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# Evaluation of Commercial Reverse Osmosis and Nanofiltration Membranes for the Removal of Heavy Metals from Surface Water in the Democratic Republic of Congo

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**Abstract:** This study evaluates the performance of commercial reverse osmosis (RO) and nanofiltration (NF) membranes for the removal of metal ions from synthetic water and surface water carried from the north-west of Lake Tanganyika in the city of Uvira, in the east of the Democratic Republic of Congo. Metal ion analyses were performed by the standardized ICP-MS and ICP-OES methods. The RO membrane showed higher metal ion rejection in high-concentration solutions (synthetic samples) prepared in the laboratory as well as in low-concentration samples from real raw water collected near Lake Tanganyika. Rejection levels were higher than 98% for $Cr^{3+}$, $Pb^{2+}$, $Cd^{2+}$, $As^{3+}$, $Ni^{2+}$, and $Sb^{+3}$ ions in the synthetic solutions, and 99.2, 98.8, 98.6, 99.2, 98.4, and 98.8%, respectively, in the real samples. The concentrations of metals in the permeate varied depending on the feed concentration and were 0.15 to 1.02 mg/L, 0.33 to 22 mg/L, and 0.11 to 22 mg/L in RO, NF90, and NF270 membranes, respectively. Regarding the NF membranes, the rejection of Cr, Ni, and Cd ions was interesting: 98.2, 97.8, and 92.3%, respectively. However, it was lower for Pb, As, and Sb ions: 76.9, 52.5 and 64.1%, respectively. The flux of NF was 329 to 375 L/m$^2$.h, much higher than for RO membranes, which had a flux of 98 to 132 L/m$^2$.h. The studied membranes are thus a feasible solution to remove the studied metals from real water sources at low concentrations since they meet the standards of the World Health Organization on specific values assigned to chemicals from industrial sources and human habitation areas where these ions are present in drinking water.

**Keywords:** reverse osmosis; nanofiltration; wastewater; heavy metals; Lake Tanganyika

## 1. Introduction

Pollution of water by heavy metals is a critical global environmental problem [1–6]. Millions of tons of toxic metals such as As, Cd, Cr, Cu, Hg, Ni, Pb, Se, V, and Zn, delivered by manufacturing and mining activities, are annually released or transported to the environment and may undergo transformations that have great environmental, public health, and economic impacts [7–10]. In addition, drinking water scarcity is one of the problems caused by population growth, global industrialization [11–15], mining, and agricultural shortages. The application of environmentally unfriendly techniques in industry and waste management, and natural forces such as volcanoes, earthquakes, or storms [16–18], have caused heavy metals to reach alarmingly toxic levels in the environment [19–21].

Due to their high solubility in aquatic environments, toxic metal ions can be easily adsorbed and concentrated in the tissues of living beings as their progress through the trophic chain can cause considerable health problems [22]. The removal of heavy metals is of particular concern because of their recalcitrant nature and persistence in the environment [23]; moreover, the removal of heavy metals from wastewater is an active area of research with many gaps [24].

Unlike many other pollutants, removing heavy metals from the environment is difficult because they cannot be chemically or biologically degraded and are ultimately indestructible [25]. Heavy metals released into water bodies by waste have an incorrigible impact on the aquatic system and destroy the self-purification capacity of an aquatic body [26], as well as posing a carcinogenic risk to public health [27]. This situation affects freshwater bodies, such as the case of Lake Tanganyika, given its lower altitude compared to that of the agglomerations erected on its banks. Unfortunately, it constitutes the main receiving environment for the pollution generated in Bujumbura (on the Burundian side), Uvira, Kalemie, and Moba (on the DR Congo side), Kigoma and Kipili (on the Tanzanian side), and Mpulungu (on the Zambian side). These agglomerations are also home to a diversity of chemical and agrifood industries and activities (markets for example) that generate enormous amounts of wastes in both liquid and solid forms [28], which if not treated will constitute a potential danger to the lake.

The World Health Organization (WHO) produces a series of water quality guidelines covering drinking water, the safe use of wastewater, and safe bathing water [29]. Table 1 presents the latest version, updated in 2017, of the surface and drinking water quality guidelines, which are the benchmarks for the safety of drinking water.

**Table 1.** World Health Organization's quality guidelines for drinking water [29].

| Element | Concentration Normally Found in Surface Water [28] | WHO Guidelines |
|---|---|---|
| Sb | <4 μg/L | 0.02 mg/L |
| As | - | 0.01 mg/L |
| Cd | <1 μg/L | 0.003 mg/L |
| Mn | - | 0.1–0.2 mg/L |
| $Cr^{3+}$, $Cr^{6+}$ | <2 μg/L | Chrome total: 0.05 mg/L |
| $Cu^{+2}$ | - | 2 mg/L |
| Pb | - | 0.01 mg/L |
| Hg | <0.5 μg/L | 0.01 mg/L |
| Ni | <0.02 μg/L | 0.07 mg/L |
| Zn | - | 3 mg/L |

Several processes can be used to remove metal ions from wastewater, in particular precipitation, adsorption on activated carbon, ion exchange, membrane filtration, and hybrid systems [30–35]. Membrane separation technology is the most cost-effective and widely applied technology for water purification. Generally, the membrane acts as a se-

lective barrier that allows for the passage of certain constituents while inhibiting the passage of other constituents based on different mechanisms depending on the type of membrane (e.g., size-exclusion, diffusion). Filtration-based membrane processes have been classified into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), and forward osmosis (FO), in which the first two often use a microporous membrane while the last three use a thin composite membrane with a selective dense layer [36]. In order to remove ions, NF and RO membranes have demonstrated good retention capacities. Boussouga et al. [37] showed that the variations in water salinity had no impact on the high rejection of As (V) in nanofiltration, while the variation of pH significantly affected the rejection of As (V) and the presence of humic acid in the water increased the overall release of As (V). In the presence of humic acids, an improvement in As (V) rejection occurred with the NF270 membrane by 82–94%. At different salinity conditions (0.58–20 g $L^{-1}$), rejection of As (V) with both NF270 and NF90 membranes showed a consistently high removal of 82–88% and 93–98%, respectively. The elimination of Sb compounds did not depend on the pH of the solution since the oxidation state of antimony changes from Sb (III) to Sb (V) in a very short time. The elimination of Sb (III) and Sb(V) is nearly constant over a pH range of 3–10, unless Sb (III) removed with the NTR-729HF membrane greatly decreases by 60.2 to 45.7% with a reduction in pH from 7 to 10 [38]. Experiments conducted by Qdais et al. [39], show that RO and NF membranes can remove synthetic wastewater containing 250 ppm of $Cu^{2+}$ and $Cd^{2+}$ ions at 98% and 99% and the permeate concentration would be reduced to 3 ppm. Using NF, more than 90% of the copper ions were removed. Mohsen-Nia et al. [40] examined the effect of increasing ion size on membrane rejection; $Cu^{2+}$, and $Ni^{2+}$ ions were chelated by the disodium salt of ethylenediaminetetraacetic acid ($Na_2EDTA$), a commonly used chelating agent. The results obtained indicate that $Na_2EDTA$ can increase the size of the chelated $Cu^{2+}$ and $Ni^{2+}$ ions, and therefore, their rejection in the RO removal process was 99.5%.

