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Geochemistry of Groundwater in the Semi-Arid Crystalline Terrain of Sri Lanka and Its Health Implications among Agricultural Communities

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Abstract: Chronic kidney disease with uncertain etiology (CKDu) is an emerging health problem in Sri Lanka, particularly among the dry-zone farming communities that use groundwater for drinking. We investigated the quality of groundwater in an area where both high- and low-prevalence clusters of CKDu have been recorded. Eighty-four groundwater and five surface water samples, covering the selected region, were collected and analyzed for both major anions and cations. The groundwater in the region is mainly of the Ca-Mg-HCO_3 type, probably due to the long residence time in fractured hard rock aquifers in this region. Irrespective of the CKDu prevalence, over 50% of samples exceeded the recommended limits for EC/TDS, alkalinity, hardness, and Mg^{2+} content in groundwater. Water hardness in CKDu clusters was dominated by Mg^{2+}. High fluoride content up to 4.0 mg/L was also found in most groundwater samples from the region. The water quality index (WQI) values indicated that 42% of the groundwater samples in regions with no or low CKDu prevalence and 49% of the samples in regions with high prevalence were poor in quality. The spatial distribution of WQI and fluoride concentration overlapped, indicating the direct influence of fluoride on the groundwater quality in the study region. In addition, regions with higher WQI values overlapped with the CKDu hotspots, indicating the direct impact of groundwater quality on the disease prevalence in the studied river basin. The WQI can be used to effectively demarcate areas with possible groundwater-related health effects in the dry-zone regions of Sri Lanka.

Keywords: CKDu; water quality index; water hardness; fluoride; groundwater; Yan Oya river basin

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Citation: Udeshani, W.A.C.; Koralegedara, N.H.; Gunatilake, S.K.; Li, S.-L.; Zhu, X.; Chandrajith, R. Geochemistry of Groundwater in the Semi-Arid Crystalline Terrain of Sri Lanka and Its Health Implications among Agricultural Communities. *Water* **2022**, *14*, 3241. https://doi.org/10.3390/w14203241

Academic Editor: Guy Howard

Received: 19 September 2022
Accepted: 10 October 2022
Published: 14 October 2022

1. Introduction

Water is one of the most important substances on Earth, since every living organism relies on water for survival. Around the world, groundwater has become the only reliable resource if surface water is not available, sufficient, convenient, or feasible for consumption [1]. Particularly in arid and semi-arid regions, groundwater is indispensable due to the serious shortage of surface water and precipitation [2]. The quality of water resources is as important as their quantity, determining their suitability for various purposes. The hydrogeochemical properties of groundwater determine the quality of groundwater, which is highly influenced by geological formations, sub-surface geochemical processes, the quality of recharged water, and anthropogenic activities [3–5]. In the recent past, the demand for water consumption has increased drastically all over the world due to the rapid growth of populations, urbanization, and the development of agricultural and industrial activities [6]. With the increased demand, significant pressure has been put on groundwater resources, leading to overexploitation, which has caused the depletion and deterioration of water quality, thereby making it unfit for domestic use. The quality of drinking water is a vital factor, since the deterioration of water quality threatens public health and thereby potentially severely affects the social and economic development of a country [7,8].

Over the past three decades, chronic kidney disease of uncertain origin (CKDu) has become one of the most serious national health issues in Sri Lanka [9–11]. The disease is common among male paddy farming communities in the dry zone in the age group of 40 to 60 years [9,12]. Recent epidemiological investigations on CKDu showed that serum creatinine is higher among the affected population compared to non-affected communities with the same socioeconomic background [13]. The regional clustering of the disease is an important feature of CKDu [11]. Thus, it seems likely that CKDu in Sri Lanka is an environmentally induced problem. However, even within the endemic hotspots, the disease shows different phases of dispersion that can be categorized as high- (over 30%), moderate- (20–30%), and low- (less than 10%) prevalence clusters [13].

Similar kinds of kidney diseases with undetermined origins have been reported not only in Sri Lanka but also in India, Egypt, China, Nepal, and some Central American countries such as Nicaragua and El Salvador, particularly among farming communities and manual laborers who work and live in hot, arid climatic regions [14]. Due to its spatial distribution, CKDu is widely believed to be associated with some kind of unknown toxin, infectious agents, dehydration, or possibly a synergetic effect of several factors [14]. However, the factors that contribute to CKDu are yet to be conclusively identified, unlike the more traditional factors linked to known-origin chronic kidney disease (CKD), such as hypertension and diabetes.

In Sri Lanka, CKDu is more prevalent in the rural dry-zone regions where the majority of people use groundwater as their primary source of drinking water [11]. Although the root cause of the disease is still uncertain, a direct or indirect influence of drinking groundwater has been highly suspected either for the etiology or the progression of the disease [15,16]. Therefore, many studies have been carried out in CKDu regions searching for possible etiological factors related to drinking water quality and the disease. Through extensive water quality studies, high fluoride, high hardness [15–18], and heavy metals (Cd and As) [19–21] that exceed the international and local drinking water quality standards, as well as the application of agrochemicals such as glyphosates [22], have been identified as potential risk factors for CKDu. Some later studies do not support the involvement of heavy metals as an etiological factor for CKDu [23–26]. Another recent study in Sri Lanka showed that 68% of households with CKDu cases (n = 262) used groundwater for domestic purposes, and these cases indicated significantly lower ($p = 0.0184$) estimated glomerular filtration rates (-6.7 mL/min/1.73 m^2) compared to the population who did not use groundwater as their primary drinking water source [9]. Therefore, drinking groundwater is one of the most important risk factors for CKDu in the dry zone of Sri Lanka.

Another major characteristic of CKDu is the remarkable geographical distribution in the dry-zone region of Sri Lanka [11]. The disease appears in several isolated clusters in the dry-zone regions, particularly around the north-central region, where the annual rainfall is about 1000 mm/a (Figure 1a). However, the disease has not been recorded in the wet zone of the country, where the annual rainfall is higher than 2500 mm/a.

Groundwater quality and its suitability for drinking can be assessed effectively using the water quality index (WQI), which is widely employed as a water quality monitoring tool [27]. It represents the composite influence of water quality parameters on the overall figures rather than the individual parameters that mostly compare with available drinking-water quality standards. The WQI summarizes solute composition into meaningful, simple terms that make it easy to report, analyze, and compare. Therefore, this study was carried out by assessing the hydrogeochemical composition of the groundwater in a region where both high-prevalence areas of CKDu (hotspots) and areas with low or no CKDu (cold spots) have been reported.

