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Mapping the potential human health implications of groundwater pollution in southern Sri Lanka

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ABSTRACT

In southern Sri Lanka, irrigation influences the concentrations of faecal bacteria and inorganic toxic contaminants in groundwater. We develop a groundwater vulnerability map describing the potential human health implications of harmful constituents in the Uda Walawe Basin, by overlaying geological and land use data with information describing the irrigation system, the oxygen isotope composition of water bodies, and the concentrations of selected contaminants. Given the limited data available, we examine the spatial distribution of harmful constituents and the potential human health risks. Fluoride poisoning from groundwater is the greatest health threat in our study area, where fluoride concentrations ranging from 0.1 to 9.2 mg/L are associated with a geologic origin. Arsenic occurs in high concentrations, up to 0.4 mg/L, in areas with low recharge, although the source of arsenic is not clear. Nitrate concentrations are low, ranging from 0.4 to 23 mg/L, despite high fertilizer inputs, except in areas with low recharge and non-favourable reducing conditions, where concentrations

Abbreviations: DO, dissolved oxygen; DRASTIC, model for vulnerability mapping that considers Depth to water, net Recharge, Aquifer media, Soil media, Topography, Impact of vadose zone media, and hydraulic Conductivity of the aquifer (Aller et al., 1987); EC, electrical conductivity; Eh, redox potential; GIS, Geographical Information System; ICP-AES, inductively coupled plasma atomic emission spectroscopy; IWMI, International Water Management Institute; LMWL, Local Meteoric Water Line; m bgl, metres below ground level; pH, potential of hydrogen, an indicator of acidity; ThCU/100 mL, number of thermotolerant coliform units per 100 mL; TISAB, Total Ionic Strength Adjustment Buffer; VSMOW standard, Vienna Standard Mean Ocean Water; WHO, World Health Organization; WMWL, World Meteoric Water Line [15].

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up to 136 mg/L are found. Faecal bacteria decrease from surface water via shallow groundwater to deep groundwater. Irrigation water appears to play a major role in increasing microbial contamination and diluting inorganic constituents in groundwater. Hence, the most important determinants for mapping groundwater vulnerability are local geology and infiltration of irrigation water. The method we present provides a qualitative, yet practical, alternative to commonly used vulnerability mapping techniques for countries where high human health risk via consumption of groundwater is inevitable, and thus acts as a tool for selecting preventive and curative measures.

Introduction

Groundwater plays a crucial role in rural livelihoods by meeting the need for drinking and domestic water supply in Sri Lanka, as in many developing countries [25]. Protection of this valuable resource demands safeguarding groundwater quality against undesirable changes that occur directly when natural or artificial substances such as fertilizer are discharged into aquifers. Indirect effects due to the leaching of ions from soil or rock also must be addressed [34].

We examine the determinants of groundwater quality in the Uda Walawe Basin, Sri Lanka, and create maps depicting human health threats. In particular, we develop a groundwater vulnerability map using local geology and infiltration of irrigation water with its own isotopic signatures and aquifer properties, in relation to the most dangerous inorganic toxic substances. The map can be used for environmental risk communication and to develop preventive and curative measures for future communities of Sri Lanka as an alternative to groundwater monitoring that is considered too costly to define the geographic extent of contamination. Our approach to vulnerability mapping can support the development of highly needed groundwater management policies in Sri Lanka [47,32].

The number of wells in rural Sri Lanka is increasing, especially in irrigated dry zone areas where shallow wells are usually dug by the users beside irrigation canals or other water bodies to benefit from infiltration of surface water. In addition, the Government of Sri Lanka has implemented several rural groundwater supply programs by drilling deep tubewells to help meet the rural drinking water demands. However, such projects have provided limited benefits to communities where groundwater is contaminated [22]. Some tubewells have been abandoned due to bad taste or high salinity, iron and hardness levels. Other wells containing high levels of hazardous substances such as fluoride, which are not detectable by taste, colour or odour of the water, have been in use since the time of their construction.

Groundwater vulnerability mapping is an interdisciplinary approach to identifying regions that are likely to be affected by contamination. The widely used DRASTIC model for vulnerability mapping considers Depth to water, net Recharge, Aquifer media, Soil media, Topography, Impact of vadose zone media, and hydraulic Conductivity of the aquifer [2,40], usually applied as weighted layers in a geographic information system (GIS). However, this model is less suitable for pollutants of geological origin and cannot be justified in our study area, where most of the layers of the GIS would be incomplete. Similarly, maps based on statistical modelling techniques may not reflect actual vulnerability unless several options are checked and compared for their predictive values before dissemination [45]. Recent studies on vulnerability assessment incorporate geochemical and isotopic analysis along with GIS and remote sensing in complex heterogeneous geological environments [19]. We extend this earlier work, but take a more qualitative approach. We discuss groundwater vulnerability in the crystalline rock aquifers in rural southern Sri Lanka in terms of inorganic (principally fluoride and arsenic) and microbiological contamination, which is harmful to human health.

Exposure to high levels of fluoride may lead to dental and skeletal symptoms in humans, and high arsenic may induce skin cancers [52]. Both contaminants are known to have negative impacts on children's development [48]. In addition, there might be a risk of polluted surface water in the region, such as waste disposal containing aluminium, flushed into the irrigation canals by surface runoff. High aluminium may cause permanent neurotoxic effects leading to brain damage [8]. Health risks of nitrates in drinking water, often associated with the use of agrochemicals, include the 'blue baby syndrome' in bottle-fed infants, although impacts on adults have not been conclusively assessed [33,50]. Our vulnerability map integrates findings on these major toxic constituents in groundwater.

For consumers of groundwater in southern Sri Lanka, fluoride poisoning probably holds the greatest health risk. Fluoride is beneficial to bone and dental development in humans but the consumption of high levels of fluoride may cause severe health hazards such as dental mottling, fluoride depositions on the bones (skeletal fluorosis), skeletal deformities, and reduced growth and intelligence in children [48,51]. The prevalence of dental fluorosis in the study area is 43% among 518 14-year old school children [46]. The consumption of high fluoride water is the major risk factor for dental fluorosis and increases the risk of caries here and in other regions in Sri Lanka [20,46]. The consumption of 3 L of water with the national maximum fluoride concentration of 0.8 mg/L (2.4 mg of fluoride) already approaches the WHO-recommended maximum intake of 0.05 mg/day/kg body weight (3 mg for a 60 kg person) and a few cups of tea may easily add another milligram [3,29,50]. It is important to know where groundwater consumption might present a health risk and thus threaten livelihoods and hinder human development.

