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Spatiotemporal Variation of Groundwater Quality in North Central Province, Sri Lanka

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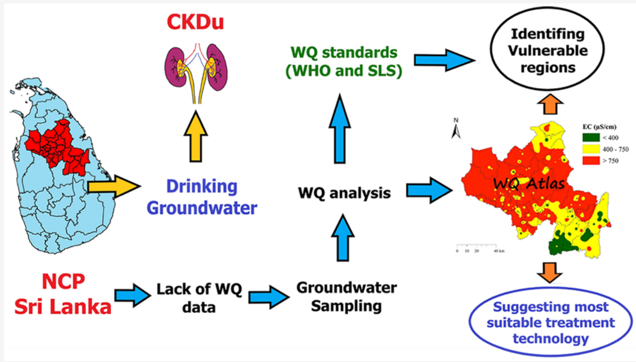
ABSTRACT: This study focused on fulfilling the demand for a comprehensive investigation of groundwater quality in the North Central Province (NCP) of Sri Lanka to achieve United Nations Sustainable Development Goal 6 (Ensure availability and sustainable management of water and sanitation for all). The NCP was selected as the study area due to its highest prevalence of chronic kidney disease with unknown etiology within Sri Lanka. Here, 334 shallow and deep groundwater samples were collected at the end of wet and dry seasons, respectively. Results revealed that elevated and unexpected levels of salinity, hardness, fluoride, nitrate, sulfate, and dissolved organic carbon and the alkaline nature of groundwater were the main issues, and the Ca-HCO_3 type was dominant in the groundwater. Water quality index analyses showed that 8.9% and 13.8% of wells had poor quality undrinkable groundwater in wet and dry seasons, respectively. Only 7% of samples were susceptible to sodium and salinity hazards for irrigational use. Reverse osmosis technology with a softening and activated carbon pretreatment process was identified as the most suitable way to treat groundwater with high salinity for many regions of the NCP. The groundwater quality atlas for the NCP created by this study was very useful for making a master plan of safe drinking water supplies and developing and implementing cost-effective water purification technologies in the NCP.



KEYWORDS: *Shallow groundwater, Water quality atlas, Geogenic contaminants, Reverse osmosis, Nanofiltration*

1. INTRODUCTION

Groundwater has been identified as a vital source of drinking water with the depletion of surface water bodies due to climate change. In Sri Lanka, especially in the dry zone, people used to consume raw groundwater for a few centuries.[1] The extraction of groundwater in this dry zone drastically increased with the increase in population demand for drinking water, irrigation water, and other domestic water needs. With the discovery of different groundwater quality issues in rural Sri Lanka, the importance of regular monitoring of drinking water (groundwater) sources became essential.[2−6] Thus, plenty of studies on groundwater quality investigations have been conducted in the past two decades to identify health issues such as chronic kidney disease with unknown etiology (CKDu) in rural areas including the NCP.[7−10] To achieve the sixth goal (clean water and sanitization) of the United Nations (UN) Sustainable Development Goals (SDG), Sri Lanka made considerable efforts and progress by giving 93.2% of the population (Target 6.1) access to safe drinking water.[1,11] Even though the groundwater supply for human consumption has been reached for 39.6% of the population, a proper water quality monitoring strategy has not been established yet for these rural regions of the dry zone.[1] See Text S1 for more information regarding groundwater quality highlights in the NCP based on previous studies.

The shallow and deep groundwater in the dry zone of Sri Lanka has been used as the main drinking water source for centuries without proper treatment.[12,13] Different communities in CKDu regions tend to try different technologies (conventional, not well established, and locally produced) on the domestic scale because of lack of knowledge and experience. However, with the improvement of research and development in the water sector in Sri Lanka, advanced treatment technologies such as ions exchange (softening), activated carbon filtration, and reverse osmosis (RO) membrane

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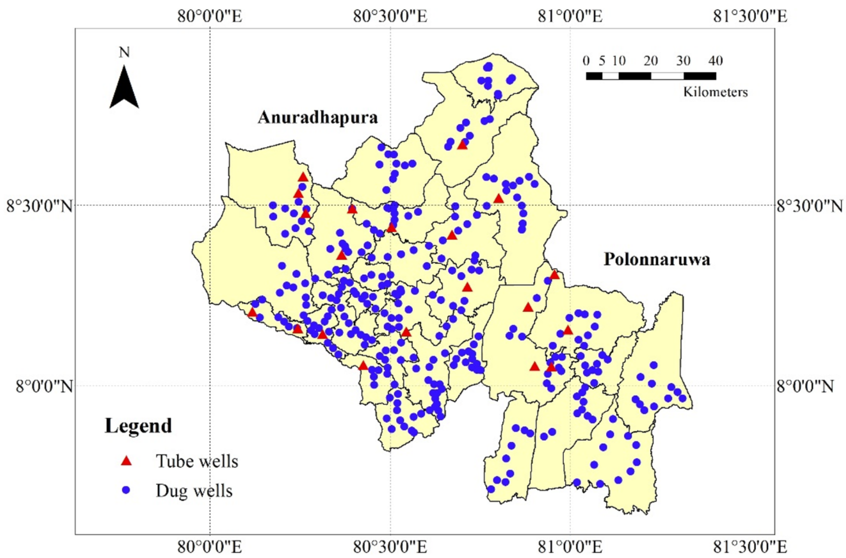


Figure 1. Distribution of sampling locations of dug wells and tube wells in this study.

filtration were introduced to these rural areas with groundwater quality issues.[14,15] This huge step in the development of the Sri Lankan drinking water production sector could improve the drinking water in rural regions at a significant level. Now, this RO drinking water has been widely spread throughout the dry zone of Sri Lanka as the major groundwater purification technology. It has been quite popular among rural communities because of the drastic reduction in the annual and newly reported CKD/CKDu incidence after introducing these RO stations which produce safe drinking water to their villages.[16]

Hence, a proper statistical geochemical evaluation of the groundwater qualities in the NCP should be carried out to further understand the natural and anthropogenic contamination sources as well as the contaminant transport mechanism. It will be beneficial for making important decisions in the drinking water sector and health sector as well as for agricultural practices in the country.

As the major objective of this study, for the first time, creating a detailed groundwater quality atlas illustrating different critical water quality parameters was carried out for the whole NCP in Sri Lanka with ArcGIS software. Meanwhile, identification of critical and vulnerable regions for groundwater contaminations were carried out as another objective. These objectives will be beneficial for making drinking water supply schemes in the future.

2. MATERIALS AND METHODS

2.1. Study Area. The major focus of this study was to investigate the groundwater quality in the CKDu prevailing regions in the dry zone of Sri Lanka because the majority of the CKDu cases were reported from the NCP. Thus, in this study, groundwater sampling sites were selected from each divisional secretariat division (DSD) of the two districts (Anuradhapura and Polonnaruwa) that belong to the NCP. Twenty-two DSDs from Anuradhapura and seven from Polonnaruwa were covered to collect groundwater samples from shallow and deep wells in both wet and dry seasons. Usually, this dry season lasts from April to September, and the wet season lasts from October to March in the NCP, in Sri Lanka.[17]

2.2. Sample Collection. A total of 334 wells were selected including 254 wells from the Anuradhapura district and 80 from the Polonnaruwa district to collect groundwater samples from different DSDs with the highest prevalence of CKDu based on the CKDu data of the Ministry of Health, Sri Lanka (Table S1).