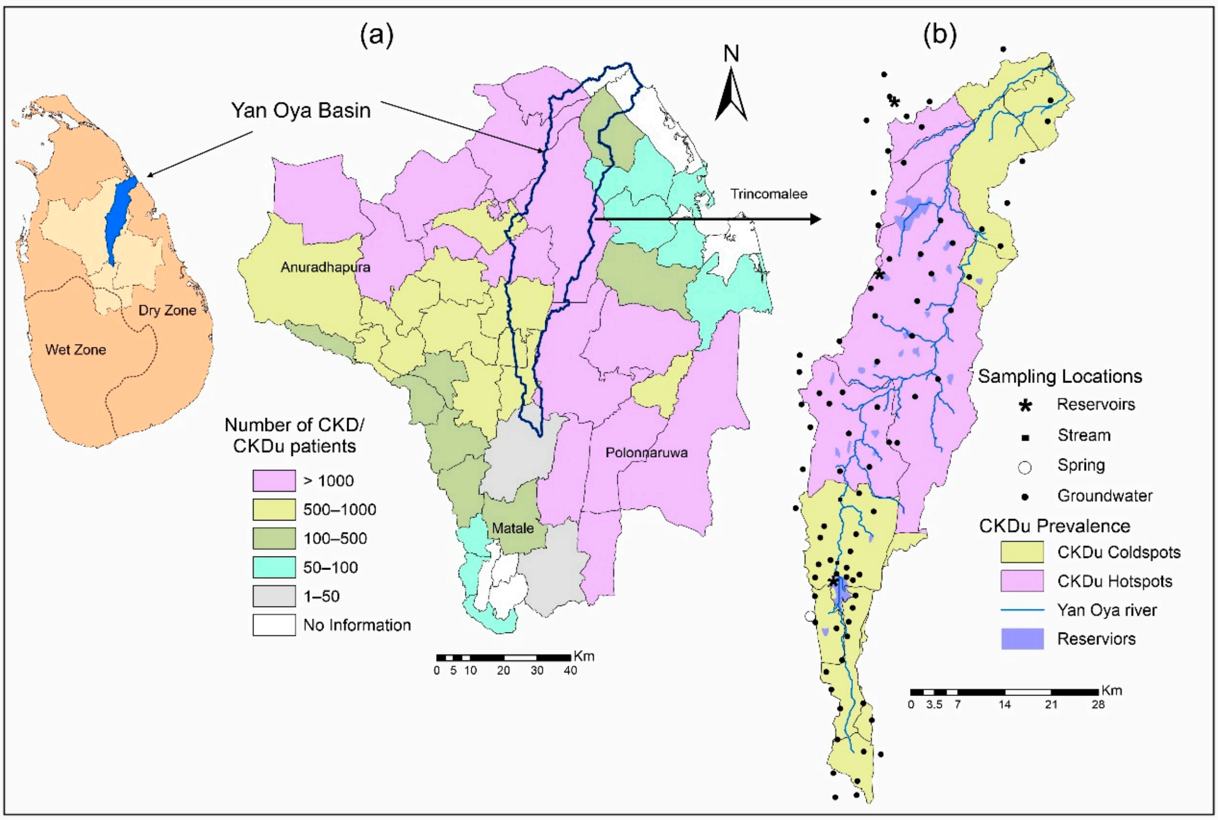

Figure 1. (**a**) CKD/CKDu prevalence in the north-central region of Sri Lanka (modified after Ranasinghe et al. 2019 [12]) and (**b**) sampling locations in the Yan Oya river basin.

2. Study Region

The Yan Oya river is the fifth longest river (142 km) in Sri Lanka, flowing towards the north-eastern region of the country with a total catchment area of 1538 km^2 that is entirely located in the dry zone of Sri Lanka (Figure 1a). Nearly one sixth of the basin area (14,281 ha) is under paddy cultivation, while almost 64% is covered with natural forests and grasslands. One tenth of the basin area is occupied by homesteads, and nearly 8% of the area is covered by water bodies, especially man-made reservoirs that irrigate the paddy lands in the catchment [28]. Geologically, Precambrian high-grade metamorphic rocks underlie the studied river basin. This crystalline basement complex is well-known for its very limited availability of groundwater. The groundwater in the region is made up of shallow "regolith" aquifers and deeper "fracture-zone" aquifers, which occur in the underlying fracture zone at depths of more than 30 m [29]. The thickness of the regolith aquifers in the region is variable but is no more than 10 m. The water-holding and transmissivity capabilities of regolith aquifers are limited compared to fractured crystalline aquifers [30]. The most important feature of this basin is the alternative occurrence of CKDu hotspots (high-prevalence areas) and cold spots (low- or no-CKDu-prevalence areas). Therefore, the region is ideal for demarcating and identifying the effect of hydrogeochemistry on the etiology of the disease. Among the 12 administrative divisions in the river basin, Padaviya, Kebitigollewa, Horowupothana, and Kahatagasdigiliya were identified as CKDu hotspots with the highest number of CKDu patients according to the reports from the Ministry of Health, Sri Lanka.

3. Materials and Methods

3.1. Sampling and Analyses

The environmental sampling program was carried out during the post-monsoon season (February to April) of 2021, covering the entire Yan Oya river basin. Samples were also collected from areas adjacent to the river basin, since the disease was not restricted to the river basin. Eighty-four groundwater samples, one spring water sample, and five surface water samples were collected for the geochemical investigation (Figure 1b). Most shallow wells were protected from stormwater runoffs that occur during the monsoon period. The depth of the shallow wells ranged from 2.8 to 12 m, while deep wells showed depths around 20 to 30 m. Out of 84 groundwater samples, 39 samples were collected from CKDu hotspots, while the others were obtained from CKDu cold spots. The overflowing natural spring located in the Mahadiwulwewa area (Figure 1b), which supplies drinking water for over 10 nearby villages, was also sampled.

Samples were collected into high-density polyethylene bottles, which were soaked in 10% HNO_3 overnight, washed thoroughly with distilled water, and oven-dried. For cation analysis, samples were filtered through 0.45 µm cellulose acetate membrane filters and preserved by adding several drops of HNO_3 to lower the pH (<2). Filtered and non-acidified samples were collected for anion analysis. All the samples were immediately stored in a storage box with ice bags after sampling and transferred to the refrigerator within a few hours until laboratory analyses were performed.

Water temperature, pH, and electrical conductivity/total dissolved solids (EC/TDS) were measured in situ using a pre-calibrated multi-parameter kit (SANXIN Model SX751; Shaanix, China). The total alkalinity (TA) of the samples was determined as early as possible via the H_2SO_4 titrimetric method using an auto-titrator (OrionStar T-910; Thermo Fisher, Bremen, Germany), from which ±2% relative standard deviation was obtained for repeated measurements. Bicarbonate (HCO_3^-) concentrations in samples were then calculated using total alkalinity. Major anions, including nitrate (NO_3^-), phosphate (PO_4^{3-}), fluoride (F^-), and sulfate (SO_4^{2-}), were measured using a spectrophotometer (Hach DR-2400; Hach, Loveland, CO, USA) according to the standard procedures approved by the US EPA. The method detection limits of these measurements were 0.01, 0.2, 0.02, and 2.0 mg/L, respectively. Titrimetric methods were used to determine total hardness (TH) and chloride (Cl^-) content. Major cations (Ca^{2+}, Mg^{2+}, Na^+, and K^+) in water samples were measured by atomic absorption spectrophotometry (Varian 240FS; Mulgrave, Australia), for which replicate sample measurements showed a ±1% relative standard deviation. Analytical precision was determined by calculating the ion balance error using PhreeqC version 3 (USGS, Denver, CO, USA), and of all the investigated samples, 83% were within ±10% error, while 17% showed a higher ion balance error up to ±20%. The higher ion balance may have been due to unmeasured constituents such as organic matter and/or due to higher concentrations of fluoride, Fe, Sr, and Mn in the samples.