Materials and methods

Study area

The Uda Walawe Basin is in the southern lowlands of Sri Lanka, extending south from the south-eastern corner of the Central Highlands (Figure 1). Our study area includes about 350 km^2, divided into

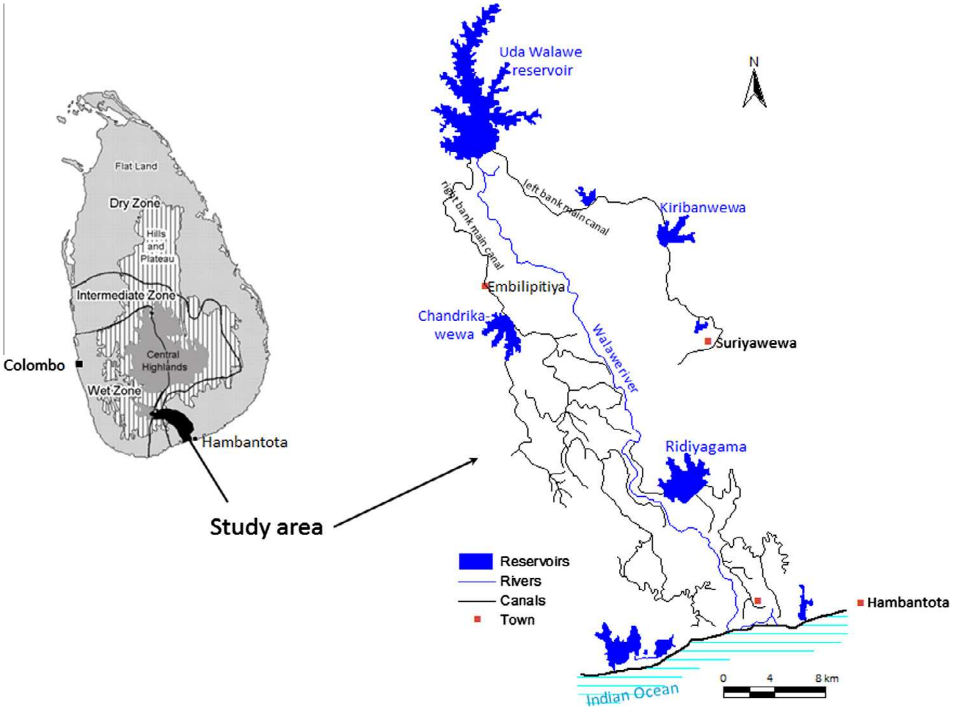

Figure 1. Location map of the study area centred on the lower Uda Walawe Basin in Southern Sri Lanka.

the Uda Walawe (UW), Suriyawewa (S) and Ridiyagama (R) catchments. The region experiences hot and humid conditions with an average annual temperature of 28 °C, humidity of 77%, average annual rainfall of 1979 mm and average annual potential evapotranspiration of 1988 mm. Rainfall has high temporal and spatial variability, with most of the rain falling from November to early March, during the south–west monsoon. The north-western region, at higher elevation, tends to receive more rain (>2000 mm) than the rest of the area (<1500 mm).

More than 50 years ago, a large dam and reservoir were constructed on the Walawe River, feeding a canal irrigation system that covers almost 20,000 ha [36]. The irrigation system incorporates traditional surface water reservoirs and has unlined and concrete-lined canals that are used for various domestic and productive purposes in addition to irrigation. The irrigated area has known strong economic development and is now more prosperous and healthier than the nearby areas under rain fed agriculture [1,31]. A close relationship was found between water flows in unlined irrigation canals and nearby shallow groundwater levels [35]. Shallow wells were the safest source of drinking water in the area, considering microbiological pollution and salinity [43]. When the canals were lined to increase the efficiency of transport of irrigation water to the downstream areas, less water seeped from canals into the groundwater and shallow wells became dry within a few months after lining [35].

Geology is an important source and controlling factor of the concentration of contaminants such as fluoride and arsenic in groundwater [28,44]. The Uda Walawe Basin is made-up of two major geologic formations (Figure 2). The Highland Series is characterized by metamorphosed sediments and various charnoketic and biotite gneisses, inter-banded with schists and granitic gneisses. The Vijayan complex is a varied group of gneisses and granites, separated into regions by linear curved folds of the Highland Series [22].

Across the study area, and the region as a whole, two major aquifers occur: regolith (weathered) and hard rock (fractured), with variation in hydraulic properties in relation to major geologic formations. Accordingly, hydraulic conductivity (rate of movement of water through a porous medium) of the Highland Series and the Vijayan Complex averages about 0.2 m/day and <0.1 m/day respectively,

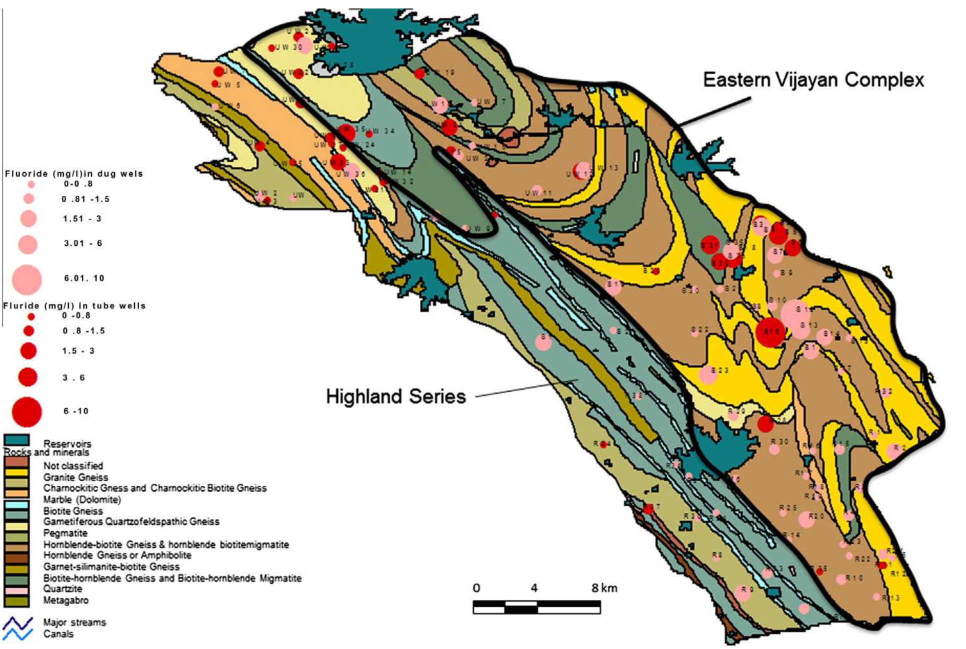

Figure 2. Fluoride distribution in relation to geology in the lower Uda Walawe Basin during the wet season, January–February 2001. Scale is indicative.

