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Using water quality and isotope studies to inform research in Chronic Kidney Disease of unknown aetiology endemic areas in Sri Lanka.

Keywords: groundwater, water quality, CKDu, Sri Lanka, isotopes

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Abstract

Chemistry of groundwater in dry climatic, agricultural regions of Sri Lanka studied intensively, in relation to the occurrence of Chronic Kidney Disease of Unknown Cause (CKDu). This paper investigates water quality studies published in CKDu affected areas in Sri Lanka and also present new data set of 27 hydrochemical and isotopic samples collected from shallow groundwater wells from four CKDu hotspots in Sri Lanka. Samples were analysed for major and trace elements, and environmental isotopes including ^{18}O , ^2H and ^{13}C in dissolved inorganic carbon. The concentration of heavy metals including Cd, U, Cr and Pb was below the detection limit. Only two samples had As concentration of $1\mu\text{g/L}$ and the rest were also below the detection limit. Further, there were no evidence of elevated anions such as nitrates and phosphates. The concentration of Si ranged from 10.6 - 64.8 mg/L. The compositions of $\delta^2\text{H}$ and $\delta^{18}\text{O}$ ranged from -13.7‰ to -37.7‰ and from -2.12‰ to -5.99‰ respectively. The average $\delta^{13}\text{C}$ in dissolved inorganic was -17.3‰. The available water quality data, including this study, is not sufficient to answer questions on whether the chemistry of shallow groundwater is related to the CKDu occurrence. However, the study identifies the importance of detailed investigation into degradation products of the agrochemicals, organic matter and Si concentration. The study also provides guidance to the type of isotopic tracers and the frequency of sampling that is needed to capture potential pollutants in future groundwater quality research in CKDu endemic areas in Sri Lanka.

1 Introduction

Endemic chronic kidney disease characterised by chronic interstitial nephritis without fully established cause(s) has been reported in geographically distinct areas throughout many parts of the world, including Eastern Europe, sub-Sahara, Japan and Meso-America. Since generally accepted factors known to increase the risk of chronic kidney failure, such as diabetes, hypertension or glomerulonephritis (Abraham et al., 2019; Webster et al., 2017) are not readily

identifiable in some cases, new tentative classifications for endemic chronic kidney disease have been introduced. These include kidney failure of uncertain aetiology (termed chronic kidney disease of non-traditional causes, or CKDnT or Mesoamerican nephropathy) (PAHO, 2017) and in Sri Lanka, chronic kidney disease of unknown aetiology (CKDu). The degree that these disease entities overlap in their causality or how they reflect similar pathophysiology remains to be established.

Possible aetiology of CKDu in Sri Lanka has been extensively discussed in the scientific literature (Gamage and Sarathkumara, 2016; Gamage et al., 2017; Jayatilake et al., 2013; Kafle et al., 2019; Makehelwala et al., 2020; Nanayakkara et al., 2014; Wanigasuriya et al., 2008; WHO, 2016). Proposed risk factors range from the chemistry of drinking water (Chandrajith et al., 2011b; Cooray et al., 2019; Fernando et al., 2019; Kafle et al., 2019; Rango et al., 2015; Wasana et al., 2016), the effect of agrochemicals (Gunarathna et al., 2018; Gunatilake et al., 2019; Jayasumana et al., 2015a; Jayasumana et al., 2014b; Jayasumana et al., 2015b; Jayasumana et al., 2015c) the effect of mycotoxins and cyanotoxins (Desalegn et al., 2011; Wanigasuriya et al., 2008), (Liyanage et al., 2016; Manage, 2019; Mohamed Zakeel, 2015) heat stress (Jayasekara et al., 2019; Siriwardhana et al., 2015), genetic susceptibility (Friedman and Luyckx, 2019; Nanayakkara et al., 2014), the use of local herbal medicines (Wanigasuriya, 2014), infections from virus (Gamage and Sarathkumara, 2016; Gamage et al., 2017; Sarathkumara et al., 2019; Yoshimatsu et al., 2019), socioeconomic disadvantage including nutrition (Jayasekara et al., 2019; Senevirathna et al., 2012; Wimalawansa, 2016) and lifestyle conditions (Levine et al., 2015; Senevirathna et al., 2012). The complex interactions between human health and the environment, notably the quality of drinking water, has been discussed and partially investigated but widely accepted outcomes have not been made in Sri Lanka (Levine et al., 2015; Rango et al., 2015).

Sri Lanka is divided into three climate zones; the wet, dry and intermediate. CKDu is found particularly in the dry zone with average rainfall amounts of 1250 mm/year (Fig. 1). Communities living in the dry zone depend heavily on shallow groundwater for their drinking water requirements (Panabokke and Perera, 2005). The quality of groundwater is influenced by anthropogenic activities such as agricultural practises occurring in the catchment and sanitation practices such as the use of wet latrines, and environmental factor such as lithology, hydrogeology and also climatic conditions such as alternating dry and wet seasons. Specifically, dry zone catchments face many environmental issues such as over-extraction of groundwater resources due to increased demand placed on the system over the past 20 years,

contamination from agricultural runoff containing fertilisers and pesticides that are high in heavy metals, nitrogen and phosphorous. The groundwater of the dry zone also has higher hardness and high fluoride (F) concentrations sourced from the bedrock (Dissanayake, 1991; Young et al., 2011).

Despite the plethora of environmental issues that can influence groundwater quality in these regions and the apparent correlation between increased occurrence of CKDu in the dry zone since 1990s when use of groundwater become the dominant water resource, very limited research into the hydrogeology of Sri Lanka has been undertaken.

The objective of this paper is to investigate shallow groundwater quality studies published in CKDu affected regions and also to provide a new data set of 27 isotopic and hydrochemical samples collected from shallow groundwater wells from hotspots in dry zone CKDu endemic regions in Sri Lanka. This is the first carbon isotope study using dissolved inorganic carbon ($\delta^{13}\text{C}_{\text{DIC}}$) values for the region. This study aims to identify directions for future hydrological and hydrogeological research. Furthermore, the paper also discuss how this new dataset can be used to characterise shallow groundwater quality and identify potential contamination pathways in CKDu affected regions in Sri Lanka.

1.1 Water quality studies in CKDu affected regions of Sri Lanka

This section discusses previous literature related to water quality studies in CKDu affected regions of Sri Lanka. Occurrences of CKDu in the dry zone of Sri Lanka form mosaic-like patterns and have been interpreted to be related to areas with fluoride concentrations above the World Health Organisation (WHO) safe limit for tropical countries, which is 0.6 mg/L (WHO, 1994). Formation of toxic aluminium fluoride complexes (AlF_x) in the reaction between F in groundwater and low-grade aluminium cooking utensils has been suggested as a causal factor for the disease (Ileperuma et al., 2009). However, this does not seem a likely cause because CKDu is not evenly distributed over regions with high F concentrations in groundwater, and the use of low-grade aluminium utensils is widespread throughout Sri Lanka.

The lack of geographical correlation with ion toxicity in groundwater has prompted research into more complex ion interaction in water chemistry. For example, Chandrajith et al. (2011a) found that the sodium (Na) to calcium (Ca) ratio of groundwater in areas with no CKDu occurrences were higher (34 – 469) than in CKDu regions (1.6 – 6.6), although F concentrations remain constant. They argue that activities of Na and Ca in shallow groundwater govern the

nephrotoxicity of F, hence the prevalence of CKDu. However, this has not been tested extensively, and its relationship in shallow groundwater in most of the CKDu regions remains unknown (Wimalawansa, 2016).

The hardness of the water has also been linked to CKDu. It has been found that shallow groundwater in the dry zone of Sri Lanka is very hard. The hardness of the water together with a high concentration of F has been linked to CKDu (Wickramarathna et al., 2017). Once again this is not conclusive because the southern and northern part of the country located in the dry zone also have very hard, F-rich groundwater (>250mg/L) and do not report CKDu occurrences.

