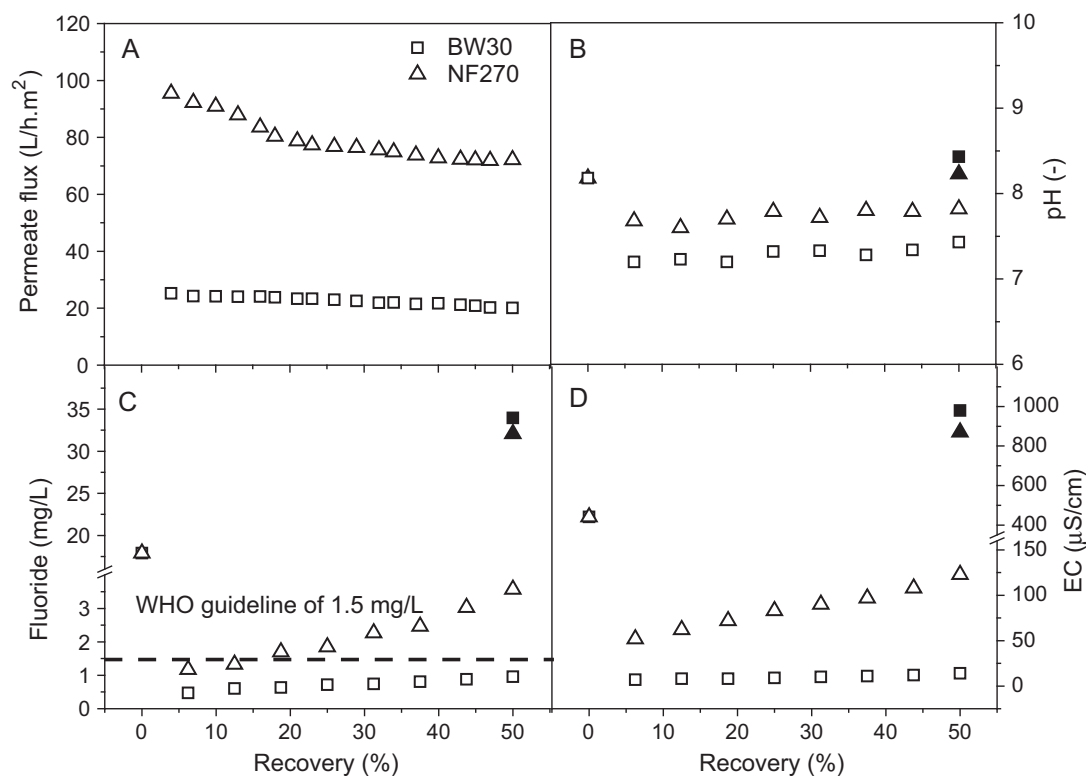


Fig. 6. Permeate (A) flux, (B) pH, (C) fluoride, (D) EC of Site 4 water by NF/RO as a function of recovery (for comparison, concentrate pH, fluoride and EC are shown in solid shapes).

solar-powered systems [15]. In consequence, a photovoltaic-powered NF/RO system was tested with Site 4 water at the NDRS. The system was previously tested in the field in central Australia and found to perform very well in removing a wide range of contaminants including fluoride (98.5% retention for water containing only 1.1 mg/L F^-) [56].

6.1. System design

Details of the system (Fig. 7) were published by Schäfer et al. [57], although some modifications have taken place with time. In brief, the system consists of two stages: the pre-treatment stage with an ultrafiltration (UF) membrane is followed by a desalination or defluoridation stage using NF/RO. The submerged ultrafiltration was a GE Water Zenon ZW230 module with a membrane surface area of 21 m². The NF/RO pressure vessel holds one



Fig. 7. Photo of the solar-powered hybrid UF-NF/RO system at the NDRS to where Site 4 water was transported for experiments.

