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Research paper

# A comparative study of community reverse osmosis and nanofiltration systems for total hardness removal in groundwater

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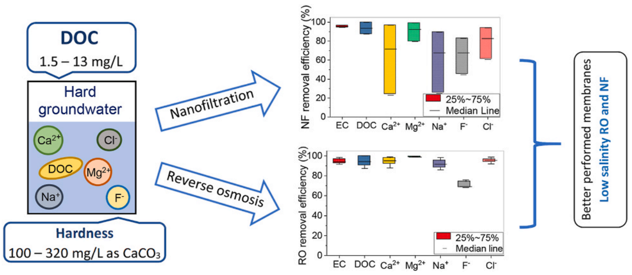
## HIGHLIGHTS

- Seventy percent of field reverse osmosis plants (RO) behave as low-pressure RO systems.
- Groundwater treatment units are designed and installed without proper investigation on feed water quality.
- Nanofiltration (NF) has a better potential to solve the mineral deficiency issue in treated water.
- NF technology is a suitable alternative for the treatment of groundwater with high hardness and DOC.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In this research, the efficiency of reverse osmosis (RO) and nanofiltration (NF) treatment methods installed in the dry zone (Sri Lanka) were examined as a function of operation parameters, feed and product water quality, membrane type, user maintenance, and wastes handling. The quality of the feedwater varies as electrical conductivity (265–1329 mg/L), total hardness (97–318 mg/L as $CaCO_3$) and fluoride (0.58–2.93 mg/L), often exceeding WHO or SLS tolerance limits. About 77% of the feedwater contains dissolved organic carbon (DOC) above 4.00 mg/L that requires control to avoid membrane fouling and other harmful effects. The salt rejection ratio, pressure flux and transmembrane pressure of the RO/NF membranes vary widely with the feedwater quality. The operation parameters of the membrane plants vary significantly; eight RO plants use low-pressure (0.5–1.0 MPa) similar to the NF pressure requirement, but they desalinate water at 95% salt rejection efficiency. Most RO membranes deviate (>50%) from the recommended transmembrane pressure and specific permeate flux values. However, such deviations are not observed in NF treatment plants. RO membranes remove solutes excessively, but post mineralization step is currently not practiced in the study area. The water recovery

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by the NF membranes is highest (>60%) compared to RO plants. The wastewater resulting from the NF plants is handled satisfactorily compared to RO-generated wastes. In the area examined, the salinity occurs due to water's permanent hardness. Therefore, NF membrane-based technology is suitable to desalinate groundwater in the dry zone (Sri Lanka).

## 1. Introduction

The worlds' only available water resource in arid and semi-arid regions is groundwater which fulfils almost half of the drinking water requirements. However, over 60% of the aquifers in these regions exhibit high salinity due to lithogenic sources (World Water Quality Alliance, 2021). In the arid or dry areas in Sri Lanka, the lack of safe drinking water due to high salinity is further aggravated by excess dissolved organic matter (Indika et al., 2021; Xu et al., 2021). Both excess or low water salinity induces palatability issues rendering low water consumption, particularly among farmers frequently exposed to heated environments resulting in health problems such as dehydration and chronic kidney disease of unknown etiology (CKDu) (Chandrajith et al., 2011; Imbulana and Oguma, 2021).

To resolve water salinity issues in Sri Lanka, the State and private organizations have introduced several interim measures, such as the provision of truck-driven water, and distribution of bottled water and softeners, with limited success. In 2013, the Sri Lankan water authorities introduced reverse osmosis desalination plants to the regions where water-related health problems are prevalent with a lack of pipe-borne water (Indika et al., 2021; Zhong et al., 2019). In 2018, nanofiltration (NF) was introduced to Sri Lanka, yet they are not common compared to the RO systems (Zhong et al., 2019). Over 2000 RO treatment units are in operation in the dry zone (Sri Lanka) (Indika et al., 2021), but only 8 NF plants have been established in Sri Lanka to date.

Both RO and NF technologies operate on pressure-driven water desalination. The operating pressure of the NF membranes is lower than RO. However, the RO plants can operate under different pressures; high pressure (>2 MPa), low pressure (0.7–2 MPa), and ultra-low pressure (<0.5 MPa) (Shon et al., 2013). However, the low-pressure reverse osmosis (LPRO) membranes perform at 98.5% solutes rejection (Wang et al., 2015), and they are comparable to NF (0.7–3 MPa) (Bellona et al., 2008; Tagliabue et al., 2016). The RO membranes efficiently remove monovalent ions, whereas NF membranes are selective for removing di- or higher valence ions. Therefore, RO membranes are well suited to desalinate seawater, and a nanofiltration is a viable option for hardness removal. The appropriate membrane topology should be judiciously introduced after characterizing the feedwater since desalination efficiency largely depends on its quality (and also membrane type, pressure, flux, temperature) (Tian et al., 2021). However, most of the treatment plants were installed without paying much attention to feed groundwater quality and quantity. For example, only four studies are available in the region to probe the treatment efficiency of pressure-driven membranes and treatment quality maintenance practices (Imbulana et al., 2020; Indika et al., 2021; Jayasumana et al., 2016; Zhong et al., 2019). Most of the membrane treatment plants show similar technical problems, namely, declined water recovery (19%–64%), membrane fouling and scaling, concentrate disposal issues, lack of technical knowledge together with the absence of a proper plant efficiency monitoring program (Imbulana et al., 2020; Indika et al., 2021). Apart from these constraints, high operational and maintenance cost is another considerable disadvantage associated with RO technology. Several studies have suggested the advantages of NF membranes compared to RO in treating groundwater with high hardness and DOC in terms of energy efficiency, cost conservation, and waste generation (Bellona et al., 2008; Tagliabue et al., 2016). But the rejection of other water contaminants, such as monovalent ions, is comparably low in NF than in RO (Taky et al., 2021). Sometimes this property of the NF membrane helps retain solutes in the treated water that favours good health.

In essence, both RO and NF treatment methods in Sri Lanka are inappropriately used. Mostly the membrane treatment units are considered “black boxes”. The selection of RO/NF membranes was carried out arbitrarily, which resulted in over or under exploitation of the technology. Therefore, the study area provides to assess the robustness of the membrane technology for critical operation parameters (Transmembrane pressure, specific permeate flux, water recovery and cleaning frequency) and feedwater quality in a high DOC zone. Here, we evaluated the performance of RO and NF community-scale plants installed in the dry zone (Sri Lanka) using 10 RO and 3 NF plants. We discussed the major drawbacks of both techniques to suggest possible solutions to overcome the existing barriers. The results will complement to enhance the efficiency of groundwater desalination programs those are now in operation in tropical regions.

