The influence of water–rock interactions on household well water in an area of high prevalence chronic kidney disease of unknown aetiology (CKDu)

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Poor drinking water quality in household wells is hypothesised as being a potential contributor to the high prevalence of chronic kidney disease of uncertain aetiology (CKDu) in Sri Lanka. One of the natural processes that can affect water quality is the dissolution of minerals contained within an aquifer by water–rock interactions (WRIs). Here we present a comprehensive assessment of WRIs and their influence on the water chemistry in household wells and spring waters in the Medawachchiya area by combining measurements of environmental isotopes, such as strontium, lithium and stable carbon isotopes and inorganic chemistry parameters, and modelling geochemical mass balance reactions between rainfall and groundwater samples. Our results reveal the presence of strontium, dissolved from both silicate and carbonate minerals, with high isotopic (87Sr/86Sr) ratios of up to 0.7316. Geochemical mass balance modelling and prior 87Sr/86Sr studies on the Wanni Complex bedrock suggest these strontium values may be the result of biotite dissolution. We also identify lithium and uranium contributed from the dissolution of silicates, albeit at concentrations too low to constitute a known health risk. In contrast, the levels of magnesium and calcium in our samples are high and demonstrate that, despite the felsic bedrock, well water chemistry in the Medawachchiya area is dominated by carbonate dissolution.

INTRODUCTION

Chronic kidney disease of uncertain aetiology (CKDu) in Sri Lanka was first reported in 1996. The disease currently has a multilevel clinical case definition, whereby disease conditions known to be associated with kidney failure, such as diabetic nephropathy, infectious disease, genetic conditions and others, are excluded. One hypothesis is that drinking water contaminants may be a causal or contributing factor to the disease. These include natural drinking water contaminants and contaminants from pesticides – pathogens and microbial toxins and inorganic chemistry parameters, and modelling geochemical mass balance reactions between rainfall and groundwater samples. Our results reveal the presence of strontium, dissolved from both silicate and carbonate minerals, with high isotopic (87Sr/86Sr) ratios of up to 0.7316. Geochemical mass balance modelling and prior 87Sr/86Sr studies on the Wanni Complex bedrock suggest these strontium values may be the result of biotite dissolution. We also identify lithium and uranium contributed from the dissolution of silicates, albeit at concentrations too low to constitute a known health risk. In contrast, the levels of magnesium and calcium in our samples are high and demonstrate that, despite the felsic bedrock, well water chemistry in the Medawachchiya area is dominated by carbonate dissolution.

Studies into natural groundwater contaminants in CKDu-affected regions of Sri Lanka have identified higher concentrations of fluoride (F), calcium (Ca) and magnesium (Mg) in Anuradhapura groundwater and lower levels of heavy metals such as cadmium (Cd), arsenic (As) and lead (Pb) compared to the World Health Organisation (WHO) drinking water guideline value and the Sri Lankan potable water standard. Groundwater chemistry in Sri Lanka is largely determined by water–rock interactions (WRIs), ion exchange and precipitation processes. Natural solutes enter groundwater through dissolution processes, which are enhanced by the input of carbon dioxide, sulfur dioxide and nitrogen oxides into infiltrating rainwater. The concentrations and composition of major ions in groundwater are dependent upon the type of minerals the water interacts with, their resistance to weathering, water residence times and the chemistry of the initial water. Exchange processes also alter the concentrations of major ions. Within aquifers, Ca and Mg take the place of adsorbed Na on aquifer solids through cation exchange processes since the interaction between sorption sites and divalent ions is typically much higher than monovalent ions. This results in an increase in dissolved Na concentrations and a decrease in dissolved Ca and Mg concentrations.