3.2. Data Analysis

The geochemical parameters were compared with the Sri Lanka standards for potable water quality (SLS 614:2013) [31] to identify whether the water samples were compatible with the standards. As most of the parameters rejected normality in the normality test, a non-parametric Mann–Whitney test was performed to identify the difference in water quality parameters between CKDu hot and cold spots. Significant differences were determined based on the p values. The Spearman correlation coefficient (r) was calculated to identify the correlations between major ions in groundwater. The major cationic (Ca^{2+}, Mg^{2+}, Na^+, and K^+) and anionic (CO_3^{2-}, HCO_3^-, Cl^-, and SO_4^{2-}) constitutions of groundwater samples, expressed in milli-equivalent units, were plotted on a Piper trilinear diagram [32] to identify the dominant groundwater types in the region. A Gibbs diagram [33] was used to determine the major mechanisms that were responsible for the dissolved chemical constituents in the groundwater.

In the present study, the water quality index (WQI) was calculated using the weighted arithmetic index method. Twelve major water quality parameters (pH, TDS, alkalinity, hardness, chloride, sulfate, nitrate, phosphate, fluoride, calcium, sodium, and magnesium) were considered for the calculation of the WQI based on the following equations:

$$WQI = \sum_{i=1}^{n} w_i q_i / \sum_{i=1}^{n} w_i \quad (1)$$

where w_i = unit weightage of i^{th} water quality parameter, q_i = quality rating value of the i^{th} parameter, and n is the number of water quality parameters considered for the calculation.

$$w_i = \frac{k}{v_s} \quad (2)$$

$$k = \frac{1}{\sum_{i=1}^{n} \frac{1}{v_s}} \quad (3)$$

$$q_i = \frac{(v_a - v_o)}{(v_s - v_o)} \times 100 \quad (4)$$

where k = proportionality constant, v_a = estimated concentration of the i^{th} parameter in the laboratory analysis, v_o = ideal value of the i^{th} parameter in pure water (v_o = 7 for pH and zero for all other parameters), and v_s = standard value recommended by Sri Lanka standards for potable water [31]. The water quality of groundwater samples was classified into five categories based on the computed WQI values (Table 1).

Table 1. Classification of groundwater quality according to the water quality index (WQI) values.

Class	WQI Value	Water Quality
1	<25	Excellent
2	26–50	Good
3	51–75	Poor
4	76–100	Very poor
5	>100	Unsuitable for drinking purposes

4. Result and Discussion

4.1. Variations in Hydrogeochemical Parameters

The descriptive statistics of the water quality parameters in both the cold spots and the hotspots and the results of the statistical comparison (Mann–Whitney test) of the variables between the two regions are presented in Table 2. In addition, each parameter of all the groundwater samples was compared with the maximum permissible levels (MPLs) described in the Sri Lanka standards for potable water (SLS 614:2013). The mean pH values complied with the permissible range for drinking (6.5–8.5) in both the cold spots (6.98) and the hotspots (7.07). Although no sample exceeded the maximum standard (8.5), seven groundwater samples did not meet the minimum standard for pH (6.5). In general, the groundwater in the study area could be categorized as "neutral" in nature. Alkalinity essentially becomes a measure of the buffering capacity of water for CO_3^{2-} and HCO_3^- ions and to some extent OH^- ions. The alkalinity based on the $CaCO_3$ content of the studied samples ranged from 33.0 to 713 mg/L (mean = 318 mg/L) for CKDu cold spots and 99.0 to 691 mg/L (359 mg/L) for hotspots. Except for one groundwater sample in a hotspot, all the others in both regions showed pH values below 8.3, indicating that the total alkalinity was mainly due to bicarbonate. In addition to the atmospheric contribution, the dissolution of carbonates and silicate minerals with carbonic acid accounted for the addition of HCO_3^- into the groundwater [34].

Table 2. Descriptive statistics of groundwater quality parameters in the Yan Oya river basin [EC—electrical conductivity; TDS—total dissolved solids; TA—total alkalinity; TH—total hardness; MPL—maximum permissible level recommended for drinking under the Sri Lanka standards for potable water (SLS 614: 2013)]. * WQI value for poor quality; ** % samples exceeding WQI value for poor quality; "-" indicates "not applicable".

Parameter	Unit	MPL (SLS 614:2013)	Cold Spots (n = 45)					Hotspots (n = 39)					Mann–Whitney U Test		
			Mean	Median	Min	Max	Samples Exceeding MPL (%)	Mean	Median	Min	Max	Samples Exceeding MPL (%)	*p*-Value	U Value	Z
Temp.	°C	-	29.3	28.8	27.2	32.2	-	30.2	30.2	27.5	33.3	-	0.003	549	−2.944
pH	-	8.5	6.98	7.10	6.12	7.62	0	7.07	7.07	6.38	8.42	0	0.84	855	−0.202
EC	µS/cm	750	833	720	167	2720	49	856	865	188	1875	56	0.667	829	−0.431
TDS	mg/L	500	725	626	145	2366	69	745	753	164	1631	69	0.667	829	−0.431
TA	mg/L	200	318	334	33	713	73	359	358	99	691	77	0.278	756	−1.09
TH	mg/L	250	302	296	104	615	60	321	308	100	632	69	0.427	789	−0.794
HCO_3^-	mg/L	-	387	407	40	868	-	436	436	120	841	-	0.286	758	−1.067
Cl^-	mg/L	250	107	72	26	518	9	96	86	18	302	5	0.986	880	0.018
SO_4^{2-}	mg/L	250	39	28	1	200	0	46	34	5	148	0	0.093	690	−1.678
F^-	mg/L	1	0.75	0.55	0.02	2.26	27	0.87	0.66	0.02	4	31	0.918	866	−0.103
NO_3^-	mg/L	50	2.68	0.88	0.44	24.8	0	3.66	1.77	0.44	14.62	0	0.284	759	−1.07
PO_4^{3-}	mg/L	2	0.32	0.26	0.02	0.84	0	0.36	0.26	0.09	3.54	3	0.76	912	0.305
Na^+	mg/L	200	64	48	5	341	2	66	50	11	216	3	0.441	791	−0.771
K^+	mg/L	-	2.67	1.32	0.32	16.4	-	2.71	0.94	0.18	34.29	-	0.137	1044	1.489
Ca^{2+}	mg/L	100	81	84	24	181	27	85	81	18	183	33	0.594	818	−0.534
Mg^{2+}	mg/L	30	33	31	5	105	51	37	36	6	123	62	0.358	775	−0.92
WQI	-	50 *	52	44	2	141	42 **	61 *	49	3	244	49 **	0.66	828	−0.439

Higher EC/TDS values were observed in the groundwater in the study area compared to the wet zone of Sri Lanka. As both of these parameters represent the dissolved constituents in the aqueous solution, their values are interconnected. The ions that leached into the groundwater via rock–water interactions may have been further concentrated due to the high evaporation that prevails in the dry climatic zone, providing a higher EC (and TDS) [17]. According to the mean concentrations, cations were in the order $Ca^{2+} > Na^+ > Mg^{2+} >> K^+$, while the anion contents were in the order $HCO_3^- > Cl^- > SO_4^{2-} >> NO_3^- > F^- > PO_4^{3-}$ in both regions, highlighting the dominance of HCO_3^- and Cl^- and the lower concentrations of NO_3^-, F^-, and PO_4^{3-}. The high concentrations of Ca^{2+}, Mg^{2+}, and Na^+ in the groundwater may have been associated with water–rock interactions and ion exchange processes.