while transmissivity values (the rate at which groundwater can flow horizontally through an aquifer) of the Highland Series vary from 1.4 to 6.4 m^2/day and those in the Vijayan complex range from 0.4 to 1.6 m^2/day (Kulatunga, 1988). Our own pumping tests [38] in two wells of the Uda Walawe catchment show transmissivity values, calculated using the software package AQUATEST, of 5.52 and 4.39 m^2/day, coupled with hydraulic conductivity values of 0.12 and 0.16 m/day. In the Suriyawewa catchment, four wells have transmissivity values between 0.57 and 0.87 m^2/day with hydraulic conductivity values ranging from 0.02 to 0.09 m/day. These results for the Suriyawewa catchment indicate longer groundwater residence times and consequently greater opportunity for rock–water interactions to occur.

The degree of variation in climate and physiography across the area makes quantitative estimates of recharge for the dry zone quite problematic [17]. Determined with chlorine profiling, mean annual groundwater recharge varies between 66.0 ± 22.0 mm/year in the middle of the study area, and 9.0 ± 3.7 mm/year towards the south [17]. Groundwater levels are closely related to changes in canal water levels [35] and average water levels in the dug wells fluctuate in wet and dry spells ranging from 0.24 to 8.26 m below the ground level (m bgl) and 0.27 to 8.68 m bgl, respectively. In deep tubewells, groundwater levels range from 0.50 to 26.3 m bgl.

Leached human and animal waste matter, and industrial and agricultural inputs or wastes are major sources of groundwater pollution in many parts of the world [21]. Probable pollution sources in the study area are pesticides and fertilizers used in the intensive irrigated agriculture and widespread onsite disposal of untreated solid wastes and domestic sewage. In the irrigated area, fertilizer input is high: up to 5000–10,000 kg/ha on rice and banana fields, mainly as urea (40% nitrogen). At the time of study, an estimated 70% of residences had water-flush pit latrine systems, while the remaining 30% had no human waste disposal system.

When land is brought under cultivation and irrigation, the composition of groundwater tends to change, as irrigation water has a different composition to that of groundwater and leaching may increase. In southern Texas, low recharge rates, irrigation return flow and natural constituents of water-bearing rock formations contributed to high fluoride levels [28]. An examination of the impacts of irrigation water on the groundwater composition in our study area has limitations, as crops in the Uda Walawe Basin receive both seasonal rainfall and irrigation water. Both are considered as sources of recharge in developing the groundwater vulnerability map.

Water sampling and analysis

In 2001, two groundwater sampling rounds were conducted during the wet and dry seasons in 112 localities. Static water levels and the depths of shallow dug and deep tubewells were measured at each sampling point. Measurements of well-head parameters, including pH (potential of hydrogen), Eh (redox potential), DO (dissolved oxygen) and EC (electrical conductivity) were taken prior to sample collection. Where possible, a flow-through cell was used at tubewells fixed with hand pumps.

Fluoride and alkalinity were analysed within a day after sample collection using unfiltered samples at a field laboratory in Uda Walawe. Fluoride determinations were made using an Orion 96-09 fluoride combination electrode with TISAB III buffer solution with a reproducibility for duplicate samples yielding an error of <2%. Total alkalinity, determined as bicarbonate, was measured by titration to a pH of 4.5. Samples for analysis of major and minor anions (e.g. nitrate) were collected in 100 ml polyethylene bottles and filtered using 0.20 μm cellulose acetate filters prior to analysis. Samples for major and minor cation analysis (e.g. arsenic, aluminium) were collected in 200 mL polyethylene bottles, acidified to 4% (by volume) using nitric acid in the field, and filtered using 0.45 μm cellulose acetate filters. Samples for major and minor ions were shipped to the UK and chemical analyses performed at the University of East Anglia by ICP-AES for cations and by ion chromatography for anions. Electrical charge balances of ions calculated using the software package PHREEQC [37] were <5% in most of the samples.

For analysis of oxygen isotopes, water samples were collected from reservoirs, irrigation canals, shallow open wells and deep tubewells in June and July 2002. Sample preparation was conducted according to the standard carbon dioxide equilibration method [23] by mass spectrometry at the University of East Anglia. The isotopic composition of each water sample was measured relative to the VSMOW standard (Vienna Standard Mean Ocean Water), originally established by Craig [15] and expressed using the standard δ-notation, with a precision of 0.02‰. Using this presentation, $\delta^{18}O$ is the ratio of oxygen-18, a natural, stable isotope of oxygen, over oxygen-16, the most abundant oxygen isotope, compared to VSMOW that, by definition, has a $\delta^{18}O$ value of zero.

For preliminary screening of the water quality for the presence of coliform bacteria, a bacteriological field kit (Colilert® Test Kit, IDEXX, US) was used in 2002. All samples were submitted for analysis within 6 h from collection. The trays with samples were incubated for 24 h over a 2 L water bath. Results for total coliform bacteria were then obtained by counting the number of yellow wells in each tray, while *Escherichia coli* results were obtained by exposure to UV light in a dark chamber and counting the number of fluorescent blue wells. For each sample, a conversion table (MPN Table, IDEXX, US) was then used to find the most probable number of colonies, according to the number of positive (yellow or blue) wells.

Data analysis

Chemical modelling, electrical charge balances and calculations related to aqueous speciation, saturation indices and partial pressures of carbon dioxide for all samples were undertaken with the software packages WATEQ4F [6] and PHREEQC [37]. Results obtained from PHREEQC were read into the Waterloo Hydrogeologic AquaChem 3.7 package [12] for data interpretation and presentation.