## 2. Materials and methods

### 2.1. Study area and selection of membrane systems

Groundwater sources located in the North Central Province (NCP) of Sri Lanka were chosen for this work. It is the largest district with a 10,472 km$^2$ area, where 31.4% of the land area is used for agriculture (Department of Census and Statistics, 2013/2014). NCP belongs to the country's dry zone with a mean annual temperature ranging from 26.5 to 28.5 °C and a mean annual rainfall of <900 mm, varied with monsoons. October to February is considered the wet season, while March to September is regarded as the dry season (Department of meteorology, 2019). The province's total population is around 1.3 million in 22 secretariat divisions covering 6.2% of the country's total population (Department of Census and Statistics, 2012) where 1–4% of the people reported for CKDu prevalence (Ranasinghe et al., 2019). Hardness, fluoride, salinity, and DOC are major problems restricting water consumption. Over 2000 reverse osmosis plants have been installed by State and Private Sector organizations. Around 450 RO plants are operated under the surveillance of the Community Based Organizations under the National Water Supply and Drainage Board (NWSDB), Government of Sri Lanka. The data available from the CBOs were chosen for this study (NWSDB,2019). Of the total of 450, only 59 RO plants were selected based on a random sampling method by covering all divisional secretariats (DS) of Anuradhapura (22) and Polonnaruwa (07). Based on population density and CKDu epidemiological data, the study locations were evenly distributed among DS divisions. Forty and nineteen RO plants from Anuradhapura and Polonnaruwa were selected for the survey. They were stratified based on the RO membrane type used in the plants. Altogether ten strata were identified (1–8 plants per strata). A single RO plant was randomly selected from each stratum (10 RO plants in total). Ten RO plants and the existing 3 NF plants in NCP were considered to compare the efficiency and suitability of RO and NF systems. The feed, permeate and concentrate water samples were collected into 50 mL polypropylene bottles during the dry season (March-2020). The locations of RO and NF plants are shown in Fig. 1, and further details, e.g., GPS coordinates, are included in the supplementary material only for the selected RO (10), and NF (3) plants (Table S1).

### 2.2. Operator survey

The differences between the operation and maintenance of the RO and NF are required to identify the selection of simpler and more affordable technology for the villagers. A questionnaire was distributed among the plant operators to receive data regarding the operational conditions, maintenance activities, cleaning routines, water quality monitoring, concentrate disposal, and difficulties experienced during the daily and continuous operation. In addition, the operating pressures, flow rates, plant configurations, pretreatments, post-treatments, membrane names, and brine disposal methods were obtained with the help of plant operator/s and in situ measurements. The technical specifications of the used RO and NF membranes were obtained from the websites of the relevant manufacturers.

### 2.3. *Water quality analysis*

The pH and electrical conductivity (EC) of the water were measured from the unfiltered samples in the laboratory within a day after collection using pH and EC probes (Thermo Scientific Orion Star A 325 Multiparameter Meter), respectively. For dissolved organic carbon measurements, the water samples were filtered through 0.45 μm polyethersulfone syringe membrane filters. The filtered water pH was adjusted (pH~2) with 0.2 M HCl while purging high purity nitrogen gas (99.996%) for 2 min to minimize $CO_2$ contamination in DOC analysis. DOC was measured using a Total Organic Carbon analyzer (TOC-L CSH/CSN, Shimadzu, Japan), and $UV_{254}$ absorbance was measured using an

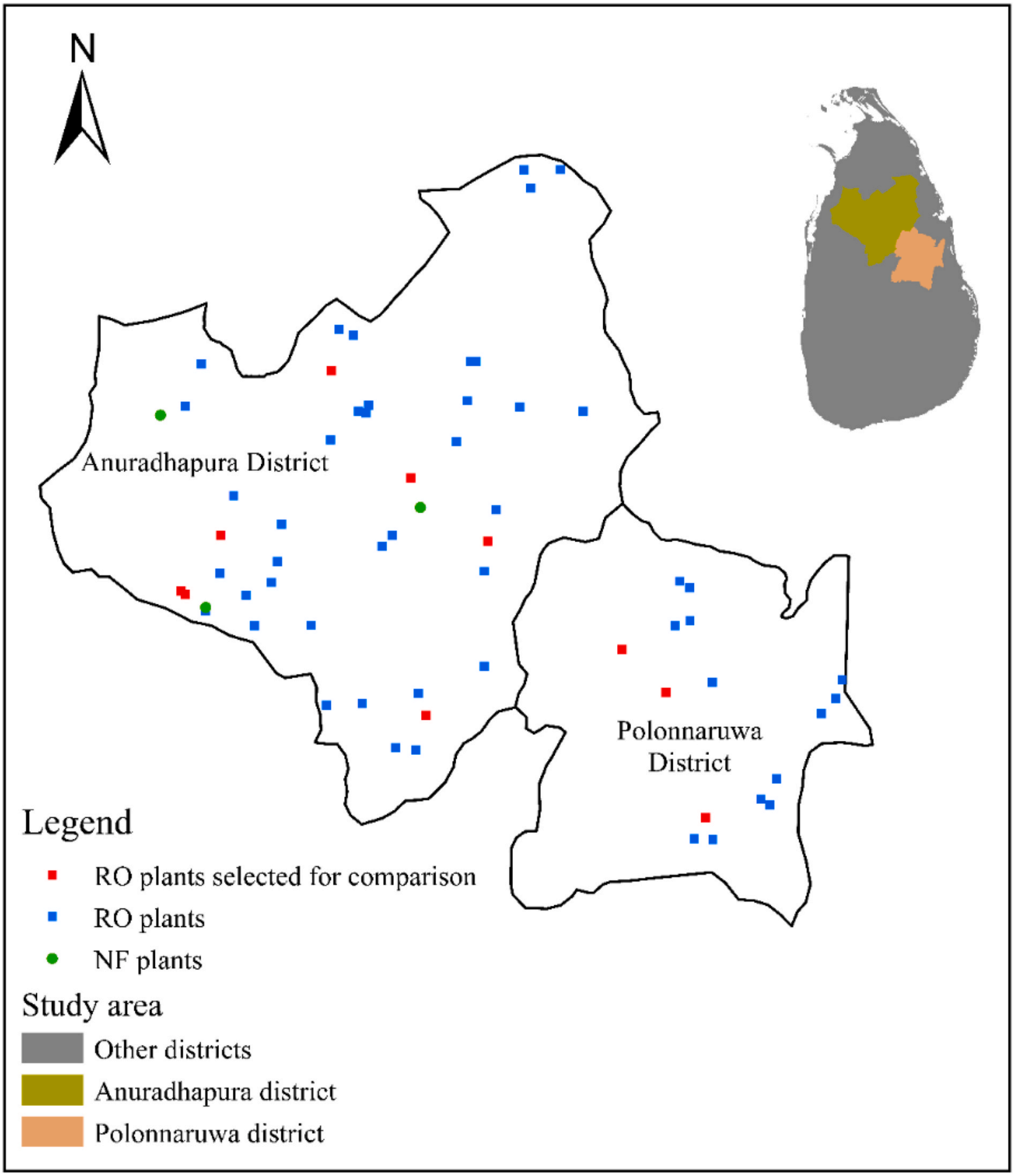

**Fig. 1.** Locations of RO and NF plants in the North Central Province, Sri Lanka.

Ultraviolet–visible spectrophotometer (UV-2700, Shimadzu, Japan). A separate portion of the sample was filtered with 0.22 μm for $Na^+$, $K^+$, $Ca^{2+}$, $Mg^{2+}$, $Co^{2+}$, $Fe^{2+}$, $Bi^{2+}$, $Ni^{2+}$, $Cu^{2+}$, $As^{3+}$, $Se^{2+}$, $Cd^{2+}$, $V^{3+}$, $Cr^{3+}$, $Mn^{2+}$, $Ba^{2+}$, $Zn^{2+}$, $Mo^{2+}$, $Al^{3+}$, and $Sr^{2+}$ analyses by ICP-MS (Thermo ICapQ analyzer, Thermo-Fisher Scientific Inc., Germany) after acidifying with 0.2 M $HNO_3$. Anions ($F^-$, $Cl^-$, $NO_3^-$ and $SO_4^{2-}$) were measured using suppressor Ion chromatography (Metrohm 930 Compact IC Flex, Metrohm AG, Switzerland) from an unacidified portion of the samples. Finally, the hardness values were calculated using $Ca^{2+}$ and $Mg^{2+}$ data. All water samples were preserved by storing below 4 °C until chemical analysis was commenced. Other standard preservation protocols like filtration and acidification of samples immediately were not followed due to the technical challenges faced during sampling in rural areas. The water quality parameters were derived as an average of 3 repetitive measurements.