Considering the uniform distribution of sampling locations and population distribution of the DSDs through the study area, groundwater wells were selected as much as possible (Figure 1). Two sets of samples were collected from each location in 2019 December (end of the wet season in NCP) and 2020 August–September (end of the dry season in NCP), respectively. These selected sample points were shallow (5–10 m shallow/dug wells) or deep (20–30 m deep hand pump and electrical pumps tube wells) wells used for groundwater extractions, and two samples were collected from natural springs. However, in the dry season, 36 wells have been dried out due to the very dry climate; thus, samples only from 298 wells from the total of 334 wells were extracted. All of the samples were collected in prewashed Teflon bottles filtered through 0.45 μm disposable polyether-sulfone membrane syringe filters (Jinteng, Tianjin, China). The samples collected for cation (such as Na, K, Ca, Mg, Al, Fe, and Si and other trace metals) analysis were acidified with ultrapure (GR) nitric acid (Sinopharm, Shanghai, China). All of the samples were stored at 4 °C for chemical analysis.

2.3. Analytical Methods. Stored samples at 4 °C were analyzed for physical–chemical water quality parameters. As onsite measurements, pH and EC of the water samples were analyzed by a Thermo Scientific Orion Star A325 multiparameter.

All the groundwater–water samples were analyzed for physical parameters (pH, EC, TDS) and chemical parameters (hardness, alkalinity, metal cations, and anions) in the Environmental Engineering Laboratory, Faculty of Engineering, University of Peradeniya; China-Sri Lanka Joint Research Demonstration Center (JRDC) for Water Technology, Peradeniya; and a laboratory in the National Institute of Fundamental Studies (NIFS), Kandy, respectively. All the dominant and trace metals (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, Li^{+}, Sr^{2+}, Ba^{2+}, Mn^{2+}, Fe^{2+}, Cd^{2+}, As^{3+}, Cu^{2+}, Zn^{2+}, Cr^{3+}, Hg^{2+}, and Si)

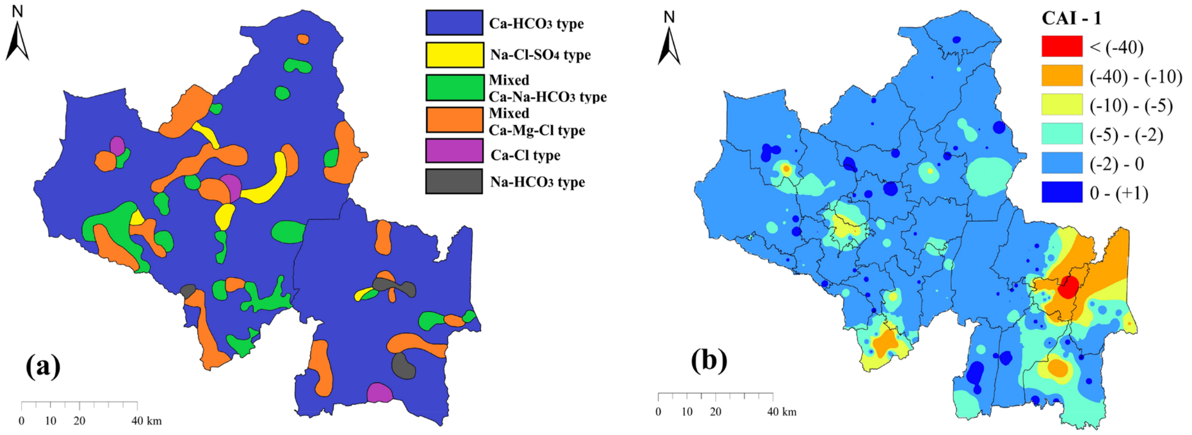


Figure 2. Spatial distribution of (a) groundwater types and (b) chloro-alkaline index (CAI 1) in the NCP.

concentrations were measured by an inductively coupled plasma optical emission spectrophotometer (iCAP 7000 Series ICP-OES, Thermo Fisher Scientific, Waltham, MA, USA) in the NIFS. The common anions (Cl^-, F^-, Br^-, SO_4^{2-}, NO_3^-, and PO_4^{3-}) were analyzed by an ion-chromatography instrument (Shine CIC D-100, China) in the JRDC. Dissolved organic carbon (DOC) concentrations of the water samples were analyzed by a TOC analyzer (Elementra, Langenselbold, Germany). The total alkalinity was tested by the titrimetric method, and the total hardness was determined by the Ca^{2+} and Mg^{2+} concentrations in the water samples using eq 1 in the Supporting Information.

HCO_3^- concentrations of the collected water samples were calculated based on the determined total alkalinity, using eq 2 in the Supporting Information. Seasonal variations of each water quality parameter were calculated using eq 3 in the Supporting Information.

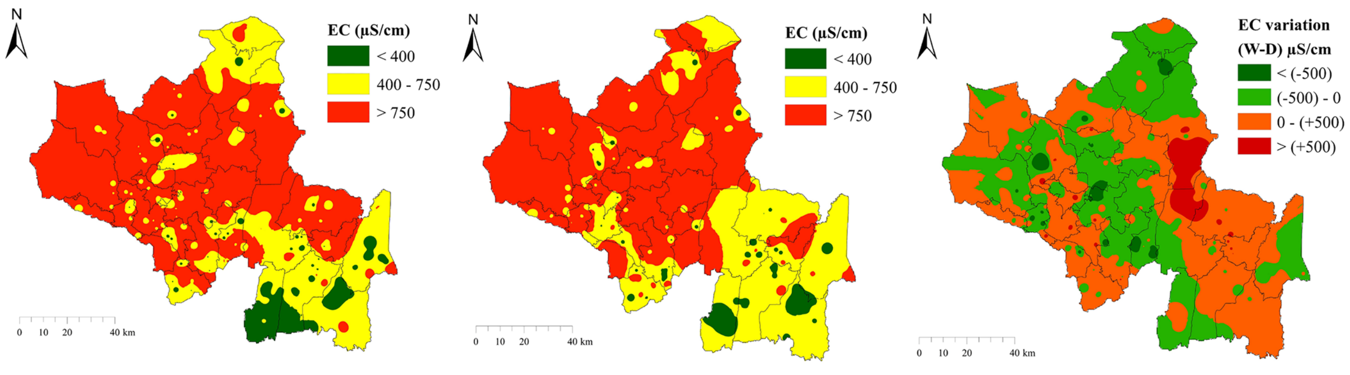
3. RESULTS AND DISCUSSIONS

3.1. Chemical Characterization of Groundwater.

3.1.1. Groundwater Classification. To geochemically classify the groundwater, piper trilinear diagrams are widely used. Six types of groundwater could be observed in the NCP, including Ca-HCO_3, Na-Cl, Ca-Na-HCO_3, Ca-Mg-HCO_3, Ca-Cl, and Na-HCO_3 types. Diagrams in Figures S1 and S2 show the dominance of the Ca-HCO_3 type groundwater throughout the NCP. However, in the wet season, the percentage of Ca-HCO_3 type groundwater (82%) showed a higher value than that in the dry season (59.4%), especially in shallow groundwater. That may be due to the dissolution of carbonate and silicate minerals in the wet season and the reduction of Ca ions by the ion exchange processes during the dry season (Text S6). Atlas of CAI 1 (chloro-alkaline index: Figure 2b) values show most of the regions in the NCP having values close to zero, except in upper region of the Polonnaruwa district (Lankapura, Welikanda, and Medirigiriya) with highly negative CAI values. This indicates the ion exchange between Na and K in the aquifer materials replaced by Ca and Mg from the groundwater in this area. This change is due to the alluvial aquifer material around the Mahaweli River basin in the Polonnaruwa district which is completely different from regolith aquifer materials in other regions of the NCP. It can be suggested that mixing with surface runoff or seepage from surface water tank systems had a possibility to change the shallow groundwater type into mixed types such as Ca-Na-HCO_3 and Ca-Mg-HCO_3, especially in wells located adjacent to the tank bunds. To illustrate these changes, for the first time, spatial distribution of groundwater types in the NCP was mapped using the ArcGIS tool. This map (Figure 2a) reveals the dominance of Ca-HCO_3 type groundwater in the majority of the areas in the NCP.