On the other hand, the low concentrations of NO_3^- and PO_4^{3-} indicated the limited impact of anthropogenic and agrogenic pollution on the study area. Nitrate is the most common human-induced pollutant in groundwater [35,36]. The major source of nitrate in groundwater in Sri Lanka is the use of nitrogen fertilizers for paddy cultivation [37,38]. In addition, the improper disposal of domestic sewage, organic and animal waste, and septic tank leaching can account for nitrate in groundwater [39]. However, the possibility of contaminating deep groundwater from such sources is fairly low compared to both shallow groundwater and surface water [38]. Phosphate in groundwater may be due to local factors such as the discharge of agriculture runoff or the mixing of sewage with the shallow groundwater and leachates from landfill sites [40]. The heavy application of P-containing fertilizers such as single and triple superphosphates in paddy cultivation leads to the saturation of sorption sites of soils and therefore increases the risk of P loss to surface water or groundwater via runoff [37]. The increase in both nitrate and phosphate concentrations together can be directly correlated with fertilizer application in agricultural fields, in particular rice paddy cultivation. Nitrogen transformation in the vadose zone could enhance mineral dissolution and influence water quality [41]. The nitrate concentration of the samples was below 15 mg/L as NO_3^-, except for one sample that was collected from a CKDu hotspot region (24.8 mg/L as NO_3^-). Except for one sample (3.54 mg/L as orthophosphate), the phosphate concentration of all other samples was below 1.0 mg/L. It was assumed that after and during the rainy season, pollutants may have been subjected to downward leaching, thus contaminating the underlying aquifers. However, paddy farmers hardly use fertilizer during the rainy period, since most of the rice fields are being prepared for the next cultivation period. This may have been another reason for the low levels of nitrate and phosphate in the collected groundwater samples.

Chloride is also an important parameter that governs groundwater geochemistry. In CKDu cold spots and hotspots, the mean Cl^- concentrations were 107 and 96.0 mg/L, respectively. Generally, most natural water contains higher concentrations of Cl^- as compared to SO_4^{2-} [40]. Halite dissolution is the most common natural process that introduces Na^+ and Cl^- into groundwater. Nevertheless, halite is not a naturally occurring mineral in crystalline rocks in the study terrain. The sources of both chloride and nitrate are generally anthropogenic inputs, most likely the disposal of domestic wastewater. In addition, aquifers adjacent to the coastal belt show high chloride contents, due to saltwater intrusion. Other than that, highly soluble chloride can be leached from agricultural activities or through mixing groundwater with irrigated water [39,42].

The F^- content in the CKDu cold spots ranged from 0.02 to 2.26 mg/L, with a mean value of 0.75 mg/L, while it ranged from 0.02 to 4.00 mg/L (mean = 0.87 mg/L) in the disease hotspots. High-grade rocks such as charnockitic, granitic gneisses, and hornblende-biotitic gneisses are abundant with fluoride-bearing minerals such as mica, pyroxene, fluorite, tourmaline, topaz, sphene, and apatite [8]. Excessive levels of total hardness in the groundwater were apparent over the entire Yan Oya basin. As to the hardness classification of the World Health Organization (WHO) [43], 82% of the samples in the basin belonged to the "very hard water" category (>180 mg/L), while 14% of the samples were categorized under "hard water" (120–180 mg/L). The presence of calcium- and magnesium-bearing minerals in the aquifer materials caused hardness in the groundwater. The mean values of all the parameters in the CKDu hotspots were higher than those of the cold spots, except for Cl^- content.

4.2. Suitability of Water for Drinking

As to the comparison of the mean concentrations of the water quality parameters with the MPLs of SLS 614, more than 50% of the samples in both CKDu and non-CKDu regions were found to exceed their MPLs for EC/TDS, alkalinity, hardness, and Mg^{2+}. In CKDu cold spots, the MPLs of EC/TDS, total alkalinity, hardness, and Mg^{2+} were exceeded by 49%, 69%, 73%, 60%, and 51%, respectively, while they were exceeded by 56%, 69%, 77%, 69%, and 62% for CKDu hotspots. These results demonstrated that a higher percentage of samples in the hotspots exceeded the MPLs of the drinking water parameters than in the cold spots, indicating that the groundwater in the hotspots was more degraded than in the cold spots. However, the mean concentrations in both regions satisfied the MPLs for drinking. Of the samples in the cold spots, 27% exceeded the MPL of F^- (1.0 mg/L), while this figure was 31% in the hotspots; 27% of samples in cold spots and 33% in hotspots exceeded the MPL of Ca^{2+} (100 mg/L); 3% of samples in hotspots exceeded the MPL for PO_4^{3-} (2.00 mg/L), while none of the samples exceeded this MPL in cold spots; and less than 10% of samples exceeded the MPLs of Na^+ (200 mg/L) and Cl^- (250 mg/L) in both regions. SO_4^{2-} and NO_3^- concentrations were well below the MPLs for drinking purposes (250 mg/L and 50 mg/L, respectively), and none of the samples in either region exceeded these MPLs. Among the drinking water quality parameters, Na^+ is considered secondary or of non-health-related importance, since higher levels have no direct impact on human health. However, excess levels of EC/TDS and hardness can cause a reduction in the palatability of water. Based on the hardness classification of WHO [43], 82% of samples in the basin belonged to the "very hard water" category (>180 mg/L), indicating the low palatability of the groundwater in the area.

Fluoride, phosphate, and nitrate content can be considered the most important primary standards, since a slight increase in their concentrations can significantly affect human health, leading to serious health issues. Excessive fluoride concentrations in the groundwater in the dry zone of Sri Lanka have been identified as a major factor that causes dental and skeletal fluorosis [18,44,45]. Although in Sri Lanka the MPL of F^- is 1.00 mg/L, previous studies have identified widespread dental fluorosis among children in the dry zone of Sri Lanka who consumed fluoride above 0.6 mg/L [46]. However, WHO has recommended 1.5 mg/L as the MPL for F^- in drinking water, with a 1.0 mg/L limit recommended for tropical countries [43]. The etiology of CKDu has also been attributed to the drinking of high-fluoride groundwater, since CKDu-endemic areas overlap with the high-groundwater-fluoride zone in Sri Lanka [11]. The results of some early studies exhibited a synergistic effect of groundwater hardness and fluoride on CKDu [15,16]. Wickramarathna et al. [17] further explained that drinking water with high fluoride content and hardness could exacerbate renal damage, but the initial damage might be caused by other factors. In contrast, the Mg^{2+}–fluoride complex could affect the kidneys, leading to end-stage renal failures [47]. In the present study, 20% of samples from cold spots exceeded the MPLs of F^- (1.00 mg/L) and hardness (250 mg/L), while 31% of samples in hotspots exceeded both levels.

In addition, the pH of the spring water sample was slightly acidic (5.45) but with low TDS values, while the other surface water samples were slightly alkaline compared to the groundwater. Of all the water samples studied, the quality parameters in the spring water were considerably lower than the MPLs for drinking water. Among all the water samples collected (n = 90), the natural spring water was the best quality. Only the EC and TDS were moderately high in surface water samples collected from reservoirs and streams, while all the other parameters remained within safe limits. With these observations, it could be concluded that the spring and surface water samples were more suitable for drinking than the groundwater in the study region when considering inorganic composition. However, more attention should be paid to trace element composition and agrochemical contaminants before the use of surface water for drinking.