### *2.4. Membrane performance evaluation*

Membrane performances were evaluated based on transmembrane operational pressure (TMP), water recovery percentage, and salt rejection ratio. Additionally, the membrane-specific permeate flux was also calculated using the field and the Manufacturer's specification data (flow rate observed during membrane testing) using the equation (4) given below (Table 1). The respective equations used for calculations of the above parameters were tabulated in Table 1. The TMP and effective membrane area were extracted from membranes specification data from the relevant manufacturers' specification sheet.

## 3. Results and discussion

### *3.1. Membrane type versus field performances*

In Sri Lanka and most other countries like Australia and Africa, the selection of RO and NF membranes in the fabrication of treatment plants is arbitrary (Burbano et al., 2007). There are no exact specifications or guidelines to select the best membrane combinations to suit a given feedwater quality. Most of the membrane treatment plants in the NCP (Sri Lanka), especially the RO plants, were installed without a reconnaissance survey into operation parameters and feedwater quality. An optimized water treatment unit also requires a critical assessment of the feedwater quality and its seasonal variations and the safe limits of groundwater extraction, which is still lacking in the country.

Both RO and NF plants selected for the study were based on the model type of the membranes. The data were examined in terms of their operational pressure, specific permeate flux, membrane age, and the salinity removal efficiency to evaluate membrane performance and suitability for groundwater treatment. According to the manufacturers' specifications, the membranes used in the field RO units are specified for various applications such as low salinity water, brackish water, harsh water, and residential drinking fountains and water dispensers (Table 2). The total dissolved solids (TDS) and DOC contents of the feedwater vary significantly from 100 to 600 mg/L and 3.9–12.7 mg/L, respectively. The RO membranes are inappropriately used. For example, ultra-low pressure low salinity membrane is suited for the purification of groundwater with high TDS (~600 mg/L), and a high-pressure brackish water membrane is suited for feedwater with low or average TDS (160–350 mg/L). However, for every plant that produces water, the TDS is less than 50 mg/L shows that all membrane types are capable of desalination. From the information acquired from the plant operators, it was identified that the seasonal variation or the available source water quantity during the dry season is not considered when installing the treatment units and it can be supported by the operator statement the feedwater intake well alteration took place in the plant. When the water level reduces in the feedwater well, the common practice is to shift to a different water source, which further signifies that the selection of membrane-type or the feedwater source is not based on any scientific factor. For a successful treatment unit operation, pilot testing for a minimum of 6 months would be beneficial to understand how the plant is operating under different water quality conditions by incorporating the plant operators and the key personnel who will be involved in the unit's operation (Burbano et al., 2007). The DOC and salinity removal depend on the pore size (Kingsbury et al., 2020; Teychene et al., 2020) and the chemical and physical properties of the membrane, such as hydrophilicity, pore structure and surface area (Yang et al., 2019); all RO and NF membranes are composed of active polyamide layer hence chemical properties are more or less similar. However, the DOC rejection by the membranes varies with physical membrane properties such as pore size and distribution and structure. The lack of details on membrane characteristics was a major limitation for a thorough understanding of the behaviour of the individual membrane in water desalination. Also, since the study was conducted on currently operating treatment units, exploring the direct membrane filtration removal efficiencies without pretreatment was challenging as there was no way to bypass the pretreatment unit. Also, no outlets were available after the pretreatment units. Hence the sole effect of the pretreatment process was not investigated. However, the majority of the NCP (Sri Lanka) field RO and NF plants have similar pretreatment units installed (Sand filter, activated carbon filter, microfilter and *anti*-scalant application). Hence, when considering the removal efficiencies, the variations were assumed to be due to the membrane effect.

**Table 1**
Membrane performance analysis parameters and equations.

| | Performance parameters | Equation | Definition of symbols |
|---|---|---|---|
| (1) | Transmembrane pressure (MPa) | $\frac{P_F + P_C}{2} - P_P$ | $P_F$ – Feed pressure (MPa)<br>$P_C$ – Concentrate pressure (MPa)<br>$P_p$ – Permeate pressure (MPa) |
| (2) | Water recovery (%) | $\frac{Q_P}{Q_F} \times 100$ | $Q_F$ – Feed flow rate (L/h)<br>$Q_P$ – Permeate flow rate (L/h) |
| (3) | Salt rejection (%) | $\frac{C_F - C_P}{C_F} \times 100$ | $C_F$ – Salt concentration in feed (mg/L)<br>$C_p$ – Salt concentration in permeate (mg/L) |
| (4) | Specific permeate flux ($Lh^{-1}m^{-2}kPa^{-1}$) | $\frac{Q_P}{A \times TMP}$ | $Q_P$ – Permeate flow rate (L/h)<br>$A$ – Membrane effective area ($m^2$)<br>$TMP$ – Transmembrane pressure (kPa) |

#### *3.1.1. RO, LPRO, or NF?*

As described by Bellona et al. (2008), specific permeate flux values can be used to categorize the membranes as RO (<0.03 $L/hm^2kPa$), LPRO (0.037–0.057 $L/hm^2kPa$), and NF systems (>11 $L/hm^2kPa$). The specific permeate flux values of the RO membranes used in NCP (Sri Lanka) were calculated using the manufacturers' technical specifications and the data collected from the field sites (Table 2). According to the Manufacturer's testing conditions, 7 out of 10 membranes have specific permeate flux values in the range of 0.037–0.057 $L/hm^2kPa$, and they can be classified as LPRO membranes. The other three have specific permeate flux values less than 0.03 $L/m^2hkPa$ and hence they are categorized as RO membranes. Under field conditions, eight membranes were performed as LPRO, while two membranes were performed as RO membranes. It is interesting to note that three membranes indexed as RO by the Manufacturer behave as LPRO membranes under field conditions in Rajanganaya, Medirigiriya, and Hingurakgoda. It may be due to defects created in the membranes from inappropriate pressures or initial flow applications that enhance higher specific permeate flux for low pressure (Indika et al., 2021). Here, it was noted that the LPRO membranes ESPA2-LD-4040 and ULP 11–4040 are contradictorily producing lower specific permeate flux similar to RO. The age of the membrane ESPA2-LD-4040 is about five years, and hence this can be attributed to membrane fouling (Belkacem et al., 2007). This also

**Table 2**
RO membranes specifications, corresponding specific permeate flux details, and LPRO/RO conditions assignments.

| Membrane model type | Membrane model type as per Manufacturer (RO/LPRO) | Purpose of application | Specific permeate flux during membrane testing[a] (L/hm$^2$kPa) NaCl$_{(aq)}$ | Nature of operation as manufacturer testing condition (RO/LPRO) | Specific permeate flux observed at field** (L/ hm$^2$kPa) | Nature of operation in NCP (RO/ LPRO) | Membrane age (years) |
|---|---|---|---|---|---|---|---|
| ESPA2-LD-4040 | Not Specified | Not specified | 0.040 | LPRO | 0.017 | RO | 5 |
| LC-LE-4040 | Low-Pressure RO | Harsh water conditions | 0.053 | LPRO | 0.089 | LPRO | 3 |
| CPA2-4040 | Not Specified | Not Specified | 0.029 | RO | 0.041 | LPRO | 1 |
| ULP 3012 | Low-Pressure RO | water dispenser and residential drinking fountain | 0.045 | LPRO | – | LPRO | 1 |
| ULP 11-4040 | Low-Pressure RO | Low salinity water | 0.051 | LPRO | 0.023 | RO | <1 |
| RE4040-BN | Not Specified | Brackish water treatment | 0.030 | RO | 0.042 | LPRO | 1 |
| RM-BW4040 | High-Pressure RO | Brackish water treatment | 0.035 | LPRO | 0.082 | LPRO | 2 |
| XLE-4040 | Low-Pressure RO | Commercial applications | 0.073 | LPRO | 0.038 | LPRO | 1.5 |
| BW30-4040 | Not Specified | Brackish water treatment | 0.034 | RO | 0.049 | LPRO | <1 |
| ULP21-4040 | Ultra-low-Pressure RO | Low salinity water | 0.044 | LPRO | 0.059 | LPRO | <1 |