Ipek et al. [41] showed that treatment of the feed solution with EDTA did not influence the removal of $Zn^{2+}$ in NF. The removal of $Ni^{2+}$ and $Zn^{2+}$ ions from the untreated solution was 99.3 and 98.9%, and after treatment, 99.7 and 99.6%, respectively. The recovery rate, rejection, and saturation factor all increased with increasing applied pressure. Increasing the pH improved the removal of $Pb^{+2}$ from 86 to 99%, but did not affect the removal of $Ni^{2+}$ from 93–99%; this is due to the differences in interaction between the surface of the membrane and the anion present in the solution [42]. The rejection of $Ni^{2+}$ and $Cr^{6+}$ ions increased with increasing pH but did not change significantly by the transmembrane pressure difference (10, 20, and 30 bar) for NF90 and NF270 membranes. The optimum conditions were found at 30 bars with a pH of 10 for the NF90 and NF270 membranes. Under optimal conditions for the NF90 membrane, the $Ni^{2+}$ and $Cr^{6+}$ rejection values were 99.2 and 96.5%, respectively. For the NF270 membrane, the $Ni^{2+}$ and $Cr^{6+}$ rejection values were 98.7 and 95.7%, respectively [43]. Lead ion rejection was greater than 98%, and increased slightly with increasing crossflow velocity and operating pressure and decreased slightly with increasing feed metal concentration. The rejection and permeate flux decreased slightly and increased sharply, respectively, with increasing feed pH [44]. The elimination of $Ni^{2+}$, $Pb^{2+}$, and $Cu^{2+}$ ions from synthetic water using nanofiltration was 85%, 78%, and 66%, respectively, under specific conditions of concentration, pH, and temperature [45]. The pH of the solution and feed concentration has a significant impact on the separation of metals, and the pressure plays a different role in the rejection and permeability of metals, depending on the kind of membrane. According to the solution diffusion theory, water and solute molecules diffuse through each dense layer [46]. The rapid diffusion of water molecules and slow diffusion of solute molecules result in an increased rejection; meanwhile, the slow diffusion of water molecules and rapid diffusion of solute molecules result in a decreased rejection. The water flux also has a common dilution effect.

Given the fact that much work has been carried out in RO and NF on the rejection of three ions at maximal in solution by adjusting the pH of the feed solution and a chemical pretreatment before filtration, the present study presents a two-fold objective: first, the

evaluation of the performance of several commercial membranes in reverse osmosis and nanofiltration in terms of retention, flow rate, and permeability, applied to synthetic water containing six different ions; and second, the assessment of the retention of several ions in real waters without prior pretreatment, which is a source of pollution to Lake Tanganyika.

## 2. Materials and Methods

### *2.1. Sampling Site of Real Samples*

Real water samples were taken at the discharge collector located at the point indicated in Figure 1. This is the last point after the households and before the real water enters the lake, and where we sampled the maximum amount of real water emitted with the coordinates 029°08′39.0″ of longitude N and 03°24′26.3″ of latitude E in the city of Uvira, in the Democratic Republic of Congo, flowing northwest into Lake Tanganyika. These samples were first filtered on Whatman MN 615. Φ 125 mm REF 431 012 filter paper model to retain large particles in solution, before being filtered through RO and NF membranes in the laboratory. Figure 1 shows the study area, as located by Google Earth and Arc GIS version 9.3 software. GPS was used to determine the geographic coordinates of the sampling sites.