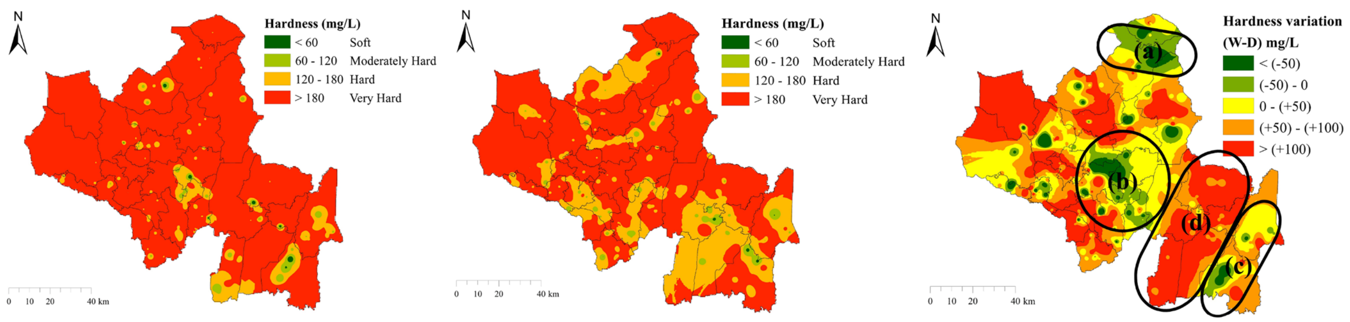
3.1.2. Geochemical Analysis of Groundwater. Figure S3 reveals that the evaporation effect was dominant on the shallow groundwater in the Anuradhapura district especially in the dry season. To identify the evolution of the groundwater, its mixing mechanisms, and the origin of dissolved minerals in the groundwater, the relationships between major ions are widely used.[18,19] Analysis shows that in the wet season, variation of the concentration of ions in the groundwater occupied significantly higher linear relationships among each other because of higher rock—water interactions in the wet season (Figure S4). On the other hand, in the dry season, the linearity of the relationships was scattered which is an indication of lower rates of dissolution of minerals in the aquifer base materials in the dry season due to the lower inflow rates. The molar ratio of Na^+/Cl^- higher than 1 indicates that Na^+ is released into the groundwater by silicate weathering which is common in the aquifer base material in the region, especially in the Polonnaruwa district (Figure S4a). It can also be confirmed by the ratio of Ca^{2+}/Mg^{2+} closely being similar to the value of 2 (Figure S4b). Similarly, the plot of $Ca^{2+} + Mg^{2+}$ vs $HCO_3^- + Cl^- + SO_4^{2-}$ had a considerably linear relationship while indicating silicate mineral weathering (Figure S4d), especially groundwater in the Polonnaruwa region which contains much higher silicates in the bedrock. To confirm Polonnaruwa silicate weathering in the aquifers, the $HCO_3^-/(Ca^{2+} + Mg^{2+})$ ratio showed a lower value than 1 (Figure S4c). Based on these results, it can be suggested that weathering of silicate minerals in the shallow regolith aquifers in the NCP was the major contributor of Ca^{2+}, Mg^{2+},and HCO_3^- of groundwater in the basin, especially in the Polonnaruwa district.

3.1.3. Correlations of Different Inorganic Ions in Groundwater. As described in the previous section, Ca-HCO_3 (type 1), Na-Cl-SO_4 (type 2), Ca-Na-HCO_3 (type 3), and Ca-Mg-HCO_3 (type 4) were the most common types of groundwater in the NCP and showed different characteristics and correlations in water quality parameters. Tables S2−S5 show the Pearson correlations of groundwater quality parameters within these types separately.



(a) Wet season (b) Dry season (c) Seasonal change

Figure 3. Spatial variation of electrical conductivity (EC) of groundwater in the NCP.



(a) Wet season (b) Dry season (c) Seasonal change

Figure 4. Spatial and seasonal variations (W-D) of total hardness in groundwater in the NCP.

The most dominant groundwater types in the NCP, type 1 ($Ca\text{-}HCO_3$) and type 4 (Ca-Mg-Cl) showed a lower correlation ($r > +0.3$; $p < 0.01$) between F^- and Br^- ions, but other types did not show any correlation between them. Similarly, Br^- showed moderate to very strong correlation ($+0.5 < r < +1.0$; $p < 0.01$) with Cl^- ions in types 2, 3, and 4 groundwater. This may be an indication of a common geogenic origin of these halides. NO_3^- can be used to predict some of the anthropogenic influences on the shallow groundwater, showing a moderate positive correlation ($r = +0.61$; $p < 0.01$) with Br^- in type 4 groundwater. However, NO_3^- did not correlate with other geogenic constituents in this type of groundwater. Hence, bromide can be suggested from either an anthropogenic origin due to the moderate correlation with NO_3^- or a natural seawater origin considering the strong correlation with Cl^-. However, the main regions having type 4 groundwater (Rajanganaya, Medawachchiya, and Horopothana) can be identified as highly cultivated areas with ongoing extensive agricultural practices. Thus, in type 4 groundwater, agrochemical-originated Br^- could be possible. Similarly, as a most controversial element in CKDu investigations, arsenic (As) is widely explored in many studies, but clear conclusions have not been made regarding CKDu or agricultural contamination of groundwater. In this study, it was observed that arsenic has a moderate positive correlation ($r = +0.613$, $p < 0.01$) with Mg, Na, and hardness levels of the groundwater, while indicating its geogenic origin, especially in the type 4 groundwater collected from heavy agricultural regions in the NCP.

3.2. Groundwater Quality Atlas of NCP, Sri Lanka. *3.2.1. Summary of Water Quality.* The compositions of groundwater samples collected in both dry and wet seasons are summarized, and statistical analysis results are given in Tables S6 and S7. Without regarding the seasonal changes, the majority of samples were reported as slightly alkaline. Some were slightly acidic (pH < 6.0), and a few were more basic (pH > 9.0) in nature. Having significantly higher dissolved mineral content or electrical conductivity (EC) values and higher hardness were the major characteristics of the groundwater in this region (NCP). More information is given in Text S2 in the Supporting Information.

3.2.2. Spatial Distributions of General Water Quality Parameters. 3.2.2.1. Electrical Conductivity (EC). EC of the groundwater is a direct indication of the level of dissolved inorganic ions, which are derived from the aquifer base materials and infiltrated anthropogenic constituents like nitrates and phosphates.

Comparatively, a higher level of EC was observed in the groundwater from Anuradhapura than Polonnaruwa (Figure 3). This is a clear indication of the spatial deviation and the stability of geogenic minerals in metamorphic terrain. The majority of the deep fractured aquifers in the Polonnaruwa region are recharged by the rainfall inflow rates in the northeastern slope of the central highland region.[20] More than a half of the lithosphere of Polonnaruwa consists of stable metamorphic minerals like quartzite and marble, which are weathering resistant and have lower solubilities in groundwater. Belonging to the highland complex[21−23] and having a recharge area in the mountain area of Sri Lanka[20] have led to lowering the dissolved mineral content in the groundwater in Polonnaruwa compared to the Anuradhapura region. Aquifers in the Anuradhapura region usually depend on the water inflow