[a] Calculated based on the information provided by the Manufacturer during membrane testing ** Calculated using the operational data acquired during the field visits.

supports the low TDS and calcium removal efficiency observed for this system (Outlier for EC in Fig. 3(a)). But the age of the membrane ULP 11–4040 is less than one year; hence the effect of membrane fouling might not be the probable cause in this case. The feedwater used for this plant has the highest EC and TDS compared to other feedwater sources, indicating that the quality of the feedwater source also has a significant impact on the system function. The low DOC removal efficiency (~70%) observed for ULP 11–4040 but not for the rest of the parameters might be due to the internal pore-blocking with the formation of compacted DOC-Na and DOC-Mg complexes, nearly the same or lesser size comparable to the membrane pore size (Xu et al., 2019).

Comparing the existing NF plants for tested and field-specific permeate flux indicated that the current field NF systems function efficiently similar to LPRO systems in terms of transmembrane pressure, ranging between 0.30 and 0.55 MPa, and produced a specific permeate flux of about 0.06 L/m$^2$hkPa (Table 3). The membrane NE 4040–90 is specified for high monovalent rejection, but the calculated monovalent ion rejection was less (25%) than the specified value (85–95%) even though the operating conditions are similar to Manufacturer tested conditions. A similar observation was noted in $Ca^{2+}$ (23%), but $Mg^{2+}$ (80%) and EC removal (97%) were observed as high. This plant (located in Netiyagama, Sri Lanka) was designed differently in which the NF concentrate water was treated with a RO membrane, and the resultant permeate was mixed with the NF treated water, and the observed deviations can be attributed to the differences in the configuration.

**Table 3**
NF membranes specifications and corresponding specific permeate flux details with membrane age.

| Membrane model type | Purpose of application | Specific permeate flux during membrane testing[a] (L/ hm$^2$kPa) | Specific permeate flux observed at field[b] (L/ hm$^2$kPa) | Membrane age (years) |
|---|---|---|---|---|
| NF90 - 4040 | Commercial purpose | 0.059 | 0.087 | 1.5 |
| DF90-4040 | Commercial purpose | 0.058 | 0.057 | 3.0 |
| NE 4040 -90 | High monovalent ion rejection | 0.060 | 0.062 | 0.5 |

[a] Calculated based on the information provided by the Manufacturer during membrane testing.

[b] Calculated using the operational data acquired during the field visits.

### 3.1.2. *Manufacturer specified vs field operational conditions*

The transmembrane pressure and the specific permeate flux values obtained for RO and NF membranes were compared against the manufacturers' recommended data, as shown in Fig. 2(a) and (b), respectively. Most RO membranes are operating at low pressures (<1 MPa) (Fig. 2(a)) even though the maximum allowable pressure is 4.14 MPa. Among the total 59 RO plants surveyed 61% operate at low TMP (0.5–1.1 MPa), and 12% operate at ultra-low TMP (<0.5 MPa) (Figure S1), and similar observations were also reported previously (Imbulana et al., 2020; Indika et al., 2021; Nanayakkara et al., 2020). Alternatively, the NF membranes operate at similar TMPs as specified by the manufacturers. In the specific permeate flux comparison (Fig. 2(b)), about half of the membranes show high deviations (50% deviation) from the Manufacturer specified values. However, membranes ULP-3012, DF90-4040, and NE 4040-90 deviate by about 20%, ensuing their proper functions. TMP and specific permeate flux deviation percentages (either negative or positive) were examined to determine the performance of the selected membranes (Figure S2). Most RO membranes showed nearly or above 50% deviation for both TMP and specific permeate flux except for a few membranes. Still, NF membranes are not observed for such variations indicating that they are functioning effectively. The system with ESPA2-LD-4040 and XLE-4040 membranes showed positive differences (TMP and specific permeate flux infield is less than the tested condition) with greater deviation indicating fouling (either organic or scaling or both) (Park et al., 2019), and it is evidenced by the lesser removal efficiency of both systems (EC- 80 and 92.5%, $Ca^{2+}$ - 88 and 89% accordingly, the data not shown) compared to the rest of the 8 RO systems. Such variations were not observed in any NF systems even though the systems' age varied between 1 and 3 years, indicating less fouling propensity of the NF membranes' than RO. However, a depth study needs to correlate fouling and membrane performances.

### 3.1.3. *Removal efficiencies of the membrane types*

The TDS, hardness, DOC, and monovalent ion rejection are also compared using various RO and NF membranes (Figure S3). All membranes showed removal efficiencies over 80% for the above parameters despite the membrane defects and fouling propensity, except for the RO membrane ULP 11–4040 (70% DOC removal efficiency) and the NF

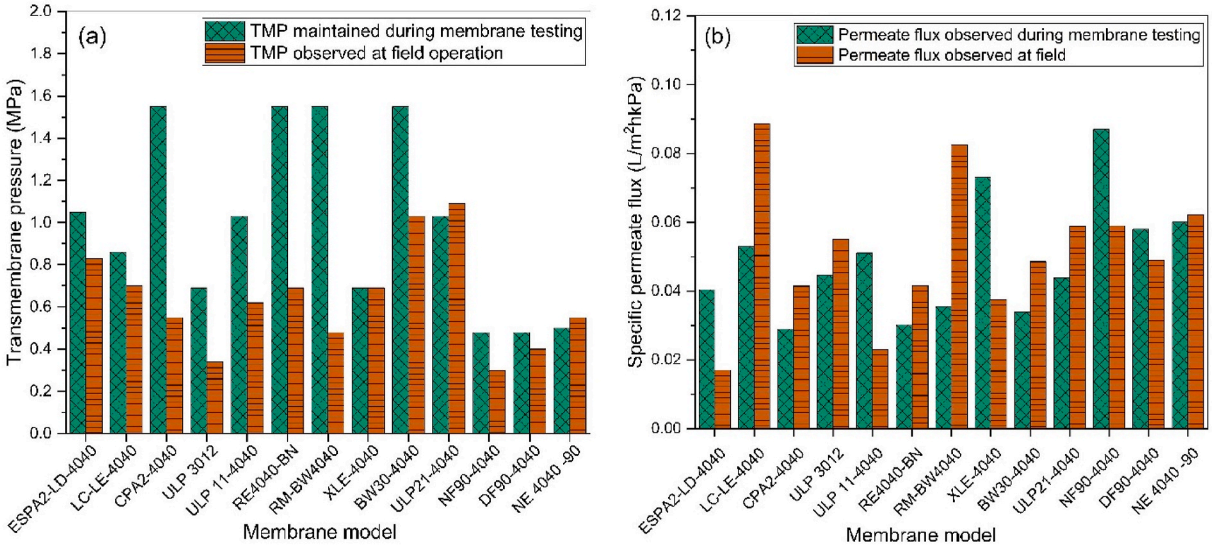

**Fig. 2.** (a) Transmembrane pressure maintained during testing and (b) Specific permeate flux obtained during field operation for different membrane model types of the selected plants.