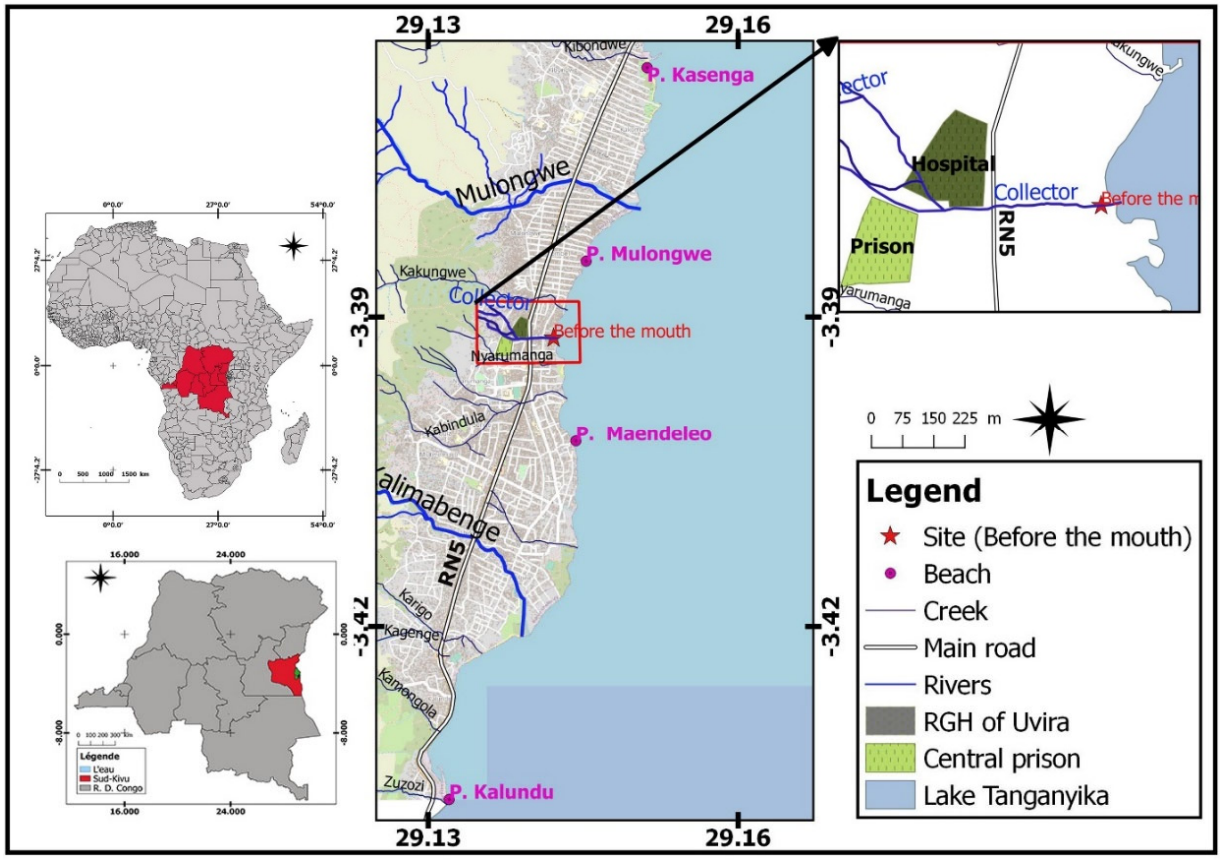


**Figure 1.** Location of sampling sites.

A chemical analysis was made to determine the concentration of each element in the actual wastewater (shown in Table 2). The initial concentrations were determined by IPC-MS and OES, measured in ppb and ppm. ICP-MS was used to detect low-concentration metals in the permeate and real waters and ICP-OES was used to determine toxic metal ions in high concentrations in synthetic waters. Real samples were filtered before being

introduced in the membrane crossflow device. Surprisingly, the initial concentration before filtration was higher than after filtration, which may be due to the adsorption of metals on the removed particles. In Table 3, the measured concentrations before and after filtration are shown.

**Table 2.** Ion concentration in the real water sample.

| Elements | Concentration Before Filtration (ppm) * | Concentration After Filtration (ppb) * |
|---|---|---|
| Co | <0.05 | 28.13 |
| Al | 0.155 | <1 |
| As | <0.05 | 6.36 |
| Ba | 0.085 | 16.96 |
| Cd | <0.05 | <0.05 |
| Cr | 0.06 | <0.05 |
| Cu | <0.05 | 5.66 |
| Fe | 0.815 | 23 |
| Mn | 0.47 | 337.27 |
| Ni | 0.03 | 6.25 |
| Pb | <0.05 | <0.05 |
| Sb | <0.05 | 1.57 |
| Sn | <0.05 | <0.05 |
| Ti | <0.05 | 26.33 |
| W | <0.05 | 5.27 |
| Zn | <0.05 | 184.26 |
| Ca | 40 | 47,390 |
| K | 66.5 | 53,920 |
| Mg | 15 | 16,900 |
| Na | 78 | 930 |
| Si | 16 | 18,700 |

* ICP-OES was used to determine the concentrations before filtration (in ppm) and ICP-MS was used to measure the concentration after filtration (in ppb).

**Table 3.** Specifications of the membranes used in the experiments (information provided by the supplier).

| Characteristics | RO X-20™ Membrane | NF90 Membrane | NF270 Membrane |
|---|---|---|---|
| Allowable operating pH range | 4–11 Continuous | NA | NA |
| Maximum operating temperature, °C | NA | 45 | 45 |
| Maximum feed turbidity, NTU | 2 | NA | NA |
| Maximum feed SDI, 15 min | 5 | NA | NA |
| Average salt discharge (%) | 99.5 | 97 | |
| Minimum salt discharge (%) | 98.5 | NA | NA |
| Filtration pressure (bar) | NA | 41 | 41 |

NA: not available.

*2.2. Synthetic Samples*

In order to determine the membrane performance under different metal concentrations in water, synthetic solutions were prepared by adding specific amounts of metals, simulating high-concentration effluents (i.e., industrial effluents) and low-concentration

effluents (i.e., water in natural water bodies) of heavy metals (chromium, arsenic, nickel, antimony, lead and cadmium). The low-concentration solutions had 15 ppm, and the high-concentration solutions had 56 ppm; the solutions were freshly prepared from an analytical-grade standard solution (Sigma Aldrich, Merck KGaA 64,271 Darmstadt, Germany) containing 1000 mg $L^{-1}$ of $As^{3+}$, $Cr^{3+}$, $Pb^{2+}$, $Cd^{2+}$, $Ni^{2+}$, and $Sb^{3+}$. A standard solution (Certipur®) was used to prepare the stock solutions.

### *2.3. Membrane Crossflow System Setup*

The Armfield FT17 lab-scale device was used for evaluating the performance of membranes in a crossflow filtration application. This device enables rapid determination of crossflow filtration performance using a range of membrane types with relatively small sample volumes (1 L). Figure 2 shows the schema of the experimental setup. Both prefiltrated real samples (Section 2.1) and synthetic solutions (Section 2.2) were used as the feed solution in the crossflow setup.