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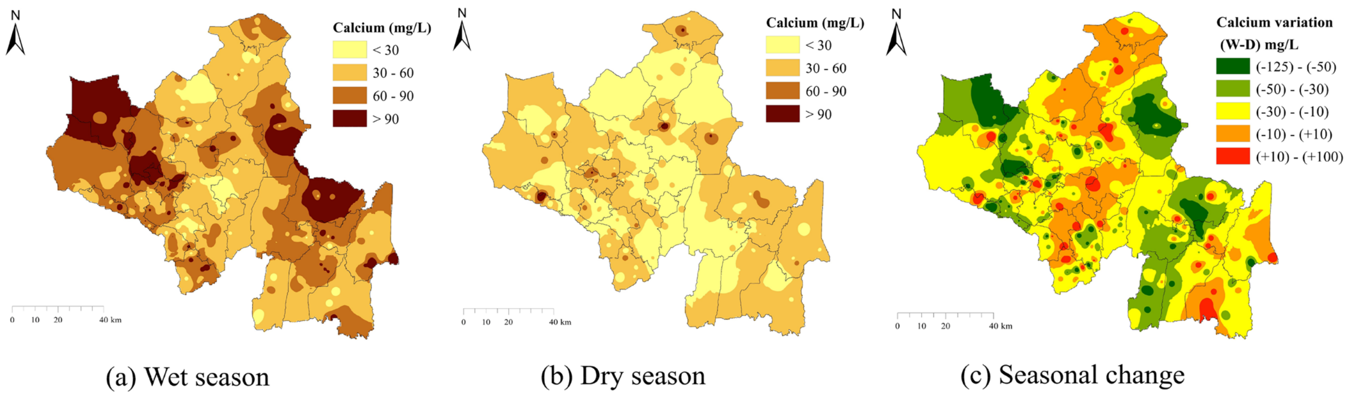

Figure 5. Spatial and seasonal variations (W-D) of calcium in groundwater in NCP.

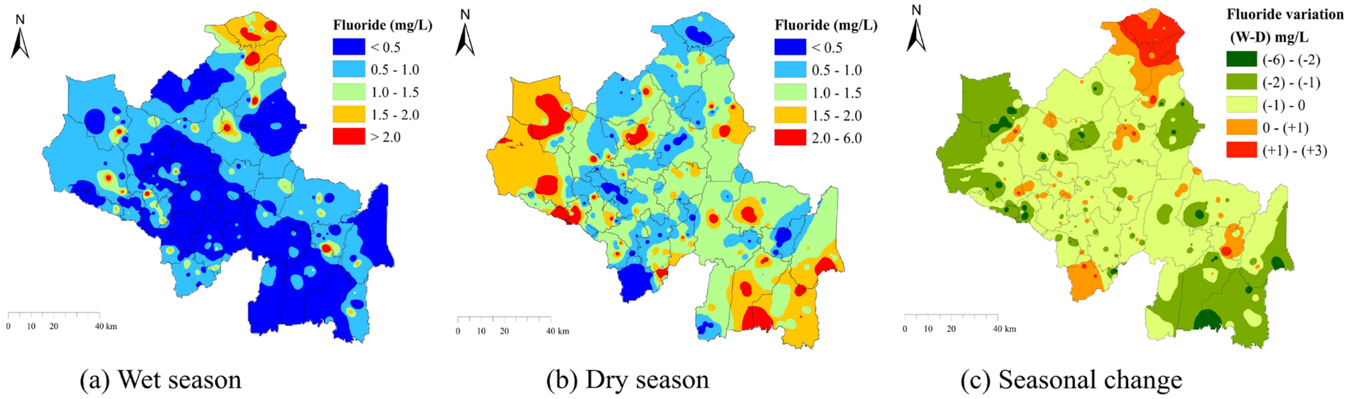

Figure 6. (a, b) Spatial and (c) seasonal variations of groundwater fluoride in the NCP (W-D).

from the ancient manmade tank cascade systems.[24,25] These aquifers have been created based on the regolith layer and deep fractured bedrock in the metamorphic terrains belonging to the Wanni geological complex.[26] Thus, this region has various silicate minerals (biotite, amphiboles, pyroxene) and carbonate minerals (calcite and dolomite) in its lithosphere which have different solubilities in groundwater. In addition, the lowest EC values (101.5 and 115.6 μS/cm) were reported from the same well located near Hingurakgoda (in the Polonnaruwa district) in both dry and wet seasons accordingly. The maximum value of EC (5479 μS/cm) was detected from a shallow well located in the Nuwaragampalatha central division in the Anuradhapura district during the dry season, but it was diluted up to 1855 μS/cm in the wet season due to a significant decrease in Mg^{2+}, Na^{+}, and SO_4^{2-} concentrations in the groundwater. See Text S3 in the Supporting Information for seasonal variations of EC.

3.2.2.2. Total Hardness. Total hardness and alkalinity represent the dominant ions (Ca^{2+}, Mg^{2+}, and HCO_3^{-}) in groundwater. Elevated levels of these geogenic minerals are inherent characteristics of the groundwater in the NCP. Generally, the groundwater hardness in the NCP during the wet season (282 mg L^{-1}) was higher than in the dry season (211 mg L^{-1}). The Anuradhapura region has a higher hardness level in the groundwater compared to the Polonnaruwa region (Figure 4). The highest hardness value in both seasons was observed from the same well located in the Horopothana division in Anuradhapura, at 1528.4 mg/L in the dry season and 974.6 mg/L in the wet season, respectively. The minimum hardness value (19.8 mg/L in the dry season and 52.2 mg/L in the wet season) was reported from the same shallow well located in the Hingurakgoda division that reported the lowest EC value. Basically, the Wanni complex which covers the majority (>90%) of the areas in the Anuradhapura region has higher calcium and magnesium minerals (calcites: dolomites, limestones) in the bedrock. The Highland and Vijayan complexes which cover the Polonnaruwa region are dominated by crystalline metamorphic carbonate minerals (marbles) and silicate minerals which are comparatively lower in Ca and Mg contents as well as having lower solubility (Figure 5).[21,22] Lower hardness levels in the Polonnaruwa region could be a result of the weak solubility of crystalline carbonate minerals in the area. Thus, the distribution of groundwater hardness seems to be affected by the geological and lithological deviations of the region.

Based on the WHO guidelines, water can be classified into four categories, according to its hardness level as follows: soft water at $CaCO_3$ hardness below 60 mg L^{-1}, moderately hard water at 60−120 mg L^{-1}, hard water at 120−180 mg L^{-1}, and very hard water at more than 180 mg L^{-1}. The majority of the groundwater samples in the NCP has hard and very hard groundwater in both wet (91%) and dry (82%) seasons (Figure S7), and a slightly lower level of hardness could be observed in the Polonnaruwa district, especially close to the border of the high land region of Sri Lanka. See Text S4 for seasonal variations of total hardness in NCP.

3.2.2.3. Fluoride (F^{-}). Fluoride is one of the major geogenic constituents in the groundwater which usually originates from natural rock weathering. Based on the Pearson correlation coefficients, a lower and positive correlation (r = +0.301; p > 0.01) was observed in fluoride with the alkalinity. Confirming the fact that fluoride-bearing minerals such as micas, hornblende, sphene, and apatite are commonly rich in