membrane NE 4040–90 (61% hardness; 25% $Na^+$). The deviations observed with NE 4040–90 could have resulted from the plant configuration, notably the concentrate recycling with RO membrane application. ESPA2-LD-4040 showed the lowest removal efficiencies (80% TDS, 92% hardness, 87% DOC, and 79% $Na^+$), presumably due to membrane aging. UPL 3012 also showed slightly low removal efficiencies (92% TDS, 96% hardness, 91% DOC, and 86% $Na^+$); this could be due to its specific applicability as automatic water dispensers and residential drinking fountains. However, our data show that the salinity removal efficiency of RO and NF membranes are comparable when membrane configurations are the same. Even though the water hardness changed between 95 and 320 mg/L as $CaCO_3$ it produces water with hardness less than 15 mg/L as $CaCO_3$ except NE 4040–90, where the RO-treated concentrate is mixed with NF-treated water. The observed efficiency variations of NE 4040–90 and ESPA2-LD-4040 are ascribed to different configurations or extensive membrane aging. Further removal efficiency is discussed under section 3.2.2 in detail in terms of water quality and membrane types. However, this part indicates that the membrane age, initial flux, and specified applications are important factors when considering the membrane performance and its successful functionality.

## 3.2. *Comparison of field RO and NF treatment systems*

### 3.2.1. *Analysis of the feedwater quality of the RO and NF treatment systems*

NCP groundwater is hard with 19–750 mg/L as $CaCO_3$ and high DOC ranging between 0 and 11 mg/L (Cooray et al., 2019; Makehelwala et al., 2019). Nonetheless, it was not indicated as brackish water, but few studies reported higher $Na^+$ concentrations greater than 1000 mg/L (Paranagama et al., 2018; Perera et al., 2020).

The feedwater quality of the selected RO and NF treatment systems was examined for its potability according to the WHO guidelines (2011) and Sri Lanka's Drinking Water Quality Standards (SLS 614: 2013) (Table 4). In general, RO is mostly suitable for monovalent ion removal. However, the $Na^+$ concentration in the feedwater ranged from 15 to 138 mg/L and this is within the acceptable limits. As shown in Table 4, the pH, cations ($Na^+$, $Cr^{3+}$, $Mn^{2+}$, $Co^{2+}$, $Ni^{2+}$, $Cu^{2+}$, $Zn^{2+}$, $As^{3+}$, $Se^{2+}$, $Cd^{2+}$) and anions ($NO_3^-$, $SO_4^{2-}$) of the feedwater were within the acceptable limits, but EC, hardness, $Mg^{2+}$, $F^-$ and $Cl^-$ have exceeded the WHO guidelines and SLS standards. Only 38% of the groundwater samples have the cation concentrations varying in the order of $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ (Cooray et al., 2019), whereas the rest of the samples showed the variation as $Na^+ > Mg^{2+} > Ca^{2+} > K^+$. Further, 85% of the samples showed hardness dominated by $Mg^{2+}$ (Imbulana et al., 2020), while the other samples showed $Ca^{2+}$ dominated hardness. There were no limits defined for the DOC level either in the WHO or SLS standards, but it was

**Table 4**
Feedwater quality of the treatment systems, including major cations.

| Water quality parameter | Groundwater quality | WHO maximum permissible level | Samples exceeding the permissible level (%) | SLS maximum permissible level | Samples exceeding the permissible level (%) |
|---|---|---|---|---|---|
| pH | 7.13–8.21 | 6.5–8.5 | 0 | 6.5–8.5 | 0 |
| EC (μS/cm) | 264.9–1329 | 400 | 77 | 750 | 38 |
| DOC (mg/L) | 1.55–12.73 | N/A* | N/A** | N/A* | N/A** |
| Hardness (mg/L) | 96.56–317.47 | 100 | 93 | 100 | 93 |
| $Ca^{2+}$ (mg/L) | 9.70–53.10 | 100 | N/A** | N/A* | N/A** |
| $Mg^{2+}$ (mg/L) | 7.70–60.38 | 30 | 31 | 30 | 31 |
| $Na^+$ (mg/L) | 14.70–138.23 | 200 | 0 | 200 | 0 |
| $K^+$ (mg/L) | 0.57–4.66 | N/A* | N/A** | N/A* | N/A** |
| $F^-$ (mg/L) | 0.58–2.93 | 1.0 | 62 | 1.5 | 15 |
| $Cl^-$ (mg/L) | 12.78–392.26 | 250 | 15 | N/A* | N/A** |
| $NO_3^-$ (mg/L) | 0.28–17.10 | 50 | 0 | 50 | 0 |
| $SO_4^{2-}$ (mg/L) | 3.95–30.60 | 250 | 0 | N/A* | N/A** |

N/A* - Not assigned, N/A** - Not applicable.

reported that the consumption of natural water with DOC levels above 4 mg/L may cause adverse effects on human health (British Columbia, 1998; Regan et al., 2017). It was observed that 77% of the feedwater samples had DOC levels above 4 mg/L, indicating the necessity of membrane water treatment applications. Particularly RO and NF are suitable as they have a better potential for DOC removal in natural water (Teychene et al., 2020; Yoon and Lueptow, 2005). NF has been identified as a suitable technique to bring the hardness and DOC levels down to the permissible limits to provide safe water (Zhong et al., 2019). Since the study area, groundwater has exceeded levels of DOC, hardness, fluoride, and chloride, and it urges the need for membrane treatment, either RO or NF, to regulate especially the DOC and hardness. The

