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Chemical characteristics and water stability evaluation of groundwater in the CKDu Zone of Sri Lanka

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ABSTRACT

Groundwater is the main source of drinking water for the rural population in the chronic kidney disease of unknown etiology (CKDu) zone of the North Central Province (NCP) in Sri Lanka. In this study, a total of 334 groundwater samples (311 dug wells, 21 tube wells and 2 springs) during the wet season from two aquifers in the NCP were collected, and investigated their chemical characteristics and evaluate their water quality, including groundwater chemistry, main ion sources, the corrosion and scaling potential of groundwater. The results showed that the two hydrochemical types of groundwater in the NCP were mainly of the Ca-HCO_3, Na·Ca-HCO_3 types, with the main HCO_3^-, Na^+ and Ca^{2+} ions in both types of groundwater originating from silicate and evaporite salt dissolution and influenced by alternating cation adsorption, while the presence of NO_3^- was mainly anthropogenic. Evaluation of water stability using namely Langelier saturation index (LSI), Ryznar stability index (RSI), Puckorius scaling index (PSI) and Larson-Skold index (LS), indicated that most groundwater presents corrosion potential and has corrosion behavior tendency of metals to some degrees. The water quality of Polonnaruwa was better than that of Anuradhapura in the

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Q3 [a] Research Interest: Membrane technology for drinking water treatment.
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NCP, and when the groundwater was worse than the "good" grade, which must be properly treated before it is used as drinking water.

Introduction

The groundwater is often of good quality and can be used safely and economically without the need for a complex processing program, thus the self-supplied water (groundwater) is considered as the most cost-effective way to provide a safe water supply in the rural area (UNESCO, 2022). In 2018, around 18% of the urban population and 34% of the rural population rely on self-supplied water (groundwater) in Sri Lanka (Foster et al., 2021), and the rest of the water supply mainly depends on surface water such as rivers, streams and tanks (National water supply and drainage board, 2019). Previous studies have already identified mainly six types of groundwater aquifers in Sri Lanka, including shallow and deep karstic aquifers in the Miocene beds, coastal sandy aquifers, alluvial aquifers in lower reaches of river basins, deep confined aquifers in the metamorphic terrain, shallow regolith aquifers of the metamorphic terrain, and lateritic (Cabook) aquifers in the south-western region (Gupta et al., 2018). Groundwater in the chronic kidney disease of unknown etiology (CKDu) prevalence zone of the North Central Province (NCP) is mainly derived from shallow weathered aquifers and deep hard rock aquifers in metamorphic terrain (Sood et al., 2015), and more than 80% of the rural population relies on groundwater as their main source of drinking water and irrigation water (Balasubramanya et al., 2020). Of these, dug well water and tube well water are the most commonly used in the CKDu affected areas (Cooray et al., 2019a; Gunawardena and Pabasara, 2016a). According to the research report of the World Health Organization (WHO), drinking water is one of the key factors affecting CKDu (WHO, 2012). Therefore, it is of great practical significance to clarify the characteristics of groundwater quality and its influencing factors in CKDu area and determine the water stability index, which can provide scientific basis for local water supply security. Moreover, in recent years, the excessive exploitation of groundwater has aggravated the change of local groundwater quantity and quality. The in-depth study of the groundwater hydrochemical will help to promote the research on the cause of CKDu.

In the rural areas of the NCP, the water supply is mainly the responsibility of the Community Based Organization (CBO) (National water supply and drainage board, 2019). As the primary source of drinking water for the NCP, groundwater quality regularly monitored and assessed is necessary to develop management plans to protect it from contamination. Generally, the chemical composition of groundwater is mainly controlled by the rock type of aquifer, residence time, flow path and recharge source (Catalán et al., 2016; Lechleitner et al., 2017; Lü et al., 2020). Medmunds et al. studied the geochemical evolution of groundwater along Lake Texcot using stable isotope and radiocarbon methods and found that groundwater recharge in the area was mainly derived from local precipitation and that the main ions in the water were mainly derived from weathering of basaltic minerals (feldspar and magnesian iron minerals), while fluoride was derived from apatite in basalt or rhyolite (Edmunds et al., 2002). Antonellini et al. found that alluvial aquifers closer to the ocean caused seawater intrusion due to excessive groundwater extraction, which changed the quality of groundwater (Antonellini et al., 2008). Sarikhani et al found that river recharge, dissolution of evaporite minerals (such as gypsum) in the formation and agricultural returned water increased the salinity of groundwater in Bushehr, southwest Iran, while the dissolution of halite caused the linear increase of sodium and chloride (Sarikhani et al., 2015). Previous study has shown that the main hydrochemical types of dug well water in the North Central Province are Ca-HCO_3 and Mg-HCO_3 (Rubasinghe et al., 2015; Wickramarathna et al., 2017), and affected by rock weathering and evaporative crystallization (Jayawardana et al., 2012; McDonough et al., 2021; Rubasinghe et al., 2015). However, most of these investigation studies revolved around the Anuradhapura area during the dry season and had fewer samples, not including the Polonnaruwa, and could not be extended to other CKDu zone in the NCP; Secondly, information on the hydrochemistry of tube well water studies is also lacking, and no systematically and in-depth analysis of its source composition, formation process and hydrochemical characteristics has been carried out (Cooray et al., 2019b; Shi et al., 2022; Udeshani et al., 2022). In addition, the application of stability indices in water distribution systems has been reported in many researches, analysed the quality of groundwater (Al-Tamir, 2021; Bum et al., 2015; Wang and Zhu, 2021). The use of groundwater with poor quality and inadequate purification methods aggravates the corrosion or scaling effect of pipelines during groundwater transportation, and pose potential threat to the safety supply of drinking water. The water stability indices (Eslami et al., 2020; Khorsandi et al., 2016; Taghipour et al., 2012) such as Langrier Saturation Index (LSI), Rezner Stability Index (RSI), Pukeles Scaling Index (PSI) and the Larsen-Scord Index (LS), allow prediction of the corrosion and scaling capacity of groundwater, which can provide guidance for water supply. Therefore, a comprehensive analysis of the mechanisms influencing groundwater hydrochemical in different wells in the NCP is needed, the water stability and water quality status are determined ensure the safe drinking water supply in the CKDu affected zone.

Through investigating the spatial distribution of basic physicochemical parameters and major ions, the purpose of this study was to comprehensively explore characteristics of groundwater hydrochemistry in the CKDu zone, and clarify the main factors influencing both types of groundwater hydrochemistry, determine their water quality stability in order to further provide a solid basis for safe drinking water supply master plan in the NCP.

1. Materials and methods

1.1. Study area and sampling

As shown in Fig. 1, the CKDu prevalence zone in the NCP was selected as the study area, including 29 District Secretariats divisions in Anuradhapura and Polonnaruwa. The samples were collected from the dug wells, tube wells and natural springs.

The NCP is in the dry zone of Sri Lanka, and the total land area is 10714.0 km^2 with a total population of nearly 1.26 million and many rivers. Its west is mainly plain and its east is hilly and plain. Where the mean annual temperature varies between 25°C and 33°C, the Southwest and Northeast monsoons bring the rainfall to this area from May to August and from October to next February, respectively. The average monthly precipitation is more than 200 mm, but less than 20 mm in the dry season, a typical tropical monsoon climate (Burt and Weerasinghe, 2014). The groundwater in the NCP existed both in the weathered rock zone and the weathered layer and deep bedrock fault zone (Gunawardena and Pabasara, 2016b). The former has a shallow depth of burial with a maximum depth of 10-20 m. As a result, the majority of dug wells are located primarily in this aquifer. In the hard metamorphic zone, deeper fracture zones occur at 30-40 m and some can reach 70-100 m, and groundwater within the fracture zones is thus mainly extracted through the use of tube wells (Gupta et al., 2018; Karunaratne and Pathmarajah, 2002).