interchangeable 4" membrane module. Three spiral wound membrane modules, namely BW30, NF90 and NF270 (Dow Chemicals, USA) were used. The NF90 membrane was selected in addition as a narrow NF membrane between BW30 and NF270. The BW30 module has an active area of 7.2 m² while the NF90 and NF270 modules have an active area of 7.6 m². Permeate and concentrate were recycled into the feed tank (400 L) to maintain a constant feed concentration. To investigate the effect of energy fluctuations on system performance at reproducible conditions, a solar array simulator (SAS, Agilent Technologies E4350B-J02, USA) was used to power the system's DC pump instead of the photovoltaic panels. The solar irradiance data (10 h, collected on a typical sunny day during the dry season at NDRS) was used as an input parameter to the SAS to allow for this solar day to be repeated for all experiments and thus allow comparability of results. Regular sampling of permeate occurred at 0.25–1 h intervals throughout the experiments for further water analyses. Online measurement of parameters such as pressure, flow, temperature and EC was recorded by a laptop using LabVIEW 8.0 software (National Instruments, USA). Detailed information regarding the sensors used in the system can be found in Richards et al. [58].

6.2. Membrane performance

The performance of three membranes is summarized in Table 3, including the specific energy consumption (SEC). The system runs about 9–10 h at a feed flow of about 600 L/h with variable flux and hence recovery. Given that the solar irradiance was the same for all experiments, the water production and SEC of the system changed significantly with the membrane type. The BW30 membrane produced a total of 1000 L drinking water at an average SEC of 2.2 kW h/m³, the NF90 produced 2444 L water at an average SEC of 0.9 kW h/m³, while the NF270 produced 2656 L water at an average SEC of 0.8 kW h/m³. Although the NF270 was the most

Table 3

Comparison of average performance over a typical and identical solar day of NF/RO membranes for Site 4 water (TMP: Transmembrane Pressure; Perm: Permeate; Conc: Concentrate, Ret: Retention).

Membrane	Flux (L/h m ²)	Recovery (%)	Daily production (L)	SEC (kW h/m ³)	TMP (bar)	Perm EC (μS/cm)	Conc EC (μS/cm)	EC Ret (%)	Perm F ⁻ (mg/L)	Conc F ⁻ (mg/L)	F ⁻ Ret (%)
BW30	13.8	17.3	1000	2.2	4.5	10	558	97.7	0.6	31.2	96.6
NF90	32.3	40.5	2444	0.9	4.4	12	810	97.3	0.4	24.6	97.8
NF270	34.9	45.0	2656	0.8	4.3	120	881	72.8	3.7	37.5	79.3

energy-efficient membrane, its fluoride concentration of 3.7 mg/L failed to meet the WHO guideline, although it still complied with the current Tanzanian guideline. The BW30 and the NF90 membranes both produced excellent permeate fluoride at 0.6 and 0.4 mg/L, respectively. The noticeably low permeate EC (10 and 12 μS/cm for BW30 and NF90 respectively) confirmed the potential need for remineralization, while for the NF270 membrane this would not be required (120 μS/cm).

Assuming a generous potable (drinking and cooking only) water consumption of 5 L per person, this system (equipped with the NF90 membrane) is in principle able to supply a remote community of almost 500 people with drinking water. It should be noted that the recovery was relatively high at 40%. This means that a further 7 L of fluoride-rich water are available per person for washing. Due to the high recovery this is lower than the usual design of 5 L of drinking and 20 L of washing per person.

6.3. System robustness toward energy fluctuations

The solar-powered system in this study has no batteries for energy storage and hence the power delivered to the pump fluctuates with solar irradiance. The solar irradiance varies with the rising and setting of the sun and also fluctuates due to cloud cover. This can be seen in Fig. 8A where the light passing cloud at 08:42 had a slight effect on the power generated by the photovoltaic panels, while the two lots of heavier clouds (13:36 and 13:46, and 14:02 and 14:12) drastically reduced the amount of power available. It is noteworthy that the system did not stop running during this time, even when the solar irradiance dropped rapidly from 1000 to 200 W/m².