The study of certain environmental isotopes can further enhance the understanding of these WRI processes and provide valuable information on the water source, the types of minerals groundwater has interacted with, the duration of interaction and information on processes, including evaporation, sorption and biological respiration. Strontium isotopic ratios (87Sr/86Sr) in groundwater reflect the 87Sr/86Sr ratios of the rock source, since dissolution and other natural processes do not cause fractionation. The 87Sr/86Sr of carbonates reflects that of Sr contained in the water that they were precipitated from. Over the past 500 million years, the 87Sr/86Sr of seawater has varied approximately from 0.7068 to 0.7092, thus the dissolution of carbonates results in groundwater Sr ratios close to these values. In igneous rocks, the 87Sr/86Sr ratio is heavily dependent upon the concentration of rubidium (Rb) incorporated into the rock, due to the decay of 87Rb to 87Sr over time. Old silicate minerals can, therefore, contain high 87Sr/86Sr ratios, with dissolved 87Sr/86Sr values reaching well above seawater ratios and up to 0.7625 previously identified in fresh waters.

Stable lithium isotopes (δ7Li) provide additional information on silicate weathering, as silicate minerals have been identified as the predominant source of Li in waters, even in carbonate-dominated environments. The δ7Li values in continental crust ranges from approximately −5‰ to +5‰; however, preferential dissolution of Li and preferential adsorption and incorporation of Li into secondary minerals results in a higher and much broader range of values in river water (+5‰ to +43‰) and groundwaters (+7‰ to +37‰). Values as high as +1225‰ have also
been reported in groundwater from a peat bog influenced by
agrochemicals.\textsuperscript{44}

The aim of this research is to contribute to a more detailed
description of the WRI in spring waters and domestic well waters in
CKDu-affected regions in order to obtain a better understanding of
how specific hydrological and hydrogeochemical factors affecting
groundwater might contribute to poor drinking water quality and
distinguish them from other non-hydrogeological causes of
drinking water contamination. A comprehensive isotopic analysis
of the spring water and domestic well waters in CKDu-prevalent
regions of Sri Lanka has not yet been undertaken. Here, using
$^{87}$Sr/$^{86}$Sr, $\delta^{7}$Li and dissolved inorganic carbon ($\delta^{13}$C\textsubscript{DIC}) combined
with major ion analyses, we show how WRIs affect water quality of
30 domestic well and spring waters used for drinking water
purposes in a CKDu-impacted region, Medawachchiya, Sri Lanka.

RESULTS

Geochemistry

Water chemistry data are presented in Supplementary Tables 1
and 2. Well waters were less fresh and contained higher electrical
cconductivity (EC, between 519 and 2095 $\mu$S cm$^{-1}$, $n =$
28) than spring waters (between 154 and 467 $\mu$S cm$^{-1}$, $n =$
2). Average dissolved oxygen (DO) and alkalinity measurements
were 3.45 mg L$^{-1}$ $\pm$ 2.60 (1σ) and 376.3 mg L$^{-1}$ $\pm$
134.39 (1σ), respectively, for the well waters, and 2.4 mg L$^{-1}$ $\pm$
0.93 (1σ) and 122.8 mg L$^{-1}$ $\pm$ 76.93 (1σ), respectively, for the
spring waters. The lowest EC measurement was observed in sample
30 spring water, and the highest EC measured in sample 27 (2096 mg
L$^{-1}$) well water. pH measurements ranged from acidic (4.92) in sample 30 to mildly
alkaline (8.06) in sample 27.

Cluster analysis was used to group the samples for anion and
cation data. The analysis shows three distinct clusters of samples
(Fig. 1a). Cluster 1 is comprised of 21 samples representing the
highest Sr/Cl ratio, Sr/Na ratio and K concentrations, and the
lowest Li concentrations (see Supplementary Table 3). Cluster 2
contains 6 samples representing the highest Sr, Cl, Na, Ca and Mg
concentrations and lowest Li/Na ratios (Supplementary Table 3).

The high concentrations of major ions in samples from Cluster 2
relative to the other cluster groups suggests that these samples
may have undergone a higher relative amount of WRI than the
other samples. Cluster 3 is comprised of 3 samples representing
the lowest Cl, Sr, Na, Ca, Mg, Si and K concentrations and the
highest Li concentrations and Li/Na ratios (Supplementary Table 3).
This group contains the two spring water samples (samples 13 and
30, Fig. 1a), and likely represent samples that have undergone
the least amount of WRI due to the low concentrations of major
ions relative to the other groups. These groupings are also
confirmed using a principal component analysis (Fig. 1b).