The correlation coefficient (Table 3) illustrates the interrelationship between each pair of geochemical parameters. EC/TDS was strongly to moderately correlated with all ions except K^+ in both regions, indicating the direct influence of ions on groundwater composition. Total hardness in CKDu cold spots was strongly correlated with HCO_3^- (r = 0.82), Ca^{2+} (r = 0.80), and Mg^{2+} (r = 0.88), indicating the contribution from carbonate and silicate weathering. Water hardness in CKDu hotspots was strongly correlated with HCO_3 (r = 0.80), Cl^- (r = 0.81), Mg^{2+} (r = 0.87), and Na^+ (r = 0.85). Moreover, total hardness was more strongly correlated with Mg^{2+} compared to Ca^{2+}, indicating the dominancy of Mg^{2+} in hardness in the Yan Oya river basin. Fluoride showed a strong correlation with Na^+ (r = 0.78) and a moderate correlation with Mg^{2+} (r = 0.66) in CKDu cold spots, while it showed moderate correlations with both Na^+ and Mg^{2+} (r = 0.62 and 0.63) in CKDu hotspots, suggesting a similar geogenic origin. These ions could be leached from fluoride-bearing silicate minerals such as amphiboles and biotite [8]. Further, in CKDu cold spots, F^- and HCO_3^- were strongly correlated (r = 0.75) with each other, whereas they were moderately correlated in hotspots (r = 0.62). This correlation could be attributed to the influence of the alkaline nature of the water on the mobilization of fluoride from minerals, leading to higher fluoride levels in groundwater [4]. Moreover, F^- was moderately correlated with total hardness in both cold spots (r = 0.63) and hotpots (r = 0.61).

Table 3. Spearman correlation coefficient matrix of the groundwater quality parameters: (a) cold spots, (b) hotspots (strong correlations (r ≥ 0.75) are shown in bold).

Parameter	pH	TDS	TH	HCO_3^-	Cl^-	SO_4^{2-}	F^-	Na^+	K^+	Ca^{2+}	Mg^{2+}	WQI
Cold Spots												
pH	1.00											
TDS	0.37	1.00										
TH	0.35	**0.84**	1.00									
HCO_3^-	0.50	**0.84**	**0.82**	1.00								
Cl^-	0.01	**0.77**	0.51	0.42	1.00							
SO_4^{2-}	0.12	**0.80**	0.68	0.59	0.72	1.00						
F^-	0.31	**0.78**	0.63	**0.75**	0.56	0.54	1.00					
Na^+	0.25	**0.87**	0.59	0.68	**0.81**	**0.81**	**0.78**	1.00				
K^+	−0.22	0.13	0.02	−0.03	0.19	0.33	0.02	0.15	1.00			
Ca^{2+}	0.28	**0.76**	**0.80**	0.61	0.52	0.62	0.45	0.55	0.08	1.00		
Mg^{2+}	0.42	**0.80**	**0.88**	**0.85**	0.48	0.52	0.66	0.56	−0.07	0.63	1.00	
WQI	0.37	**0.84**	0.69	**0.80**	0.60	0.60	**0.99**	**0.81**	0.05	0.52	0.70	1.00
Hotspots												
pH	1.00											
TDS	0.22	1.00										
TH	0.16	**0.91**	1.00									
HCO_3^-	0.26	**0.88**	**0.80**	1.00								
Cl^-	0.05	**0.84**	**0.81**	0.69	1.00							
SO_4^{2-}	0.13	**0.80**	0.70	0.61	0.66	1.00						
F^-	0.17	0.54	0.61	0.62	0.31	0.41	1.00					
Na^+	0.29	**0.95**	**0.85**	**0.90**	**0.80**	**0.75**	0.62	1.00				
K^+	0.30	0.26	0.10	0.15	0.13	0.20	0.08	0.26	1.00			
Ca^{2+}	−0.06	0.73	0.73	0.64	0.63	0.58	0.37	0.64	0.06	1.00		
Mg^{2+}	0.17	**0.88**	**0.87**	**0.79**	**0.78**	0.70	0.63	**0.85**	0.16	0.57	1.00	
WQI	0.27	0.68	0.66	0.72	0.40	0.53	**0.93**	**0.75**	0.22	0.42	0.68	1.00

4.3. Major Ion Chemistry and Hydrogeochemical Facies

The chemical composition of groundwater depends primarily on the aquifer characteristics as well as on the geochemical processes. The hydrogeochemical data of the water samples were analyzed using different graphical representative diagrams, such as a Gibbs diagram, a Piper trilinear diagram, and scatter diagrams of major ions. Gibbs diagrams are widely used in groundwater studies to determine the major geochemical processes that control groundwater chemistry (Figure 2). Accordingly, rock weathering was found to be the primary mechanism responsible for the hydrogeochemical composition of the groundwater in the Yan Oya river basin.

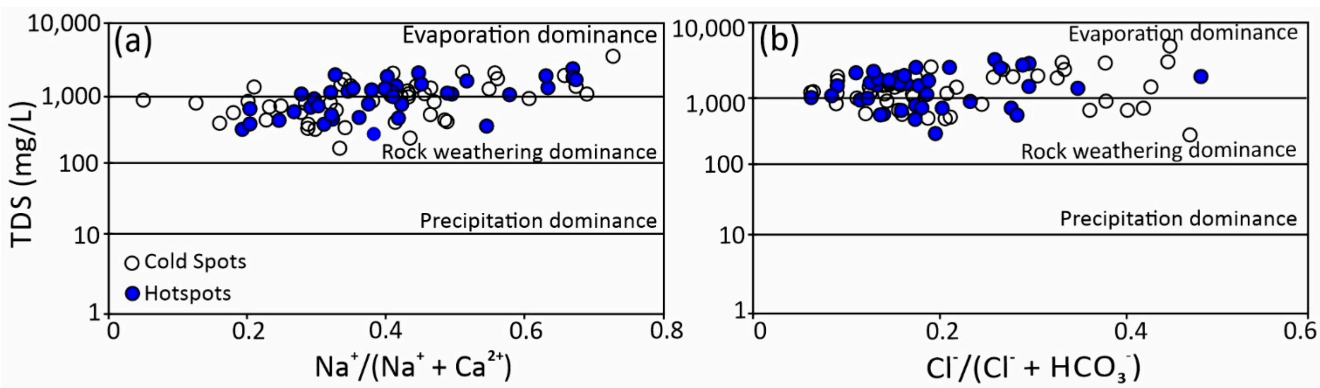

Figure 2. Gibbs plot for groundwater samples: (**a**) TDS vs. $Na^+/(Na^+ + Ca^{2+})$; (**b**) TDS vs. $Cl^-/(Cl^- + HCO_3^-)$.