A step in the data for the BW30 experiment can be seen in Fig. 8A–C and F occurred due to corrosion of the bearings in the flow meter on the feed stream (Omega model FTB9512) resulting in a jammed paddlewheel. While it was still possible to determine the feed flow from the permeate and concentrate flow, the decreased resistance across the feed flow meter resulted in a change in pressure drop of about 0.3 bar (Fig. 8B). The altered load of the system, then caused the maximum power point tracker to adjust the current and voltage to better match the output from the SAS and this is the reason for the step in the motor power data (Fig. 8A).

Power fluctuations affect the TMP (Fig. 8B) and feed flow (Fig. 8C), and hence other performance parameters such as permeate flux (Fig. 8D). Permeate flux reduced when the solar power (and hence TMP) decreased. The immediate nature of the response of permeate flux toward solar irradiance is due to the directly solar-powered pump which provides the driving force of the filtration process as TMP. Just after 14:00, when solar irradiance dropped from 1000 to 200 W/m², the permeate flux reduced significantly. In particular, the permeate flux of the BW30 module reduced from 15 L/h m² to zero, highlighting the intermittent nature of the system. In previous work, the authors have investigated the effects of including energy buffering components to overcome such short term fluctuations [58].

Similar to flux, the recovery (Fig. 8E) depends on the amount of power available to the system. The BW30 recovery (~17%) was

significantly less than the values for NF90 and NF270 (~40% and ~45%, respectively).

The SEC (Fig. 8F) is the energy consumed per m³ of water produced and is directly affected by the changes of input power and the permeate flux. Due to the low raw water salinity of the water the SEC is very low and the observed increases at low power incidences can be attributed to the small amount of water produced.

It is important to note that the feed water temperature increased by about 30% throughout a typical day (Fig. 8G) due to two reasons. Firstly, due to the nature of the experiment, whereby permeate and concentrate were recycled back into the feed tank after passing through the NF/RO membrane, a significant amount of waste heat generated by the pump is transferred to the feed flow. Secondly, the actual meteorological conditions and ambient temperature naturally affect the water temperature in the system. It is important to note that on the day when experiment NF90 was conducted a large thunderstorm started just before 15:00 and within the next hour the ambient temperature had dropped from 27.1 °C to 19.6 °C (Fig. 8H). This evidenced itself in the feed water temperature, which indeed cooled significantly over this time. In contrast, the ambient temperature on the day when the NF270 experiment was conducted was significantly warmer, peaking at 31.6 °C just after midday before cooling down in the afternoon. Both of these factors contribute to the increased feed water temperature and affect the gradual increase of permeate flux over time. Such factors will of course alter performance of such systems in real conditions.

Despite slight changes with energy fluctuations, average permeate fluoride remained well below the WHO guidelines throughout the day (Fig. 9A). This demonstrated the reliability of the NF90 membrane to produce high quality drinking water under a fluctuating weather condition at optimized water and energy efficiency. The last points in the BW30 and NF90 curves in Fig. 9A, as well as the ones just after 14:00 (large cloud passing) show that the permeate fluoride concentration increased with decreasing solar irradiance. Decreased solar power results in reduced water permeation and increased contribution of fluoride diffusion. For the BW30 experiment, the permeate flux dropped to zero for a short time after 14:00, resulting in a dramatic increase in the permeate fluoride concentration compared to the NF90 experiment (Fig. 9A). Due to concentration polarization fluoride accumulates near membrane surface [59]. In Fig. 9A it can be seen that the permeate fluoride concentration for the NF270 experiment increased throughout the day. It is hypothesized that this is also due to temperature effects. The NF270 membrane is significantly looser than the other two and does not retain monovalent ions like F⁻ well. Hence, any change in the membrane morphology, water viscosity as well as diffusivity values that are induced by increased feed water temperatures will impact strongly on the retention. In contrast, the effect is smaller for the tighter BW30 and NF90 membranes.

EC and IC behave similar to fluoride (Fig. 9B and C) throughout the solar day. The online EC measurement allows to observe EC at higher resolution which is good given the energy fluctuation. The initial peaks are caused by stagnant water in the system's dead volume.