All water types are mixed (Fig. 1c), with Cluster group 3 samples
notably comprised of waters low in Cl. The dominant anion is
HCO$_3^-$ and the dominant cation is Na. Other ions that contribute
significantly to EC in the samples are Cl, SO$_4^2-$ and Mg. There is
a clear trend of increasing Ca with increasing EC, with three outliers
present (samples 27, 16 and 18) that display reduced Ca relative to
EC compared to the other samples (see Supplementary Fig. 1a).
These three samples contain high $^{87}$Sr/$^{86}$Sr ratios, high EC, high Na
and high U compared to the other samples.

Fluoride concentrations in the Medawachchiya region are high
(min $= 0.07$ mg L$^{-1}$, max $= 3.7$ mg L$^{-1}$, average $= 1.2$ mg L$^{-1}$,
$n =$ 30) compared to the WHO guideline value for drinking water
and the Sri Lankan potable water standard\textsuperscript{48}, with high values in
the region also reported in other studies\textsuperscript{30,49}. We observe a trend of
increased F concentrations with decreased Ca/Na ($r =$ $-0.60$,
$p =$ $5.18 \times 10^{-4}$, $n =$ 30) and Mg/Na ratios ($r =$ $-0.54$,
$p =$ $1.97 \times 10^{-3}$, $n =$ 30, see Fig. 1b), suggesting a silicate F source. Reduced
Ca/Na and Mg/Na ratios associated with a silicate end-member has
been identified in other studies\textsuperscript{47,48}.

The relationship between major ions and Cl are shown in Fig. 2.
Most samples contain elevated Na, Mg, SO$_4^2-$ and HCO$_3^-$ relative to
Cl, suggesting a source of these ions in the aquifer. Elevated SO$_4^2-$
relative to Cl suggests the dissolution of gypsum or metamorphic
minerals. One sample (sample 01) shows particularly high K
relative to the other samples (Fig. 2c) and may indicate input from
an anthropogenic source, such as agrochemical application. Group
1 and Group 2 samples are characterised by low K relative to Cl,
which may indicate biotic uptake of K. Group 1 displays high Mg
(Fig. 2b), HCO$_3^-$ (Fig. 2f) and several samples with high Ca relative
to Cl (Fig. 2d) indicating likely carbonate and/or feldspar
dissolution. Two Group 2 samples in the lower right side of
Fig. 2d (samples 27 and 16) have the lowest Ca/Cl ratios (0.15 and
0.17, respectively, suggesting carbonate precipitation.

Groundwater Si concentrations usually range from 1 to
30 mg L$^{-1}$\textsuperscript{49}. We identify high Si concentrations in this study, with
Si concentrations ranging from 17.0 to 58.4 mg L$^{-1}$, with an average
of 45.1 mg L$^{-1}$ $\pm$ 10.90 (1σ) suggesting that a high level of WRI has
taken place in some samples. In the well waters, Sr concentrations
ranged from 0.21 to 1.52 mg L$^{-1}$ (n = 28), while Sr concentrations in
the spring waters were comparatively low and ranged from 0.05 to
0.23 mg L$^{-1}$ (n = 2). The Rb concentrations in all samples were low,
between $<0.001$ and 0.016 mg L$^{-1}$ for well waters (n = 28) and
0.002 mg L$^{-1}$ for the two spring water samples. The Li concentra-
tions ranged from 0.002 to 0.016 mg/L in the well waters and from
0.004 to 0.008 mg/L in the two spring waters.