Two hundred fifty-four (254) water samples were collected from Anuradhapura, including dug well (236), tube well (16), and springs (2), eighty (80) water samples were collected from Polonnaruwa from dug well (75) and tube well (5) (Table S1). In the meantime, the precipitation data of seven meteorological stations in the NCP and adjacent provinces were collected. The map of meteorological stations distribution and precipitation data are shown in Fig. S1 and Fig. S2. Major field campaigns were conducted from 2nd December 2019 to 30th December 2019, and it was covered every District Secretariats division. The sample sites were selected based on resident population, areas, availability, and accessibility to well groundwater. All of the samples were collected in pre-washed Polypropylene bottles and were stored at 4°C in the incubator. After sampling, the samples were taken back to the Research Central for Eco-environment Sciences, Chinese Academy of Sciences (RCEES, Beijing, China) for analysis.

1.2. Main ions

The pH, electrical conductivity (EC), Temperature(°C) and total dissolved solids (TDS) were determined in the field by a water quality analyzer (WTW, MultiLine Multi 3530, Welheim, Germany). The samples were stored in a thermostat at 4°C in the field and sent to the University of Peradeniya in 24 hours, and then back to China for further analysis after field sampling campaigns. Other detailed information of the ions analysis method is listed in the suport information.

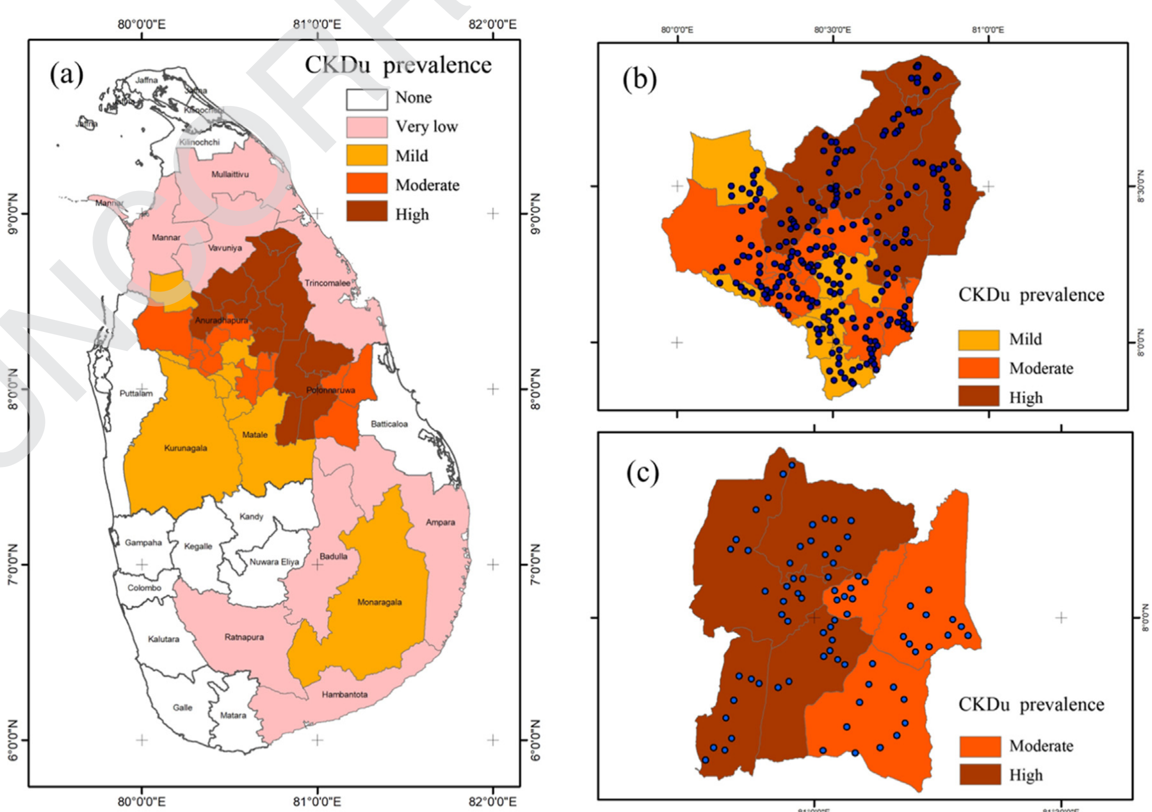

Fig. 1 – Study areas of the NCP in Sri Lanka (a) map of Sri Lanka; (b) map of Anuradhapura district, with sampling sites shown as black points; (c) map of Polonnaruwa district, with sampling sites shown as dark blue points).

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Table 1 – Water stability indices for corrosion and scaling potential.

Index	Equation	Value	Water condition
LSI	$LSI = pH - pH_S$	LSI>0	Super-saturated, Scaling potential
	$pH_s = 9.3 + A + B - (C + D)$	LSI=0	Saturated
	$A = (Log_{10}[TDS] - 1)/10$	LSI<0	Under-saturated, Corrosion potential
	$B = -13.12 \times Log_{10}(Temperature\ in\ ^{\circ}C + 273) + 34.55$		
	$C = Log_{10}[Ca^{2+}\ as\ CaCO_3] - 0.4$		
	$D = Log_{10}[Alkalinity\ as\ CaCO_3]$		
RSI	$RSI = 2pH_S - pH$	RSI<6	Scaling potential
		6≤RSI≤7	Equilibrium;
		RSI>7	Corrosion potential
PSI	$PSI = pH_{eq} - pH_s$	PSI<6	Scaling potential
	$pH_{eq} = 1.465 \times Log_{10}alkalinity + 4.54$	6≤PSI≤7	Equilibrium
		PSI>7	Significant corrosion potential
LS	$LS = (Cl^- + 2 \times SO_4^{2-})/HCO_3^-$	LS<0.2	No metal tendency
		0.2≤LS<0.4	Light metal tendency
		0.4≤LS<0.5	Low metal tendency
		0.5≤LS<1.0	Mid metal tendency
		LS≥1.0	High metal tendency

Note: pH: actual measured pH of water; pH_s: pH at the equilibrium state of $CaCO_3$; TDS: Total dissolved solids (mg/L);°C: Measuring temperature; Ca^{2+}: Calcium hardness (mg/L); Alkalinity: alkalinity (mg/L, calculated as $CaCO_3$); pH_{eq}: pH value at equilibrium; Cl^-: Chloride (mg/L); $SO_4{}^{2-}$: Sulfate (mg/L); $HCO_3{}^-$: alkalinity of hydrogen carbonate (mg/L, calculated as $CaCO_3$).

1.3. *Water stability indices*

The water stability indices such as Langrier Saturation Index (LSI), Rezner Stability Index (RSI) and Pukeles Scaling Index (PSI) can be used to predict the corrosion and scaling behavior of water (Taghipour et al., 2012; Eslami et al., 2020). The calculation of Larsen-Skold index (LS) can predict the corrosion degree of water phase on the walls of the pipelines of low carbon steel metal pipes (Khorsandi et al., 2016). Details are shown in Table 1.