Other studies have suggested the synergetic effect of F, cadmium (Cd) and hardness of shallow groundwater as causal factor (Wasana et al., 2016). They acknowledge that F alone is not causing CKDu, but the synergic effect of elements act as a triggering factor. They used multivariate statistical analysis to support their argument (Wasana et al., 2016). However, the predicted relationship between F, Cd and hardness, and CKDu is not adequately supported by the multivariate statistical analysis.

The Hofmeister series is used to classify anions and cations according to their strength to denature proteins (Yang, 2009). Dharma-wardana et al. (2015), argued that consumption of water high in Hofmeister ions as a causal factor for CKDu. Although the presence of high order Hofmeister cations like Mg and Ca is well understood, the presence of high order Hofmeister anions such as Br, NO₃, ClO₄ and SCN in shallow groundwater in CKDu affected regions is still unknown. Therefore, there is still no clear evidence as to whether the Hofmeister ions can be discounted entirely as a causal factor for CKDu.

Higher concentrations of toxic elements such as cadmium (Cd), arsenic (As), and uranium (U) were found in fertiliser used in CKDu endemic areas of Sri Lanka (Chandrajith et al., 2010). Phosphate fertilisers used in agriculture is suggested to be the main source of As in the region, and elevated levels have been implicated as a causal factor contributing to CKDu in Sri Lanka (Jayasumana et al., 2014a; Jayasumana et al., 2014b; Jayasumana et al., 2015c; Jayasumana et al., 2013). However, a number of studies found no evidences of elevated concentrations of As in drinking water sources, or in any other environmental sample such as rice, freshwater fish species or in soils (Bandara et al., 2008; Gunarathna et al., 2018; Jayatilake et al., 2013; Levine et al., 2015; Nanayakkara et al., 2019; Wickramarathna et al., 2017). Further, CKDu is not reported in Puttalam, Mannar or Jaffna where As concentration in shallow groundwater are

high (7.7 – 74.0 µg/L) (Bandara et al., 2018; Kawakami, 2012). Therefore, it is difficult to correlate the occurrence of CKDu with elevated levels of As in shallow groundwater. Glyphosate in conjunction with water hardness and elevated As concentrations have been postulated as a causal factor for CKDu (Jayasumana et al., 2014b; Jayasumana et al., 2015c). However, the detected concentrations of glyphosate and aminomethylphosphonic acid (AMPA); a major metabolite of glyphosates in groundwaters are well below the maximum contaminant levels (1-4µg/L) (Gunarathna et al., 2018). Existing evidence on As and glyphosate concentrations in groundwater make it difficult to relate between glyphosate-metal complexes and the occurrence of CKDu.

Shallow groundwater wells located close to rice fields have elevated Cd and may be high-risk for CKDu (Wanigasuriya et al., 2011). The Mahaweli River and farmland under irrigation in the north-central province (NCP), one of the highest CKDu prevalence province in the country, was found to have high concentrations of Cd (Bandara et al., 2011). However, Cd concentrations in shallow groundwater sources of CKDu endemic areas in a number of comprehensive studies were less than WHO guidelines (Chandrajith et al., 2005; Chandrajith et al., 2011b; Diyabalanage et al., 2016; Jayatilake et al., 2013; Levine et al., 2015; Nanayakkara et al., 2019; Rango et al., 2015; Wasana et al., 2016; Wickramarathna et al., 2017).

Levine et al. (2015) found chronic exposure of Cd, lead (Pb) and mercury (Hg) among north-central province (NCP) population by studying hair and urine samples. They also found shallow groundwater samples with Pb concentrations exceeding acceptable drinking water standards (0.01mg/L). Elevated concentrations of Pb (0.01-0.03mg/L) were identified in irrigation reservoirs/tanks in NCP (Bandara et al., 2008), but not in the groundwater (Chandrajith et al., 2011b; Nanayakkara et al., 2019; Rango et al., 2015). Hence, no definitive evidence has been presented to link elevated levels of As, Cd or Pb to CKDu.

Organic matter in the groundwater has also been captured the attention of CKDu researches. Nephrotoxicity of cyanotoxins is known (Piyathilaka et al., 2015) and have been discussed as a risk factor for CKDu (Manage, 2019). A high concentration of DOC (4mg/L) found in groundwater is one of the main contributors to the poor water quality index in CKDu endemic areas presenting a great health risk (Cooray et al., 2019). The toxic compounds formed by reacting DOC in groundwater with other chemical constituents such as Ca and, agrochemical and their residual products have also been discussed as a causal factor for CKDu (Makehelwala et

al., 2020; Makehelwala et al., 2019). Hence the organic matter in groundwater as a risk factor for CKDu required further investigations.

However, most of these studies are not comprehensive enough to provide a thorough water quality assessment of the discussed contaminants. A limited number of studies have investigated the seasonal variation of the water chemistry in the area (e.g. Wickramarathna et al. (2017); Cooray et al. (2019)). This type of hydrochemical assessment cannot be undertaken on a single sample round, especially in a country with extreme seasonal conditions (i.e. dry and wet seasons). Further, studies that focus on one or few elements would not lead to a complete understanding of the chemistry of the groundwater. It is also noted that the underlying hydrogeology and hydrology of the area have not been considered in any of these assessments. Hence, we suggest it is important to undertake a comprehensive geochemical assessment considering multiple hydrological factors including changes in the lithology of the aquifer, seasonal variations and recharge events such as infiltration of agricultural runoff to help understand the water quality of CKDu endemic areas of Sri Lanka.

1.2 Isotopic studies in CKDu affected regions of Sri Lanka

Isotopic tracers such as stable isotopes of oxygen (^{18}O), hydrogen (^2H), nitrogen (^{15}N) and dissolved inorganic carbon ($^{13}\text{C}_{\text{DIC}}$), can be useful for understanding fundamental hydrochemical processes in the water cycle (Clark and Fritz, 1997). They can be used to understand sources of recharge (eg. river water vs rainfall), evaporation and can be used to trace the source(s) of contaminants, carbon and nitrogen in the environment (Herrera et al., 2018; Meredith et al., 2012; Meredith et al., 2009). Application of these isotopes to water resources in Sri Lanka is in its early stages. In fact, only two studies have used this technique to provide further characterisation of water sources in CKDu affected areas (Edirisinghe et al., 2017; Wickramarathna et al., 2017).

Wickramarathna et al. (2017), was the first study to provide $\delta^2\text{H}$ and $\delta^{18}\text{O}$ results from shallow groundwater wells in CKDu areas. They found the influence of evaporated surface water on groundwater in CKDu affected areas. Edirisinghe et al. (2017), took this type of assessment further and found a relationship between recharge mechanisms with CKDu. Shallow groundwater wells were found to be recharged from direct infiltration of rainwater, or regional groundwaters were associated with high CKDu. This study was the first to suggest deep groundwater as a causal factor for CKDu. In contrast, water samples from shallow groundwater

wells which are influenced by evaporated surface were found to have no relationship with CKDu incidents.

It must also be noted that in the above mentioned studies, groundwater samples were collected from large diameter (2-3m) open wells that are exposed to evaporation. This is important for isotopic studies looking at evaporation or redox conditions of groundwater and heavy metal or trace element mobilisation. Further, understanding environmental isotopes such as ^{15}N , ^{13}C (dissolved organic carbon and dissolved inorganic carbon), and ^{87}Sr also essential for a better understanding the hydrochemical evolution of the groundwater in dry zone areas of Sri Lanka. Such studies can provide vital information on distribution and fate of agrochemicals, and other anthropogenic contaminants such as septic waste (Fiorentino et al., 2017; Lu et al., 2016; Meredith et al., 2019) and natural contaminants resulted from weathering of rocks and soils (Bain and Bacon, 1994; Meredith et al., 2013).