Using ArcView [24], a geographic information system was developed to combine a geology map with land use, main surface water bodies, and the irrigation network. All sampling points were georeferenced and the findings entered into the GIS. However, it was not possible to produce a detailed groundwater vulnerability map using standard GIS overlay methodology as applied in the DRASTIC model due to the limited information on geological structure and mineralogy, and on soils and current land use. Moreover, the spatial variation of heterogeneous aquifer and hydrochemical parameter values relevant to point source contamination patterns were unknown. Hence, the final outcome of this study, a groundwater vulnerability map, was based on water analyses and secondary hydrogeological information, with vulnerability zones demarcated using qualitative expert judgment.

Results

Groundwater and surface water contamination

Figure 2 shows the distribution of fluoride in relation to the geology of the study area. Increased fluoride concentrations are associated with a wide range of rocks and minerals but high fluoride concentrations were found in two main areas: (i) the south–east part of the study area which covers the major part of the Suriyawewa (S) catchment and the upper Ridiyagama (R) catchment; and (ii) the north–west corner of the study area parallel to the Uda Walawe reservoir and the Walawe River. In general, shallow wells close to irrigation canals have lower concentrations of ions, including fluoride, compared to wells located further from the canals (fluoride levels up to 9.2 mg/L; Table 1), which suggests that irrigation water may significantly dilute the groundwater. Figure 3 shows the distribution of fluoride in relation to the main irrigation canals. Fluoride shows positive correlations with several indicators for silicate weathering, such as alkalinity, silicon, sodium and magnesium (Figure 4). In general, as shown in Table 2, higher concentrations of fluoride, and increased alkalinity (HCO_3^-) and electric conductivity are measured in dug wells in the Uda Walawe and Suriyawewa catchments in the wet season, when the weathered materials are washed out by rain.

The concentration of aluminium is less than 0.2 mg/L in 60–70% of shallow wells and in most of the tubewells in all three catchments, although slightly higher concentrations are found in the southeast portion of the study area, hence mostly in the Ridiyagama and Suriyawewa catchments (Table 1). Sixty percent of all wells in the study area have arsenic concentrations below 0.01 mg/L, with some tubewells showing higher concentrations (0.02–0.05 mg/L). High concentrations of arsenic (0.4 mg/L) are observed in the southeast portion of the study area, similar to high fluoride, as well as high iron, manganese and sulphate concentrations. Nitrate occurs in very low concentrations, except for high concentrations in excess of 120 mg/L in the Suriyawewa and Ridiyagama catchments. In the Uda

Table 1
Selected results of water quality analysis in three sub-catchments of the lower Uda Walawe Basin, Sri Lanka in 2001 (mean values with range between brackets; groundwater from deep tubewells in bold, from shallow wells in italics and from surface water in normal font). The number of samples (*n*) is given as the product of sampling rounds and sites in the wet season.

Element	Uda Walawe		Suriyawewa		Ridiyagama	
	n	mean (range)	*n*	mean (range)	*n*	mean (range)
Fluoride (mg/L)	**24**	**0.9 (0.3–3.0)**	*9*	**2.5 (0.5–9.2)**	**7**	**0.5 (0.2–1.9)**
	13	*1.1 (0.2–2.3)*	*21*	*0.8 (0.2–6.1)*	*31*	*0.6 (0.05–2.4)*
	4	0.4 (0.32–0.62)			2	0.35 (0.23–0.47)
Arsenic (mg/L)	**24**	**0.02 (0.01–0.04)**	*9*	**0.02 (0.01–0.04)**	**7**	**0.02 (0.01–0.04)**
	13	*0.01 (0.01–0.03)*	*21*	*0.01 (0.01–0.4)*	*31*	*0.02 (0.01–0.3)*
	4	0.0056 (0.001–0.008)			2	*nil*
Aluminium (mg/L)	**24**	**0.1 (0.04–4.0)**	*9*	**0.1 (0.04–0.08)**	**7**	**0.4 (0.04–3.6)**
	13	*0.1 (0.1–0.5)*	*21*	*0.2 (0.05–2.2)*	*31*	*0.1 (0.03–13.2)*
	4	*3.54 (0.4–7.00)*			2	*1.7 (0.49–2.7)*
Nitrate (mg/L)	**24**	**1.7 (0.7–11.9)**	*9*	**12.1 (2.5–119)**	**7**	**1.3 (1.2–122.6)**
	13	*1.9 (0.4–23.0)*	*21*	*2.4 (0.9–120.0)*	*31*	*1.9 (0.9–136)*
	4	*7.5 (1.6–23.0)*			2	*1.7 (1.6–1.8)*
E. coli (units/100 mL)	**6**	**403 (0->2419)**	**15**	**2 (0–150)**[a]		
	9	*562 (0->2419)*	*60*	*334 (0–100,000)*[a]		
	8	215 (3–1553)	3	250 (14–594)[a]		
$\delta^{18}O$ (‰ VSMOW)	**6**	**(−5.78 to −3.91)**				
	9	*(−4.93 to −2.94)*			*9*	*(−3.56 to −0.01)*
	8	(−4.65 to −3.63)			3	(−1.82 to −1.15)

[a] Thermotolerant coliforms, from [43].