LSI was proposed in 1936 to describe the equilibrium relationship between water containing CO_2 and calcium carbonate solid. When LSI>0, $CaCO_3$ is saturated, calcium carbonate in water tends to deposit, and when LSI<0, water have corrosion potential; RSI is an empirical index proposed through experiments, which can relatively quantitatively predict the tendency of calcium carbonate precipitation and dissolution in water. When RSI<6, the water is easy to scale, 6≤RSI≤7, the water is relatively stable, RSI>7, the water tends to have corrosion, and as the value increase, the stronger the corrosion capacity. Based on the RSI index, the researchers also proposed the PSI, which is similar to RSI. When PSI<6, $CaCO_3$ is supersaturated, with the increase of PSI value, the water quality gradually presents corrosivity. LS index is used to evaluate the erosion of water quality on pipelines. LS<0.2, no metal tendency, 0.2≤LS<0.4, Light metal tendency, 0.4≤LS<0.5, metal tendency, 0.5≤LS<1.0, middle metal tendency, LS≥1.0, high metal tendency.

1.4. *Water quality assessment methods*

The core of the water quality assessment method is to refer to health standards as the evaluation criteria, to a certain extent to reflect the characteristics of water quality indicators constitute a set of effective comprehensive evaluation methods, and thus can objectively evaluate or judge the overall situation of water quality. In 2018, Titus (Cooray et al., 2019b) firstly used the composite index method (WQI) for a simple evaluation of groundwater in the Anuradhapura area of the NCP, exploring the possibility of a method that could be applied to the evaluation of groundwater quality in the NCP. As its small sample size and single method, it was not sufficient to extend it to other CKDu zone in the NCP. Therefore, this study uses three methods such as the Mean Composite Pollution Index, the modified Nemero Pollution Index and the Water Quality Index (WQI). Based on the results of sampling and analysis, 21 indicators such as pH, TDS, total alkalinity, hardness, Ca^{2+}, Mg^{2+}, Na^+, F^-, Cl^-, $SO_4{}^{2-}$, $NO_3{}^-$, Al^{3+}, Fe^{3+}, As, Cd, Cr, Cu, Mn, Ni, Pb and Zn were selected regarding the Sri Lankan Drinking Water Standard (SLS 614-2013) and WHO Drinking Water Guide (4^{th}), considering that DOM in drinking water can lead to the production of disinfection by-products, TOC was introduced into the calculation of indicators, for a total of 22 indicators, to evaluate the NCP groundwater.

1.4.1. *Average value synthesis pollution exponential*

Synthesis pollution exponential method is based on a single factor pollution index method based on the premise of the statistical analysis method (Shudong et al., 2022). It is a single water quality index of the measured value and the evaluation of the standard value of the ratio, used to judge the water quality indicators to meet the requirements of the corresponding standards, the expression (except for pH) is as follows.

$$P_i = \frac{C_i}{C_0} \quad (1)$$

Where: P_ii is the single factor pollution index; C_i is the actual concentration measurement of the ith indicator; C_0 is the evaluation standard value of the ith indicator (Table S3), all in mg/L.

The average value synthesis pollution exponential is based on the synthesis pollution exponential and the formula is as follows:

$$P = \frac{1}{n}\sum_{i=1}^{n} Pi \quad (2)$$

Where: P is the average value synthesis pollution exponential, which is the arithmetic mean of the single factor pollution index of n indicators; Pi is the single factor pollution index, and the pollution level is classified according to the P value is detailed in Table S2.

1.4.2. Nemerow pollution index

The Nemerow Pollution Index is an environmental quality index that highlights the maximum values, taking into account the average and maximum values of the single factor pollution index, emphasising the influence of the maximum pollution factor on water quality pollution (Wei et al., 2017). The introduction of the coefficient of variation can offset to some extent the error in the evaluation results caused by the maximum value, which in turn can reasonably reflect the comprehensive characteristics of the water quality, the formula is as follows:

$$w_i = \left(\frac{CV_i}{\sum_{i=1}^{n} CV_i}\right) \quad (3)$$

$$P_{i,max} = max\left(\frac{w_i C_i}{C_0}\right) \quad (4)$$

$$P = \frac{1}{n}\sum_{i=1}^{n} Pi \quad (5)$$

$$I = \sqrt{\frac{P^2 + (P_{i,\ max})^2}{2}} \quad (6)$$

Where: w_i is the weight of each parameter, CV is the coefficient of variation, $P_{i,max}$ is the maximum value of single factor pollution index among n indicators, P is the average value synthesis pollution exponential, I was the Nemerow Pollution Index value, and the pollution level division is shown in Table S2.

1.4.3. Water quality index (WQI)

The Water Quality Index (WQI) is based on the relative importance of each parameter, giving the weight of each parameter, to account for the synergy of individual water quality parameters, reflecting the comprehensive characteristics of water quality (Brown et al., 1972). The relative weights of each index are shown in Table S3, the formulae are as follows:

$$W_i = \frac{w_i}{\sum_{i=1}^{n} w_i} \quad (7)$$

$$Q_i = \frac{C_i}{C_0} \times 100 \quad (8)$$

$$WQI = \sum_{i}^{n} W_i \times Q_i \quad (9)$$

Where: w_i for the weight of each parameter, W_i for the relative weight of each factor index, Q_i for the single index, WQI for the water quality index value, according to the **WQI** value assessment of water quality classification, see Table S2 for details.

1.5. Statistical analysis

Statistical analysis was performed using a SPSS (21.0, IBM, U.S.A) and an Origin (2018, Originlab, U.S.A) software. ArcGIS (10.1, ESRI, U.S.A) was used to create maps of major ion distribution, water stability indices maps and water quality assessment maps.

2. Results and discussion

2.1. Groundwater chemistry

As is shown in Table 2, the pH of the dug well and the tube well are 7.81 ± 0.7 and 7.15 ± 0.76 respectively, and the TDS is 873.25 ± 531.54 and 729.48 ± 494.89 mg/L respectively. Among which, HCO_3^- was the main anion in the dug wells, accounting for 74.27% of the total anion concentration, and Na^+ and Ca^{2+} are the main cations, accounting for 79.13% of the total cation concentration. The ion concentration order was HCO_3^- > Na^+ > Ca^{2+} > Cl^- > Mg^{2+} > SO_4^{2-} > K^+ > Br^- > NO_3^- > F^-, and the order of trace ion concentration was Mn> Zn> Al> Fe> Cu> Ni> As> Pb> Cr> Ti> Cd (Table S4). In tube wells, the anion was dominated by HCO_3^-, accounting for 76.80% of the total anion concentration, the cation was dominated by Na^+ and Ca^{2+}, accounting for 83.05% of the total cation concentration, the ion concentration order was HCO_3^- > Na^+ > Ca^{2+} > Cl^- > Mg^{2+} > SO_4^{2-} > Br^- > K^+ > NO_3^- > F^-, and the trace ion concentration was Zn> Mn> Fe> Al> Cu> Ni> Ti> As> Pb> Cr> Cd. From the average values of the main ion concentrations, the cation with the highest concentration in both groundwaters was Na^+ and the anion with the highest concentration was HCO_3^-, the order of ion concentrations is the same indicating that the two groundwaters have some similarities (except for Br^-). The classification according to TDS indicates (Table S5) that both dug well water and tube well water have a high degree of mineralization, which revealing 66.89 % and 71.43 % were freshwater type (TDS<1000 mg/L) according to the TDS classification (Bouaissa et al., 2021). 99% of groundwater samples are moderately hard based on hardness classification (Gupta et al., 2018) (Fig S3), these were similar to the results of previous studies (Abeywickarama et al., 2016; Cooray et al., 2019b, 2019a; Pinto et al., 2020). The coefficient of variation (CV) reflects the degree of dispersion of each measured value, except for pH, the coefficients of variation for the major water chemical ions in both groundwaters are greater than 0.5, which were above moderate variation, indicating that there are more factors affecting the concentration of the major ions in groundwater

According to the Sri Lanka Drinking Water Standard (SLS614-2013), the TDS, total alkalinity, Mg^{2+} and total hardness in both groundwaters were exceeded to some degrees. Spatial mapping of major ions in groundwater using interpolation (IDW) as shown in Fig. S4, the concentrations of all

Table 2 – Concentrations of major water chemistry parameters and DOM in the study area (mg/L, except pH, Temperature/°C).