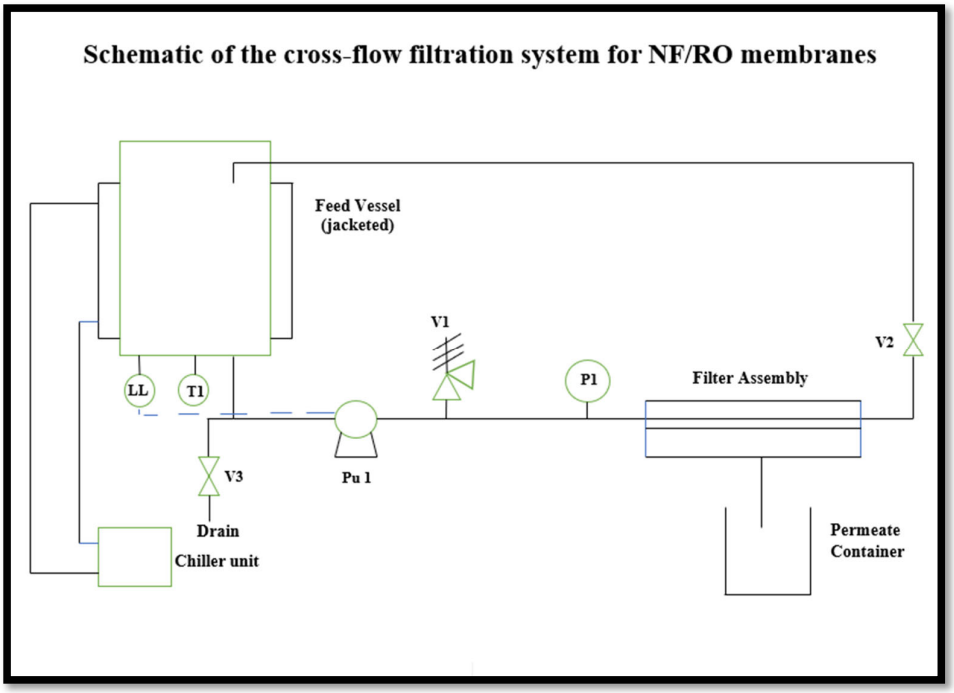


**Figure 2.** Legend: PI: pressure indicator (left); LL: low-level sensor (pump protection); LH: high level (vessel overfilling protection); P1: pressure sensor; T1: temperature sensor; V3, V2, V1: valves.

### *2.4. Commercial Membranes*

The membranes used in this work are the polyamide urea RO X-20™, NF90, and NF270, supplied by Lenntech Water Treatment Solutions (Distributieweg 3, 2645 EG, Delfgauw, The Netherlands). The selection of these membranes for this study was based on average salt removal. The specifications of these membranes are presented in Table 3.

### *2.5. Experimental Procedure*

Before the experiment, the membranes were soaked in ultrapure water for 30 min. The membranes were then inserted into the crossflow RO setup to filter the samples at different pressures, notably 50 bar for RO and 38 bar for NF. The choice of this pressure is justified by the characteristics of the nanofiltration membrane, which can be operated up to a maximum pressure of 41 bar. Referring to the literature [43], the pressure of 10, 20, and 30 bars did not significantly change the rejection at nanofiltration.

The prefiltrated real water (see Table 2) and synthetic solutions were filtered in the RO unit. The feed capacity was 1/2 L at a constant maximum pressure of 50 bar in RO and 38 bar in NF. The temperature varied from 299.14 K for ions concentrated at 15–26 ppm and 303.14 K for ions concentrated at 56 ppm in RO, and in nanofiltration was 296.14 K for ions 15–26 ppm at 297.14 K for 56 ppm concentration. After filtration, the permeate and retentate of the real waters, because of their low feed concentrations, were analyzed by the standardized method [47,48] with an inductively coupled plasma mass spectrometer (ICP-MS), model NexION 5000 Multi-Quadrupole ICP Mass Spectrometer, provided by PerkinElmer®, and inductively coupled plasma optical emission spectroscopy (ICP-OES) provided by Agilent Technology was used for the synthetic solutions because of their higher concentrations. This analytical technique allows for the identification of the atomic composition of a particular sample. It uses the unique photophysical signals of each element to successfully detect the type and relative amount of each element within the complexity of a compound. From values of composition in the retentate and permeate (Cr and Cp, respectively), the retention was calculated as indicated in Table 4. In addition, the permeation flux, J, and the permeability, Lp, were obtained using the equations also listed in Table 4. In this work, the osmotic pressure was 0.026 bar, leading to a driving pressure (transmembrane pressure) of 48.96 bar.

**Table 4.** Main parameters used to evaluate the performance of RO and NF membranes.

| Parameter | Formula | Equation | Ref |
|---|---|---|---|
| Retention (%)<br>where *Cp* and *Cr* are the permeate and retentate<br>Mass balance calculation | $R = \left(1 - \frac{Cp}{Cr}\right).100$<br>$(C.V)_f = (C.V)_r + (C.V)_p$ | (1) | Luis et al. (2018) [49] |
| Permeation flux<br>flux (*J*)<br>(L. $m^{-2}.h^{-1}$) | $J = \frac{Qp}{S}$<br>where *Qp* is the permeate flow rate (L $h^{-1}$), *S* (0.00664 $m^2$) is the membrane area | (2) | Luis et al. (2018) [49] |
| Driving pressure *DP*<br>$\Delta\pi$ is the osmotic pressure gradient (bar)<br>*C* is the total concentration of ions (mol $L^{-1}$)<br>*R* = 0.082 L atm $K^{-1}$ $mol^{-1}$<br>*T* (K) | $DP = \Delta P - \Delta\pi$<br>$\pi = RTC$<br>$C = \frac{n}{V}$<br>with *n* as the total number of moles of ions | (3) | Van der Bruggen et al. (2018) [50] |
| Membrane Permeability<br>*Lp* is the solvent (water) permeability<br>(L $m^{-2}$ $h^{-1}$ $bar^{-1}$) | $Lp = \frac{J}{DP}$ | (4) | Luis et al. (2018) [49] |

## 3. Results and Discussion

### *3.1. Effect of the Metals' Feed Concentration on the Membrane Performance*

The removal of ions by the studied membranes is shown in Figure 3 using synthetic solutions with 15, 26, and 56 ppm of metals. The synthetic solutions were filtered under a pressure of 50 bar at 303.14 K for the reverse osmosis membrane, and 38 bar at 297.14 K when using the nanofiltration membranes.