Fig. 9D shows permeate TOC. The initial TOC value of Site 4 water was 2.8 mgC/L. This value is low. The residual water in the

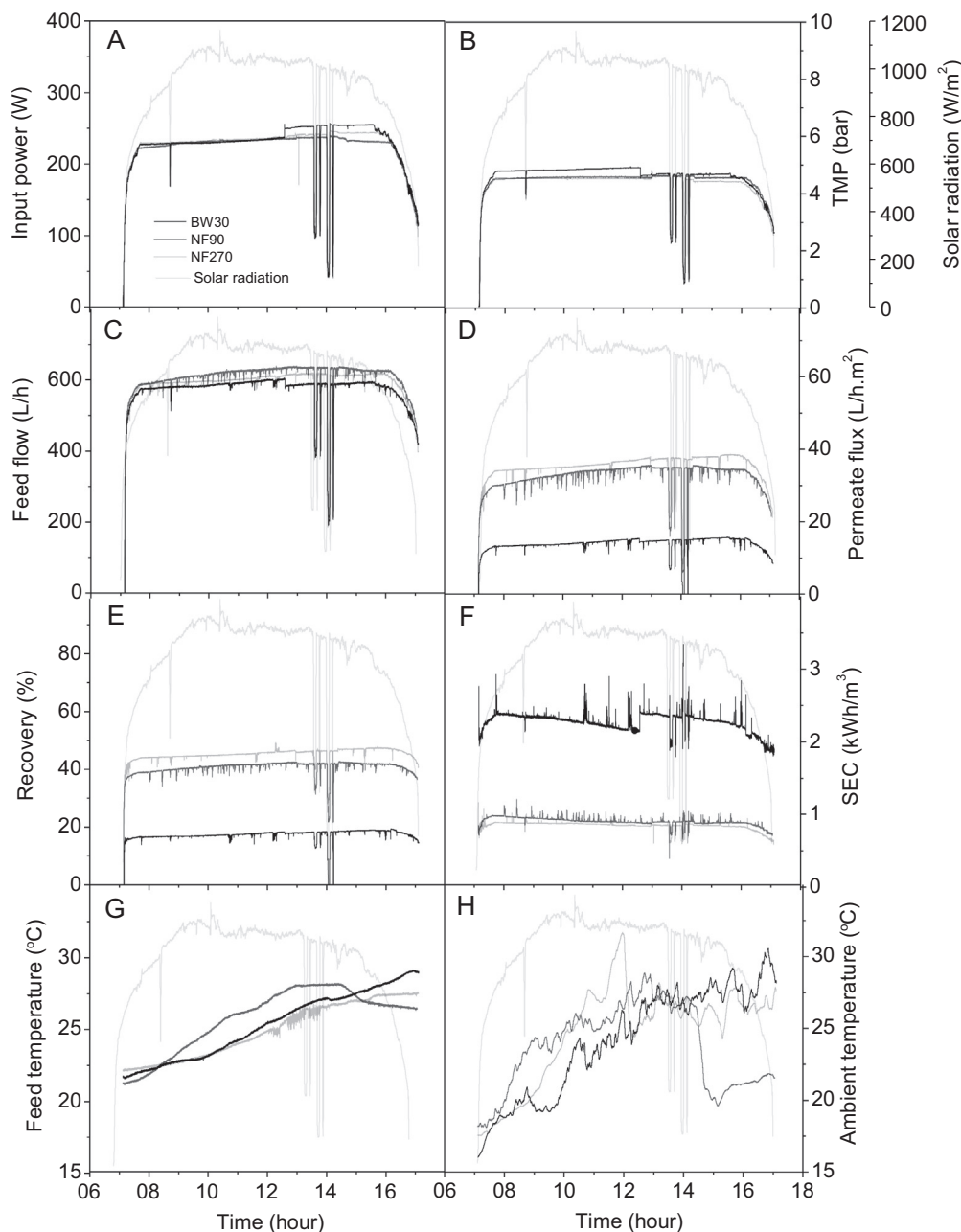


Fig. 8. System performance throughout the solar day: (A) input power, (B) TMP, (C) feed flow, (D) permeate flux, (E) recovery, (F) SEC, (G) feed tank temperature, and (H) ambient temperature.

beginning, the energy fluctuations at 14:00, and the gradual increase of feed water temperature together affect the permeate TOC concentration, which is particularly evident in the NF270 experiment. By contrast, the tight BW30 and NF90 membranes show high reliability to produce good-quality drinking water under such a fluctuating weather condition. Given the relatively low raw water TOC that will make contribute to analytical errors, the collection and transport of a large volume for pilot studies at the NDRS in plastic tanks that may have resulted in contamination, this data should be treated with care.