Parameters	Dug well			Tube well			Spring		
	Range	Mean±SD	CV%	Range	Mean±SD	CV%	Range	Mean±SD	CV%
pH	5.86-9.38	7.81±0.70	0.10	6.61-7.73	7.15±0.76	0.11	7.0-7.84	7.42±0.0.59	0.08
Temperature	22.2-32.5	27.22±0.99	0.04	24.5-29.8	27.49±1.51	0.05	26.5-26.9	26.7±0.28	0.01
TDS	116.00-4390.00	873.25±531.54	0.64	279-2270	729.48±494.89	0.68	98.50-105.00	101.75±4.60	0.05
HCO_3^-	24.00-844.00	276.38±153.71	0.56	59.00-583.00	311.09±148.10	0.48	40.00-69.00	54.23±20.27	0.61
K^+	0.31-619.86	8.49±44.62	5.23	0.88-7.09	2.88±1.75	0.61	1.44-1.61	1.52±0.12	0.08
Ca^{2+}	0.54-162.03	61.21±32.89	0.54	4.24-155.25	64.89±43.02	0.66	5.43-5.60	5.52±0.12	0.02
Mg^{2+}	0.02-179.58	30.05±23.72	0.79	8.72-46.41	41.01±22.03	0.54	1.30-3.68	2.49±1.69	0.68
Hardness	1.44-974.48	276.56±140.41	0.51	99.04-626.50	312.82±133.20	0.43	28.72-186.62	107.67±111.65	1.04
Na^+	0.03-1126.21	84.88±106.25	1.25	12.11-431.74	153.33±165.72	0.94	10.02-50.83	30.43±28.86	0.95
F^-	0.02-4.85	0.82±0.77	0.93	0.14-1.77	0.71±0.47	0.70	0-0.10	0.05±0.07	1.41
Cl^-	0.03-215.07	58.37±43.47	0.74	0.37-157.22	57.57±43.03	0.75	11.32±23.03	17.18±8.28	0.48
Br^-	0.04-112.59	7.01±10.32	1.47	2.11-21.28	8.33±4.38	0.53	6.61-7.60	7.11±0.70	0.10
NO_3^-	0.02-54.44	4.65±7.98	1.70	0.26-8.07	2.04±2.06	1.01	0-2.27	1.14±1.61	1.41
SO_4^{2-}	0.17-307.58	24.22±30.49	1.26	0.94-118.83	25.31±27.72	1.10	2.27-10.02	6.4±5.13	0.80
TOC	0-31.60	5.64±3.57	0.63	1.7-10.70	5.31±2.40	0.45	2.60-3.30	2.95±0.49	0.17

major ions in groundwater in the Anuradhapura were significantly higher than in the Polonnaruwa (except for Ca^{2+} ions). The spatial distribution of K^+, Na^+, Mg^{2+} and TDS were consistent, and F^-, NO_3^- and SO_4^{2-} were similar in their spatial distribution. Specifically, areas with TDS over 1000 mg/L are concentrated in Anuradhapura and less than 500 mg/L are concentrated in the southern part of Polonnaruwa, indicating that groundwater mineralization is higher in Anuradhapura than in Polonnaruwa. The hardness of groundwater in the NCP is high, with "very hard groundwater" in western and eastern Anuradhapura and "hard groundwater" or "Moderate hard groundwater" in the rest of the NCP. In contrast, the areas of both total alkalinity and Mg^{2+} were concentrated in Anuradhapura. In addition to the above ions, the higher concentrations of F^-, Cl^- and SO_4^{2-} are also concentrated in Anuradhapura, while the NO_3^- maximum are more scattered. Previous studies have shown that the fluoride concentration in the groundwater in the dry zone of the NCP exceeds the standard (SLS614-2013, $F^- < 1$ mg/L) (Dharmaratne, 2015; Young et al., 2011). Combined with the land use types in Fig. 2, areas with high ion concentrations in groundwater are mainly found in built-up land and cropland land, while in forest land, where there are more nature reserves, ion concentrations in groundwater are relatively low, indicating that anthropogenic factors have a greater influence on the major ions in groundwater. In addition, the main land-use type in the NCP is cropland land, with cropland land accounting for 49.5% of the total land area in the NCP. Studies have shown that groundwater is one of the main sources of irrigation water for agriculture (Athukorala et al., 2017; Villholth and Rajasooriyar, 2010), therefore, agricultural activities may have a great impact on the groundwater in the NCP. It is worthy noting that the distribution of Ca^{2+} concentration is opposite to the concentration of other cations, with higher concentrations occurring at Polonnaruwa and lower Ca^{2+} concentrations in the Anuradhapura area, the reasons for which need further analysis.

As shown in Table S6, the main ions of the dug wells, and tube wells were selected for correlation analysis. The TDS of

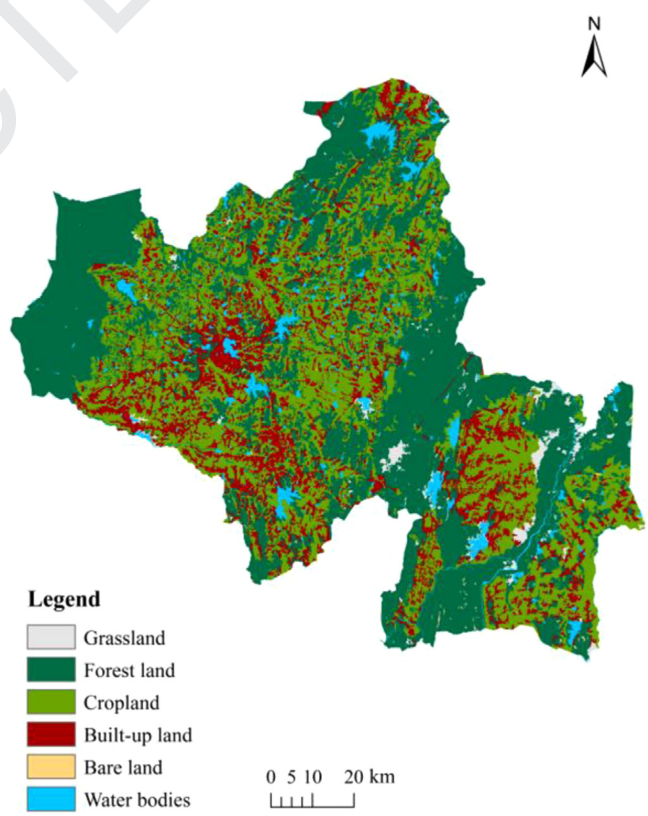

Fig. 2 – Land use spatial distributions map of NCP.

dug well groundwater was significantly correlated with K^+, Ca^{2+}, Na^+, SO_4^{2-}, Cl^- and HCO_3^- ($p < 0.01$), suggesting that they are the main ions resulting from the increase of salinity in shallow weathering aquifer groundwater. K^+, Na^+, Ca^{2+}, Cl^- and HCO_3^- have a strong positive correlation, which indicates that these chemical components may have the same source. TDS of tube well water was only significantly corre-