The majority of the groundwater samples (83%) belonged to the Ca-Mg-HCO_3 type (Figure 3), demonstrating the dominance of alkaline earth over alkali (viz., Ca^{2+} + Mg^{2+} > Na^+ + K^+) elements and weak acidic anions over strong acidic anions (i.e., HCO_3^- > Cl^- + SO_4^{2-}). The sample collected from spring water belonged to the Ca-Cl type, and the two samples collected from streams were also of the Ca-Mg-HCO_3 type. Among the samples collected from reservoirs, two samples belonged to the Ca-Mg-Cl type, while one sample belonged to the Ca-Mg-HCO_3 type. The dominance of alkaline earth elements over alkali elements in the groundwater was further verified by the ion scatter diagrams (Figure 4). The equiline of the diagrams implied 100% contribution to the total cations by the ions represented by the Y-axis, whereas the 2:1 line denoted a 50% contribution to the total cations. Almost all the samples regardless of the CKDu prevalence were plotted in between the 1:1 and 2:1 lines (Figure 4a). This indicated that around 70% of the total cations in the groundwater samples were comprised of alkaline earth metals. All samples were plotted below the 2:1 line and showed the lower contribution of alkali cations to the total concentration of cations (Figure 4b).

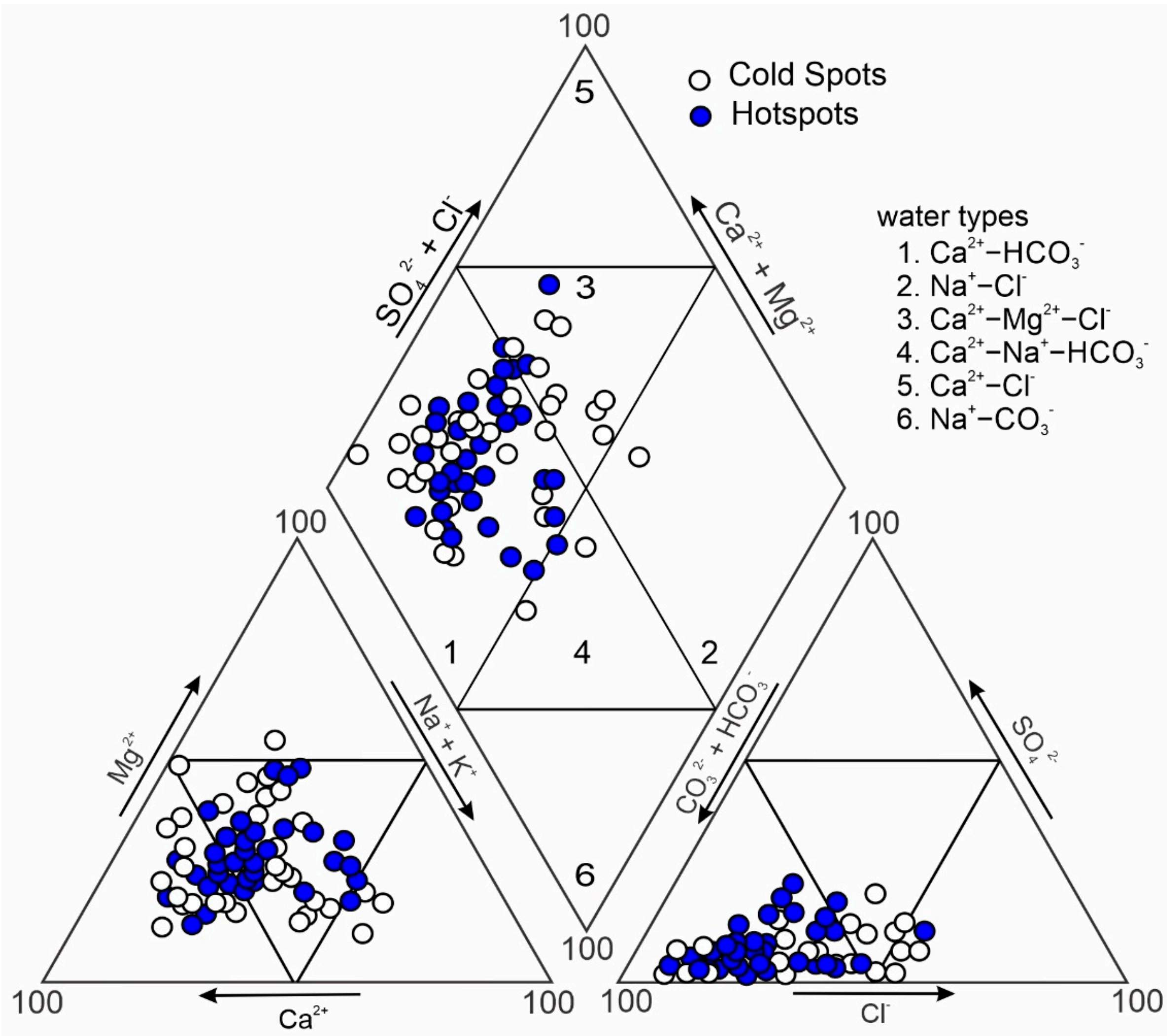

Figure 3. Piper trilinear diagram classifying major hydrogeochemical facies of groundwater in the Yan Oya river basin.

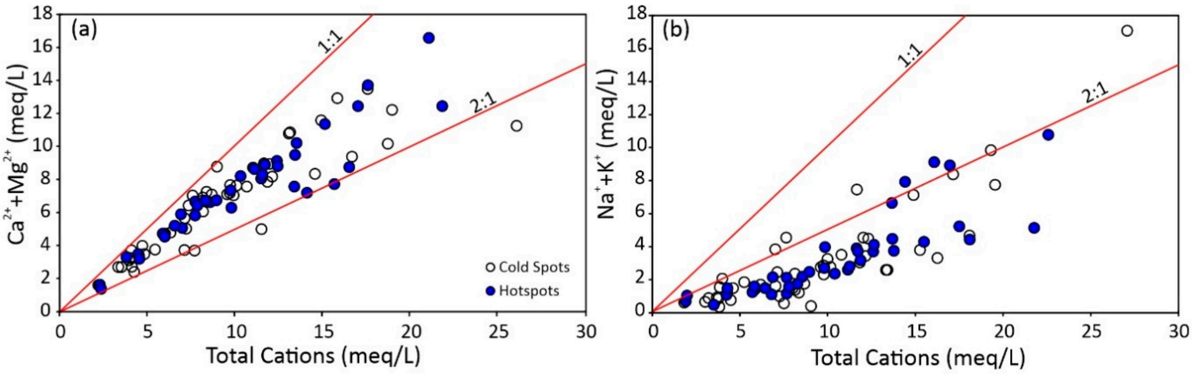

Figure 4. Scatter diagrams of: (**a**) Ca^{2+} + Mg^{2+} vs. total cations; (**b**) Na^{+} + K^{+} vs. total cations.