2. Environmental setting

The present ongoing study was designed to target high CKDu endemic dry zone areas of Sri Lanka namely, Mahiyanganaya (A) in Mahaweli basin, Padaviya (B) in Ma Oya basin, and Medawachchciya (C) and Rambewa (D) in Aruvi Aru basin.

Annual rainfall for the dry zone area is 1,250 mm per year, the temperature range from 28 - 30 °C, and humidity is around 70%. The climate of the area is governed by two distinct seasons; wet and dry. The wet season starts with inter-monsoonal rain from October to November, which is followed by north-east (NE) monsoon from December to February. The remaining

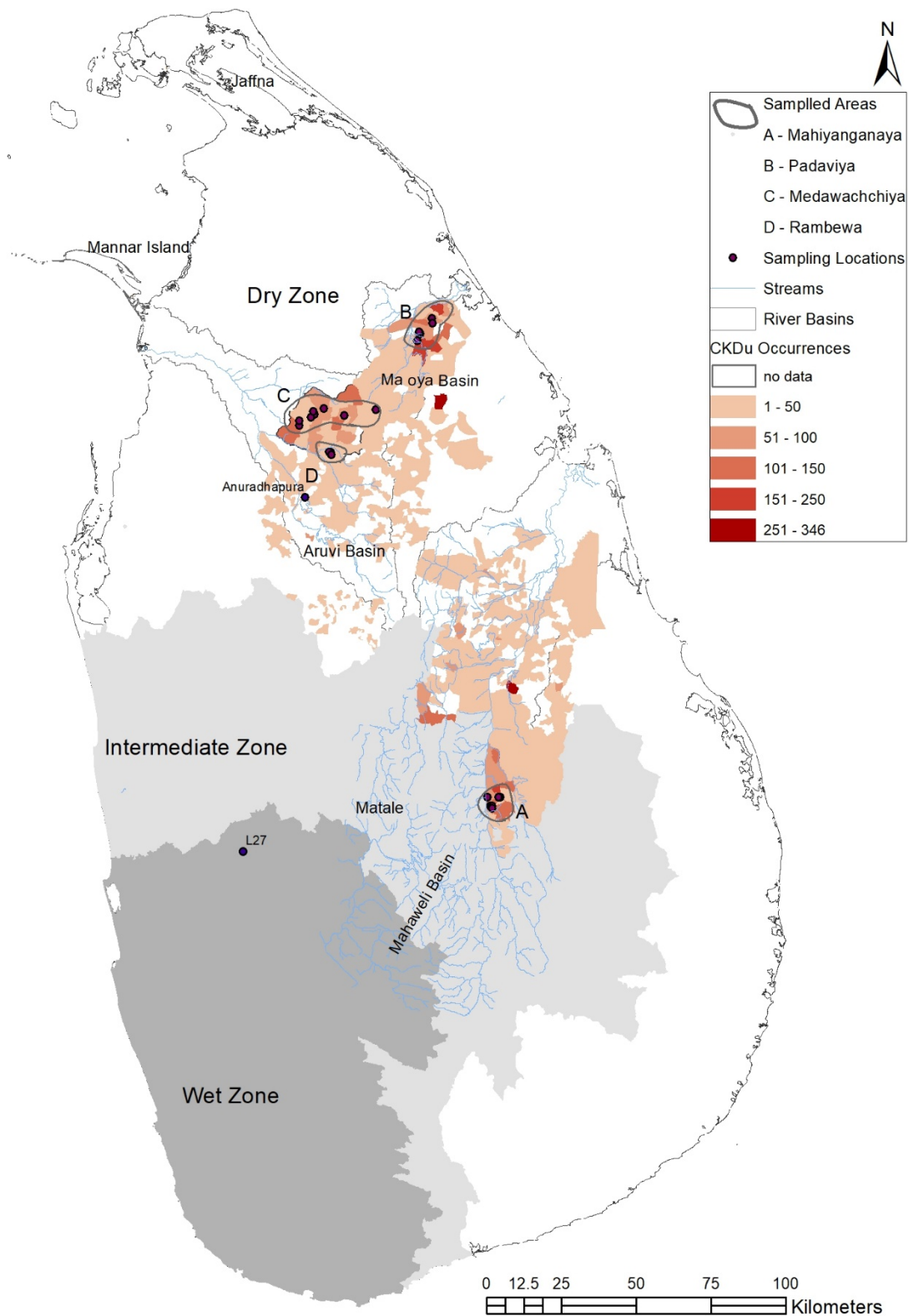


Figure 1: The Map of the sample area

months of the year are considered as the dry period, which peaks from June to July. Monthly rainfall recorded in Anuradhapura, which is the closest observation centre to the study areas B, C and D is given in Figure 2 (long term average = 1285mm). The composition of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ in monsoon rainfall in Dehiattakandiya is -6.62 ± 1.75 and -41.2 ± 11.9 ‰, respectively and the local meteoritic water line was calculated to be $\delta^2\text{H} = 7.9 \delta^{18}\text{O} + 12$ ‰ (Edirisinghe et al., 2017). The Ma Oya and the Aruvi Aru Rivers are fed by the NE monsoon rains. The Mahaweli River is fed by both NE and south-west (SW) monsoons, which produce rainfall that occurs on the central hills and carries large volumes of water throughout the year.

Paddy cultivation in the dry zone starts with the monsoon rain. Maha; the main season starts in September with the inter-monsoonal and ends by March. Yala season from March to August is considered as the minor growing season as the SW monsoon does not bring effective rainfall.

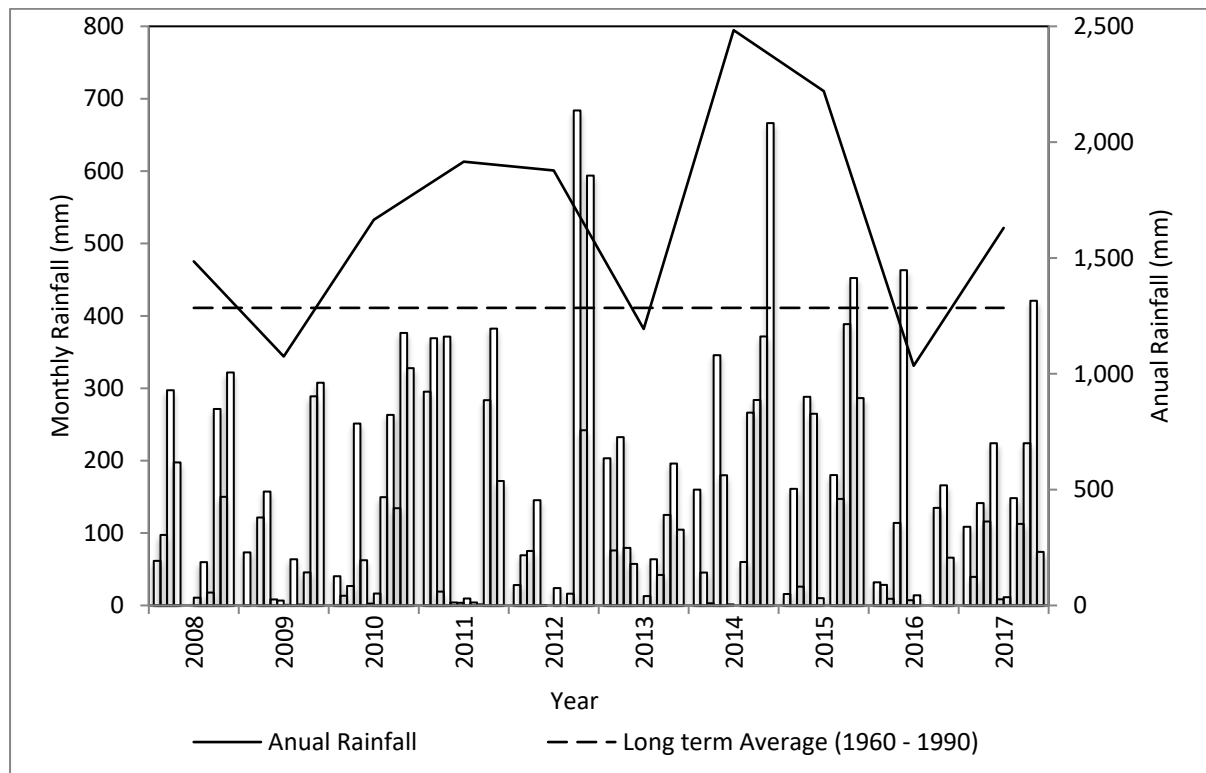


Figure 2: Monthly rainfall distribution in Anuradhapura. (source: DCSSL (2014) and DCSSL (2017)).