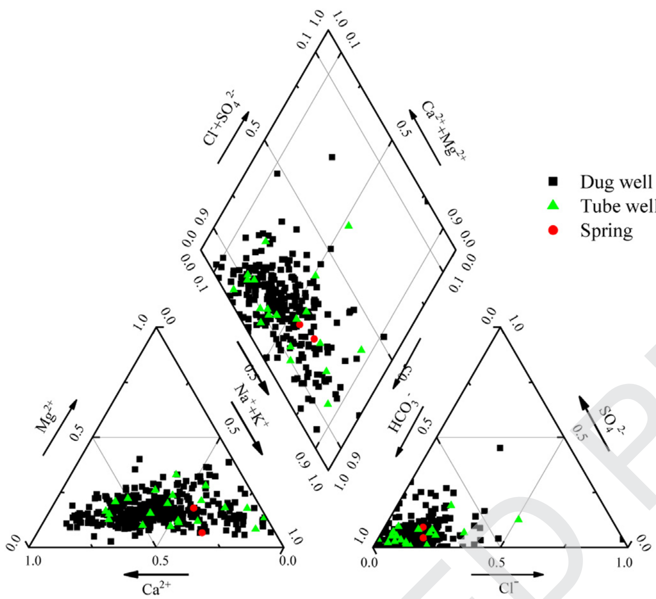

Fig. 3 – Piper diagram of groundwater samples in NCP.

lated with Ca^{2+} ($p < 0.05$), which reflects that its mineralization was mainly caused by Ca^{2+}.

2.2. *Factors influencing of hydrochemical characteristics*

The hydrochemical data were plotted in a Piper diagram (Piper, 1944), which provided an excellent method to classify the groundwater types from the different aquifers (Fig. 3).

As shown in Fig 3, both types of groundwater were predominantly Ca·Mg-HCO_3 and Ca·Na-HCO_3 types, and Spring water is Na·K-HCO_3 type. The main sources of dissolved minerals in groundwater can be shown using the Gibbs diagram (Gibbs, 1970), and it is a widely used method to describe the source of chemical components in water, which includes three sources: rock weathering, evaporation-crystallization, and atmospheric precipitation. As shown in Fig. 4**a** and **b**, the dug well and tube well groundwater samples are located between the rock weathering and evaporation-crystallization. Atmospheric precipitation did not affect the chemical composition of groundwater, which means that rock weathering and evaporative crystallization are the main sources of chemical components of these two types of groundwater sources, this is in line with previous studies (Abeywickarama et al., 2016). Spring $Na^+/(Na^++Ca^{2+})$ ratios greater than 0.5 and relatively low TDS suggest that spring groundwater chemistry is more complex than that of dug wells and tube wells, and is influenced by rock weathering, atmospheric precipitation and human activity.

HCO_3^-, Na^+ and Ca^{2+} are the main anions and cations in groundwater in the NCP and are mainly influenced by rock weathering and evaporation-crystallization. However, as the ion molar ratios of mineral dissolution products are different in different types of rocks, their sources can be resolved using the relevant ion ratio relationship (Fengjiao et al., 2017; Wei et al., 2020). As shown in Fig. 4**c** and **d**, the endmember diagram method shows that the two kinds of groundwater and spring were mainly affected by the dissolution of silicate weathering and evaporated rock salt (Li et al., 2020; Mukherjee and Fryar, 2008). $(Na^++K^+)/Cl^-$ can be used to indicate the dissolution of rock salts and silicates in groundwater (An et al., 2012). The (Na^++K^+/Cl^-) ratio in natural waters is approximately 1. Most of the dug wells in the study area, as well as all of the tube well water sample points, are distributed above the 1:1 line (Fig. 4**e**), and the concentration of Na^++K^+ is essentially greater than the Cl^- concentration, indicating that groundwater dissolves other silicate minerals containing Na^+ and K^+ as it flows through the aquifer. (Na^++K^+)-Cl^- can be used to indicate whether there is an increase or decrease in Na^+ except for dissolution of the rock salt, and $(Ca^{2+}+Mg^{2+})$-$(SO_4{}^{2+}+HCO_3{}^-)$ indicates whether there is an increase or decrease in Ca^{2+} and Mg^{2+} relative to the dissolution of the carbonate rock, when the ratio $[(Na^++K^+)-Cl^-]/[(Ca^{2+}+Mg^{2+})-(SO_4{}^{2-}+HCO_3{}^-)]$ is close to -1, indicated the presence of alternating cation adsorption. As shown in Fig. 4**f**, the dug well, tube well and spring samples were mostly around the 1:1 line in the fourth quadrant of the coordinates, suggesting that alternate cation adsorption exists to some degrees in both types of groundwater, mainly in the form of Ca^{2+} release and Na^+ adsorption(An et al., 2012). The effect of the ion exchange process on the mineralization of water can be illustrated using the Chlor-alkali index (CAII and CAI II, equations 12 and 13), and negative CAI values indicate that Na^+ and K^+ in the rock exchange ions with Ca^{2+} and Mg^{2+} in the water (Zhang et al., 2021). It can be seen that most of the tube well water, dug well water and spring water samples have negative CAI values, and a small number of dug well and tube well water samples have positive CAI (Fig. 4**g**), indicating that ion exchange between Na^+ and K^+ in the rocks and Ca^{2+} and Mg^{2+} in the water dominates, while the smaller the CAI value, the greater the degree of alternate ion sorption. Therefore, the overall intensity of alternate cation sorption in the study area shows that dug well groundwater > tube well groundwater > spring water, and the associated possible reaction equations are shown