The safe operating window (SOW) of this system when equipped with BW30 and NF90 membranes has been previously determined [60]. Although the EC is significantly lower here, it is likely that the permeate fluxes and TMP values would be within the SOW of the system.

6.4. System sustainability

In the last five decades, different research teams visited villages in this ward with the goal to alleviate health problems associated with insufficient water supply and poor water qualities [58,61–63]. So far, only the technique of bone char adsorption columns is adopted but its implementation as an appropriate technology is found to be far from satisfactory [22]. This indicates the need to ensure that the communities are fully aware of the problem and actively engaged in the solution – most importantly via committing some of their own resources – to ensure that systems are sustainable in long term operation.

Montgomery et al. [64] identified three components of sustainability for water supplies in developing countries as (1) effective community demand, (2) local financing and cost recovery, and

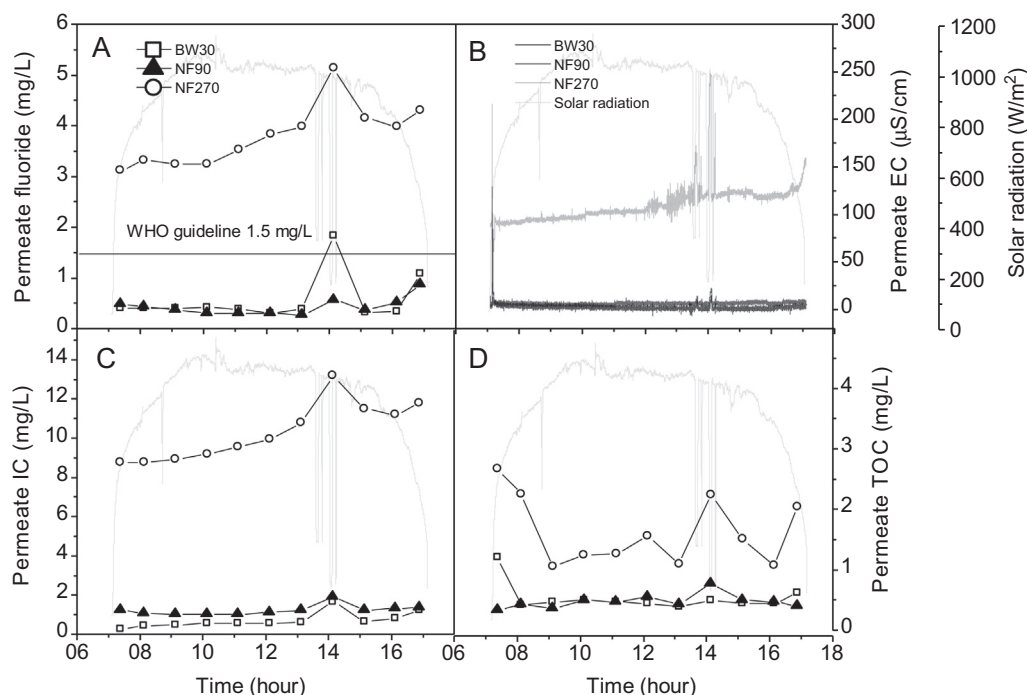


Fig. 9. Water quality throughout the solar day: (A) permeate fluoride, (B) permeate EC, (C) permeate IC and (D) permeate TOC.