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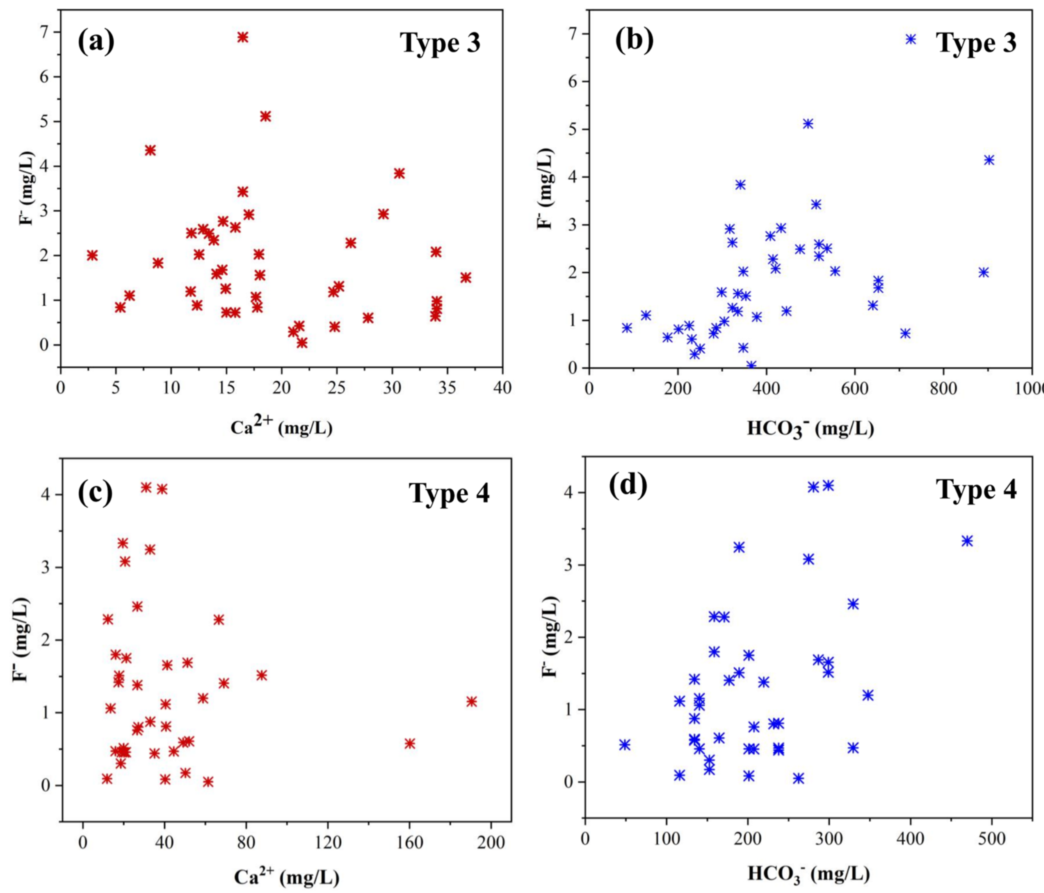

Figure 7. Variation of fluoride with calcium ions (a, c) and bicarbonate (b, d) in types 3 and type 4 groundwater in the dry season.

Precambrian metamorphic crystalline terrain in Sri Lanka, then minerals such as fluorite, tourmaline, and topaz which contribute to the general geochemical cycle of fluorine are also found in many locations in the dry zone.[10,27−29] Health-based drinking water guidelines have been provided by WHO and SLS to mitigate common disease like dental and skeletal fluoroses.[30] Results show that fluoride content in groundwater within the NCP varied from 0.02 to 4.85 mg L^{-1} in the wet season and 0.05 to 6.9 mg L^{-1} in the dry season, respectively, and a higher mean fluoride of 1.23 mg L^{-1} was observed in the dry season followed by a lower level of 0.58 mg L^{-1} in the wet season, indicating the higher evaporation effect in the dry season, under lower solubility of fluoride-bearing minerals. Here, 25.5% and 18.6% of the wells exceeded the MAL (>1 mg L^{-1}) established by WHO, in the wet and dry seasons, respectively. Similarly, 39.7% and 20.1% of the samples had lower fluoride content in both seasons in regard to the minimum required fluoride level (0.5 mg L^{-1}) given by SLS guidelines. Only 34.8% and 61.3% have been observed as acceptable for drinking purposes in both seasons.

Usually, in the wet season, shallow groundwater is likely to be diluted due to mixing with infiltrated rainwater through the soil layer while concentrated in the dry season due to high evaporation rates. But, some regions in the NCP such as the Padaviya division, showed a reduction fluoride content in the dry season (Figure 6b). This phenomenon can be explained by the solubility and the precipitation of CaF_2 (fluorite) in the aquifer or well.[31] Figure 6b shows that in the dry season F^- content was quite lower (<1 mg/L). It may be due to having a higher calcium level (>40 mg/L) in the shallow groundwater as shown in Figure 5b. Similarly, in the wet season (Figure 5a), calcium ion content was much lower in Padaviya compared to other regions of the NCP. When the season changes from wet to dry, fluoride ions tend to precipitate as CaF_2 due to the higher Ca concentrations in the groundwater which are further concentrated by the higher evaporation rates. Hence, the fluoride level in the dry season was significantly lower than in the wet season. However, calcium levels also decreased in other regions of the NCP in the dry season which may be due to the eventual flushing of minerals out by seepage from nearby surface water tanks, precipitation of Ca as calcites, or ion exchange with the aquifer materials during the dry season. See Figure 7 and discussion in Text S5 in the Supporting Information for further information on fluoride geochemistry and its fate in groundwater in the NCP.

3.2.2.4. Nitrate (NO_3^-). Nitrates and phosphates in the groundwater in Sri Lanka are mostly controlled by anthropogenic factors rather than natural factors.[2,32] It is one of the major constituents in freshwater, especially in surface water which is leached from inorganic fertilizers, septic systems, and manure storage or spreading operations.[33] It has been observed that the majority of groundwater samples collected from the NCP had lower nitrate levels compared to the drinking water quality standards (50 mg L^{-1}) given by WHO. They ranged from 0.02 to 54.4 mg L^{-1} in the wet season and from 0.05 to 90.8 mg L^{-1} in the dry season, respectively. Mean values of nitrate were ~3.05 and 6.6 mg L^{-1} accordingly. Here, 10.8% and 13.8% of water samples from the NCP in both seasons were identified as water having unacceptable levels of nitrate for drinking, based on the

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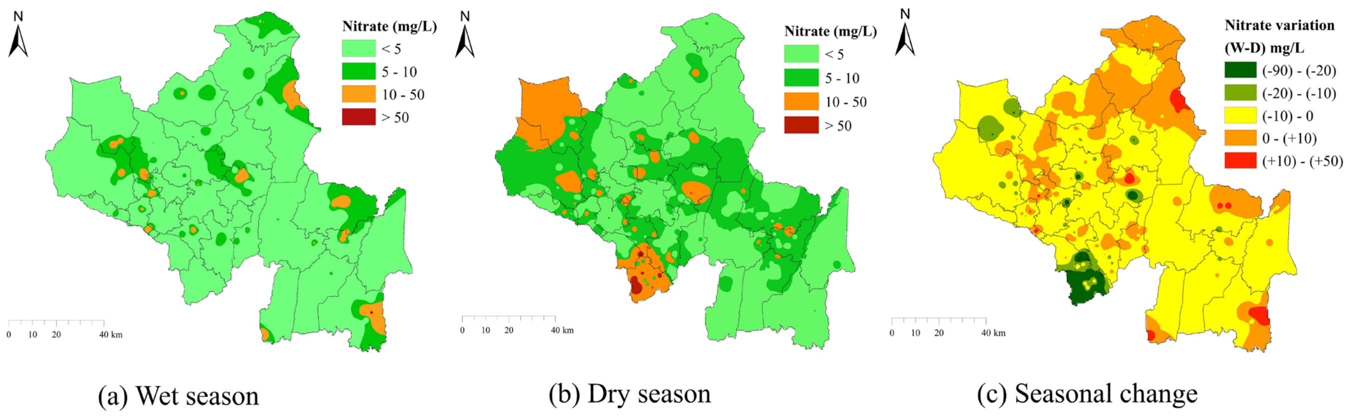

Figure 8. Spatial variation of nitrates in groundwater in the NCP.