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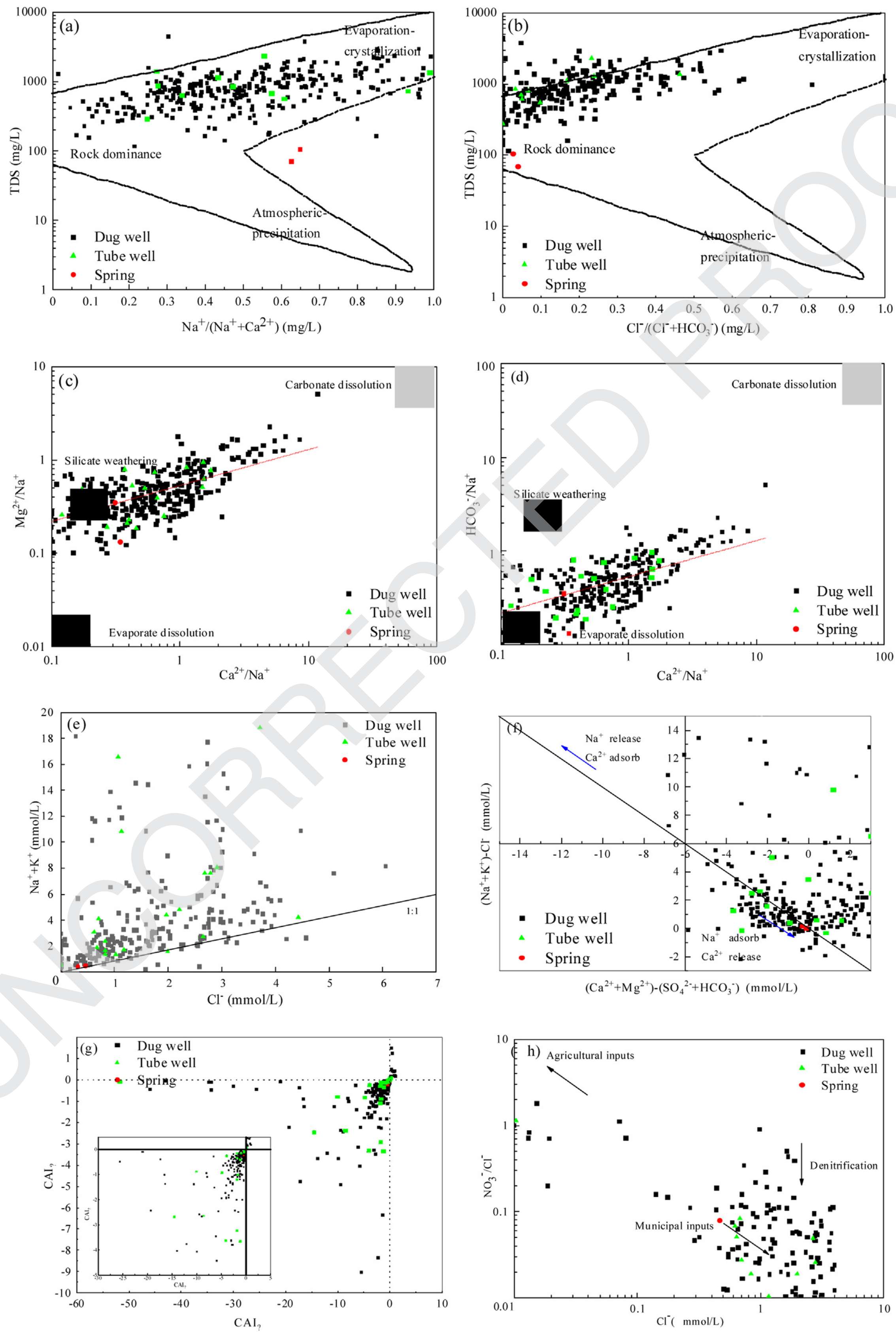

Fig. 4 – Relationship between the rate of the main ions of water samples. (a, b: Gibbs diagram; c, d: Endmember diagram; e: Ion ratio analysis of Na^++K^+/Cl^-; f: Ion ratio analysis of (Na^++K^+)-Cl^-/(Ca^{2+}+Mg^{2+}) - (SO_4^{2-}+HCO_3^-); g: Chloro-Alkali Indices; h: Variation of NO_3^-/Cl^- molar ratio with Cl^- molar concentration).

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in Eq. 14.

$$CAI_I = \frac{(Cl^- - (Na^+ + K^+))}{Cl^-} \quad (12)$$

$$CAI = \frac{(Cl^- - (Na^+ + K^+))}{\left(HCO_3^- + SO_4^{2-} + NO_3^-\right)} \quad (13)$$

$$2Na^+Rock + Ca^{2+}Water \rightarrow 2Na^+Water + Ca^{2+}(Rock) \quad (14)$$

Typically, NO_3^- ions are not produced by rock weathering processes and potential sources of NO_3^- and Cl^- in groundwater include major atmospheric rainfall, natural sources (dissolution of minerals), agrochemicals (potash or potassium chloride), animal manure, septic tank effluent and road salt (Bastani and Harter, 2019). As shown in Table 1, the concentration of NO_3^- in dug well water and the tube well water were between 0.02-54.44 mg/L and 0.26-8.07 mg/L, respectively, and the coefficient of variation were 1.7 and 1.09 respectively, belonging to strong variation and indicated that many factors are causing the existence of NO_3^- in groundwater. Fig. 4**h** shows that NO_3^- ions in groundwater are mainly of anthropogenic origin (i.e. agricultural activities and municipal inputs) (Liu et al., 2006). This is mainly due to the predominance of rice cultivation in agricultural activities in rural areas of the NCP, and the lack of effective treatment of domestic sewage. As a result, the direct discharge of domestic sewage and the fertilizers and pesticides contribute to the presence of NO_3^- in groundwater. The spatial distribution of NO_3^- in Fig. S4 is consistent with the distribution of Built-up land and cropland land, which supports the result that groundwater is influenced by human activities.

The results of the ion ratio analysis indicate that groundwater is mainly influenced by the dissolution of silicate rock salts and evaporite salts (Fig. 4**c** and **d**). Na^+ and K^+ exceeded Cl^- in 79.10% of the dug well and 80.95% of the tube well groundwater samples (Fig. 4**e**), suggesting the weathering of silicate rocks such as sodic and potassium feldspar results in elevated Na^+ and K^+ ion concentrations. In addition, the surface of rock and soil particles is negatively charged, and they can adsorb cations. Under certain conditions, some cations can be adsorbed by rock particles, and some of them can be released and re-transferring them to groundwater (Mondal, 1973; SHAINBERG et al., 1988). The adsorption capacity of different cations on rock surfaces is different, and the higher ion valency and smaller radius increase competitive adsorption ability. According to the adsorption ability, the order is as follows (Goren et al., 2011):

$$H^+ > Fe^{3+} > Al^{3+} > Ca^{2+} > Mg^{2+} > K^+ > Na^+$$

Therefore, the opposite distribution of Ca^{2+} and other cation concentrations was mainly due to the presence of alternating cation sorption (Fig. S4). In contrast, the results in Fig. 4**f** and **g** provide further evidence that ion exchange processes may be a factor influencing the chemical composition of groundwater. This is also supported by the K^+ coefficient of variation of 5.23 and the significant negative correlation between Ca^{2+} and K^+ ($p < 0.01$) for the dug well water in Table 1 and Table S6. In a word, the main hydrochemical ions in groundwater are influenced by the dissolution of silicate and evaporite salts, alternating cation adsorption and anthropogenic factors.

2.3. *Principal component analysis (PCA) of hydrochemical formation*

For the principal component analysis, four principal component factors with eigenvalues greater than 1 were selected for analysis. The maximum variance method was used to rotate the component matrix to obtain the rotation factor load matrix (Table. 3). The cumulative variance contribution rate of the four principal components of the excavated groundwater was 67.85%, and the cumulative variance contribution rate of the tube well groundwater was 78.04%. According to the factor load value, the factors were divided into three categories: "weak (0.3-0.5)", "medium (0.5-0.75)" and "strong (> 0.75)". As the load matrix of rotation factor shows, in dug well, PC1 replaces TDS, Na^+ and Mg^{2+} in the original data, with the dissolution of silicate minerals and salt rocks causing elevated Mg^{2+} and Na^+ concentrations in groundwater. In tube well, PC1 replaces Na^+ and SO_4^{2-} in the original data, indicated that in addition to the dissolution of silicate minerals, the dissolution of sulfur-containing minerals also causes the increase of SO_4^{2-}. The principal component factors PC2, PC3 and PC4 of the dug well groundwater mainly replaced the Ca^{2+}, F^- and NO_3^- in the original data, while the tube well groundwater PC2 replaces the Ca^{2+} and NO_3^- in the original data, while PC3 and PC4 represent F^- and HCO_3^- respectively, which means that except for the dissolution of silicate minerals, both groundwaters are polluted by nitrate. PC3 was considered to be the effect of the dissolution of fluorine-containing minerals in groundwater, indicating that both groundwater had the effect of fluoride. To sum up, rock weathering and evaporative crystallization have resulted in the enrichment of ions in groundwater, while some salts with weak adsorption capacity (such as sodium salt) have been separated out, resulting in the relative increase of Na^+ concentration in groundwater and occupying the main cation position. HCO_3^- is the main anion in the NCP groundwater, and finally, Ca·Mg-HCO_3 and Ca·Na-HCO_3 types of groundwater were formed.