Rainwater is stored and delivered through an interconnected network of human-made micro (water area = 0.1 – 1 ha) to major (water area > 200 ha) reservoirs that are referred to as ‘tanks’ that act as a cascade system for rice fields in the region (Bebermeier et al., 2017). Along its 320 km reach, the Mahaweli River has been dammed in several locations to provide irrigation facilities for dry zone cultivation. Water for domestic use in the region is extracted from 2-3 m

diameter wells dug into shallow regolith aquifers and on few occasions deeper wells located within underlying fractured bedrock aquifer (approximately 60 m in depth). The groundwater level in most large-diameter dug wells are at the surface during the wet season and decrease by 3-5 m during the dry season.

Based on age, metamorphic grades and geology, Sri Lankan Precambrian basement is divided into four lithological units, namely Highland, Vijayan, Kadugannawa and Wannu (Cooray, 1994). The Ma Oya and the Aruvi Aru Basins are located in the Wannu complex, whereas the Mahaveli Basin is located in the highland complex. Sampled areas in the Ma Oya and the Aruvi Aru basins are contained charnockitic gneiss, quartzo-feldspathic gneiss, biotite gneisses, hornblende biotite gneisses and quartzite bedrock (Nandadasa and Priyanga, 2010). Common rock types found in the Mahaveli Basin are quartzo-feldspathic gneiss, charnockitic gneiss, garnet sillimanite gneiss and quartzite (Nandadasa and Priyanga, 2010).

Physical and chemical properties of residual soil in the dry zone of Sri Lanka are highly influenced by the climate. Soil profiles are thin (2-3m), and immature with no clear horizons except the top layer, which is rich in organic matter (Jayawardana et al., 2014; Jayawardana and Izawa, 1994). Kaolinite, halloysite, smectite and haematite are the main minerals that make up the soils in the dry zone.

3. Methods

3.1 Sampling and analysis

Groundwater samples were collected from 26 different water sources including; shallow groundwater wells (n = 24), a spring and a deep well (~60m). A sample from a shallow groundwater well in Kurunegala (L 27) in the wet zone was also collected for comparison. Sampling was conducted during the dry season (July) in 2017. Shallow groundwater wells that have been in use for more than ten years were selected for sampling to ensure the long term usage. The depth to the water level the dug wells varied from 3 to 5 m and the well diameter varied from 2-3 m. Shallow groundwater wells were designed to collect water for domestic use and were generally not covered, these samples are not representative of deeper groundwater end-members but do represent the groundwater consumed. Samples were collected from (1) wells used by CKDu patients (n= 14) and (2) those that did not have CKDu (n=13). The data from this reconnaissance sampling round is considered as a baseline assessment in this study.

All samples were filtered through 0.45µm disposable filters. Samples were collected in 60ml high-density polypropylene (HDPE) bottles for anion analysis. A second set of 60ml samples were collected and preserved with ultra-pure HNO₃ acid for cation analysis. Samples for δ²H and δ¹⁸O were collected into 30ml HDPE bottles. Pre-conditioned 12 ml glass gas-tight vials were used to collect for inorganic carbon isotopes (δ¹³C_{DIC}) no headspace. Sample processing and preservation were performed in accordance with the United State geological society (USGS) standard procedures described in Wilde et al. (2004). Electrical conductivity (EC), pH, dissolved oxygen (DO), and temperature of samples were measured in the field using WTW Multi 3430 set G portable test kit. Meters were calibrated before each sampling day.

Major element composition of water samples were analysed by inductively coupled plasma atomic emission spectrometry (ICP-AES) while trace elements were measured using inductively coupled plasma atomic spectrometry (ICP-MS). Alkalinity of samples was measured by auto-titration at the laboratory. Anion compositions of samples were measured using ion chromatography. Charge balance errors (CBE) were calculated using major cations (Ca, Mg, Na and K) and major anions (Cl, NO₃, HCO₃, CO₃ and SO₄) in order to determine the analytical precision. Despite the fact that alkalinity was analysed after a month of sampling, 66% of samples had a charge balance error (CBE) below 5%.

The δ²H and δ¹⁸O isotopic ratios were determined using Picarro Cavity Ring-Down Spectroscopy (CRDS) method with an accuracy of ±1‰ and ±0.15‰ for δ²H and δ¹⁸/δ¹⁶O, respectively. Results were reported relative to Vienna standard mean oceanic water (VSMOW). Equilibration method on Gas Bench II coupled to continuous-flow delta V advantage isotope ratio mass spectrometer (IRMS) was used to measure δ¹³C abundance in samples with an accuracy of ±0.3‰. Results are reported relative to the international atomic energy agency (IAEA) secondary standards that have been certified relative to Vienna pee dee belemnite (VPDB) for carbon. Results of the isotope analysis are expressed in standard per mil (‰) notation. All samples were analysed in the Australia nuclear science and technology organisation (ANSTO) laboratories. The δ²H and δ¹⁸O and δ¹³C isotope methods are described in Meredith et al. (2012), and Meredith et al. (2015).

4 Results

In this study, we present hydrochemical parameters (Table 1 & 2) and environmental isotopes compositions for stable isotopes of δ¹⁸O, δ²H and δ¹³C_{DIC} (Table 3). This data will provide

Table 1: Summary of the hydrochemistry for shallow groundwater samples in CKDu endemic areas of Sri Lanka during the dry season (July 2017)