Average $^{87}$Sr/$^{86}$Sr ratios were highest in Group 2 (0.717) and
lowest in Group 1 (0.713), with $\delta^{7}$Li (‰) values highest in Group 2
(+18.9‰ $\pm$ 6.1) and lowest in Group 3 (+7.3‰ $\pm$ 8.6, see Supple-
mentary Table 4). Notably, the lowest and highest $\delta^{7}$Li values
were found in samples 27 and 16 respectively. The $\delta^{13}$C\textsubscript{DIC}
values ranged from $-0.002$ to $-14.79‰$ to
$-19.99‰$ (Supplementary Table 4). The lowest value is found
in Group 3 in the freshest spring water sample (sample 30) at
$-23.59‰$, which may indicate DIC originating from an organic source, for
example through biodegradation of soil organic matter\textsuperscript{35}.

Geochemical mass balance modelling

The NETPATH computer program was used to interpret net
geochemical mass balance reactions between initial and final
waters along a user-defined chemical evolution flow path in the
aquifer. Geochemical mass balance modelling was used to quantify
the amount of carbonate dissolution occurring after rainfall is recharged and forms the observed groundwater sample.

Samples 02, 07 and 23 were considered to be the most influenced
by carbonate dissolution. It was calculated that between 0.61 and
1.71 mmol kg$^{-1}$ of calcite was needed to produce the resultant
water chemistry, if considering rainwater was the starting water
and according to the models in Supplementary Table 5. The
model that best predicted the measured $\delta^{13}$C\textsubscript{DIC} value was sample
23. This model showed that the dissolution of 0.94 mmol kg$^{-1}$
of calcite, influx of 2.2 mmol kg$^{-1}$ of CO$_2$ dissolution of 1.3 mmol
kg$^{-1}$ of MgSiO$_3$, addition of 7.4 mmol kg$^{-1}$ of NaCl from within the
aquifer and dissolution of 0.5 mmol kg$^{-1}$ of hornblende into the
rainwater recharge and the removal of 1.9 mmol kg$^{-1}$ of Ca/Na by
exchange and removal of 2.3 mmol kg$^{-1}$ of SiO$_2$ from the water
best describes the hydrochemical evolution of the waters most
influenced by carbonate processes.

Mass balance modelling was also used to model samples 16, 18
and 27, which were considered most influenced by silicate
weathering processes. Similar starting water, assumptions, con-
straints and phases were used as describe above. From these
models, it was found that higher amounts of calcite (1–2.5 mmol
kg$^{-1}$) were needed to produce the resultant water chemistries. If
calcite was not added to the models, the calculated $\delta^{13}$C\textsubscript{DIC} values

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were between 3 and 8‰ more depleted than the measured values. The model that best predicted the measured $\delta^{13}C_{DIC}$ value for these three samples was sample 18. This model showed that the dissolution of 1.3 mmol kg$^{-1}$ of calcite, dissolution of 0.14 mmol kg$^{-1}$ of biotite, influx of 6.0 mmol kg$^{-1}$ of CO$_2$, addition of 2.4 mmol kg$^{-1}$ of Ca/Na through exchange, dissolution of 5.8 mmol kg$^{-1}$ of MgSiO$_3$ and addition of 2.9 mmol kg$^{-1}$ of NaCl to the rainwater and the removal of 5.5 mmol kg$^{-1}$ of SiO$_2$ best describes the hydrochemical evolution of silicate influenced waters. Models suggest that all waters in this region are influenced by calcite dissolution.