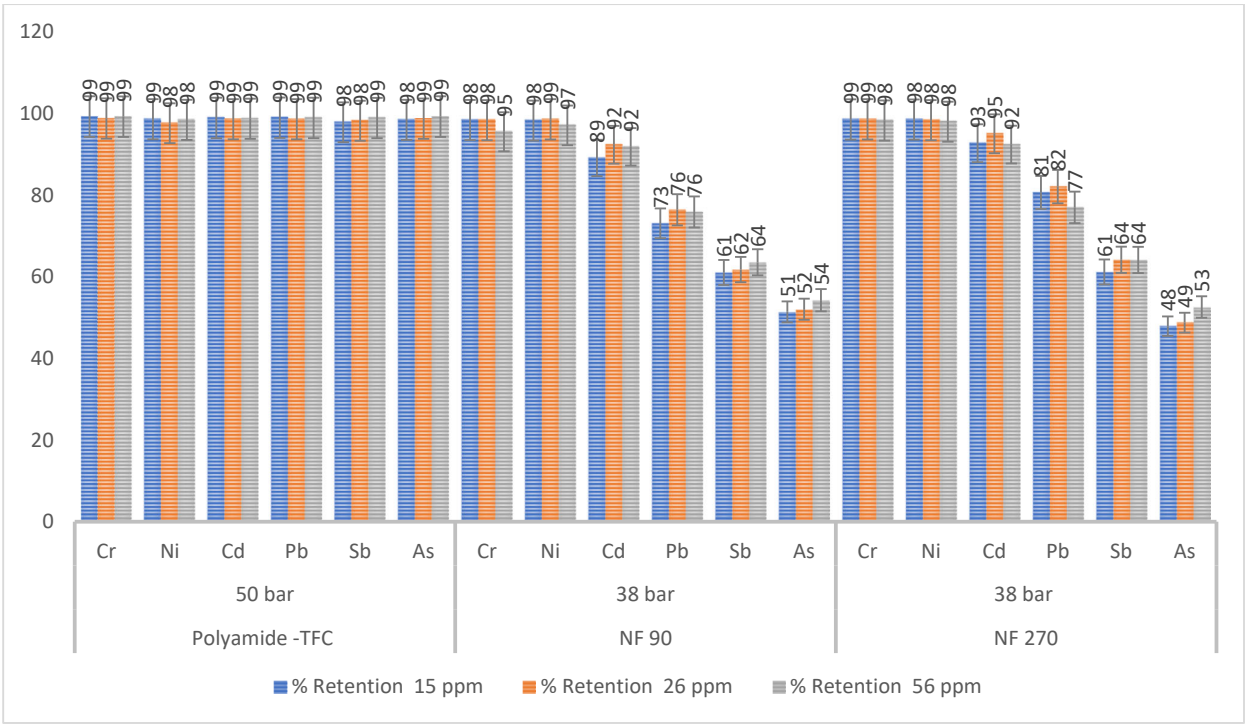


**Figure 3.** Removal of heavy metals as a function of feed concentration.

The results showed that the retention of $Cr^{3+}$, $Cd^{2+}$, $Ni^{2+}$, $Pb^{2+}$, $Sb^{3+}$, and $As^{3+}$ ions was >98% (99.2, 98.6, 98.8, 99.2, 98.4, and 98.8%, respectively) independently of the concentration for the reverse osmosis membrane.

It was observed that the rejection of $Cr^{3+}$ is similar to that of $Ni^{2+}$, although the size of $Cr^{3+}$ is larger than that of $Ni^{2+}$. These two ions are larger than $Cd^{2+}$, $Pb^{2+}$, and $Sb^{3+}$, of which the sizes are the same. Removal of antimony was found to be <65% and arsenic <55% in nanofiltration.

The RO membrane gave much higher ion rejection than the nanofiltration membranes. The rejection of ions by the NF 90 membrane was almost at the same level as that at NF270, except for the 56 ppm feed solution, which was higher. Therefore, it was considered that size exclusion is not the only key factor in ion rejection. However, NF270 gave a lower ion rejection than NF 90 due to its larger pore size and also due to the interaction with the membrane. The t-test on single samples shows that the difference is not significant between NF90 and NF270 ($p$-value: 0.6 to 0.8) for all concentrations, but it is highly significant between RO and NF ($p$-value: 0.001 to 0.0001). The structure of the RO and NF membranes would be the factor to influence the p-value. Very few studies have been conducted on the removal of antimony by membrane processes, unlike arsenic. Kang et al. [37] achieved a rejection of 60% Sb (III) and 95% Sb (V) by reverse osmosis using the ES-10 membrane, and 45–60% Sb (III) and 95% Sb (V) rejection using the NTR-729HF membrane. These values are lower than our research results using the reverse osmosis membrane.

Previous studies showed that 99% rejection As (V) could easily be achieved with NF and RO membranes. The Pb retention is low and ranges from 73 to 82% using nanofiltration. However, despite this difference in sizes between the ions, the retention is very high and is almost similar in reverse osmosis, due to the interaction between the ions. Similar results have been reported by [51,52], who mentioned that $Cd^{2+}$ can be removed at a slightly higher efficiency compared to $Cu^{2+}$.

In recent years [53] many works have been devoted to the removal of heavy metal ions using nanofiltration. It was reported the application of a thin-film composite nanofiltration polyamide membrane for the rejection of nickel ions from aqueous wastewater [54]. It was observed a nickel rejection of 98% and 92% for a feed concentration of 5 and 250 mg/L respectively, and they studied the binary separation capacity of heavy metals (cadmium and nickel) of a commercial NF membrane starting from solutions [55]. The maximum solute rejection observed for nickel and cadmium ions was 98.94% and 82.69%, respectively, for an initial feed concentration of 5 mg/L. The nickel rejection is similar to our results, unlike cadmium, which varies between 89 to 95%, and it is greater than their result. This difference would be due to the initial concentration and the pressure applied. Household and industrial wastewaters contain other heavy metal ions at various concentrations.

Several research studies have been conducted on the removal of As (III) and (V) by NF [53], and the removal of As (V) and As (III) was comparable. The low rejection of As (III) suggests that size exclusion governed their separation behavior and not charge interaction.[53]. Boussouga et al. [37] confirmed the efficiency of NF to remove As (V), unlike As (III), which is much more difficult to remove, except when tight-pore NF membranes are used. Furthermore, NF membranes can only reject <50% of As (III) at pH ≤ 8 [56,57]. Sato et al., Sen et al., and Tay et al. [58,59] recommended pretreatment by oxidation prior to filtration to improve As (III) rejection. These results highlight the role of charge interactions between the membrane and ions in determining the rejection performance of NF membranes, particularly for As species [60]. The X-20TM type TFC PA RO membrane was efficient in removing heavy metals (at low- or high-feed solution concentration), but the permeate flux was low and the applied pressure was very high. The nanofiltration membranes showed good ion rejection at low concentrations and the permeate flux was very high, but the retention was low for high-concentration feed solutions. Many low-income countries have very limited energy resources; consequently, the application of nanofiltration becomes relevant to lift this heavy metal challenge. On the other hand, despite the excessive energy consumption in RO, it still has many advantages to removing inorganic, organic, and microplastic micropollutants [61].

### *3.2. Ion Removal in Real Waters*

It was observed that the retention of ions in the real wastewater, filtered by reverse osmosis, at 50 bar pressure at 303.14 K, for Co, Al, As, Ba, Cr, Cu, Ni, Sb, Sn, Ti, and Zn ions was 100% and for Fe, Mn, W ions, was 94.5, 99.2, and 88.7, respectively.