	DO	pH	EC ($\mu\text{S/cm}$)	Ca (mg/L)	Mg (mg/L)	Na (mg/L)	K (mg/L)	F ⁻ (mg/L)	Cl ⁻ (mg/L)	SO ₄ ²⁻ (mg/L)	NO ₂ ⁻ (mg/L)	NO ₃ ⁻ (mg/L)	HCO ₃ ⁻ (mg/L)	CO ₃ ²⁻ (mg/L)
A: Mahiyanganaya (n=7)														
Min	1.58	6.15	132.40	7.66	4.68	6.93	0.34	0.15	4.96	4.53	0.03	0.05	15.86	0.30
Max	5.12	7.02	426.00	107.00	22.90	59.00	33.20	1.18	26.10	18.20	0.03	0.05	187.88	0.30
Mean	3.32	6.45	262.24	26.09	12.57	26.50	5.42	0.65	15.71	8.97	0.03	0.05	86.27	0.30
B: Padaviya (n=6)														
Min	1.37	6.41	428.00	28.00	20.40	29.90	0.52	0.33	17.80	9.26	0.05	0.05	222.04	0.30
Max	5.71	7.47	1029.00	75.00	54.50	94.40	0.86	1.01	80.40	32.90	2.05	7.20	516.06	1.20
Mean	3.66	6.87	674.67	49.33	31.03	52.32	0.66	0.67	46.55	19.98	0.38	1.38	317.61	0.50
C: Medawachchiya (n=7)														
Min	1.37	6.91	587.00	20.70	15.80	37.00	0.60	0.04	37.90	11.30	0.03	0.10	275.72	0.30
Max	7.32	7.36	1377.00	150.00	63.20	104.00	2.29	1.27	265.00	42.40	1.45	2.90	575.84	1.80
Mean	4.12	7.14	1049.71	86.00	36.91	65.53	1.45	0.68	111.64	22.40	0.41	1.03	436.06	0.77
D: Rambewa (n=4)														
Min	2.58	6.69	702.00	63.40	15.40	35.20	0.73	0.66	21.20	9.75	0.03	0.20	295.24	0.30
Max	4.37	6.92	1020.00	130.00	31.20	50.10	1.64	1.10	87.30	16.40	0.26	1.50	480.68	0.30
Mean	3.12	6.82	812.25	94.13	24.73	44.20	1.04	0.91	56.23	14.16	0.11	0.80	399.25	0.30
DGW	3.74	6.81	615	73.90	23.40	23	1.17	0.72	15	8.91	0.38	2.50	350.14	0.30
Spring	2.48	5.55	170.00	7.52	5.34	11.20	1.94	0.06	21.50	0.95	0.03	0.05	34.16	0.30
WZGW	4.63	6.07	197.60	16.40	7.24	8.70	0.64	0.07	16.70	3.20	1.31	16.80	64.66	0.30

DGW= deep groundwater WZGW= wet zone groundwater

further insight into the quality of the shallow groundwater in CKDu endemic areas, which may help identify contaminants of concern in the shallow groundwater .

Over 60% of the shallow groundwater samples from CKDu areas had F concentrations greater than 0.6 mg/L, which is the safe limit for tropical countries (WHO, 1994). Average Mg concentration in the shallow groundwater in Medawachchiya and Padaviya exceed Sri Lankan drinking water guidelines (30 mg/L) (SLIS, 2013). This finding is consistent with (Wickramarathna et al., 2017). Besides this, a number of samples exceed Sri Lankan safe drinking water limits for Mn, Ca and HCO_3 (Table 1). Except for K, shallow groundwater from Mahiyanganaya had the lowest concentration of all major cations and anions, while Medawachchiya had the highest. Despite variable lithologies and catchments, water types in all study areas were Mg – HCO_3 type waters in the piper plot (not given).

The EC of analysed the shallow groundwater samples from studied CKDu areas varies from 134 to 1,377 $\mu\text{S}/\text{cm}$ with the lowest average of 262 $\mu\text{S}/\text{cm}$ from Mahiyanganaya (n=7). Average EC of water from Medawachchiya (n=7) is four times greater than that of Mahiyanganaya. All shallow groundwater samples had pH values ranging from 6.45-7.14. Both samples from Kebithigollawa spring and wet zone were slightly more acidic (5.55 and 6.07 respectively). The EC of these two samples (170, and 198 $\mu\text{S}/\text{cm}$) were also lower than the average of samples from four CKDu endemic areas studied (Table 1).

The dissolved oxygen (DO) varies in concentration from 1.37 mg/L to 7.32 mg/L with an average of 3.06 mg/L (n = 27). These results suggest the waters are in equilibrium with the atmosphere.

All shallow groundwater samples from Mahiyanganaya (n=7) and the spring sample had nitrate concentrations below the detection limit (0.1 mg/L). The highest nitrate concentration of 16.8 mg/L was observed in the shallow groundwater sample from the wet zone. The highest average nitrate level in CKDu endemic areas was observed in Padaviya (1.38 mg/L; n = 6). A quarter of the samples exceed WHO guideline limits for nitrite for long-term consumption (0.2 mg/L) with the highest value of 2.05 mg/L (L17). Except for one sample from Medawachchiya (L8), all samples had phosphates levels below the detection limit (1 mg/L).

There was no evidence of high concentrations of trace elements in these samples (Table 2). Concentrations of Cd, U, chromium (Cr), and selenium (Se) in all samples were below the detection limit, which is 1 $\mu\text{g}/\text{L}$ and Pb concentration was less than 10 $\mu\text{g}/\text{L}$ (Appendix 1).

Table 2: Trace element and heavy metal concentration of the shallow groundwater samples

Sample	Area	As (µg/L)	Cu (µg/L)	Mn (µg/L)	Fe (mg/L)	Ni (µg/L)	Rb (µg/L)	Zn (µg/L)	Si (mg/L)	Sr (mg/L)
L1	A	<1	<1	33	13	<1	<1	7	52.0	0.034
L2		<1	<1	420	27	<1	1	9	41.3	0.064
L3		<1	<1	251	14	3	<1	16	19.2	0.060
L4		<1	<1	17	<5	<1	<1	67	10.6	0.079
L5		<1	<1	57	12	<1	1	25	43.4	0.082
L6		<1	<1	108	34	5	<1	24	64.8	0.075
L7		<1	<1	75	10	<1	16	40	44.8	0.766
L8	C	<1	<1	32	16	<1	<1	11	58.0	0.168
L9		1	<1	6	13	<1	<1	55	49.3	1.36
L10		<1	<1	<1	<5	<1	<1	35	41.3	1.27
L11		<1	<1	64	<5	<1	<1	57	48.0	1.01
L12		<1	<1	8	8	4	<1	23	52.7	0.363
L13		<1	<1	2	11	<1	<1	13	41.7	1.01
L14		<1	<1	138	8	3	<1	11	24.5	0.641
L15	D	1	<1	5	10	<1	<1	19	46.1	0.715
L16		<1	<1	169	12	6	<1	28	42.5	0.292
L17		<1	<1	54	12	4	<1	35	55.7	0.259
L18		<1	<1	33	21	1	<1	70	53.6	0.175
L19	B	<1	<1	12	9	1	<1	9	49.7	0.395
L20		<1	<1	120	14	4	<1	29	55.0	0.154
L22		<1	1	81	18	2	2	14	48.0	0.487
L23		<1	<1	3	13	<1	<1	10	50.3	0.804
L24		<1	<1	56	12	<1	<1	9	51.6	0.646
L25		<1	<1	5	8	<1	1	10	49.9	0.453
L26	DGW	<1	10	<1	9	1	2	11	49.7	0.402
L27	WZGW	<1	<1	3	<5	<1	<1	6	18.1	0.190
L21	Spring	<1	1	2	6	3	2	9	15.8	0.045

Higher Si concentrations were observed in CKDu endemic areas compared to the wet zone and spring water. Average Si content of observed shallow groundwater samples in CKDu endemic areas (n = 25) was 45.75 mg/L which, was 2.5 – 3 times greater than that of wet zone and spring samples.

The $\delta^2\text{H}$ compositions ranged from -13.7‰ to -37.7‰ and $\delta^{18}\text{O}$ compositions ranging from – 2.12‰ to -5.99‰. These results are consistent with findings of Wickramaratna et al. (2017), which reported an average of -29.7‰ and -4.88‰ for $\delta^2\text{H}$ and $\delta^{18}\text{O}$, respectively (Table 3).