**DISCUSSION**

Strontium typically retains the same ratio of $^{87}$Sr/$^{86}$Sr during dissolution into groundwater as its source mineral. The $^{87}$Sr/$^{86}$Sr ratios in minerals are dependent upon the mineral type, age and rubidium content of the rock, as well as the proportion of crustal and mantle sources for igneous rocks and amount of alteration of carbonates and evaporites$^{50}$. In groundwater, the $^{87}$Sr/$^{86}$Sr ratios represent the relative proportions and $^{87}$Sr/$^{86}$Sr ratios of the dissolved input. For example, where Sr input is 50% from carbonate dissolution where the carbonate $^{87}$Sr/$^{86}$Sr ratio is 0.705 and 50% from silicate dissolution where the $^{87}$Sr/$^{86}$Sr ratio is 0.729, the groundwater $^{87}$Sr/$^{86}$Sr ratio observed will be approximately 0.717 (see Fig. 3). The $\delta^{7}$Li, however, fractionates more readily in groundwater through dissolution and adsorption processes, with $^7$Li preferentially dissolved$^{51}$ and $^6$Li preferentially adsorbed to clay mineral surfaces$^{52}$. This results in increasingly positive $\delta^{7}$Li values with water-sediment processing (Fig. 3). Strontium isotopes can therefore be used to understand silicate weathering processes, and when used with the $\delta^{7}$Li values, the secondary processes can be defined. For example in sample 30
The freshest spring water sample, the \(^{87}\text{Sr}/^{86}\text{Sr}\) ratio of 0.713 is observed, along with a very low \(\delta^{7}\text{Li}\) value for fresh water \((-1.8\,\text{‰})\). This value could be indicative of 50% Sr from carbonate dissolution and 50% from silicate weathering as well as the dissolution of weathering products containing high amounts of \(^{6}\text{Li}\), such as clay minerals from nearby soils.

Figure 4a shows that the increasing Sr concentrations associated with both increasing and decreasing \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios are likely the result of silicate and carbonate weathering, respectively. Currell and Cartwright\(^{53}\) identified carbonate and silicate weathering in groundwater samples by comparing \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios and \(\delta^{13}\text{CDIC}\) values. Increasing enrichment in \(\delta^{13}\text{CDIC}\) without an increase in \(^{87}\text{Sr}/^{86}\text{Sr}\) suggested carbonate input, while increasing \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios not associated with enrichment in \(\delta^{13}\text{CDIC}\) \(\approx (-1.8\,\text{‰})\) indicated silicate inputs. This agrees with our findings which demonstrate that samples trending towards silicate input in Fig. 4a display increased \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios without an increase in \(\delta^{13}\text{CDIC}\). Figure 4b also shows that samples trending towards carbonate input in Fig. 4a display an increase in \(\delta^{13}\text{CDIC}\) without an increase in \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios. While carbonates are not expected to comprise a large portion of the geology in the region, they weather more readily with the input of the naturally weakly acidic rainwater, and thus can be a large source of Sr. Dissanayake and Weerasooriya\(^{54}\) report weakly acidic rainwater pH values of \(<5.5\) in the study region, which is supported by the pH value of 4.9 observed in sample 30 from

![Fig. 2 Bivariate plots. C1 vs a Na, b Mg, c K, d Ca, e SO\textsubscript{4} and f HCO\textsubscript{3}. The black line on each plot indicates the ratio of Cl to each ion present in rainwater from Vavuniya, north of the study site. Points above the line indicate enrichment in the major ion compared to chloride, suggesting that the source of the ion is from mineral weathering rather than rainwater. Points below the line suggest depletion of the ion compared to Cl, indicating removal through an exchange process or mineral precipitation process.](image)

![Fig. 3 Conceptual figure of an aquifer showing processes impacting on \(\delta^{7}\text{Li}\) and \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios in groundwater. The \(^{7}\text{Li}\) is preferentially dissolved into groundwater from Li bearing minerals such as granite, with \(^{6}\text{Li}\) preferentially adsorbed to mineral and clay surfaces, resulting in increasingly high \(\delta^{7}\text{Li}\) values in groundwater with processing. The \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios do not fractionate with dissolution, therefore ratios in groundwater are the result of the proportion of Sr from the dissolution of silicates and carbonates.](image)
the HCO₃- feldspathic gneiss and granites containing 1% carbonate, 82% of the hypothesis. resistant to weathering; however, a groundwater residence time and 18, allowing for the dissolution of silicates that are more could be a result of longer water residence times for samples 27, 16 carbonate inputs in samples affected by silicate weathering. This the additional weathering of silicates, rather than a lack of that the differences observed between the two groups are due to result of silicate weathering have also been demonstrated in both Blum et al.24 demonstrated that, in quartz- samples, suggesting that it has not been buffered to a higher pH by carbonate dissolution. The 87Sr/86Sr ratios of 0.705 have been reported in whole-rock flux was derived from weathering of the carbonates. Similarly, our mass balance modelling results reveal that the samples most influenced by carbonate and silicate weathering, respectively, have all undergone carbonate dissolution processes, with calcite inputs between 0.61 and 2.54 mmol kg⁻¹. This suggests that the differences observed between the two groups are due to the additional weathering of silicates, rather than a lack of carbonate inputs in samples affected by silicate weathering. This could be a result of longer water residence times for samples 27, 16 and 18, allowing for the dissolution of silicates that are more resistant to weathering; however, a groundwater residence time indicator such as tritium would be required to confirm this hypothesis.