2.4. *Water stability indices map*

Numerous literature have been reported the application of water stability indices in scaling and corrosion potential of the drinking water pipelines (Li et al., 2016; Tong et al., 2019;Tan et al., 2020). Therefore, the stability index of water quality of calculation (Fig. 5, Fig. S5 and Fig. S6) could help better determine the chemical integrity of groundwater. The Langelier Saturation Index (LSI) results show that most of the groundwater is under saturated, indicating that there is a corrosion potential. The water samples with LSI>0 account for 47.04% and 23.81% in the dug well and tube well respectively, while the water samples with LSI<0 account for 52.96% and 76.19% respectively. As clearly shown in Fig. 5**a**, the groundwater with scaling potential mainly existed in Polonnaruwa, and the groundwater with corrosion potential mainly existed in Anuradhapura. However, this index cannot determine the node where the corrosion tendency of water quality appears.

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Table 3 – Rotated factor loading matrix.

Parameters	PC1		PC2		PC3		PC4	
Well	Dug well	Tube well	Dug well	Tube well	Dug well	Tube well	Dug well	Tube well
TDS	0.749	0.382	0.359	0.721				
HCO_3^-	0.518		0.381		0.512			0.959
K^+	0.316		-0.443	0.495		0.707	0.389	
Ca^{2+}			0.863	0.893				
Mg^{2+}	0.818	0.452				0.682		0.3
Na^+	0.861	0.804						
F^-					0.947	-0.794		
Cl^-	0.593	0.692	0.402					
NO_3^-				0.854			0.883	
SO_4^{2-}	0.584	0.914						
Variance contribution /%	30.498	24.452	14.284	23.611	12.125	17.78	10.947	12.194
Cumulative variance contribution /%	30.498	24.452	44.782	48.063	56.907	65.843	67.855	78.037

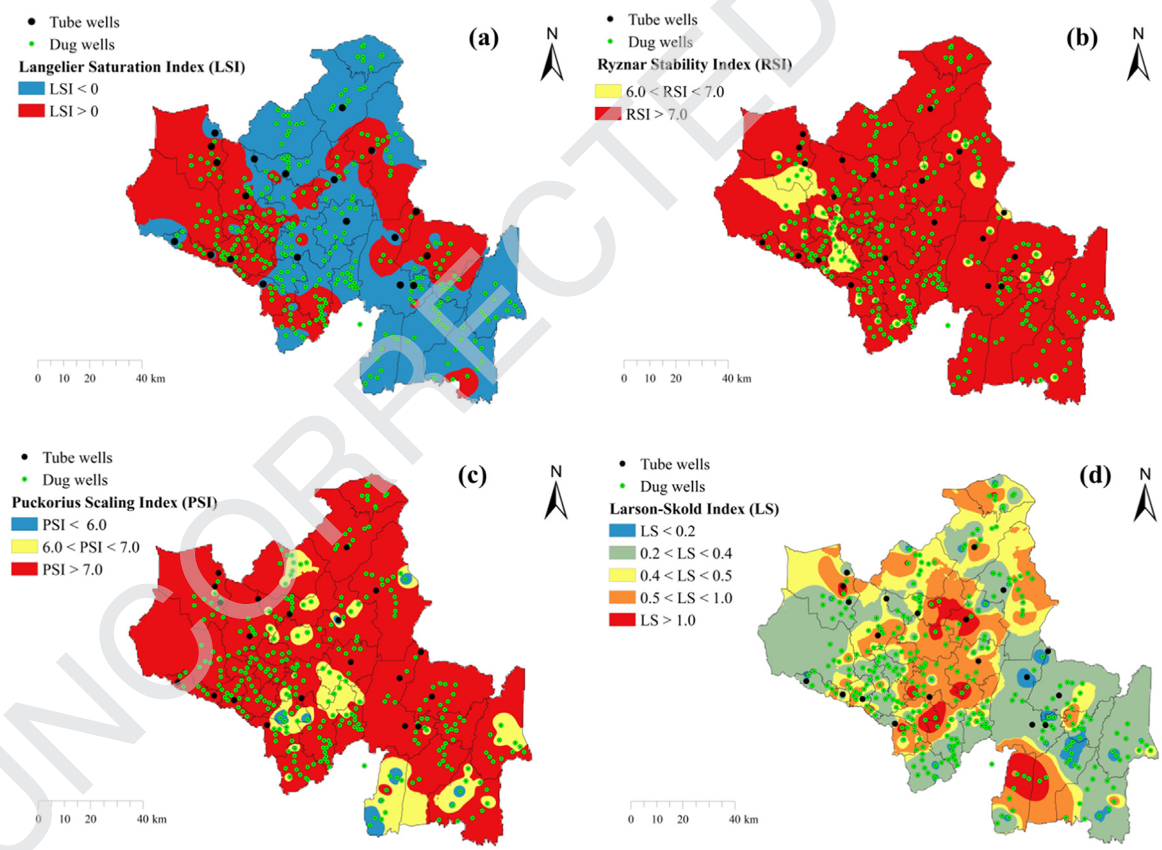

Fig. 5 – Spatial temporal distribution of the indices (a:LSI; b:RSI; c:PSI & d:LS).

Therefore, researchers have proposed the Ryznar Stability Index (RSI) based on experiments. The results show that most groundwater in the NCP has corrosion tendency. The groundwater with RSI value <6 (scaling potential) only exists in the dug well, accounting for 2.30%. The groundwater samples with 6≤RSI≤7 (Equilibrium) accounted for 22.04% and 9.52% of the groundwater in the dug well and tube well, respectively. The groundwater samples with RSI>7 accounted for 75.66% and 90.48% respectively in the dug well and tube well. However, RSI, as an empirical index, ignored the buffering capacity of water. Therefore, after further optimization based on RSI, the Puckorius Scaling Index (PSI) was proposed. This index uses equilibrium pH instead of the measured pH to account for the buffering effects, quantifying the relationship between the saturation state of water and scaling, which can be closer to the actual situation. The results of PSI was in concordance with RSI, indicating that most groundwater has corrosion potential. Among them, the groundwater with scaling potential (PSI<6) accounts for 8.88% and 4.76% respectively in the dug well and tube well, the groundwater in equilibrium state

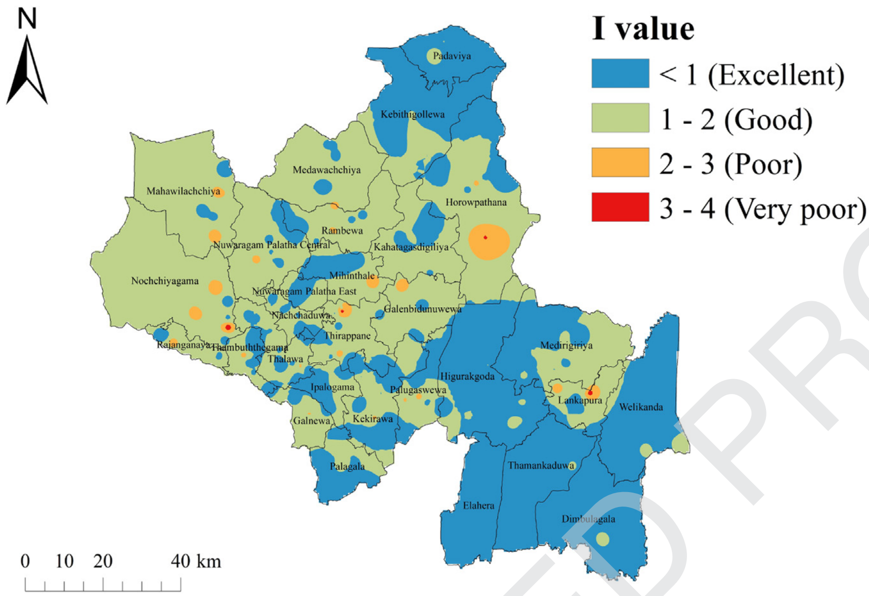

Fig. 6 – Water quality map (Modified Nemerow Pollution Index).