The reactions between groundwater and aquifer materials have a significant role in determining water quality and are also useful for understanding the genesis of groundwater geochemistry. The results of this study showed that the groundwater in the study region was dominated by high amounts of Ca^{2+}, Mg^{2+}, and HCO_3^- ions. This suggested that the weathering of carbonate and silicate minerals in the high-grade metamorphic basement in the region is responsible for the geochemical composition of the groundwater. The dissolution of carbonate minerals such as calcite ($CaCO_3$) and dolomite ($CaMg(CO_3)_2$) releases Ca^{2+} and Mg^{2+} ions into groundwater. The weathering of silicate minerals such as pyroxene ($CaMg(Si_2O_6)$) and amphibole ($Ca_2Mg_5Si_8O_{22}(OH)_2$) produces a lower amount of Ca^{2+} and Mg^{2+} ions while releasing an exorbitant amount of HCO_3^- ions into groundwater. Moreover, albite ($NaAlSi_3O_8$) dissolution contributes equal amounts of Na^+ and HCO_3^- to groundwater [48]. In addition to the chemical weathering of minerals, subsequent ion-exchange reactions also control groundwater geochemistry. Ion exchange occurs more commonly in clay minerals in the soil and weathered aquifer materials. Na^+ and K^+ in rocks participate in a direct exchange, while Ca^{2+} and Mg^{2+} add to groundwater through reverse ion-exchange processes [48]. The scatter plots of major ions could be further employed to describe the ion-exchange process in the groundwater. The slight deviations in the slope of the trend lines from −1 (in hotspots = −1.08 and in cold spots = −1.47) and the low r^2 values (hotspots = 0.6 and cold spots = 0.7) indicated that ion-exchange processes were only somewhat responsible for the groundwater geochemistry in the study region (Figure 5). Meanwhile, the majority of samples were plotted close to the intersecting point, indicating the restricted ion-exchange processes and the contribution of major ions to the groundwater by the dissolution of minerals. However, ion exchange was more prominent than reverse ion exchange in both regions due to the long residence time of the groundwater in fractured hard rock aquifers in this dry zone.

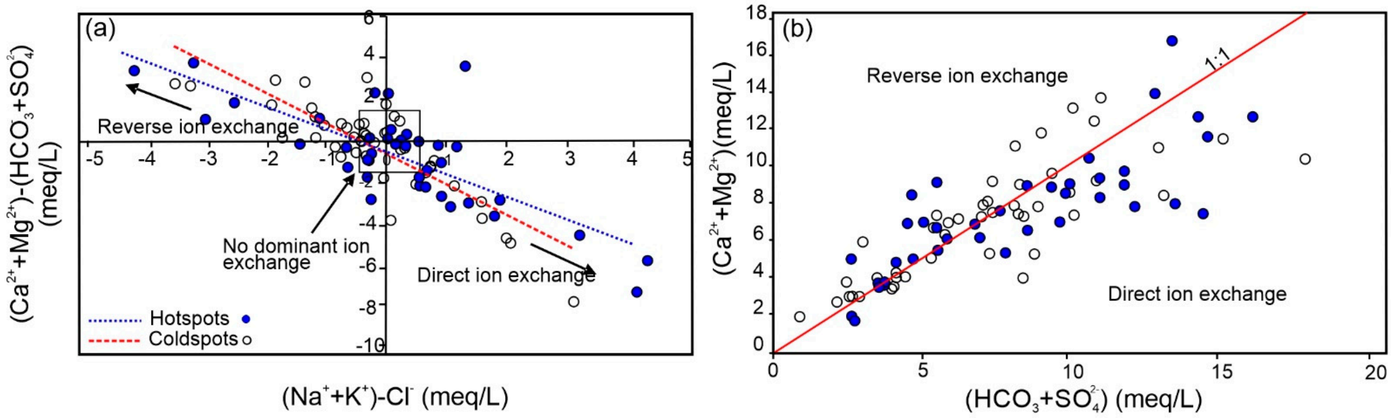

Figure 5. Scatter plots indicating the presence of ion-exchange process in groundwater: (**a**) (Ca^{2+} + Mg^{2+}) − (HCO_3^- + SO_4^{2-}) vs. (Na^+ + K^+) − Cl^-; (**b**) (Ca^{2+} + Mg^{2+}) vs. (HCO_3^- + SO_4^{2-}).

To determine the chemical equilibria between the minerals in solid rock and groundwater, saturation indices (SIs) were calculated for several minerals. Nearly half of the water samples in the basin were saturated with respect to calcite (54%) ($SI_{calcite}$ = −1.34 to 0.51); dolomite (51%) ($SI_{dolomite}$ = −2.98 to 1.17); and aragonite (40%) ($SI_{aragonite}$ = −1.48 to 0.37). On the other hand, under-saturation was observed in terms of gypsum (SI_{gypsum} = −3.63 to −1.34); anhydrite ($SI_{anhydrite}$ = −3.83 to −1.53); halite (SI_{halite} = −8.47 to −5.40); and fluorite ($SI_{fluorite}$ = −4.87 to −0.03).

4.4. Water Quality Index (WQI)

The computed WQI values of the Yan Oya basin ranged from 2.00 to 244, with a mean value of 56. Of the 84 groundwater samples, 46 were within the safe limits (WQI < 50) for drinking, while 26 were in the "poor" and "very poor" (50 < WQI < 100) categories. Furthermore, 12 samples exceeded the WQI limit of 100 whcic were unsuitable for drinking. The water quality of the natural spring (WQI = 2) and other surface water samples was excellent (WQI < 25), while the Hurulu-wewa reservoir sample belonged to the "good" category (WQI = 33).

In the CKDu cold spots, the WQI values ranged from 2 to 141 (mean = 52), while they ranged from 3 to 244 (mean = 61) in the CKDu hotspots (Table 2). Of the 45 groundwater samples collected from CKDu cold spots, 58% were in the "excellent" and "good" categories. However, 42% of the samples exceeded the WQI value of 50, indicating poor to not suitable for drinking. Of the 39 groundwater samples collected from CKDu hotspots, 51% were within the safe limits (WQI < 50), while 49% exceeded a WQI of 50. These results implied that the groundwater quality in the CKDu hotspots had deteriorated greatly compared to that in the CKDu cold spots (Table 2).

Interestingly, the WQI in both the CKDu cold spots (r^2 = 0.987) and hot spots (r^2 = 0.966) was dependent on the fluoride concentration of the water samples. The WQI was strongly correlated with F^- in cold spots (r = 0.99) and hotspots (r = 0.93), indicating that the quality of the groundwater in the Yan Oya basin is determined directly by the fluoride concentration. A similar relationship was also observed in another dry-zone region in Sri Lanka, where the WQI was significantly correlated with the fluoride content in the groundwater [38]. The relationship between the WQI and the fluoride concentration in the groundwater samples was further confirmed by the overlap in their spatial distribution (Figure 6). The strong relationship between the WQI and the fluoride concentration proved that the water quality in the study region was determined directly by the underlying geology of the region, as the fluoride in groundwater is mostly contributed by the weathering of fluoride-bearing minerals such as biotite and amphiboles, which are abundant in aquifer materials. Furthermore, high WQI values and high fluoride concentrations could be observed in the middle part of the basin where the CKDu hotspots were located (Figure 6).

Moreover, "excellent" and "good" (WQI < 50) types of water sources were observed within the CKDu hotspots (51%), while water sources belonging to the "not suitable" category (WQI > 100) were observed within CKDu cold spots (11%) (Figure 7). This could be attributed to the CKDu absence areas within the hotspots. In many cases, CKDu non-prevalence regions can occur even within high-prevalence pockets in the dry zone of Sri Lanka. Furthermore, this may have been due to the heterogeneous distribution of F^- in the groundwater in the study area. Previous studies have identified a strong variation in F^- distribution over lateral distances of only a few tens of meters [12]. The uneven distribution of F^- in the groundwater in the study area was possibly related to the alternative occurrences of CKDu cold spots and hotspots in the dry zone of Sri Lanka. A recent animal model experiment indicated that exposure to groundwater from Sri Lanka with high hardness and F led to kidney damage, pronephric duct obstruction, and abnormal behaviors among zebrafish [49].