The 87Sr/86Sr ratios of 0.705 have been reported in whole-rock carbonatite samples from the Wanni Complex, with relatively low ratios (0.710) also identified in the felsic fraction of migmatite gneiss in the region56. In contrast, 87Sr/86Sr ratios from biotite in carbonatite rock from the Wanni Complex contain much higher 87Sr/86Sr ratios (approximately 0.729), which our mass transport modelling suggests is likely to be contributing to the high 87Sr/86Sr ratios in samples 21, 6, 24, 18 and 16 as shown in Fig. 4a. These samples also contain higher U concentrations (Fig. 4c) indicating potential hydrolysis of silicate minerals due to carbonic acid to produce soluble uranyl carbonate complexes or assimilation of uranium in the bedrock46. Increasing 87Sr/86Sr ratios as a result of silicate weathering have also been demonstrated in both river catchments33 and groundwaters53. Figure 4d shows a corresponding increase in Li concentration with increasing 87Sr/86Sr ratios (rₛ = 0.76, p = 3.85 × 10⁻⁴) indicating Li input from weathering of silicates.

Dissolution of Li in the absence of adsorption mechanisms would be expected to result in a linear correlation between δ¹³Li and Li concentration. We do not observe a significant correlation between δ¹³Li and Li concentration (rₛ = −0.27, p = 0.22) in the current study, which suggests that there are multiple processes affecting the δ¹³Li values in groundwater in the Medawachchiya region. Notably, the sample with the highest silicate input and highest 87Sr/86Sr ratio observed in Fig. 4a (sample 16) has the highest δ¹³Li value and highest EC value but not the highest Li concentration (Fig. 4d), suggesting that Li adsorption may have taken place.

The 87Sr/86Sr ratios found in our study indicate that silicate weathering appears to be the main source of U and Li input in household well waters in the Medawachchiya region. As the water chemistry of most samples in this study appears to be controlled by carbonate dissolution, U and Li concentrations are low (<0.001 mg L⁻¹ for 83% of samples and <0.01 mg L⁻¹ for 77% of samples, respectively). In contrast, we observe concentrations of F that are higher than guideline values, with 57% of samples over the suggested optimal level of 1.0 mg L⁻¹ of F in Sri Lankan drinking water (SL614:2013)55. High F levels are known to result in skeletal fluorosis in the dry zone of Sri Lanka57 and are hypothesised to contribute to CKDu58. The significant negative correlation between F and Ca/Na and Mg/Na ratios suggests a silicate source. Experimental data from Chae et al.59 provide evidence that the concentrations of F in groundwater are also highly dependent on Ca concentrations, with increasing Ca inputs resulting in decreasing

![Fig. 4 Biplots.](https://example.com/fig4.png)