The retention of Co, Al, Ba, Cr, Cu, and Sn ions was also 100% by NF90 and NF270 membranes, while for As, Fe, Mn, Ni, Ti, and Zn ions, it was 99.8, 82.2, 88, 94.9, 99.2, 96.6% and 98.3, 90, 92.5, 98.6, 99.1, 95.9%, respectively. Tungsten was not retained by NF90 but NF270 had retained it by 27.2%. Cd, Pb, and Sn were below the ICP-MS detection limit.

The complete elimination of toxic ions and low level of heavy metal concentration in the permeate, below the WHO standard, means that good-quality water can be obtained. Figure 4 below, shows the ion retention of real waters.

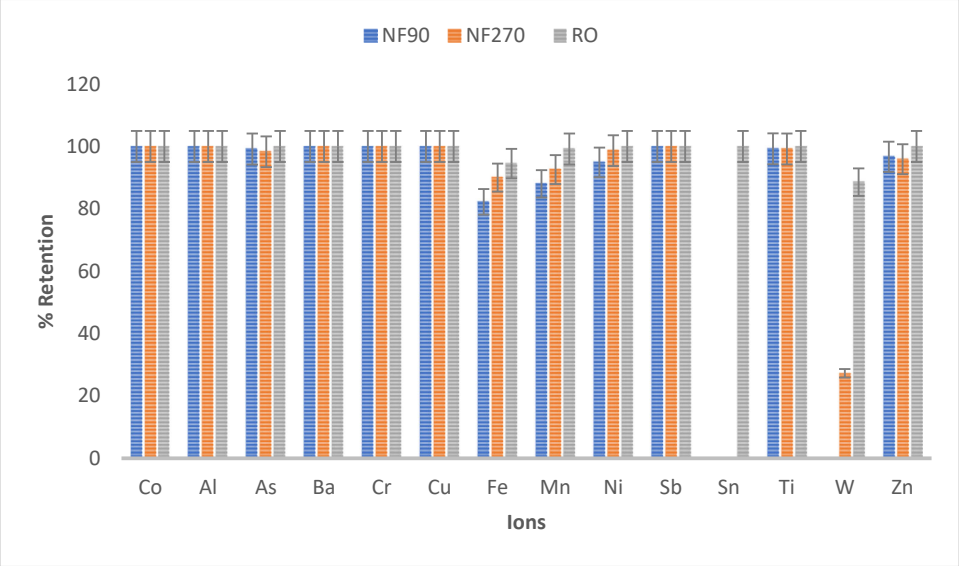


**Figure 4.** Retention of charged ions in real waters; analysis by ICP-MS.

The filtration of real wastewater with RO and NF eliminated the ion concentration to the WHO standard, as shown in Table 5.

**Table 5.** Ion concentrations in permeate.

| | Ions Conc (ppb) | | |
|---|---|---|---|
| **Elements** | **NF90** | **NF270** | **RO** |
| Co | 0 | 0 | 0 |
| Al | 0 | 0 | 0.00 |
| As | 0.30 | 0.36 | 0.00 |
| Ba | 0 | 0 | 0 |
| Cd | <0.05 | <0.05 | <0.05 |
| Cr | 0 | 0 | 0 |
| Cu | 0 | 0 | 0 |
| Fe | 64.66 | 17.96 | 6.54 |
| Mn | 78.10 | 90.70 | 0 |
| Ni | 1.30 | 0.91 | 0 |
| Pb | <0.05 | <0.05 | 0 |
| Sb | 0 | 0 | 0 |
| Sn | <0.05 | <0.05 | 0 |
| Ti | 0.33 | 0.41 | 0 |
| W | 5.06 | 3.80 | 1.51 |
| Zn | 12.90 | 17.85 | 0.00 |

### *3.3. Comparison of Real and Synthetic Water Retention*

Figure 5 shows the results of retention of the RO and NF membranes for real and mimicked surface water. It is observed that the removal of metals from the real water was higher compared to the synthetic solution. In real water, under the same feed pressures of 50 bars at 303.14 K in reverse osmosis and 38 bars at 297.14 K in nanofiltration, the Cr and Sb ions were completely eliminated from the permeate filtered by the three membranes. The rejection of $Ni^{2+}$ and $As^{3+}$ ions for NF90 was 99.20% and 99.18%, and for NF270, it was 98.68% and 98.3%, respectively. The $Cd^{2+}$ and $Pb^{2+}$ values in the permeate were be-

low the detection limit by ICP-MS. No trace of these ions had been identified in the permeate after filtration by reverse osmosis. The retention of $Ni^{2+}$ by NF90 (99.2%) agreed with the results obtained by Basaran et al. [44], for the same membrane.

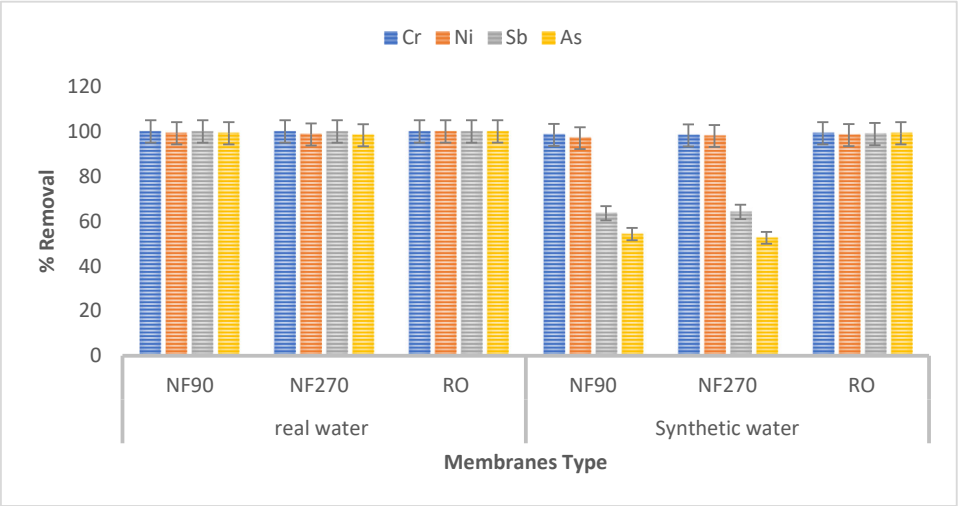


**Figure 5.** Comparison of % removal between synthetic waters and real water.

On the other hand, for the synthetic solution, the rejection of $Cr^{3+}$, $Ni^{2+}$, $Sb^{3+}$, and $As^{3+}$ ions were 98.49, 97.06, 63.53%, and 54.25%, respectively, for NF90, followed by 98.25, 97.97, 64.11%, and 52.59% for NF270 and finally 99.2, 98.41, 98.87%, and 99.21% for reverse osmosis. Those values are the same as the ones obtained for real wastewater, except for $As^{3+}$ and $Sb^{3+}$.