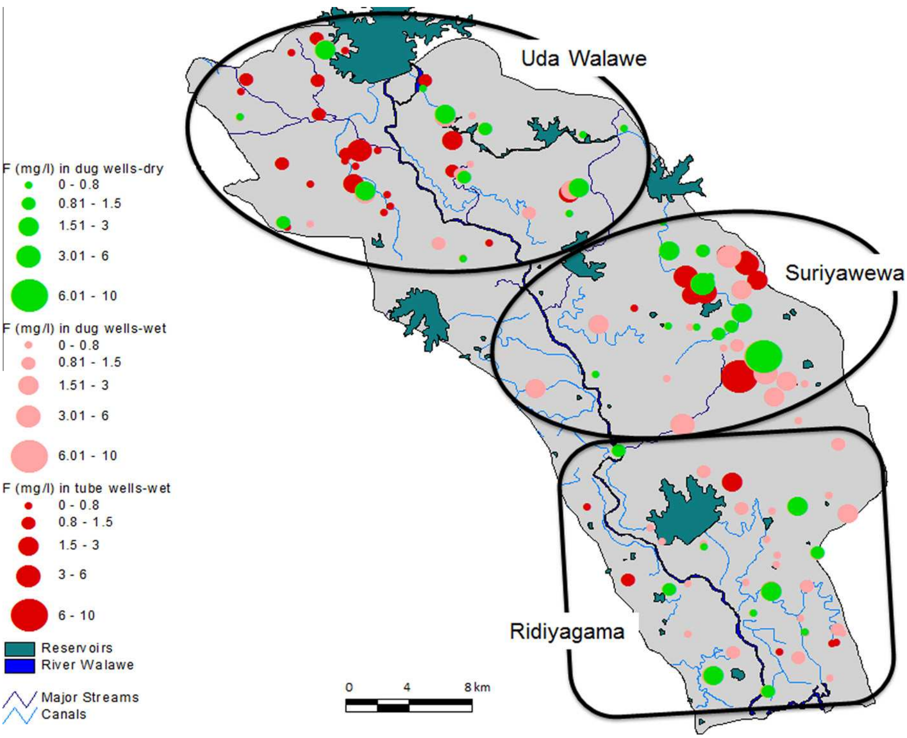

Figure 3. Fluoride concentration in relation to the irrigation system in the three catchments of the lower Uda Walawe Basin, Sri Lanka during the wet (January–February) and dry (July–August) seasons 2001. Scale is indicative.

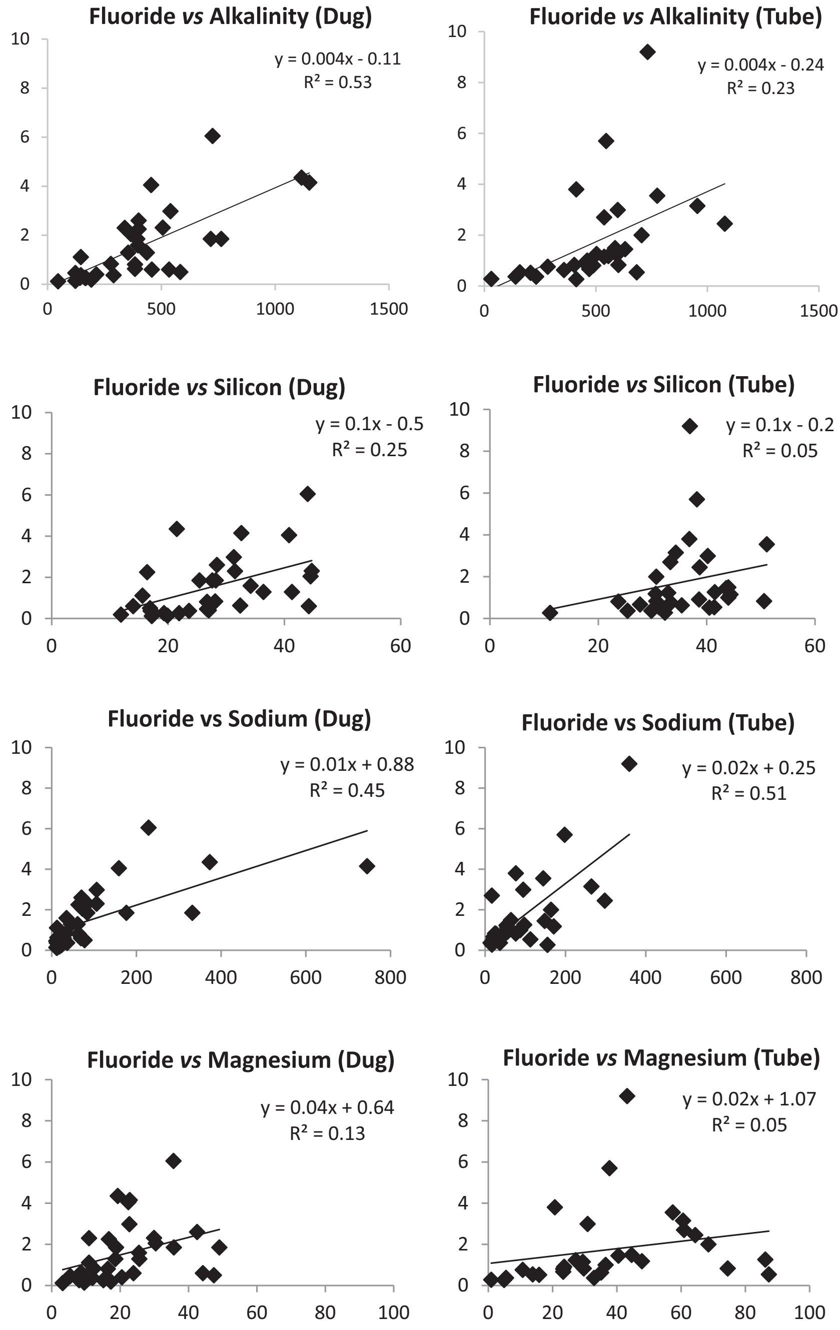

Figure 4. Comparison of fluoride and the principal weathering products associated with dissolution of crystalline bedrock aquifer formations in the Uda Walawe Basin. Data are for the Uda Walawe and Suriyawewa catchments collected in January–February 2001 and divided into dug wells and tubewells. Concentration units are mg/L. Note that samples containing outliers in the dataset have been removed.

Table 2
Seasonal variation and minimum, maximum and median values of hydrochemical parameters in dug wells and tubewells in catchments of the Uda Walawe Basin during the wet (January–February) and dry (July–August) seasons 2001.