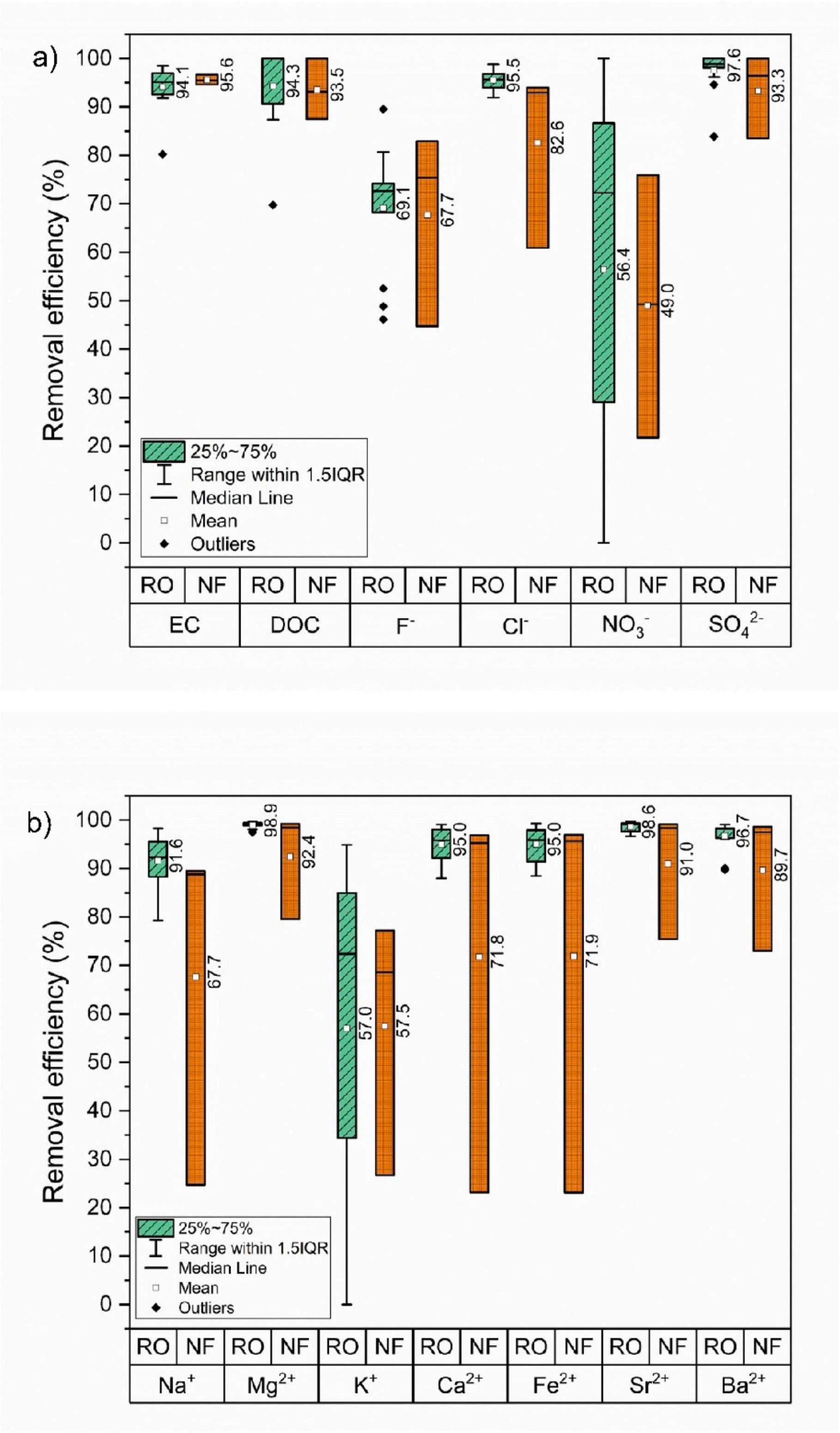

**Fig. 3.** Removal efficiency ranges of the RO and NF systems; (a) EC, DOC and dominant anions, (b) Dominant cations (>100 μg/L). The ranges and mean values are calculated from the data obtained for the 10 selected RO plants and 3 NF plants.

current study was conducted during the dry season. Since significant variations, especially the TDS, are reported in the groundwater quality during different seasons, a seasonal study on the RO/NF system performance is recommended (Imbulana et al., 2021).

### 3.2.2. *Groundwater treatment efficiency of RO and NF plants*

The groundwater treatment efficiencies of the selected field RO and NF plants were investigated by measuring the pH variation, the mean removal efficiencies for EC, DOC, and other primary cations ($Ca^{2+}$, $Mg^{2+}$, $Sr^{2+}$, $Ba^{2+}$, $Na^{+}$, $K^{+}$, $Al^{3+}$), heavy metals ($V^{3+}$, $Cr^{3+}$, $Mn^{2+}$, $Fe^{2+}$, $Co^{2+}$, $Ni^{2+}$, $Cu^{2+}$, $Zn^{2+}$, $As^{3+}$, $Se^{2+}$, $Cd^{2+}$) and anions ($F^{-}$, $Cl^{-}$, $NO_3^{-}$ and $SO_4^{2-}$) of the treated water was calculated using the equation (3) mentioned in Table 1. A comparison of the pH values of the treated water by the RO and NF systems indicated that the NF treated water has pH values ranging from 6.1 to 7.5, whereas for RO treated water, the pH was mostly less than 6.5 (5.7–7.5) with an average of 5.8. The SLS guidelines for drinking water specify a pH range of 6.5–8.5. Therefore, NF seems more favourable than RO in terms of the pH of the treated water. The lesser pH values in the RO treated water could be due to the enhanced removal of the electrolytes from the system. Figs. 3 and 4 presents the removal efficiencies of the principal water quality parameters. The average removal efficiencies were nearly or above 90% and higher in RO systems compared to the NF systems. Both methods' mean DOC removal efficiencies were comparable (RO - 94.3% and NF - 93.5%). As observed, both RO and NF systems effectively remove groundwater DOC below 2 mg/L levels (Permissible DOC level for treated water as per Ministry of Environment, 1998) that will not impair human health. However, one RO plant produced water with 2.8 mg/L DOC, which was higher than that observed for the other plants. The DOC removal efficiency of this particular RO plant was only 69.7%, which is significantly lower than the rest. Feedwater quality of the plant showed the highest EC (1329 μS/cm) dominated by $Na^{+}$ (138 mg/L) together with higher DOC (9.4 mg/L) and $UV_{254}$ (0.023 $cm^{-1}$). The highest $Na^{+}$ of the feedwater with low molecular weight DOC fraction could result from compaction of DOC via formation of neutral DOC-Na complexes less than membrane cut off level and it is following the experienced specific permeate flux deviation with the internal pore-blocking scenario (Adusei-Gyamfi et al., 2019; Makehelwala et al., 2019; Xu et al., 2019). Even though the DOC removal is influenced by the membrane pore size that governs the DOC removal mechanism by diffusion/convection (Yoon and Lueptow, 2005), comparatively similar removal efficiency ranges were observed in both NCP RO and NF systems. Both systems were incorporated with a granular activated carbon (GAC) filter in the pretreatment step. The GAC filters are known to remove feedwater DOC to a certain extent via adsorption (Monnot et al., 2016). Therefore, it can be assumed that the GAC filters adsorb most DOC present in the feedwater. However, complete removal of DOC is unlikely, and the remaining DOC can be a potent foulant for both RO and NF membranes. Since the studied treatment plants were not designed to bypass the pretreatment unit, the sole effect of the membrane in removing DOC could not be identified. Nonetheless, DOC removal efficiency requires a further in-depth study on the impact of DOC's molecular size distribution in groundwater and the influence of the pretreatment step.

The comparison of the removal efficiencies of the constituents in the feedwater revealed that the RO systems have higher removal efficiencies as compared to that of NF for most of the elements, but the major cations and anions' mean removal efficiencies of RO were comparable with NF as observed by Wafi et al. (2019) since these RO systems are LPROs. The mean EC removal efficiency of RO was acceptable even though the higher EC removal observed for RO compared to the NF due to the insignificant deviation (1.5%) and the range of removal efficiencies observed. The EC removal efficiencies of the studied 10 RO plants ranged from 91.8 to 98.5 with an outlier of 80.0 (membrane age 5 years), while that of the 3 NF plants varied from 94.6 to 96.7 and it was similar in the range observed for RO/LPRO and NF/LPRO membranes in Bellona et al. (2011) study. Since this comparison was based on the existing systems, the EC removal efficiency deviation could be due to the variation in feedwater quality and operational and maintenance conditions (Indika et al., 2021; Shen and Schäfer, 2014). Here the 3 NF feedwater sources had EC values above 800 μS/cm, but the RO feedwater EC values mostly ranged from 250 to 500 μS/cm, and only 3 were above 800 μS/cm. The operating pressures also varied significantly from the manufacture specified values as shown in Fig. 2 (a). In addition to this the ions in the water together with DOC can affect the removal efficiency via DOC-ion complexation and its fouling on membrane surfaces (Adusei-Gyamfi et al., 2019; Xu et al., 2019). The removal efficiency of $Cu^{2+}$ was low in both systems, 11.4% and 35.6% in RO and NF, respectively, and this could be due to low concentrations of $Cu^{2+}$ present in the groundwater (about 3 μg/L on average). Additionally, a wider range of removal efficiencies was observed for $Al^{3+}$ (7–80%), $Mn^{2+}$ (30–85%), $F^{-}$ (45–83%), and $NO_3^{-}$ (22–76%) and among them, $Mn^{2+}$, $F^{-}$ and $NO_3^{-}$ significantly varied to the concentrations of the respective element in the feedwater with the Pearson correlation