(6≤ PSI ≤7) accounts for 22.04% and 9.52% respectively, while the groundwater with corrosion potential (PSI>7) accounts for 74.34% and 85.72% respectively. Compared with LSI, PSI index shows that groundwater with corrosion tendency mainly in Anuradhapura, and in the south of Polonnaruwa with equilibrium, which deviating from LSI in-terpretations.

LS index results show that the metal corrosion tendency of Anuradhapura groundwater was higher than that of Polonnaruwa. Among them, no metal corrosion tendency (LS<0.2) is 22.4% and 33.33% in the dug well and tube well, respectively, which are mainly distributed in Polonnaruwa, including Thamankaduwa, Welikanda, Dimbulagala and Higurakagoda. The groundwater with light metal tendency (0.2≤LS<0.4) was 36.80% and 33.33% respectively, mainly in Anuradhapura, including Nochchiyagama, Thalawa, Galnewa, Palagala, Kekirawa, Palugaswawa and Padaviya, as well as Medirgirya in Polonnaruwa. The groundwater with low metal tendency (0.4≤LS<0.5) was 9.20% and 4.76% respectively, which were mainly distributed in the north of Anuradhapura, including Mahawilachchiya, Medawachchiya, Kebithigollewa, Horowpathana and Nuwaragam Palatha Central. The groundwater with mid metal tendency (0.5≤LS<1.0) was 24.00% and 14.29% respectively, which were mainly located in Rambewa, Mihinthale, Thirappane and Ipalogama in Anuradhapura, and Elahera in Polonnaruwa. The groundwater with high metal tendency (LS≥1) was 6.00% and 14.29% respectively, which only existed in Mihinthale. It should be noted that the distribution of groundwater with no metal and light metal tendency was consistent with that of forest land in land use types, and mid metal and high metal tendency was consistent with that of built-up land in land use types, its further indicated that the chemical components in groundwater are affected by human factors.

Therefore, most of the groundwater in the NCP with corrosion potential. However, in the personnel gathering area, the metal corrosion capacity of groundwater was relatively high, which should be considered in the process of water supply.

2.5. *Water quality assessment map*

As listed in Table 2, the main hydrochemistry ions in both types of groundwater in the NCP are exceeded the maximum values of Sri Lanka drinking water standards to some degree. The use of three methods such as the average value synthesis pollution exponential, the modified Nemerow Pollution Index and the water quality index (WQI) can provide a more comprehensive understanding of groundwater quality in the NCP and explain the interactions between different water quality parameters. As is shown in Fig. 6 and Fig. S7, the water quality of Polonnaruwa was better than that of Anuradhapura under different water quality assessment methods. Districts of poor water quality were mainly distributed in Rajanganaya, Rambewa and Horowpathana in the Anuradhapura, and in Lankapura of Polonnaruwa. Based on the results of the average value synthesis pollution exponential, groundwater in the NCP can be classified into five types (“Excellent”, “Good”, “Poor”, “Very poor” and “Unsuitable”), of which 10.84% are of “Excellent” quality, while the others have different degrees of pollution, “Good”, “Poor”, “Very poor” and “Unsuitable” groundwater quality accounted for 43.67%, 37.95%, 6.33% and 1.20%, respectively. The results of the modified Nemerow pollution index show that there were four types of groundwater quality (“Excellent”, “Good”, “Poor” and “Very poor”) in the NCP, accounting for 53.61%, 37.65%, 7.53% and 1.20%, respectively. The WQI results of the water quality index method show that none of the WQI values exceeds the limit of 300, so there are three main types (“Excellent”, “Good” and “Poor”). Among them, “Excellent” groundwater quality occupies the main position (37.12 %), others are “Good” and “Poor” groundwater quality (Fig. S8). A comparison of the results of these three

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methods shows that in the average value synthesis pollution exponential, the larger the exceedance of the concentration of a single indicator, the greater its impact and the more pessimistic the evaluation results tend to be. And the WQI results, although the weight values of different pollutants are different, when the key indicators (toxicological indicators) exist as evaluation factors cause the WQI values to be lowered, making the evaluation results too optimistic and contrary to the actual situation, and the WQI will have a certain degree of unreasonableness in practical application. Therefore, in the evaluation of water quality, both the maximum pollution value and the influence of key indicators need to be taken into account. Modified the Nemerow pollution index method weakens the influence of the maximum value and incorporating the coefficient of variation makes its evaluation results more objective and accurate compared to the average value synthesis pollution exponential and WQI. At the same time, in the process of water quality evaluation, different evaluation methods have their advantages and disadvantages, should not overly magnify the advantages and disadvantages of a certain method, should be combined with the actual local conditions, choose the appropriate groundwater quality evaluation methods, that the water quality evaluation results are more accurate, and thus can be a comprehensive and realistic reflection of the study area groundwater quality conditions.

Therefore, when the water quality is lower than the “good” grade, the groundwater needs to be treated before drinking, which conforms to the requirements of the “water purification plan” in the WHO report (WHO, 2012). In summary, when groundwater quality is below the “good” grade, it must be treated appropriately before it can be used as drinking water.

3. Conclusion

This study provides a comprehensive analysis of the spatial distribution of the main ions in groundwater in the NCP during the wet season, as well as studying the groundwater chemical characteristics and influencing factors based on PCA and correlation analysis. Major conclusion are made as the following:

a) Over 95% of groundwater is high hardness water, anions in both dug well water and tube well water is dominated by HCO_3^-, accounting for over 70% of the total anion concentration, cations are dominated by Na^+ and Ca^{2+}, accounting for over 75% of the total cation concentration, with the main ion concentrations showing $HCO_3^- > Na^+ > Ca^{2+} > Cl^- > Mg^{2+}$.
b) Rock weathering and evaporative crystallization affect the hydrochemical ions of the two kinds of groundwater, and both groundwater hydrochemical types are predominantly Ca·Mg-HCO_3 and Ca·Na-HCO_3 types. The main HCO_3^-, Na^+, Ca^{2+} and Mg^{2+} ions in both groundwaters originate from the silicate and evaporative rock salt dissolution and are affected by alternate cation adsorption, with the intensity of adsorption generally showing dug well groundwater > tube well groundwater > spring.
c) The water quality stability indices (LSI, RSI, PSI and LS) shows that most of the groundwater in the NCP has corrosion potential, and the metal corrosion capacity of groundwater was high in the personnel gathering area.
d) Based on the different water quality assessments, water quality of Polonnaruwa is generally better than that of the Anuradhapura, with poorer areas being found in Rajanganaya, Rambewa and Horowpathana in the Anuradhapura, and Lankapura in Polonnaruwa, which must be treated appropriately before it can be used as drinking water.

Declaration of competing interest

All authors have approved to submit to your journal. All the authors claim that none of the materials in the manuscript has been published or is under consideration for publication elsewhere, and all the authors listed have approved the manuscript is enclosed. The authors declare no conflict of interest.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2023.05.034.

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