Season	Parameter	Suriyawewa			Uda Walawe			Ridiyagama		
		Min	Max	Median	Min	Max	Median	Min	Max	Median
Dug wells										
Wet	Temp. (°C)	25.1	28.4	27.0	26.0	28.0	27.5	25.5	30.1	27.8
Dry		26.9	30.9	28.9	26.1	30.0	27.3	26.9	29.1	27.5
Wet	pH	6.5	7.8	7.4	6.5	7.7	7.0	6.5	7.9	7.1
Dry		6.2	8.2	6.9	6.4	7.8	6.9	6.9	8.3	7.4
Wet	EC (μS/cm)	230	4950	700	100	2080	600	50	5600	800
Dry		207	1427	414	267	1676	282	377	10580	981
Wet	D.O. (mg/L)	3.8	7.7	5.0	2.6	5.6	4.1	1.6	8.1	4.9
Dry		0.3	5.8	2.9	0.9	7.5	3.9	1.1	6.4	2.7
Wet	HCO_3^- (mg/L)	142	1150	400	46	763	384	73	671	436
Dry		168	699	306	134	537	298	177	762	463
Wet	SiO_2 (mg/L)	11.9	44.5	27.0	15.6	44.7	26.7	5.4	33.2	15.1
Dry		7.6	44.5	25.2	13.8	49.1	35.5	8.0	26.0	17.3
Wet	F^- (mg/L)	0.2	6.1	0.8	0.2	2.3	1.1	0.05	2.4	0.6
Dry		0.2	3.2	0.8	0.1	2.4	0.9	0.4	1.9	1.0
Tubewells										
Wet	Temp. (°C)	26.0	30.0	29.0	28.0	29.5	29.0	28.3	30.2	28.9
Dry		28.9	31.1	30.5	28.0	30.1	29.0	29.0	30.5	–
Wet	pH	6.2	7.0	6.6	5.5	7.9	6.7	6.4	7.9	6.6
Dry		5.7	7.3	6.8	5.9	7.8	6.8	6.4	8.1	–
Wet	EC (μS/cm)	1200	6200	1890	100	4800	850	200	13500	825
Dry		1246	6710	1752	100	4950	778	288	14110	–
Wet	D.O. (mg/L)	3.4	4.7	4.3	3.3	5.2	3.9	3.6	6.6	4.2
Dry		1.7	5.4	2.6	2.4	5.7	3.3	2.9	3.1	–
Wet	HCO_3^- (mg/L)	546	1078	705	31	689	466	128	677	360
Dry		500	1049	732	49	653	453	162	659	–
Wet	SiO_2 (mg/L)	30.6	51.1	34.8	11.1	50.6	35.4	11.8	34.5	21.0
Dry		26.5	51.7	33.5	12.4	45.7	33.3	31.4	34.6	–
Wet	F^- (mg/L)	0.5	9.2	2.5	0.3	3.0	0.9	0.2	1.9	0.5
Dry		0.9	6.0	3.9	0.3	3.1	0.9	0.7	2.5	–

Walawe sub-catchment, colonies of *E. coli* are observed in all of the surface water samples, in six out of nine samples from shallow dug wells and in a single deep tubewell (Table 1). Along irrigation canals in the Suriyawewa sub-catchment, increasing numbers of coliforms were measured in the downstream direction [43].

The $\delta^{18}O$ ratios for surface water range from −4.65 to −1.15‰, corresponding to the Uda Walawe reservoir and an unlined irrigation canal in the Ridiyagama catchment, respectively. Along this line, roughly from north to southeast, we observe a gradual enrichment in the $\delta^{18}O$ values of surface waters. Within the Uda Walawe catchment, $\delta^{18}O$ ratios in shallow groundwater range from −4.93‰ next to the Uda Walawe reservoir to −2.30‰ at the southeast corner of this catchment (Table 1). The isotopic signature of the shallow groundwater in the Ridiyagama catchment is significantly enriched, presenting $\delta^{18}O$ ratios that range from −3.56 to −0.01‰, close to the VSMOW standard of zero, and thus showing the influence of seawater intrusion on water quality in the south of the study area. The deep wells in the Uda Walawe catchment have the most depleted $\delta^{18}O$ signatures, ranging from −5.78 to −3.91‰.

The slope of the relationship between $\delta^{18}O$ and $\delta^{2}H$ is controlled by local climatic factors such as the origin of the water vapour mass and the seasonal variations of precipitation, humidity, temperature, salt concentration and other factors. As shown in Figure 5, a Local Meteoric Water Line (LMWL) with the equation $\delta^2H = 7.9\delta^{18}O + 8$ was established for the present study area based on the isotopic composition of the deep groundwater samples, which are expected to represent more closely the local precipitation isotopic composition. Interestingly, the LMWL plots just to the right of the World Meteoric Water Line (WMWL, $\delta^2H = 8\delta^{18}O + 10$; [15]).

As illustrated in Figure 5, evaporation from surface water bodies is a non-equilibrium process that enriches the residual water body such that the $\delta^2H/\delta^{18}O$ slope is less than 8 [14]. The evaporation trend observed in the study area is depicted by the evolution of the isotopic composition of surface waters in the irrigation canals (Figure 5). Samples collected in upstream areas plot next to the LMWL, showing little evaporation. In downstream areas, however, particularly in the Ridiyagama catchment, the water samples plot to the right of the LMWL, defining an evaporation line with the equation $\delta^2H = 3.5\delta^{18}O + 8$.

Groundwater vulnerability map

Our vulnerability map is based primarily on the health risk parameters of fluoride, nitrate, and arsenic in groundwater and, to a lesser extent, the degree of microbiological pollution. We consider other factors also in assessing and ranking qualitatively the groundwater vulnerability (Table 3). The local geology is classified into two major groups depending on mineralogy and geological structure. We also use a hydrogeological classification divided into shallow regolith (weathered) aquifers

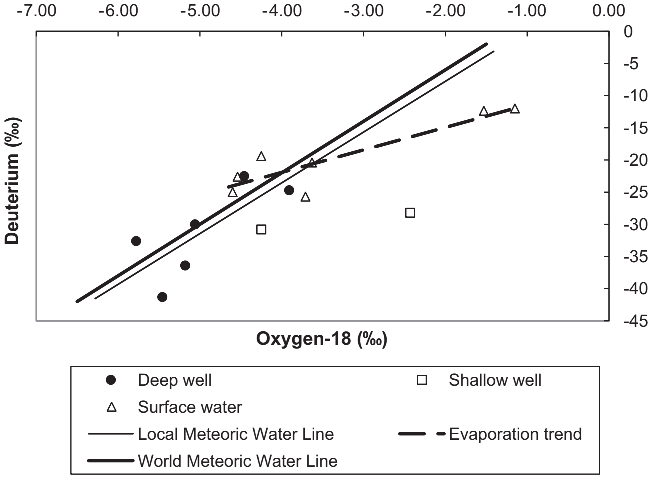

Figure 5. δ^2H versus $\delta^{18}O$ cross-plot of surface waters and groundwater in the study area in relation to the World and Local Meteoric Water Lines.

Table 3
Parameters included in the development of a groundwater vulnerability map for the Uda Walawe Basin, Sri Lanka (based on [43,49,50,52] and primary data from this study).