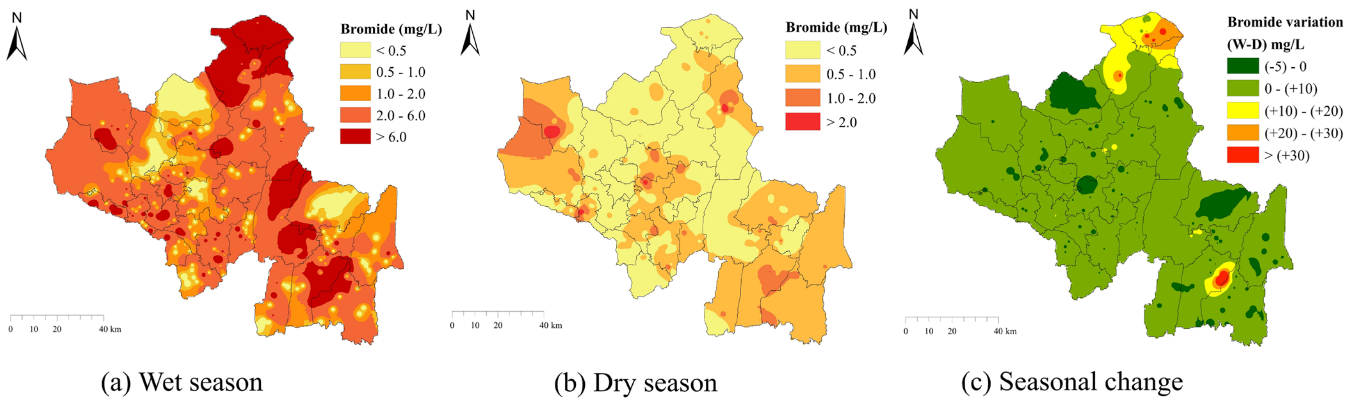

Figure 9. Spatial variation of bromide in the (a) wet season and (b) dry season and (c) its seasonal variation in shallow groundwater in the NCP.

WHO standards (MAL: 10 mg/L). Thus, the general outlook of the nitrate levels in groundwater (Figure 8a, b) revealed that in the dry season water was more polluted compared to the wet season due to the evaporation effect in the dry season (Figure 8c).

Seasonal variation of nitrates throughout the NCP showed that rainfall encouraged the increase of pollutants, especially in Horopothana and Hingurakgoda regions, while the nitrate level was diluted in the wet season and concentrated under high evaporation rates in the dry season in the Palagala and Mahavilachchiya regions (Figure 8b). According to the spatial distribution map, several locations (Palagala, Mahavilachchiya, Nochchiyagama Galenbindinuwewa, Horopothana, Medirigiriya, and Lankapura), especially regions with extensive paddy cultivation practices, showed significantly higher level (10−50 mg L^{-1}) nitrate increases compared to other regions. They were identified as groundwater nitrate hotspots in the NCP. Anthropogenic nitrate sources are further discussed in Section 3.4.

3.2.2.5. Bromide (Br^-). Bromide is a trace nonmetallic element in the groundwater in the NCP. Basically, bromide is not considered a toxic element for human consumption. However, the WHO has recommended that it is safe to drink water at 6 mg L^{-1} bromide for adults and 2 mg L^{-1} for children. In the NCP, groundwater bromide concentration spatially varied from 0.04 to 112.6 mg L^{-1} in the wet season and from 0.008 to 5.57 mg L^{-1} in the dry season while having mean values of 7.12 and 0.56 mg L^{-1} accordingly. The wet season showed the highest levels of bromide in the groundwater in the NCP, while 77.6% of samples exceeded the WHO MAL (2 mg L^{-1}).

Usually in coastal regions, groundwater contains a significantly higher Br^- concentration due to the seawater intrusion into the coastal aquifers. Similarly, anthropogenic sources such as potassium mining, fossil fuels (coal), acid mine drainage, coal-bed methane, oil and gas brine (up to 1287 mg L^{-1}), antiknock additive in leaded gasoline, agricultural pesticides, and some biocides have been reported in many studies around the world.[34,35] The NCP is located in the inland region of Sri Lanka and also belongs to the dry zone; seawater intrusion is thus not possible. Bromide can also be leached from the regional Precambrian metamorphic bedrock which consists of bromide-bearing minerals such as halite. The Pearson correlation coefficient showed that Br^- has a moderate positive correlation (r = +0.606; $p < 0.01$) with Cl^- and a lower correlation (r = +0.357; $p < 0.01$) with SO_4^{2-} ions indicating the common geogenic origin. Figure 9 shows the partial distribution of bromides in the wet and dry seasons and its seasonal change. Further, discussions regarding anthropogenic bromide contamination of groundwater in the NCP are discussed in Section 3.4.

3.3. Feasibility as Drinking Water and Irrigation Water.

3.3.1. Water Quality index (WQI). WQI is a comprehensive measure of the feasibility of water for drinking purposes.[11,36] It is a combination of many different water quality criteria with their weighted risks for health concerns, and this index let the decision makers and consumers understand the current situation of the water quality. This was calculated based on 11 different water quality parameters:

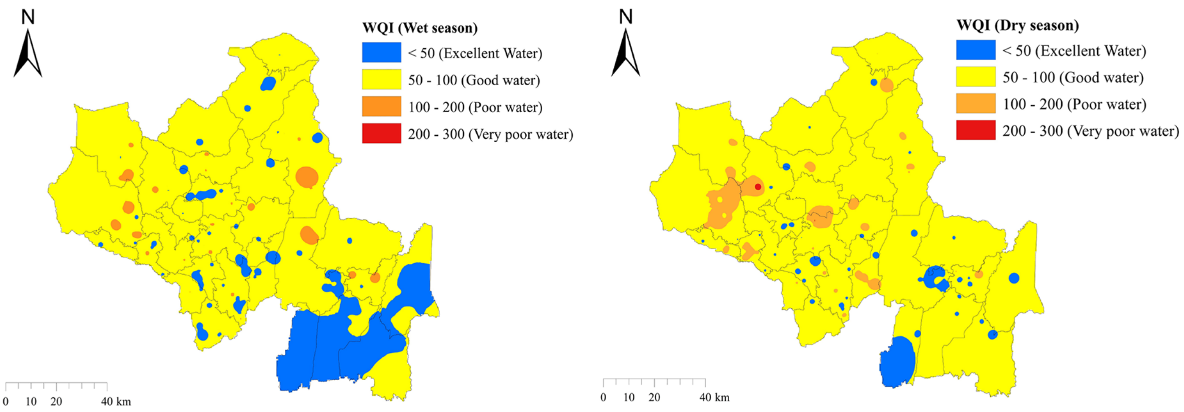

Figure 10. Spatial variation of WQI values in the NCP.

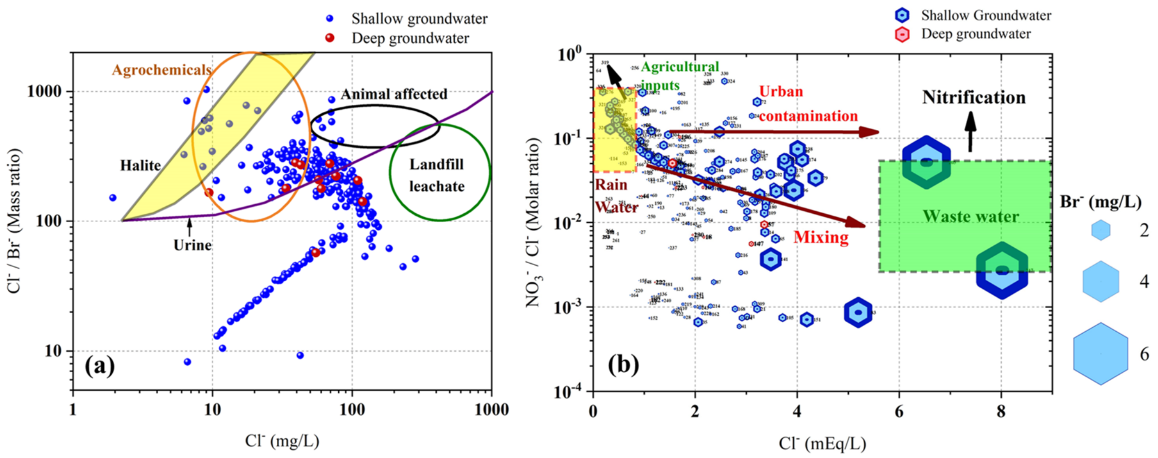

Figure 11. Anthropogenic impacts on shallow groundwater: (a) Cl^-/Br^- mass ratios with Cl^- concentration in groundwater and (b) variations of NO_3^-/Cl^- molar ratios with Cl^- molar concentrations of shallow groundwater.