(3) dynamic operation and maintenance. Membrane technologies are superior to meet community water demands as modular membrane systems can be easily sized to different scales, and can be coupled with hybrid power systems. Costs of such advanced membrane systems have always been regarded as a main limitation for rural areas [65]. However, considering the cost of unsuccessful boreholes, it could be worthwhile for the government and NGOs to invest in the capital costs of water treatment systems instead [65]. In addition, previous studies indicated that in developing countries, the general costs estimated for decentralized membrane systems are within the range of some untreated water costs such as tanker trucks and water vendors, and well below infrastructure costs of centralized systems with medium range distribution systems [13,65]. Setting the optimum price for the treated water is of crucial importance for cost recovery as many villagers preferred to use free untreated water sources when the prices are too high. Distinguishing between high-grade (potable) water and low-grade (washing) water at different prices appears a logical solution and this is indeed already practiced by many villagers.

Operation and maintenance is probably the most significant obstacle for adopting defluoridation systems in remote locations [22]. While training of local technicians should be a mid- to long-term goal, technical support from the suppliers must accompany the early adopters of such technologies and this support has to be included in the system cost to ensure feasibility and sustainability. The long-term integration of such renewable energy powered membrane systems requires comprehensive assessment from techno-socio-economic perspectives and solid strategies at a local level. Research into suitable business plans and sustainable operation and maintenance regimes is currently under way to develop creative and responsible solutions.

In view of previous important lessons learnt, and in view to increase the sustainability of the membrane system, the recommendations of the authors would be to:

- (1) approach the problem of rural communities forced to source fluoride-rich waters through establishing a service provider of fluoride-free water which, at an affordable cost, could

provide safe water in particular for the most affected youngest individuals;

- (2) create a fluoride-committee able to monitor the provision of safe water to the smaller children combined with nutritional circumstances;
- (3) increase further the awareness of the effects of excessive fluoride exposure in children and create by laws to promote safe water consumption; and
- (4) involve Village Authorities in the implementation of the systems by laws.

7. Conclusions

Fluorosis is a major health problem affecting a large part of the population in the Oldonyosambu Ward in northern Tanzania, which is the site of this case study. Fluoride concentration in the 7 out of 10 available water sources is >10 mg/L and up to 12 times higher than the WHO guideline value of 1.5 mg/L. The naturally occurring dissolved fluoride is found to be positively correlated with the Na-HCO_3 type of water caused by local geology. Observed health effects are severe and range from dental to skeletal fluorosis.

Bench-scale experiments indicate that NF/RO is capable to remove fluoride from these waters that other than for fluoride are of very high quality. Naturally, higher retention means lower water productivity, higher energy costs and higher likelihood for remineralization requirements.

Pilot-scale experiments show the possibility of supplying safe drinking water using a solar-powered decentralized membrane system with a capacity of about 1000–2500 L of drinking water per solar day and a further 3500–5000 L of disinfected non-potable water for irrigation or washing. System robustness toward energy fluctuations was confirmed. Both technology and methodology in this study are transferable to any remote location in the world where dissolved contaminants are of concern. This study is intending to encourage further work to adapt such decentralised and advanced technologies for those rural locations where water quality is posing particularly difficult challenges.

Acknowledgements

The authors would like to thank Leverhulme Royal Society Africa Awards SADWAT-Tanzania and SUCCESS for project funding. PhD studentship for Junjie Shen is provided by Energy Technology Partnership (ETP) Scholarship with the Drinking Water Quality Regulator for Scotland (DWQR) being the industrial sponsor. Special thanks are extended to the Oldonyowas village executive officer and the Oldonyosambu Ward Councilor for their cooperation, guidance and permission to carry out this research. We thank Dr. Edda Vuhahula (Muhimbili University of Health and Allied Sciences, Tanzania) for discussions on dental health. Prof. Harald Horn (KIT, Germany) for critical comments on the manuscript. Prof. Dr.-Ing. Felix Mtalo at UDSM is appreciated for his hosting the bench-scale testing in his laboratory, accommodation for the team and logistics support. We also thank Geert Aschermann (TU Berlin, Germany) for helping with field experiments and Elly Karle and Reinhard Sembritzki (KIT, Germany) for IC and ICP sample analysis. Dow Chemicals are appreciated for kindly providing membrane flat sheet samples and modules, and GE Power & Water (Zenon) for the generous provision of the UF module.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.seppur.2015.05.027>.

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