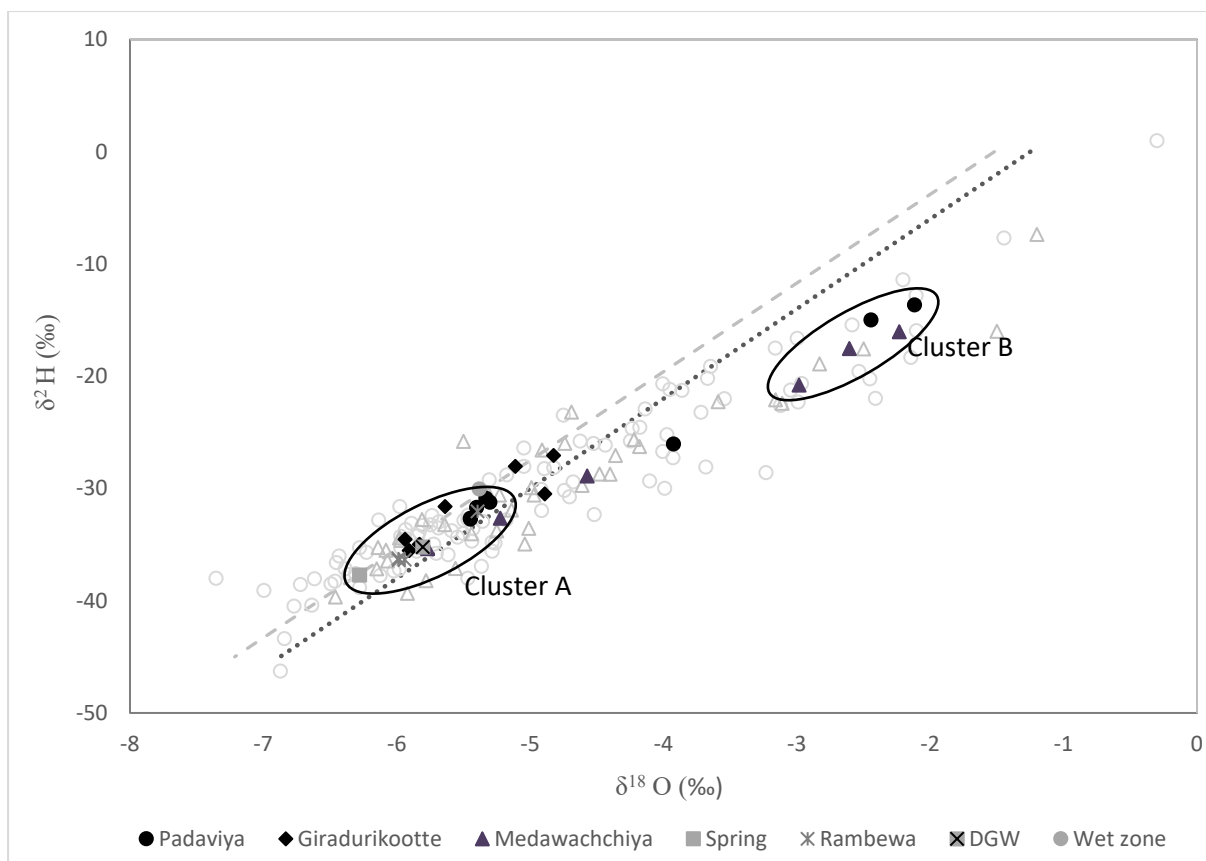


Figure 3: $\delta^{18}\text{O}$ vs $\delta^2\text{H}$ in CKDu endemic areas; solid symbols represent the results from the present study, and light coloured symbols represent the findings of the previous studies. Light colour circles represent the findings of Edirisinghe et al. (2017). Light colour triangles represent the findings of Wickramaratna et al. (2017). The Global meteoritic water line (GMWL) is given in light dotted line, and local meteoritic water line (LMWL) is given in light dotted line.

Two distinct clusters of samples can be identified from the bivariate plot of $\delta^2\text{H}$ vs $\delta^{18}\text{O}$ values; (A) ($n = 17$) and (B) ($n = 5$) (Fig. 3). Cluster A waters plot above the global meteoritic water line (GMWL) and below the local meteoric water line (LMWL) described in Wickramaratna et al. (2017). Cluster B plot below both GMWL and LMWL, and are enriched in $\delta^2\text{H}$ vs $\delta^{18}\text{O}$ compared to cluster A. The sample from the spring had the most depleted isotope composition compared to the other samples ($\delta^{18}\text{O} = -6.28\text{‰}$ and $\delta^2\text{H} = -37.7\text{‰}$) and the wet zone sample had $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values of -5.38‰ and -30.1‰ , respectively. The sample from the deep well and the spring plot within cluster A, therefore we suggest this group is representative of the groundwater in the area.

Significant variation was observed in $\delta^{13}\text{C}_{\text{DIC}}$ values in CKDu endemic areas from -13.9‰ to -21.9‰ with the lowest average of -19.9‰ in Mahiyanganaya ($n = 7$) and the highest average $\delta^{13}\text{C}_{\text{DIC}}$ of -15.4‰ in Padaviya ($n = 6$). The spring had the lowest $\delta^{13}\text{C}_{\text{DIC}}$ value of -23.3‰ and DIC concentration of 0.006 mmol/L .

Table 3: Environmental isotope results for groundwater samples

Area	ID	$\delta^2\text{H}(\text{‰})$	$\delta^{18}/^{16}\text{O} \text{ (‰)}$	$\delta^{13}/^{12}\text{C}_{\text{DIC}} \text{ (‰)}$
A: Mahiyanganaya	L1	-34.5	-5.94	-20.6
	L2	-27.1	-4.82	-17.2
	L3	-28.0	-5.11	-21.9
	L4	-26.0	-3.92	-18.7
	L5	-31.6	-5.64	-21.0
	L6	-35.5	-5.91	-20.2
	L7	-35.0	-5.83	-19.6
B: Padaviya	L15	-15.0	-2.44	-15.7
	L16	-30.9	-5.33	-19.2
	L17	-32.7	-5.45	-14.5
	L18	-31.2	-5.30	-17.7
	L19	-13.7	-2.12	-13.9
	L20	-31.7	-5.40	-20.1
C: Medawachchiya	L8	-16.0	-2.23	-14.3
	L9	-30.5	-4.89	-15.1
	L10	-35.4	-5.77	-14.7
	L11	-32.7	-5.22	-15.8
	L12	-28.9	-4.57	-17.1
	L13	-17.5	-2.61	-14.8
	L14	-20.8	-2.98	-16.0
D: Rambewa	L22	-36.2	-5.99	-17.3
	L23	-36.3	-5.94	-17.1
	L24	-36.4	-5.99	-17.3
	L25	-32.0	-5.39	-17.6
DGW	L26	-35.2	-5.80	-14.0
WZGW	L27	-30.1	-5.38	[NT]
Spring	L21	-37.7	-6.28	-23.3

5 Discussion

5.1 Hypotheses related to major ion chemistry

The quality of the shallow groundwater in CKDu affected regions in Sri Lanka has been studied for more than 30 years by various researches (Bandara et al., 2008; Chandrajith et al., 2011b; Cooray et al., 2019; Ileperuma et al., 2009; Rango et al., 2015; Wanigasuriya et al., 2008; Wickramaratna et al., 2017). As part of our discussion, we draw upon previous studies and incorporate recently collected hydrochemical and isotopic results from shallow groundwater wells that were part of a reconnaissance survey in CKDu affected regions of Sri Lanka. This

work will provide further clarity to help discount or identify hypotheses associated with water quality in CKDu affected regions that require further research.

Fluoride

An elevated F concentration in the groundwater has been considered a causal factor for CKDu. It has been suggested that the source of F is from bedrock weathering processes (Dissanayake, 1991; Jayawardana et al., 2014; Young et al., 2011). In our study, ~70% of samples had F concentrations above the Sri Lankan guideline (BOI-SL, 2013). The maximum F concentrations observed was 1.27 mg/L (L12) which is much less than in many parts of the world, specifically in areas impacted by limited or overallocated groundwater resources such as in; India (11mg/L; Raju (2017)), northwest China (6.33 mg/L; Chen et al. (2017)), Iran (1.99 mg/L; Dehbandi et al. (2017)), Tunisia (2.4 mg L/1; Guissouma et al. (2017)) and Pakistan (10.8 mg/L; Rahman et al. (2017)) and Ethiopia (15 mg/L; Rango et al. (2017)). Extreme dental and skeletal fluorosis are common in these areas but no reported CKD/CKDu as a result of F toxicity.