The high rejection of the real waters can be explained by the low concentration of the load in ppb, unlike the concentration of the synthetic solution, which was 56 ppm. Lower concentrations in the feed thus involve higher rejection. This is in agreement with the results obtained by Shigidi et al. [62], whereby the removal of ions at 99.8% was possible in a 5 ppm feed solution and decreased to 94.3% in a 30 ppm feed solution and finally to 77.2% for the 100 ppm feed solution. Boussaga et al. [37] found that the presence of humic acids in wastewater can influence the removal of ions. [37].

Table 6 reviews the developments and potential applications related to nanofiltration and reverse osmosis, with a particular focus on heavy metal removal. Results from the present study show that the application of RO without chemical pretreatment achieved a more superior removal of toxic metal ions than that obtained by Chan et al. [52], Algureiri et al. [45], and Shigidi et al. [62], and was similar to other works, such as Ipek et al. [41] and Mohsen-Nia et al. [40], whose feed solutions were chemically treated. Removal of Cr, Ni, and Cd ions was excellent in nanofiltration and like previously treated feed solutions.

**Table 6.** List of key studies on heavy metal removal by using reverse osmosis and nanofiltration.

| Type | Membrane | Element | Initial Conc | Type of Water | Removal Efficiency (%) | Conditions | Conc Permeate | Ref |
|---|---|---|---|---|---|---|---|---|
| NF | NF270 | As (V) | 250 µg/L | Synthetic | 82–88 | Feed pressure of 20 bars | | Youssef et al., 2021 [37] |
| | NF90 | | | | 93–98 | | | |
| RO | ES-10 | As III | 50 µg/L | Drinking water | 75 | 0.75 MPa | ND | Kang et al., 2000 [38] |
| | | Sb III | | | 60 | | | |
| | | As V | | | 95 | | | |
| | | Sb V | | | 95 | | | |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | NTR-729Hf | As III | | | 20–43 | | | |
| | | Sb III | | | 45.7–60.2 | | | |
| | | As V | | | 80–95 | | | |
| | | Sb V | | | 95 | | | |
| RO | Polyamide | $Cd^{2+}$ | 250 ppm | Synthetic | 98.5 | ND | 3.2 ppm | Qdais et al., 2004 [39] |
| NF | | | | | 82–97 | | 13.16 ppm | |
| | | $Ni^{2+}$ | 500 mg/l | | 99.5 | Operation pressure 5 atm | 2.01 ppm | Mohsen-Nia et al., 2007 [40] |
| | | Cu II | | | | | | |
| | | As III | <500 μg/L | | 20–55 | 3 atm | ND | Chan et al., 2008 [52] |
| | | As V | | | 91–99 | | | |
| RO | | $Ni^{2+}$ | 44–169 mg/L | | 99.3 | Operational pressure 1100 kPa | | Ipek et al., 2005 [41] |
| | | As III | ND | | 99 | ND | ND | |
| | | As V | | | 99 | | | Jekel et al., 2006 [42] |
| NF | NF70 | As III | | | <30 | | | |
| | | As V | | | 60–90 | | | |
| | | Pb II | | | 86 | 0.75 MPa | ND | |
| | | Ni II | | | 93 | | | |
| | | Cu II | | | 98.72 | | | |
| NF | Polyamide | Pb II | 1 ppm | Drinking water | 99.61 | | | Maher et al., 2014 [43] |
| | | Mn II | | | 99.31 | | | |
| | | Ni II | | | 99.11 | | | |
| | | Zn II | | | 99.51 | | | |
| | NF90 | Ni II | | | 99.2 | | ND | |
| NF | | Cr VI | | Wastewater | 96.5 | 10, 20, and 30 bars | | Basaran et al., 2015 [44] |
| | NF270 | Ni II | | | 98.7 | | | |
| | | Cr VI | | | 95.7 | | | |
| NF | AFC 80 | Pb II | 5–250 mg/L | Synthetic | >98 | 25 bars | <0.05 ppm | Gherasim et al., 2014 [45] |
| | | Pb II | 15 mg/L | Wastewater | >99 | | | |
| | | Cd | 5 mg/L | | >99 | | | |
| | | Ni II | | | 98.5 | | ND | |
| RO | | Cu II | | | 97.5 | 1 and 4 bars | | |
| | | Pb II | | | 96 | | | |
| | | Ni II | 50 ppm | Synthetic | 85 | | | Algureiri et al., 2016 [46] |
| NF | | Cu II | 100 ppm | | 66 | 1, 2, 3 and 4 bars | | |
| | | Pb II | 150–200 ppm | | 78 | | | |
| RO NF | | Cu II | 2 g/L | Synthetic | >95 | Operating pressures 35 bar | ND | Cséfalvay et al., 2009 [51] |
| | | Co II | 39.4 | | 98.6 | | | |
| RO | TFC PA | Ni II | 214.9 | Synthetic | 98.1 | 41 bars | ND | Ricci et al., 2017 [63] |
| | | Mg II | 242.9 | | 98.6 | | | |
| | | Pb II | 0.034 | | <1 ppb | | <1 ppb | |
| | | Zn II | 0.153 | | <0.002 ppb | | <0.002 | |
| RO | | Cd II | 0.025 | Real water | <0.1 ppb | 1.76 MPa | <0.1 ppb | Thaçi, et al., 2019 [64] |
| | | Co II | 0.018 | | <0.2 ppb | | <0.2 ppb | |
| | | Mn II | 1.146 | | 99.48 | | 0.006 ppb | |
| | | Ni II | 0.004 | | <0.5 ppb | | <0.5 ppb | |
| RO | BW30XFR | Cr VI | 5 | Synthetic | 99.8 | | ND | |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | | 30 | | 94.3 | 10, 30, and 45 bars | | Shigidi et al., 2022 [62] |
| | | | 100 | | 77.2 | | | |
| RO | X-20TM | Cr III | 15, 26, 56 ppm | Synthetic | 99 | 50 bars | 0.15–1.02 | |
| | | Ni II | | | 98–99 | | | |
| | | Cd II | | | 99 | | | |
| | | Pb II | | | 99 | | | |
| | | Sb III | | | 98–99 | | | |
| | | As III | | | 98–99 | | | |
| NF | NF90 | Cr III | 15, 26, 56 ppm | Synthetic | 95–98 | 38 bars | 0.33–22 | This study |
| | | Ni II | | | 97–99 | | | |
| | | Cd II | | | 89–92 | | | |
| | | Pb II | | | 73–76 | | | |
| | | Sb III | | | 61–64 | | | |
| | | As III | | | 51–54 | | | |
| | NF270 | Cr III | 15, 26, 56 ppm | Synthetic | 98–99 | 38 bars | 0.11–22 | |
| | | Ni II | | | 98 | | | |
| | | Cd II | | | 92–95 | | | |
| | | Pb II | | | 77–82 | | | |
| | | Sb III | | | 61–64 | | | |
| | | As III | | | 48–53 | | | |

ND: No data.