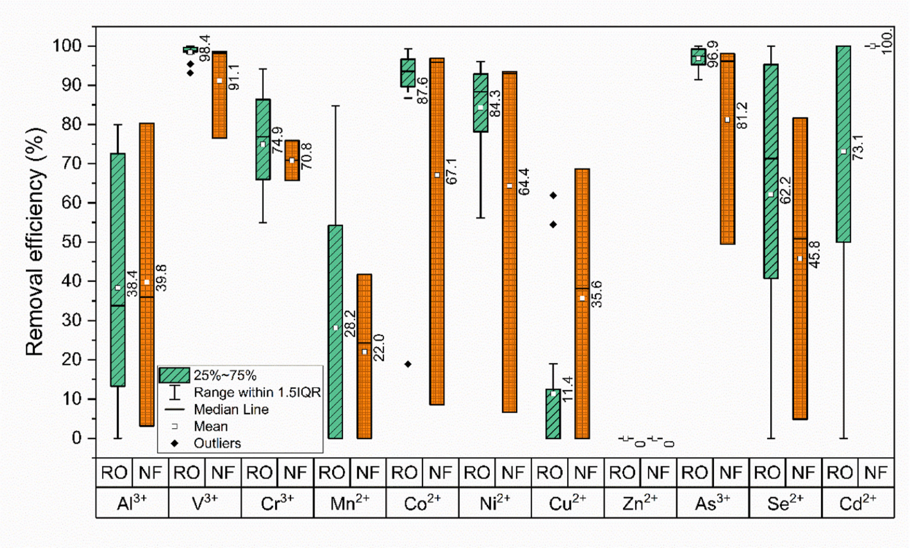

**Fig. 4.** Removal efficiency ranges of submissive cations in the RO and NF systems. The ranges and the mean values are calculated from the data obtained for the 10 selected RO plants and 3 NF plants.

coefficients of 0.545, 0.850 and 0.503, indicating that $Mn^{2+}$and $NO_3^-$ rejection moderately (>0.5) influenced by its concentration and $F^-$ rejection highly (>0.8) influenced by the concentration. Also, the removal efficiencies of $Zn^{2+}$ were nearly zero for both RO and NF systems, presumably due to their lower concentration and low-pressure application. However, despite the presence of lower concentrations of certain other heavy metals, such as $Cr^{3+}$, $Mn^{2+}$, $Co^{2+}$, and $As^{3+}$, these showed higher removal efficiencies with both techniques. Moreover, the comparison between two mean removal efficiency values based on P-values obtained from two samples' t-tests also indicated that there was a considerable deviation between these RO and NF removal efficiencies. Statistically significant difference observed only for $Cd^{2+}$ and $Pb^{2+}$ (P-value $< 0.05$), and considerable difference observed for EC, $Na^+$, $Cr^{3+}$, $Cu^{2+}$, $Cl^-$ (P-value 0.15–0.20) and DOC, $Ca^{2+}$, $Mg^{2+}$, $Sr^{2+}$, $Ba^{2+}$, $V^{3+}$, $Fe^{2+}$, $As^{3+}$ (P-value 0.20–0.25). Others were not shown any considerable statistical differences while considering the obtained P-values (>0.30). Hence, both statistically and experimentally proved that removing the essential elements' efficiencies was considerably affected by these two different membrane applications and RO exhibits exaggerated removal. In contrast, NF exhibits an acceptable removal level. Therefore, increasing the TMP of the RO systems could be a suitable solution for the existing RO systems. The efficiency will be increased in terms of higher productivity with lower removal efficiencies leading to more mineral's retention in produced water (without exceeding the WHO and SLS standard values) since the existing values were too low compared to standard values.

### 3.2.3. *Health consequences of produced water quality*

The chemical composition of drinking water plays a significant role in human health, not only in terms of not exceeding the levels of potentially toxic elements but also in containing sufficient levels of essential elements such as $Ca^{2+}$, $Mg^{2+}$, and $F^-$. Treatment methods such as RO and NF both remove essential and non-essential minerals from water during the treatment process. Particularly in RO, mineral removal is comparably high due to the superior rejection ability of the membrane. However, prolonged consumption of RO-treated water may cause adverse health effects (Janna et al., 2016; Rosborg et al., 2015). Although $Ca^{2+}$ can be taken via dairy products at the same level as drinking water, greater attention must be given to $Mg^{2+}$ due to its lower bioavailability (Barbagallo and Dominguez, 2018; Greupner et al., 2017).

The $Ca^{2+}$, $Mg^{2+}$, and $F^-$ levels were measured in RO and NF treated water of the selected plants (Table 5). The values were compared with the minimum required levels in demineralized/softened water established by the European Union member states due to the absence of specific minimum levels of these minerals in SLS standards and WHO guidelines (Kozisek, 2020). As can be observed, even the maximum concentration values of $Ca^{2+}$ and $Mg^{2+}$ in both RO and NF produced water were below the minimum level. Still, NF showed relatively higher values as compared to RO. The $F^-$ concentration should be maintained above 0.5 mg/L as per WHO guidelines. But in both permeate water, the $F^-$ concentration was below that recommended level. When considering the essential mineral content in the treated water, NF can provide a better solution for the mineral deficiency issue. If RO water is rich in crucial minerals post-mineralization step would be essential; however, in none of the plants examined, this step was not performed, presumably due to the lack of proper knowledge and lack of resources. Since both RO and NF produced water mean mineral content is less, the higher productivity NF membrane could be applicable to enhance the mineral concentration with proper periodical monitoring. Also, recycling treated concentrate water seems a better solution.

## 3.3. *Operation and maintenance practices*

During continuous operation of the membrane-based treatment systems, the corresponding specific permeate flux of the system can vary due to membrane fouling, as experienced in systems with ESPA2-LD-4040 and XLE 4040. Membrane fouling is inevitable but can be removed to a certain extent by following proper chemical cleaning practices and reduced with appropriate inexpensive pretreatments. However, most RO/LPRO membranes used in the NCP treatment systems are not subjected to periodic chemical cleaning and end up with fouling and lower removal efficiencies. Instead of cleaning the membranes, those are replaced by the plant's installation company upon their performance reduction (permeate flow/treated water quality variation). But chemical cleaning is performed with NaOH and HCl in the NF treatment plants on a semi-annual basis to recover the specific permeate flux loss and improve the membrane performance. However, the pretreatment processes were well maintained in both NCP RO and NF systems to reduce the fouling formation. Pretreatment modules are occasionally replaced with new modules by maintaining routine EC/TDS monitoring. Pretreatment methods such as sand filtration, microfiltration, and GAC filtration are generally used in every RO/LPRO plant and NF plants with antiscalant dosing to reduce the scaling effect that severely hinders the membrane performance can be removed only through extensive cleaning with strong cleaning solutions. Most plants perform daily backwashing of the pretreatment filters, if not once per 2–3 days. The replacement of microfiltration is performed occasionally within three months to maintain the produced water quality and fouling reduction.