Aluminium fluoride complexes (AlF_x) formed by the reaction between F in groundwater and low-grade aluminium cooking utensils has been suggested as a causal factor for the disease. Long term exposure to toxic AlF_x can damage the liver (Strunecká et al., 2002), and blood cells (Rendu et al., 1990). However, there are no reports on damaged liver or blood cells among CKDu patients. Further, CKDu is not evenly distributed over regions with high F concentrations in groundwater, and the use of low-grade aluminium utensils is widespread. The northern part and the southeastern part of the country where F content in the groundwater is greater than 0.6 mg/L, CKDu is not observed. All these evidence suggest high F in the groundwater is not the only factor influencing the occurrence of CKDu.

The formation of NaF_x complexes, which reduced nephrotoxicity of F in the groundwater is limited when Na/Ca ratios are low (1.6 – 6.6), making low Na/Ca ratios with the presence of high F, a risk factor for CKDu (Chandrajith et al., 2011a). The samples collected in this study had very low Na/Ca ratios (0.1 – 1.9) and plotted within the endemic cluster on the Na/Ca vs F graph described in Chandrajith et al. (2011a). However, other groundwaters in Sri Lanka such as those from the north in Jaffna (Chandrajith et al., 2016), and from the wet zone in Matale (Chandrajith et al., 2015), where CKDu is not reported at endemic levels, also plotted inside the endemic cluster (graph not provided) making it difficult to draw conclusions from this assessment.

Hardness

The hardness of the shallow groundwater has been suggested to be a causal factor for CKDu. Most studies, including this one, found hard to very hard groundwater but, this is not exclusive to all CKDu endemic areas. For example, only 14% of samples from the Mahiyanganaya site had hard to very hard groundwater. Similarly, Edirisinghe et al. (2017), also found only 25% of samples from this area to be hard to very hard. Furthermore, groundwaters from the other areas in Sri Lanka not impacted by CKDu had very hard waters such as the intermediate climatic zone with hardness ranging from 15.6 – 1246 mg/L (Rubasinghe et al., 2015), and the Jaffna Peninsula were double the dry zone areas (Bandara et al., 2018; Chandrajith et al., 2016). These results make it difficult to correlate hardness with CKDu prevalence.

Hofmeister series ions

Hofmeister series cations (Ca and Mg) were major cations observed in most of water quality studies in CKDu endemic areas, including our study. However, Ca and Mg concentrations in Jaffna, Mannar and Matale, where CKDu is not reported in epidemic level, are nearly two folds greater than that of CKDu endemic areas (Bandara et al., 2018; Chandrajith et al., 2016; Chandrajith et al., 2015). Furthermore, Hofmeister series anions such as Cl and NO₃ were higher compared to our results in CKDu endemic areas, in Jaffna, Mannar, Huruluwewa and Wellawaya, where there were no records on CKDu (Bandara et al., 2018; Chandrajith et al., 2011a; Chandrajith et al., 2016; Chandrajith et al., 2015; Vithanage et al., 2014). This suggests that the effect of Hofmeister series ions on CKDu prevalence is not obvious.

Silica

It is notable that we found very high silica (Si) concentrations (average 45.7 mg/l) in CKDu endemic regions compared to the wet zone sample (18.1 mg/l). Elevated Si concentrations suggest silicate weathering processes are important, which are likely to be associated with water-rock interaction processes. A number of studies discuss nephrotoxic effects of Si as a result of occupational exposure and injection through drinking water (Ghahramani, 2010; Mascarenhas et al., 2017; Ng et al., 1992). However, Si concentrations in studied waters were less than half of the nephrotoxic Si level (100 – 120 mg/L) observed by Mascarenhas et al. (2017), (e.g. Mahiyanganaya average = 39.44 mg/L and Padaviyaaverage = 50.43 mg/L). Further, the bioavailability of Si is believed to be governed by Mg and Ca content in the water. In the presence of high concentrations of Mg and Ca, Si tends to form insoluble complexes limiting its bioavailability (Mascarenhas et al., 2017). From our initial results, it appears that

elevated Si concentrations need further investigations to understand how Si is affecting the quality of the shallow groundwater in CKDu affected regions.

5.2 Hypotheses related to metals

Metals such as As, Cd and Pb have been identified as contaminants of concern in CKDu - water quality studies. These metals can be found naturally in the groundwater as a result of mineral weathering and can also be induced from anthropogenic activities such as the use of agrochemicals, especially when using triple superphosphate (TSP) which is used extensively in paddy agriculture (Chandrajith et al., 2010).

In our study, Cd concentrations in the shallow groundwater samples were below the detection limit (1µg/L). A number of other studies also found similar results (Levine et al., 2015; Nanayakkara et al., 2019; Rango et al., 2015). We did not analyse the chemistry of surface water samples. However, Diyabalanage et al. (2016) reported over 90% water samples from the Mahaweli River and its tributaries to have Cd concentrations below Sri Lankan drinking water guideline values (SLIS, 2013).

Elevated concentrations of As in the shallow groundwater has been proposed as another causal factor for CKDu (Jayasumana et al., 2013). However, a number of studies including our study reported low concentrations of As in water in CKDu endemic areas (Edirisinghe et al., 2017; Levine et al., 2015; Rango et al., 2015; Rubasinghe et al., 2015). In contrast, high concentrations of As was found in karstic aquifers in Jaffna (15.1 mg/L) (Chandrajith et al., 2016) and Mannar island (Bandara et al., 2018), where paddy and vegetable cultivation is the main livelihood, with no records on endemic CKDu. Furthermore, there was no evidence of Pb in the samples we analysed. Similar results were found by Rango et al. (2015) in the shallow groundwater in CKDu endemic areas. Based on previous data together with this dataset, no evidence suggests metals to be the causal factor for CKDu.

5.3 Hypotheses related to agrochemicals

The use of agrochemicals has been hypothesised as a cause for CKDu. Jayasumana et al. (2015c) argued that chelates formed by nephrotoxic elements, especially As with glyphosate, in hardwater is a causal factor for CKDu. However, low concentrations of heavy metals, glyphosate and absence of CKDu in agricultural regions in the northern part of Sri Lanka that have hardwater and higher concentrations of As (Bandara et al., 2018; Chandrajith et al., 2016;

Gunarathna et al., 2018; Jayasumana et al., 2014b) compared to CKDu endemic areas, do not provide evidence of this hypothesis.

Other geochemical indicators of agrochemical pollution in the groundwater are elevated concentrations of nitrate and phosphate. In this study, 96% of samples from CKDu endemic areas had phosphates concentrations below the detection limit (1 mg/L). Much higher concentrations of phosphate were found by Rubasinghe et al. (2015) and Chandrajith et al. (2011a) as a result of intense use of phosphate containing fertiliser such as single and triple superphosphates. Nitrate was low in our study; water from Mahiyanganaya had nitrate concentrations below 0.1 mg/L and the other three areas also had nitrate concentrations below the maximum allowable limit in drinking water (50mg/L).