pH, TDS, alkalinity, EC, Na, Ca, Mg, F^-, Cl^-, NO_3^-, and SO_4^-. The formula for the determination of this WQI is extracted from previous studies.[37] Based on the WQI results from this study (Table S8), it was observed that 99.3% of groundwater has a WQI value below 200. However, only 28% and 16.3% of the water samples had excellent water quality in wet and dry seasons, respectively. Poor water quality (WQI > 100) was observed in 8.9% and 13.1% of water samples from the NCP in two seasons, respectively. Thus, the percentage of wells that can be used to produce drinking water without proper treatment is more than 85%. The rest of them need advanced water treatment technologies which can remove dissolved constituents to produce safe drinking water.

The spatial variation of WQI values showed that Polonnaruwa had higher quality groundwater compared to the Anuradhapura district (Figure 10). However, groundwater quality in the dry season showed a slight deterioration which could be due to the rise in dissolved mineral content.

3.3.2. Groundwater as Irrigation Water. The sodium adsorption ratio (SAR) can be used to determine how irrigation water affects the soil texture.[38] It was calculated by eq 4 (Text S7). Usually, irrigation water with a higher SAR value has the ability to reduce the permeability of the soil and change its granular structure into a quite hard nonporous nature. In the dry season, groundwater in the NCP usually has higher SAR and EC values. SAR varied from 0.1 to 13.1, while EC ranged from 102 to 5479 μS/cm. Most of them (99.3%) had a low to medium salinity risk except only two wells which showed a high sodium risk. Similarly, only 2.4% of shallow groundwater samples showed a very high salinity hazard, and 5.5% of them had a lower salinity risk The rest of them (92.1%) showed a medium to high salinity risk. Deep groundwater lies in between a medium to high salinity risk section while lying in a low to medium sodium hazard risk. Though the sodium risk is not a significant issue in many water samples, 7% of them were identified as unsuitable for direct use in irrigation purposes (Figure S8). The residual sodium carbonate (RSC) index (eq 5) in this shallow groundwater ranges from −28.4 to 11.2 while at an average of 0.72. The results revealed that 58%, 26.6%, and 15.4% of groundwater samples from the NCP were safe (RSC < 1.5), marginal (1.5 < RSC < 2.5), and unsuitable (RSC > 2.5) for irrigation purposes, respectively.

The use of these high sodium waters requires special soil management methods, good drainage, a high leaching ability, and high organic matter conditions.[38] These high sodic waters can be used for soil with a high calcium content or soil containing gypsum ($CaSO_4{\cdot}2H_2O$). However, the use of gypsum would be a cost effective way to treat soil where high sodic groundwater was used.[38] However, another effective way to use these high risk source waters is by diluting with groundwater with lower salinity and SAR values from nearby wells or surface water from tanks before use as irrigation water.[38,39]

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3.4. Anthropogenic Impacts on Groundwater and Its Solution for Safe Drinking Water. *3.4.1. Anthropogenic Influence.* Halides (Br^-, Cl^-, and I^-) are widely used for the identification of possible contamination sources due to their conservative natures resulting in lower levels of interactions with the subsoil of groundwater flow. The mass ratio between chloride and bromide (Cl/Br ratio) can be used as a tool to determine the origin and evolution of groundwater concerning anthropogenic activities and natural contaminations such as seawater intrusion.[40] In this study, the Cl/Br mass ratio for all the groundwater samples ranged from 27.7 to 3470 with an average of 694. It was found that the highest value of Cl/Br occurred in the Nachchadoowa division located in the central Anuradhapura district with a mass ratio over 3000, while the lowest Cl/Br mass ratio occurred in the Thamankaduwa division in Polonnaruwa district with a Cl/Br mass ratio around 27.7. The Cl^- concentrations in these groundwaters were well below 285 mg L^{-1}. Usually, for seawater with a Cl^- concentration well above 3000 mg L^{-1}, a Cl/Br mass ratio for seawater-intruded water would be 610−680. Typically, Cl^- concentrations in groundwater are the highest near continental coastlines and drop rapidly toward the inland.[41] Thus, these observations would give a clear indication of not having the effect of seawater intrusion because this region is located in the inland area which is far away from the coastal side. Based on the Cl/Br mass ratio analysis (Figure 11a), 3.6% of wells showed the characteristics of recharge water in pristine conditions with certain trends of water−rock interactions (evaporite dissolution such as halite). They have groundwater with Cl^- levels below 20 mg L^{-1} and a Cl^-/Br^- mass ratio between 100 and 1000. Only five wells (1.8%) were identified with a possibility of having animal-affected groundwater. Most of these wells are open wells that do not have a proper cover for the opening. Thus, contamination by animals and plant residues could be possible. However, 65 (23.3%) of the wells showed a possibility of contamination by agrochemicals, because a paddy cultivation field covering a huge area of the open land of the NCP gives a hint for the possible connection between agrochemical and the quality of underlying groundwater bodies. For further confirmation of these observations and facts, an isotopic analysis should be conducted for the groundwater. As Figure 11b illustrates, along the mixing direction of the groundwater with different sources like wastewater runoff, the bromide (Br^-) level had an increasing trend toward the wastewater characteristics, while the highest Br^- level was reported from the samples which were affected by wastewater. Thus, it is clear that the source of bromide could be wastewater runoff.

According to the variation of the NO_3^-/Cl^- molar ratio with a Cl^- concentration, 12.2% of shallow wells were identified as groundwater with minimal impacts from water−rock interactions and recently recharged with infiltrated rainwater (Figure 11b). Those wells were mostly domestic shallow wells which have a huge possibility to recharge by precipitation due to the sandy soil type with higher permeability around the wells. Having a significantly higher level of Cl^- with much lower ratios of NO_3^-/Cl^- is a specific characteristic of municipal sewage-contaminated groundwater.[40,41] It was observed that few groundwater samples showed clues of contamination and mixing with municipal sewage or wastewater with shallow groundwater (Figure 11a). Samples with much lower Cl^- concentrations and higher NO_3^- levels could be ascribed to nitrate contamination of recently infiltrated rainwater by agricultural runoff in the wet season.[42,43] Thus, nitrate could be leached from the inorganic fertilizers from extensive agricultural lands, urban runoff, and municipal sewage or wastewater in the NCP. Anthropogenic pollution of groundwater in the NCP was confirmed by the cumulative probability analysis as well (Text S8 and Figure S9).

3.4.2. Selection of Different Water Treatment Technologies for Different Areas of the NCP Based on the Specific Water Quality Issues. Elevated and unexpected levels of different water quality parameters and their spatial variations in groundwater could be observed throughout the NCP. Thus, the technologies required for the removal of those contaminants should be different by region. Especially, unaccepted levels of EC, hardness, alkalinity, fluoride, nitrate, and TOC were major health concerns in water qualities that were identified in many different places of the NCP (Figure S10). Based on these water qualities, it was realized that a single type of process or technology cannot be recommended for every vulnerable region. Thus, different types of drinking water treatment technology implementations or their combinations should be considered.