Parameter	Vulnerability		
	Low	Medium	High
Chemical			
Fluoride	<0.8 mg/L	0.8–1.5 mg/L	>1.5 mg/L
Arsenic	<0.01 mg/L	0.01–0.05 mg/L	> 0.05 mg/L
Aluminium	<0.2 mg/L	>0.2 mg/L	> 0.2 mg/L
Nitrate	<10 mg/L	10–50 mg/L	> 50 mg/L
Faecal	>95% Negative samples	80–95% Negative	<80% Negative
bacteria	<100 ThCU/100 mL	100–1000 ThCU/100 mL	>1000 ThCU/100 mL
Geology	Highland Series		Eastern Vijayan Complex
Minerals	Not fluoride-bearing		Fluoride-bearing
	Not arsenic-bearing		Arsenic-bearing
Salt-mixing	>10 km from coast		<10 km from coast
Aquifers	Non-fractured bed rock	Fractured bed rock	Shallow weathered bed rock (regolith)
Transmissivity	<1 m^2/day	1–10 m^2/day	>10 m^2/day
Recharge	High: irrigation plus rainfall	Medium: ∼2000 mm annual rainfall	Low: ∼1500 mm annual rainfall

and deep hard rock (fractured) aquifers, as well as into high, moderate and low transmissivity aquifer zones. Another group of parameters considers recharge, distinguishing between areas subjected to high rainfall and irrigation recharge and areas subject only to low rainfall recharge. This is based on a map of the irrigation network, on groundwater table measurements [35], and on the results of the stable isotope analysis. Finally, two major groups of salt water mixing, most likely related to sub-surface seawater intrusion, are classified as areas near and away from the coast.

By combining the above factors, areas of low, medium and high groundwater vulnerability to pollution are defined qualitatively using expert judgment. The results of the groundwater vulnerability classification are shown in Figure 6. The dominant determinant of contamination risk is geology, influenced by groundwater recharge from irrigation. This explains why the map has only four distinct areas, demarcated by geological groups and irrigation infrastructure. The Eastern Vijayan Complex is underlain by more fluoride-bearing minerals and is subject to limited fracture continuities, limited transmissivities and more clay formations. This allows more rock–water interaction and greater release of fluoride. However, areas subject to irrigation recharge in the Eastern Vijayan Complex show dilution effects and less fluoride in general (Table 1). Rocks in the Highland Series contain more fracture continuities, higher transmissivities and less clay formations, and are subject to a dense irrigation network. In places, fractures tend to increase fluoride concentrations due to groundwater flow from the interconnected fractures or aquifers but, in general, geological structures and high irrigation recharge control the risk of fluoride and arsenic in the Highland Series zone.

Discussion

Impact of geology on groundwater quality

In the Uda Walawe Basin, geology is the major factor that determines the quality of groundwater, especially fluoride occurrence. Both geological regions in the study area are underlain by fluoride-bearing rocks but the Vijayan Complex is especially abundant in granites that are rich in fluoride-bearing minerals [22]. Fluoride has been found in similar geological environments in many parts of the world, as well as in sedimentary formations such as alluvial deposits, sandstone, siltstone and conglomerate sediments [9,16,30] where fluoride is found in association with desorption from hydrous metal (iron) oxides. In India, China, Korea, Norway and Ghana, fluoride has been reported in association with metamorphic and crystalline rocks, in charnokites, granites, pegmatites and gneisses, usually together with biotite and hornblende that are common in these areas [3,7,10,13,26,39,41].

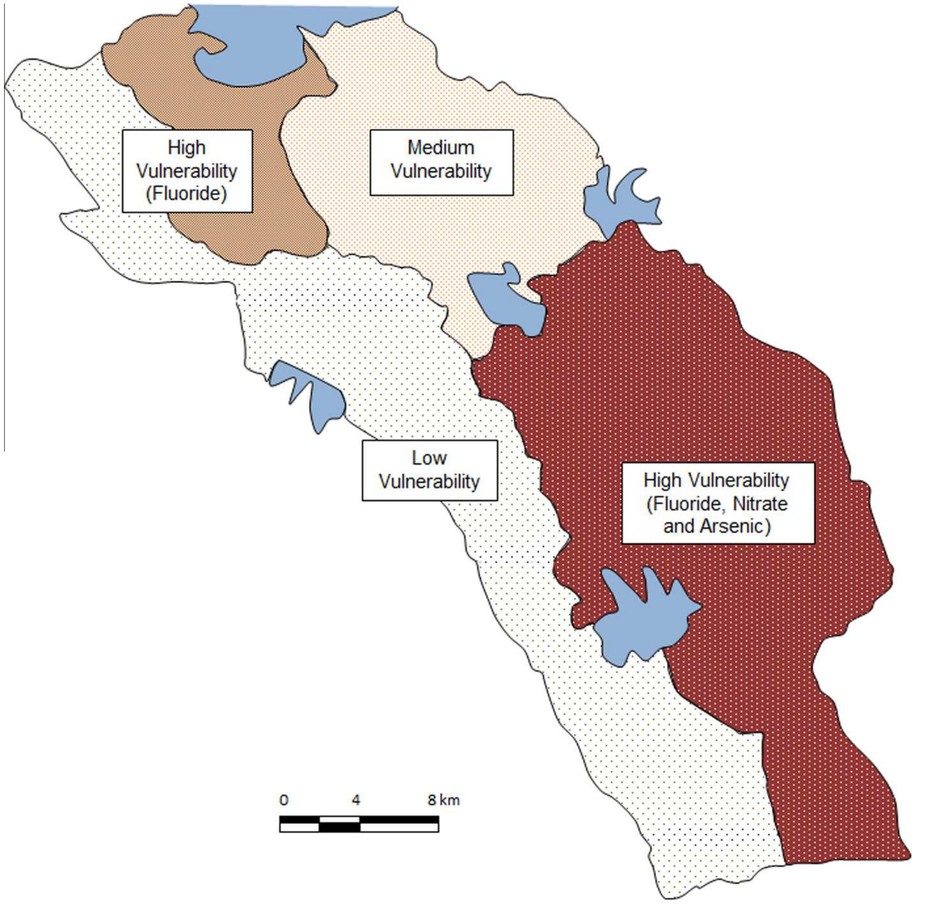

Figure 6. Groundwater vulnerability map for the lower Uda Walawe Basin, Sri Lanka. Scale is indicative.