**Fig. 4 Biplots.** a Biplot of 87Sr/86Sr ratio (n = 28) on y-axis with the inverse of Sr concentration (1/Sr (mmol L⁻¹)) on the x-axis. Upward and downward trends on the y-axis with increased Sr concentration indicate silicate and carbonate inputs, respectively. b 87Sr/86Sr ratios vs δ¹³C DIC. Plot highlights that samples displaying increased 87Sr/86Sr with minimal enrichment in δ¹³C DIC are associated with silicate weathering. Increasing enrichment in δ¹³C DIC with minimal increases in 87Sr/86Sr are associated with carbonate input. c Biplot with samples trending towards silicate inputs are associated with increased U concentrations. d Biplot of δ¹³Li on y-axis (n = 21) with the inverse of Li concentration (1/Li (mmol L⁻¹)) on x-axis. Scale bar represents 87Sr/86Sr ratios with dark blue points indicating lower 87Sr/86Sr (carbonate inputs) and higher Li concentrations and lighter blue points indicating higher 87Sr/86Sr (silicate inputs) and lower Li concentrations. NB: no 87Sr/86Sr value available for the grey data point (sample 29) shown to the lower left.
dissolved F concentrations due to the precipitation of fluorite. We identify high water hardness in the groundwater from the study area, which we propose are not only due to carbonate inputs but also from high silicate weathering, both natural hydrogeochemical processes. We identify only one sample that exceeded the most stringent drinking water limit available of 1.5 mg L\(^{-1}\) Sr. The 1.5 mg L\(^{-1}\) limit is based on the United States Environmental Protection Agency’s preliminary regulatory determinations for contaminants on the third drinking water contaminant candidate list (www.govinfo.gov/app/details/FR-2014-10-20/2014-24582) and is based on safe limits for consumption in sensitive populations such as those with renal failure and other health issues. Our results suggest that U, Li and Sr are unlikely contributors to CKDu due to their low concentrations.

The \(^{87}\text{Sr}/^{86}\text{Sr}, \delta^7\text{Li} \text{ and } \delta^{13}\text{C}_{\text{DIC}} \text{ isotopes enhance the understanding of WRI, which cannot be understood by using major ions alone. This study uses }^{87}\text{Sr}/^{86}\text{Sr}, \delta^7\text{Li} \text{ and } \delta^{13}\text{C}_{\text{DIC}} \text{ values to demonstrate WRI in drinking water wells in a CKDu-affected region of Sri Lanka. Our study demonstrates that }^{87}\text{Sr}/^{86}\text{Sr} \text{ ratios and } \delta^{13}\text{C}_{\text{DIC}} \text{ provide clear indication of the input of silicates and carbonates, with carbonate dissolution predominantly controlling the water chemistry in the region. The } \delta^7\text{Li} \text{ isotopes, in contrast, are more complex and relatively unstudied in groundwater. We conclude that } \delta^7\text{Li} \text{ data in household well waters in Medawachchiya are impacted by several different processes, which results in a lack of clear correlation between } \delta^7\text{Li} \text{ and Li concentration. The Li and U input from silicate minerals, however, is observed through the positive correlation between }^{87}\text{Sr}/^{86}\text{Sr} \text{ ratios and Li concentration, and the increase in U concentration in samples with high }^{87}\text{Sr}/^{86}\text{Sr} \text{ ratios. These results highlight the importance of isotopic analyses in determining WRI and sources of solutes in drinking water. While our results suggest that U, Li and Sr are unlikely contributors to CKDu due to their low concentrations, this could be confirmed with time-series analyses to determine the seasonal variability of these parameters.}

MATERIALS AND METHODS

Environmental setting

Three main climatic zones are observed in Sri Lanka, defined based on the mean rainfall quantities received per year. These include the “Dry Zone” (<1750 mm year\(^{-1}\)), “Intermediate Zone” (1750–2500 mm year\(^{-1}\)) and “Wet Zone” (>2500 mm year\(^{-1}\)) (Fig. 5). Medawachchiya is in the Dry Zone and receives an average rainfall of 1240 mm year\(^{-1}\), largely in October, November and December where long-term (1960–1985 CE) average monthly rainfall rates are between 206 and 247 mm\(^{60}\). Temperatures are relatively stable throughout the year, with an average minimum monthly temperature of 20.3 °C in January and a maximum average monthly temperature of 34.0 °C in April in Vavuniya\(^{60}\), which is located approximately 22 km north of the Medawachchiya sampling region.