Nochchiyagama and Mahawilachchiya were identified as regions with elevated levels of EC (>1000 μS/cm), hardness (>250 mg/L), alkalinity (>200 mg/L), fluoride (>1.5 mg/L), nitrate, and silicon (>100 mg/L) as well as TOC (>5 mg/L). Similarly, Galenbindinuwewa and Thirappane divisions showed similar groundwater compositions except for lower fluoride contents (<1.5 mg/L). Thus, advanced water treatment technologies such as RO, nanofiltration (NF), and electrodialysis reversal (EDR) are required to purify these groundwaters with high mineral content. RO membrane technology has been already implemented in many villages of these regions,[1,13,15] RO systems can handle feedwater with higher EC and hardness to produce drinkable water with lower mineral content. However, the copresence of hardness cations Ca and Mg with TOC at elevated levels can be a major threat to membrane durability and efficiency due to the organic−inorganic synergistic fouling phenomenon.[44,45] This heavy fouling can be irreversible and thus reduce the life span of the RO membranes while increasing its maintenance cost. Similarly, the elevated levels of silicon originating from the weathering of silicate minerals could be a critical issue when the groundwater has higher levels of hardness, because membrane fouling by silicon polymerization can be enhanced with the presence of these cations, which could lead to a drastic reduction in the performance of the RO membrane system and its life span.[46] Hence, when RO membrane technology is used to purify the groundwater in this region, a proper softening process should be carried out to reduce the hardness of metal cations, and an activated carbon filtration step should be used to remove the DOC or natural organic molecules in the RO feedwater to mitigate irreversible fouling.[47,48]

In the southern Polonnaruwa district, Thamankaduwa and Dimbulagala divisions were only with high levels of fluoride and alkalinity (Figure S10c, d). Thus, fluoride removal should be the main focus in this region. This can be easily achieved by the RO filtrations because RO technology shows excellent removal of fluoride and HCO_3^- ions in this type of groundwater.[13,15] However, this groundwater does not need an advanced and expensive purification technology with a higher salt rejection capability, because the total mineral content (EC < 500 μS/cm) was significantly lower in the

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shallow groundwater in this region. If RO filtration is used, produced drinking water will have quite lower mineral content (EC < 20 μS/cm) which could lead to mineral deficiencies due to its higher mineral rejection rates (>96.6%);[13] thus, a post mineral addition process should be carried out. However, the NF technology showing excellent performance in producing drinking water while retaining essential minerals would be a much better option for treating this low mineral groundwater, because NF shows lower rejection (71.8% and 92.4%) of essential minerals (Ca^{2+} and Mg^{2+}) compared to the RO rejection performance (95% and 99%) while showing a sufficient level of removal rates for fluoride (67.7%) as well.[15]

Shallow groundwater in the northern tip of Anuradhapura (Padaviya division) at a moderate level of dissolved minerals (EC < 1000 μS/cm) showed a higher level of fluoride, hardness, and DOC concentrations (Figure S10b, d, f). The use of pressure-driven membrane technologies such as RO and NF would not be necessary, and organic−inorganic combined synergistic fouling could be a major issue in RO and NF when treating groundwater with higher hardness and DOC levels. Thus, the removal of DOC by activated carbon (AC) filtration should be carried out before the membrane process. EDR could be a better process for treating these types of groundwater conditions because EDR has the ability to remove ions selectively, and rejection rates can be adjusted to produce drinking water with the required levels of essential minerals like Ca and Mg, as well as fluoride.[49,50] The major advantage of this technology is that a significant amount of inorganic scaling can be removed by a reversal of the electric field while the treatment process continues. However, feedwater pretreatment should be a necessary step to mitigate organic fouling as well as boron fouling which could be irreversible later.[51]

The southern tip (Palagala division) of the Anuradhapura district showed comparatively better groundwater quality. The surrounding area is rich in the availability of natural springs with excellent water quality in this region. Except for the nitrate level, EC, hardness, alkalinity, fluoride, and TOC were at acceptable levels in the shallow groundwater in this division (Figure S10e). Due to a lower dissolved mineral content, the ion exchange process can be suggested as an efficient and cost-effective way for nitrate removal without much interruption of other constituents in the groundwater compared to membrane-based technologies. This process has long been used for nitrate removal in drinking water production using nitrate selective or nonselective resins which are commercially available and show higher performances.[52−54]

Implementation of existing RO drinking water stations in the NCP has not been carried out considering the regional and specific water quality issues. Thus, a considerable number of RO stations have been shut down and stopped their services due to a lack of consumers and commonly available good quality natural drinking water sources such as spring water in the Palagala and Kebithigollawa divisions.[13,15] Similarly, RO stations in some regions suffer from heavy fouling problems due to higher organic matter with very hard water. Hence, it is important to consider the source water quality and availability of good quality natural water before implementing any new technologies in a location (Figure S10h).

CONCLUSIONS AND PERSPECTIVES

A comprehensive investigation of groundwater quality in the NCP was conducted to fulfill the current demand for clean drinking water. The shallow and deep groundwater samples were collected from two districts (Anuradhapura and Polonnaruwa) of the NCP including CKDu prevailing areas. Silicate weathering was identified as the main source of dissolved minerals in the NCP, which was followed by the cation exchange process and anthropogenic activities, especially for shallow groundwater. Unexpected levels of salinity (EC), hardness, fluoride, nitrate, sulfate, dissolved organic carbon (DOC), and alkalinity of the groundwater were the common issues in the majority of rural areas in the NCP. Heavy agricultural regions like Palagala, Mahawilachchiya, Nochchiyagama, Galenbindinuwewa, Horopothana, Medirigiriya, and Lankapura divisions are identified as nitrate hotspots that could originate from inorganic fertilizers.

Results revealed that, in both wet and dry seasons, Ca-HCO_3 type groundwater dominated in both shallow groundwater (82% and 59.4%) and deep groundwater (66.7% and 58.3%), respectively. The composition of the groundwater in the NCP is directly controlled by rock−water interactions, surface water interactions, and the evaporation effect. According to the WQI analyses, 8.9% and 13.8% of wells with poor quality groundwater have been identified in wet and dry seasons, respectively. SAR and salinity hazard analysis revealed that only 7% of the groundwater samples from the NCP could be harmful for soil fertility. This was the first time that groundwater quality and geochemical atlases were created for the entire region of the NCP, including the high risk CKDu region within the province.

In the past, implementation of decentralized RO drinking water stations in the NCP did not consider regional water quality issues. However, RO technology with a pretreatment process of activated carbon filtration and softening (ion exchange) was identified as the most suitable way to treat groundwater for many regions of the NCP with the groundwater having higher mineral content, while NF and EDR are suggested for low salinity groundwater with contamination issues. Regions with higher nitrate ions (Palagala division) could use adsorptions methods for treating groundwater rather than using membrane-based technologies. Hence, these findings reveal the importance of a source water quality survey before making water supply master plans and implementing water treatment technologies to a specific region of the country.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestwater.2c00490.

> Literature review of groundwater quality highlights in the NCP, Sri Lanka. Equations used in the paper. Basic data of sample location. Piper trilinear diagrams and Gibbs plots for groundwater in the NCP for both wet and dry seasons. Groundwater geochemical relationship illustrations. Correlation among water quality parameters. Summary of water quality. Seasonal variation information on groundwater quality. Groundwater quality atlas for major ions and trace elements. Mineral weathering information in Sri Lanka. Hardness classification information. SAR and salinity hazard analysis. Cumulative probability analysis of anthropogenic groundwater nitrate in the NCP. Atlas of regional

water quality issues and suggested advance treatment processes in the NCP. (PDF)

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Notes

The authors declare no competing financial interest.

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