The RO/LPRO membrane replacement frequency is around 2–3 years. However, membrane cleaning and factors such as operating pressure increase or specific permeate flux reduction and change of treated water quality are rarely considered when replacing the membranes. It is more or less of a routine maintenance step. This indicates the requirement of proper guidelines for the operation and maintenance of the treatment units to increase the effective usage of membranes. Adopting automatic control and maintenance facilities with smartphones developed by Wu et al. (2022) can be adopted for better operation of these plants. The NF membranes last for three years without any unusual changes in their performances since they are subjected to proper cleaning periodically every six months or at least once a year. The expected lifetime of the NF membranes is five years regarding the system design and configurations. The backwashing in NF is carried out with treated water, whereas in RO/LPRO systems, raw water is used for backwashing. Therefore, proper cleaning practices and establishing guidelines for membranes systems monitoring will help to increase membrane durability. Frequent replacement of RO membranes (once every 2–3 years) in combination with the need for post-mineralization of RO treated water (though it is not currently practiced in the studied

**Table 5**
$Ca^{2+}$, $Mg^{2+,}$ and F- levels in RO and NF treated water.

| Minerals | RO system treated water | | | NF system treated water | | | Required minimum concentration in softened water |
|---|---|---|---|---|---|---|---|
| | Min | Average | Max | Min | Average | Max | |
| $Ca^{2+}$ (mg/L) | 0.14 | 0.88 | 2.44 | 0.61 | 6.89 | 18.83 | 30[a] |
| $Mg^{2+}$ (mg/L) | 0.07 | 0.30 | 1.44 | 0.25 | 2.27 | 6.21 | 10[a] |
| $F^-$ (mg/L) | 0.30 | 0.32 | 0.37 | 0.35 | 0.38 | 0.40 | 0.5[b] |

[a] European Union member states.
[b] WHO and SLS standards.

plants) makes RO a costly process as compared to NF. The lack of technical knowledge of the plant operators was also a major disadvantage for the improper functioning of the treatment units. Hence, educating and training the plant operator is vital for an effectively functioning treatment unit.

### 3.4. *Reject water management*

Among the 59 NCP RO plants considered for the study, nearly 90% have maintained water recovery percentages below 50%, as shown in Fig. 5, but the NF plants have maintained the recovery percentages nearly at 60%. Even though the wastewater generation in NF plants is less than that of the RO plants, it can be further reduced by increasing the water recovery percentage up to 80% as in practice globally (Jones et al., 2019) by limiting the feed and reject water flow rate while maintaining the optimum TMP. Increasing the recovery percentage will ultimately decrease the negative environmental impacts caused by the brine discharge to the ecosystem.

During the field survey, NF plants followed better reject water management techniques by associating the reuse of reject water for cement block manufacturing and toilet flushing. In one plant, the concentrate water was recycled through a RO membrane, and the treated water was then allowed to be mixed with NF treated water and the concentrate water obtained from RO was used for toilet flushing purposes. However, almost all the rejected water was discharged into the ground in RO plants without any safety measures. In some cases, the rejected water was used for crops, and no differences were observed between other crops grown with normal groundwater. Discharge of the concentrate to the land area adjacent to the treatment plant may affect the indigenous plant species due to the resulting changes in soil properties (Nanayakkara et al., 2020) in the short-term and the long term. This will cause groundwater quality degradation since reject water is concentrated 1.2–7.6 times more than the feedwater. Also, the application of reject water for different reuse purposes was verified by referring to the ambient water quality standards of Sri Lanka (2019) and pH range, $F^-$, $Cl^-$, $NO_3^-$-N and $SO_4^{2-}$ were accepted for irrigation and aquatic life purposes (Table S 2). However, half of the RO plant concentrates exceeded the limit of EC values of 700 μS/cm. The derived SAR values of the reject water ranged from 0.9 to 4.3 for RO and 1.0–2.5 for NF, also indicating the potential of utilizing reject water for irrigation. Still, the use of rejected water for agriculture purposes is not recommended due to the possibility of contamination of food products and health-related problems added due to the soil salinity increment. As per Sri-Lankan ambient water quality standards, water is suitable for aquatic life, bathing, and recreation. In addition, the available economic and sustainable brine management opportunities such as salt and metal recovery by converting the waste into a resource through combining different processes, use for fish and halophyte production systems, incorporating reject water treatment or resource recovery technologies with renewable energy from solar, wind or thermal power sources could be considerable (Giwa et al., 2017; Jones et al., 2019).

### 3.5. *Future projections*

According to the past RO and NF comparison studies (Elazhar et al., 2015; Pontié et al., 2013), moving towards NF technology will be advantageous for a tropical country like Sri Lanka with low TDS water (<3000 mg/L) with high yield and solutes along with the optimized brine production and energy requirement (Wu et al., 2022). The present RO/LPRO systems result in excess removal of solutes with voluminous waste production under ineffective operation at low pressures (~1 MPa). As discussed earlier (Tian et al., 2021), specific characteristics of feedwater and the purpose of the treatment require consideration to increase the sustainability of the RO/NF plants. Practicing combined applications such as NF for hardness reduction and RO for treating concentrate and recycling with feedwater with near-zero waste (5%) is a suggestible option as at the Netiyagama plant (Wu et al., 2022). Increasing the TMP of RO systems is a temporary solution to increase productivity to make these RO plants more efficient than existing RO systems. However, a cost comparison must be done by considering operational, maintenance, and cleaning costs to evaluate the economic efficiency of these systems. Fabrication of new membranes to target contaminants will direct towards cost-effective technology that will enhance membranes' permeability, selectivity, and stability (Tian et al., 2021). Practicing chemical and physical cleaning methods could be more effective for better removing membrane fouling layers (Jiang et al., 2017). Also, as with the NF plants, chemical cleaning will be beneficial for NCP RO plants to increase the membrane lifetime with acceptable performance and cost conservation by extending membrane replacing intervals (Jafari et al., 2021). Also, seasonal variation in feedwater quality and their fouling mechanisms have to be considered when selecting a suitable membrane to ensure the sustainability of the plant (Othman et al., 2022). Even though the CKDu reduction was observed with these membrane treatment methods, there will be a possibility of other illnesses anticipated with prolonged low mineral intakes. Therefore, a comparative study is recommended to evaluate the health consequences of treated water consumption.

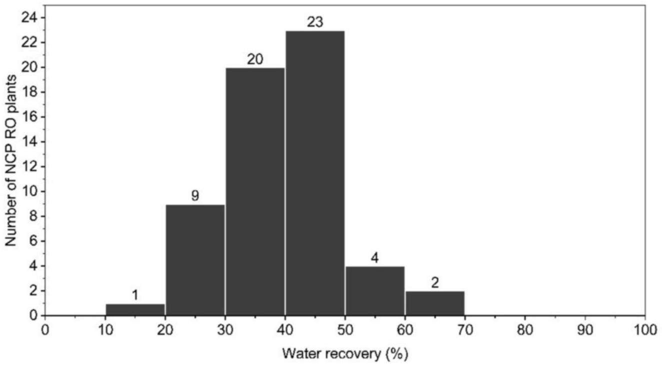

**Fig. 5.** Number of NCP RO plants and their maintained water recovery percentage ranges.

## 4. Conclusions

The efficiency of the field RO and NF treatment plants established in the NCP, Sri Lanka, was compared with plant operating conditions and feedwater quality. The pH of the NF-produced water was within the acceptable limits, whereas RO systems produce water with relatively low pH. The mineral content in NF-treated water was higher (19 mg/L $Ca^{2+}$) than in the RO treated water (2.5 mg/L $Ca^{2+}$); hence, it offers an alternative solution to the mineral deficiency issues. However, the increased productivity of NF membranes recycling the concentrate with RO membrane application could be a better solution for NCP groundwater treatment. Future studies are required to elucidate seasonal variation of feedwater and the dissolved organic and complexes effect on membranes and systems.

Nonetheless, proper guidelines and awareness must be introduced to reject water disposal. In addition, low-cost reject water reuse methods must be adopted for the NF and RO systems. In conclusion, the NF system can be considered as a more suitable option for the treatment of NCP groundwater compared to the existing RO systems when considering productivity, waste generation, and energy efficiency.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.gsd.2022.100800.

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