### *3.4. Transmembrane Flux*

The results in Sections 3.1–3.3 show that the performance of membranes is evaluated based on the concentrations of the feed solutions. Permeate flux is one of the key factors demonstrating the effectiveness of membranes in resisting fouling [49].

The permeate flux of synthetic solutions and real waters was determined on the three membranes. The experimental results are presented in Figure 6 below, under operating pressures of 48.9 bar in reverse osmosis and 36.9 bar in nanofiltration. The nanofiltration membranes exhibited a high flux and a more severe flux drop in reverse osmosis.

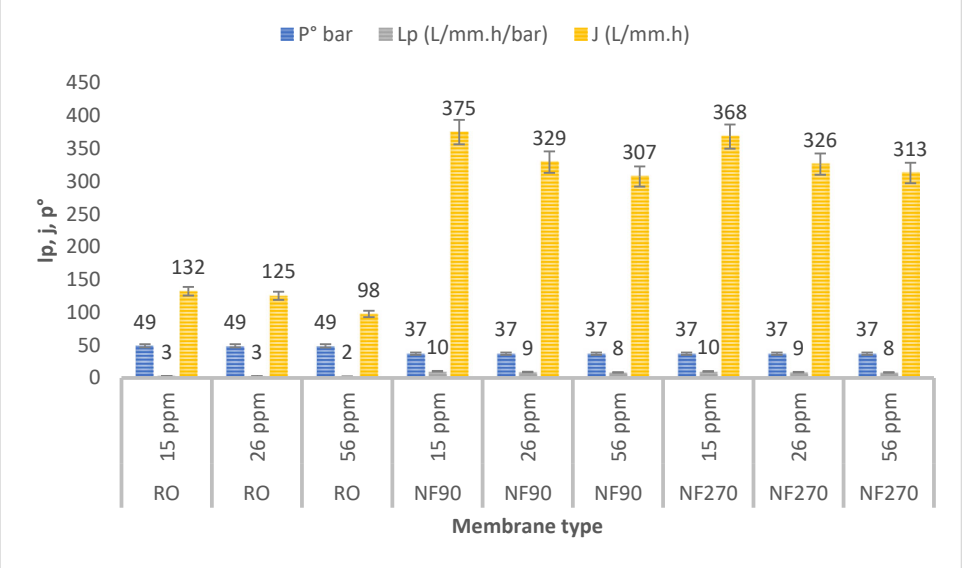


**Figure 6.** Permeability and permeate flow at constant pressure.

The water permeabilities of the nanofiltration membranes were much higher than those of the osmosis membranes and are, respectively, 2.7, 2.5, and 1.9 L/m².h/bar for RO, 10.1, 8.9, and 8.3 L/m².h/bar for NF90, and finally 9.9, 8.8, and 8.4 L/m².h/bar for NF270.

A study [65] showed that the water permeability of NF90 and NF270 membranes were 6.05 and 14.86 L/m².h/bar and that of seawater reverse osmosis (SWRO) was 5.91 L/m².h/bar. Another study [66] showed that pure water permeability and salt rejection of both NF90 and BW30 membranes were measured by crossflow permeation tests with pure water and an aqueous solution of 1000 mg/L of NaCl, and were found to be 9.4 and 4 L/m².h/bar, respectively. The difference in permeability is explained by the crossflow filtration system, the applied operating pressure, and the solute concentrations.

The increase in the concentration of solute slightly decreased the membrane permeability, and the temperature was raised from 299.14 to 303.14 K for RO, and from 296.14 to 297.14 K for NF. Low permeability due to solute concentration was observed at 15 ppm nanofiltration and 56 ppm reverse osmosis. Nanofiltration membranes had the highest water permeability and the rejection was lower than for reverse osmosis.

## 4. Conclusions

Three commercial nanofiltration and reverse osmosis membranes were successfully tested using synthetic solutions prepared in the laboratory and real water. The results showed that among the studied membranes, the reverse osmosis membranes had a lower permeability compared to nanofiltration membranes and the ion rejection was higher for the synthetic solutions. The ion release rate in the real water was higher and met the WHO standard.

The elimination of heavy metals at low concentrations in the permeate implies that good-quality water can be transported to Lake Tanganyika without disturbing the ecosystem, as well as perhaps the possibility of reusing it by the local population.

In conclusion, nanofiltration is recommended for the removal of heavy metals from low-concentration wastewater, and reverse osmosis would be the best choice for a very high concentration of heavy metals in wastewater. However, commercial membranes are very expensive and present some challenges in terms of excessive energy consumption, are sometimes inefficient for removing some heavy metals, and a low flux is observed. It would therefore be interesting to develop specific low-cost membranes such as mixed-matrix membranes (MMM) based on a biodegradable polymer and nanoparticle, which combine several advantages in terms of high permeate flux, good selectivity, low manufacturing cost, and low energy consumption.

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## Abbreviations

| | |
|---|---|
| Conc | Concentration |
| DRC | Democratic Republic of Congo |
| FO | Forward osmosis |
| ICP-MS | Inductively coupled plasma mass spectrometer |
| ICP-OES | inductively coupled plasma optical emission spectroscopy |
| Init | Initial |
| MF | Microfiltration |
| MMM | Mixed-matrix membranes |
| Na2EDTA | Disodium salt of ethylenediaminetetraacetic acid |
| NA | not available |
| NF | Nanofiltration |
| ND | No data |
| PA | Polyamide |
| RO | Reverse osmosis |
| SWRO | Seawater reverse osmosis |
| TFC | Thin-film composite |
| UF | Ultrafiltration |
| WHO | World Health Organization |

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