The geology in Sri Lanka is mainly comprised of three major high-grade metamorphic rock units: the Highland Complex, Wanni Complex, Vijayan Complex, and the Kadugannawa Complex (KC), and the Mesozoic unit to the north-west. Map adapted from Cooray\(^{66}\). Inset shows the sample locations (black diamonds) within the Medawachchiya district (black outline). The geology in the region is predominantly comprised of gneiss including charnockitic gneiss and biotite gneiss, with granitic gneiss, microcline, quartzite and quartz schist\(^{67}\). Samples 1–12 and 14–29 represent well water samples, while samples 13 and 30 represent spring water samples. Dry (<1750 mm year\(^{-1}\)), intermediate (1750–2500 mm year\(^{-1}\)) and wet (>2500 mm year\(^{-1}\)) zones are defined by the blue and orange lines.
Hydrogeochemical models were constructed using C, Mg, Cl, Ca, Na, redox and Si as constraints. The modelled phases were organic matter (CH_{2}O), CO_{2}(g), hornblende, Mg/Na Ex, MgSiO_{3}, SiO_{2}, NaCl (from atmospheric sources), biotite, Ca/Na exchange and calcite. The pH and major ion (Ca, Mg, Na, K, Cl and SO_{4}) concentrations from a rainwater sample from Vavuniya, also located within the Dry Zone approximately 23 km north of the study area, was used as the initial water. The system is assumed to be receiving piston flow recharge and the groundwater system is in chemical equilibrium. Furthermore, carbon mass balance using Rayleigh distillation equations for all incoming carbon sources and all isotopically fractionating outgoing carbon phases were calculated. Models were assessed for accuracy by comparing the observed and computed δ^{13}C values. Results produced by each model and most fell within ±2‰ for the difference between modelled and measured δ^{13}C values. Models that failed to predict the δ^{13}C value were not used in the interpretation.

DATA AVAILABILITY

The full data set used in this study is provided in Supplementary Information.

CODE AVAILABILITY

The code used to develop individual figures is available upon request to the corresponding author.

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hydrogeochemicalmodelswereconstructed using C, Mg, Cl, Ca, Na, redox and Si as constraints. Themodeled phases were organic matter (CH_{2}\text{O}), CO_{2}(g), hornblende, Mg/Na Ex, MgSiO_{3}, SiO_{2}, NaCl (from atmospheric sources), biotite, Ca/Na exchange and calcite. The pH and major ion (Ca, Mg, Na, K, Cl and SO_{4}) concentrations from a rainwater sample from Vavuniya, also located within the Dry Zone approximately 23 km north of the study area, was used as the initial water. The system is assumed to be receiving piston flow recharge and the groundwater system is in chemical equilibrium. Furthermore, carbon mass balance using Rayleigh distillation equations for all incoming carbon sources and all isotopically fractionating outgoing carbon phases were calculated. Models were assessed for accuracy by comparing the observed and computed δ^{13}C values. Results produced by each model and most fell within ±2‰ for the difference between modelled and measured δ^{13}C values. Models that failed to predict the δ^{13}C value were not used in the interpretation.

DATA AVAILABILITY

The full data set used in this study is provided in Supplementary Information.

CODE AVAILABILITY

The code used to develop individual figures is available upon request to the corresponding author.

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AUTHOR CONTRIBUTIONS
Sampling design and the suite of water quality analyses were organised by C.N. and K.T.M. Field sampling was undertaken by C.N. and R.B.B. L.K.M. drafted the main manuscript, undertook statistical associations between water quality parameters and environmental isotopes and prepared figures under the guidance of K.T.M. C.N. provided input regarding the analysis of water chemistry data and sample site details. All authors reviewed and provided comments on the manuscript.

COMPETING INTERESTS
The authors declare no competing interests.

ADDITIONAL INFORMATION
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