The low concentration of nitrate and phosphate does not necessarily indicate that groundwaters are not impacted by agrochemicals. For example, these nutrients may bind to organic matter, be processed by algae present in the wells or be denitrified (Ahearn et al., 2005; Gardner et al., 2011; Royer et al., 2004). A number of samples (n=7) had nitrite concentrations that ranged from 0.03 – 2.05 mg/L. This may indicate denitrification processes. In the presence of dissolved oxygen such as the waters we sampled, aerobic denitrification bacterias such as *Pseudomonas stutzeri*, *Pseudomonas mendocina* and *Pseudomonas putida* can initiate the denitrification process (Lv et al., 2017). Sampling was completed during the dry season (July) when agricultural activities were not taking place, which may also result in low nitrate and phosphate concentrations in water. Chunnakam aquifer in Jaffna (Vithanage et al., 2014) and tank/irrigation water in NCP (Wijesundara et al., 2012; Wijesundara et al., 2013) also reported lowest nitrate and phosphate concentrations during the month of July reflecting the influence of agriculture and rainfall in ground and surface waters.

Apart from denitrification algae can produce cyanotoxins. Manage (2019) discussed cyanotoxins in surface and the groundwater in CKDu endemic areas as a risk factor for CKDu. DOC is another causal factor discussed in relation to the CKDu (Makehelwala et al., 2020). However, in our study we did not measure the organic matter in the groundwater. Significant quantities of DOC can be added into groundwater in areas like dry zone of Sri Lanka, where intense agricultural activities, high groundwater level fluctuations and wet laterins observed (Clark and Fritz, 1997). Therefore, organic matter in the groundwater needs further investigations to understand possible contaminations and nutrient cycling in the groundwater and finally to understand the quality of the groundwater in CKDu affected regions.

5.4 Hypotheses relating to Water sources

The stable isotopes of ^{18}O and ^2H have been used in CKDu affected regions to infer the source(s) of groundwater water. Edirisinghe et al. (2017), found that the groundwater in the dry zone is recharged either by (1) rainwater or (2) both rainwater and surface water sources. They argue that the groundwater in CKDu affected regions are replenished by rainwater and not subjected to evaporation. In contrast Wickramarathna et al. (2017), argued that the groundwater in CKDu endemic areas is mixing with evaporated surface water sources as stable water isotope composition of the groundwater is similar to that of surface water sources.

In our study, we found two distinct water types, cluster (A), which are isotopically depleted waters plotting close to the LMWL and the GMWL representing rainfall recharge. The deeper groundwater sample (>60 m) and the Kebithigollewa spring plot within this group, representing the groundwater signal for the area (Fig. 3). Waters in cluster A have been recharged by rainfall and have not experienced evaporation. The second group (B) plot below both LMWL and GMWL on a regression line of $\delta^2\text{H} = 5.36 \delta^{18}\text{O} - 3.38\text{‰}$ suggesting they have experienced evaporation. Isotopic enrichment due to evaporation is largely dependent on the humidity (h). In high humid conditions (>70%) like in the dry zone of Sri Lanka, the slope of the graph, $\delta^{18}\text{O}$ vs $\delta^2\text{H}$ is greater than 5 (Clark and Fritz, 1997). We observed a slope of 5.36 in the $\delta^{18}\text{O}$ vs $\delta^2\text{H}$ graph consistent with evaporation. These results are consistent with findings of Edirisinghe et al. (2017).

In their study, Edirisinghe et al. (2017), observed some of the shallow groundwater wells in CKDu endemic areas with isotopically enriched waters compared to the LMWL. In contrast, Wickramarathna et al. (2017) found that all studied groundwaters in CKDu endemic areas are isotopically enriched compared to the LMWL (Fig. 3). They argued that mixing of the groundwater with infiltrating isotopically enriched surface water creates the enriched isotopic signature. These values could also be a result of evaporation in the open well, which is directly in contact with the atmosphere. It is worth noting that samples collected in all three studies were from shallow groundwater wells, which are open to atmosphere via a 2 to 3 m diameter well head hence, in-situ direct evaporation should be considered when using stable water isotopes for understanding water sources.

6. Further Recommendations

All previous studies have shown that the physicochemical properties of the groundwater vary between sample locations in CKDu affected regions. Waters from CKDu affected regions do

not have a distinct hydrochemical fingerprint. This seems reasonable when considering the different recharge mechanisms, anthropogenic activities such as agrochemical application and/or irrigation practice, sanitation practices and different hydrogeological environments. In order to understand the water quality of the groundwater of the area, it is essential to design studies that capture the dynamics of potential contaminants of concern. For example, nitrate in river water is highly dynamic. A weekly sampling of the river Thames shows that nitrate peaks are missed. The elevated concentration of nitrate in the river was only captured at daily sampling frequency. In Sri Lanka, hydrology is even more complex where the water table fluctuates 2-5 m seasonally due to monsoonal conditions. For example, nitrate and phosphate in tank/irrigation water in NCP closely resemble the rainfall pattern of the area and peaked during April to May and October to November (Wijesundara et al., 2012; Wijesundara et al., 2013). Studies designed to measure chemistry of the groundwater as a single sample round or snap-shot will not capture the true complexity of the system, therefore regular sampling campaigns are advised. It is also important to note that the groundwater samples need to be collected from monitoring wells designed to sample a targeted section of the aquifer (i.e. with short less than 3 m screened intervals).

Not only does sample design need to be considered but a more comprehensive suite of analysis be measured, such as organic matter character especially carbon isotopes of organic and inorganic carbon to understand sources and how carbon is cycled (Meredith et al., 2016) and is utilised by the microbes in the water. We measured the $\delta^{13}\text{C}_{\text{DIC}}$ values, and they ranged between -13.9‰ and -21.9‰ with an average of -17.3‰, suggesting carbonate mineral dissolution is not a major process in these waters, but the dissolution of soil CO_2 . Dissolution of carbonate minerals such as calcite and dolomite can decrease the $\delta^{13}\text{C}_{\text{DIC}}$ values to around 0‰. The observed range of $\delta^{13}\text{C}_{\text{DIC}}$ values is likely to relate to the sediment interaction processes for each site, therefore identifying how varying climatic conditions influence these sites will give us further information on the processing of the water. Measuring groundwater residence time tracers such as ^3H , ^{14}C of both inorganic and organic carbon to understand the transit time within the wells would also be beneficial. Additionally, time-series analysis using ^3H data to monitor the variability in water isotopes will also provide additional information on contaminant dynamics and residence times.

The use of agrochemicals has been of concern in CKDu affected regions, therefore using isotopic and geochemical tracers such as nitrogen and sulphur isotopes together with Rare Earth Elements (REE) to identify sources and transport of agrochemicals through the environment is

suggested. Other stable isotopes such as Li, Sr and B will also be useful in understanding water sediment processes (Meredith et al., 2013). From our initial results, we identify that Si needs further investigation, and we suggest the use of Sr and Si isotopes to understand silicate weathering processes. Many fundamental water quality tracers have not been measured in CKDu affected regions, and comprehensive time series analysis has not been published.

7 Conclusions

In this paper, updated water quality research related to CKDu in Sri Lanka is presented. We identified the gaps in the literature, and draw inferences from previous and current water quality datasets to guide future collaborative efforts in water quality research in CKDu affected regions in the future. A comprehensive water quality study including fundamental isotopic tracers, considering natural and anthropogenic influences on the groundwater quality such as seasonal conditions, varying lithologies, recharge events and agriculture is needed.

Although most of the previous studies show similar results to this study, available temporal, and spatial scale of the water chemistry datasets, including this study, is not sufficient to support any postulated hypothesis. Our results confirm no evidence of heavy metal contamination, including As, Cd, and also Pb or agrochemical pollution in the dry season sampled from the groundwater sources. The study emphasises the importance of environmental isotopes and provides guidance on the type of isotopic tracers and the frequency of sampling that is needed to capture potential pollutants. Furthermore, detailed investigation into the degradation products of the agrochemical, organic matter and Si concentrations is needed. Outcomes of this research will encourage consistent sampling guidelines and protocols in CKDu affected regions to better understand degradation pathways and residence times of contaminants.

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