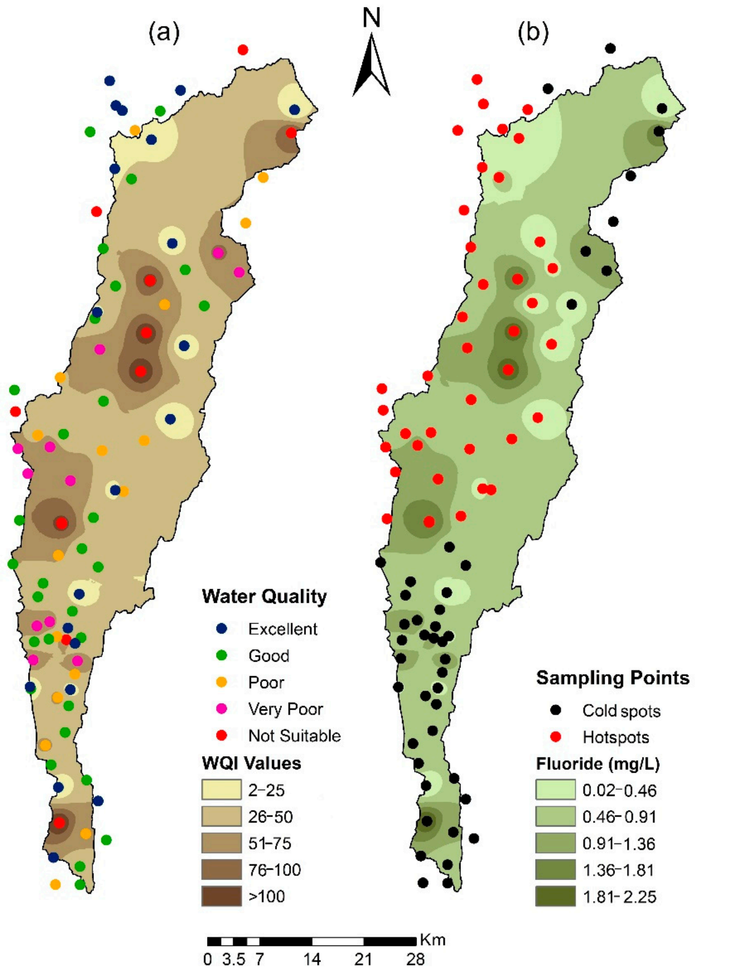

Figure 6. Spatial distribution of (**a**) WQI and (**b**) fluoride concentration of groundwater samples in Yan Oya Basin, Sri Lanka.

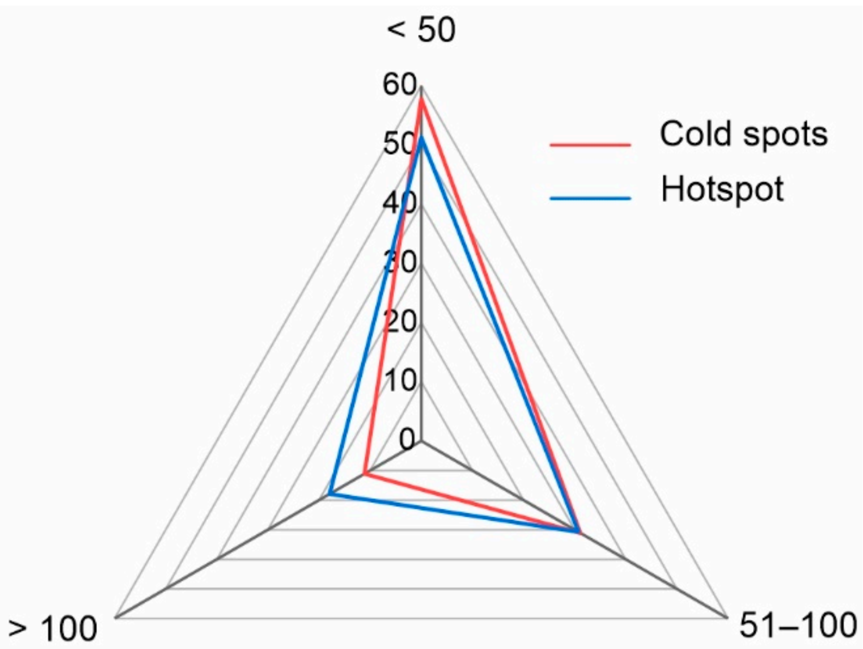

Figure 7. Variation in WQI in CKDu cold spots and hotspots in the Yan Oya river basin.

5. Conclusions

In this study, a general groundwater quality investigation was carried out in the dry climatic conditions of the Yan Oya river basin to determine the impact of groundwater quality on CKDu prevalence in the region. More than 50% of the samples in both CKDu cold spots and hotspots exceeded the MPLs of EC/TDS, total alkalinity, total hardness, and Mg^{2+}, indicating the deteriorated quality of the groundwater in the entire region. Although none of the differences were statistically significant, the mean concentrations of all hydrogeological parameters were higher in hotspots than in cold spots, implying their potential impact on the high prevalence of the disease in hotspots. The palatability of the groundwater across the entire region was reduced mainly due to high EC/TDS and hardness levels. The groundwater chemistry in the region was mainly governed by water–rock interactions. Meanwhile, the high ambient temperature enhanced mineral dissolution and increased the hardness of the water. Furthermore, Mg^{2+}-dominant hardness in the groundwater was identified across the entire basin. However, the presence of permanent hardness in the CKDu hotspots suggested that this may have a considerable impact on the higher prevalence of the disease in hotspots than in cold spots. The results suggested the synergetic toxicological impact of high fluoride and hardness on the onset of CKDu in the dry zone of Sri Lanka. Moreover, the WQI was directly determined by the F^- concentrations in the groundwater in both regions, highlighting that the underlying geology is responsible for the groundwater quality in the region. With the careful inspection of the aforementioned facts, it could be assumed that the long-term use of quality-degraded groundwater may be the primary causative factor for the presence of CKDu in both regions. Furthermore, the elevated levels of hydrogeological parameters in the hotspots compared to the cold spots and the presence of permanent hardness may have been the factors that increased the disease prevalence in the hotspots. Therefore, the assurance of a safe drinking water supply would be a positive intervention for the mitigation of the disease. Since wells with good water quality were noted within the CKDu hotspot regions, these wells could be used to supply water for the surrounding communities.

Author Contributions: Conceptualization: R.C. and S.-L.L.; data curation, analysis, and writing of the original draft: W.A.C.U.; methodology and resources: N.H.K., S.K.G., X.Z. and R.C.; resources and funding acquisition: R.C. and S.-L.L.; review and editing of the final version: N.H.K., R.C., S.-L.L. and X.Z. All authors have read and agreed to the published version of the manuscript.

Funding: This work was carried out as collaborative research, funded by the National Natural Science Foundation, China (Grant No. 41861144026), and the National Science Foundation, Sri Lanka (Grant No. ICRP/NSF-NSFC/2019/BS/02).

Institutional Review Board Statement: Not Required as no human or animal subjects were involved.

Informed Consent Statement: Not Required.

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

Acknowledgments: The authors are grateful to Chamindra Vithana and Sachintha Senarathne for their help in the field investigations. We thank Eileen Richardson from the University of Glasgow for polishing the English of this manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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