High fluoride concentrations in groundwater in Sri Lanka are known to occur in areas with bedrock such as charnokitic gneisses associated with pegmatite and intrusive granites, hornblende-biotite gneisses, marble and calk gneisses [5]. Dharmagunewardene [18] found fluoride in abundance associated with biotite, occurring widely among all granites and gneisses in the north-central province of Sri Lanka. The presence of alkalinity (as bicarbonate) with high partial pressure of carbon dioxide indicates carbonic acid as the main acid for silicate weathering and the release of major cations (calcium, magnesium and sodium), bicarbonate, silicon and fluoride, as illustrated by the positive correlations between all but one of these parameters and fluoride (Figure 4). The occurrences of accessory minerals such as carbonates, apatites and fluorite are very limited and are not considered an important source of fluoride given that groundwater in the discharge areas of the basin are saturated with respect to these accessory minerals [38]. Hence, silicates rich in magnesium, such as biotite or hornblende, are the most likely sources of fluoride in the study area. The increased leaching process in the wet season, indicated by the higher fluoride concentrations and increase in electrical conductivity and bicarbonate (Table 2), leads to a potentially higher health risk from shallow groundwater consumption in the wet season than in the dry season, specifically in the Suriyawewa and Uda Walawe catchments.

We note that the distribution of fluoride does not show a similar geological pattern throughout the study area, due to a marked variation in the structure and hydrogeological conditions found in the two geological zones. Also, there is little evidence to show that land use, or use of fertilizer, has an influence on fluoride or metal concentrations across the area. Unlike fluoride, arsenic concentrations could not be clearly associated with geology. Although there is no literature available on arsenic geochemistry in the study area to help explain its occurrence, it is possible that arsenic occurs in a reduced form in mineral veins and as an oxide. Arsenate (AsO_4^{3-}) occurs in some metallic ores and can replace phosphate in apatite [27]. Arsenic may also substitute in pyrite (FeS_2) or be found as the separate mineral arsenopyrite (FeAsS) [4].

The high vulnerability area in the south–east of the study area is underlain by the Eastern Vijayan Complex, which is well known for the nature of its geological fractures as observed in the associated aquifer properties and well yields. This area was largely covered by shrubs and forest at the time of study and thus lacked an irrigation network, making it totally dependent on rainfall or leakage from seasonal surface water bodies to recharge the aquifers. Hence, longer groundwater residence times could occur in this area, leading to more rock–water interaction, pyrite oxidation and high releases of most elements into the system, as indicated by the dangerously high concentrations of fluoride and arsenic.

The other area of high vulnerability located in the northwest corner (Highland Series) of the Uda Walawe Basin shows patterns of higher hydraulic continuity with aquifer properties indicating quite substantial groundwater flow between interconnected fractures. Mixing of water from interconnected fractures may be one of the major factors determining the high concentrations of fluoride in this area, similar to the situation found in India [41]. The northwest corner of the area is not irrigated, such that the effect of dilution from leaking canal water is reduced and fluoride concentrations increased. The isotopic composition of the deep groundwater also suggests an origin of water from less evaporated rain water.

Impact of irrigation on groundwater quality

The irrigation system has a significant influence in regulating the concentrations of ions of geological origin, such as fluoride, contrary to results from a study in Andhra Pradesh, where no difference was found between groundwater in domestic areas and irrigated fields [11]. Shallow wells close to irrigation canals have lower concentrations of ions compared to wells located further from the canals, which suggests that irrigation water may dilute the groundwater. This is consistent with recharge measurements that demonstrate how the groundwater table fluctuates with water levels in unlined canals nearby [35] and with the results from the stable isotope analysis. The dilution effects depend on several factors, such as initial concentrations of the chemicals in both surface water and groundwater at individual localities, the amount of recharge, evaporation, local geology and hydrochemistry of the area. Unfortunately, these dilution effects do not necessarily reduce the risk of fluoride in the study area, as even in most of the wells with diluted groundwater, fluoride concentrations are still above the Sri Lankan health advisory limits of 0.8 mg/L.

Irrigation water also plays a role in determining the distribution of microbial contamination. Irrigation canals are used by the villagers for domestic purposes and by animals, mainly cattle and have high levels of faecal coliform bacteria [43]. In addition, we show that the number of *E. coli* increases in the direction of water flow in the irrigation canals. In both studies the numbers of coliforms decrease from surface water to shallow groundwater to deep groundwater. In deeper aquifers, the hydrogeology co-determines microbiological contamination. Confined or semi-confined aquifers are less vulnerable to microbial pollution as they are not affected by direct surface water infiltration. This is confirmed by the stable isotopic composition of the deep groundwater that shows less influence of evaporated rainwater, as well as coliform pollution observed in few deep wells only, at much lower concentrations. Microbial pollution is significantly less in lined wells than in unlined wells [43] but no relationship is found between the level of faecal pollution in wells and their relative distance from irrigation canals. This suggests both a pollution migration from surface water into groundwater and a predominance of point source pollution from sewage and other waste disposal activities.

Impact of land use on groundwater quality

We expected to see pollution from nitrate in the study area due to high fertilizer inputs and poor human and animal waste disposal systems, along with shallow water tables. Nitrate occurs in very low concentrations in most of the groundwater samples, probably because nitrates are reduced in the water-logged conditions under paddy fields [42,53]. In the southeast part of our study area, nitrate concentrations are high, due possibly to improper human and animal waste disposal, without dilution. Aluminium concentrations in shallow wells are higher in the wet season than in the dry season, which supports the idea of surface pollutants being washed out into shallow groundwater. However, we identified no clear sources of aluminium pollution in the field.

Conclusions

The combination of prevailing geological and mineralogical conditions, high recharge from irrigation and rainfall, shallow groundwater tables, local waste disposal, and intensive agriculture with high use of fertilizers and agrochemicals leads to serious health risks from groundwater consumption in the Uda Walawe Basin. In the study area, the role of irrigation and its resulting groundwater recharge in determining the concentrations of ions is clearly observed. However, irrigation water returns cannot always minimize the risk of groundwater contamination, as in the case of fluoride, where the concentrations in groundwater close to the irrigation canals are still too high for safe consumption.

The western half of the study area, which is underlain by the Highland Series rocks, is a good example of this situation. In general, this area exhibits low groundwater vulnerability except in the northwest corner, where fracture continuities control the hydrochemistry, resulting in high concentrations of fluoride. Even so, the problems related to high concentrations of fluoride are limited in the irrigated areas, especially in shallow groundwater. Any attempt made to control the seepage from canals leads to reduced groundwater recharge [35] and may result in a hazardous hydrogeological and hydrochemical situation with high exposure to substances such as fluoride.

Irrigation water management and maintenance of shallow aquifer wells might improve the inorganic quality of shallow groundwater in the Uda Walawe Basin, but might also result in increased microbiological pollution. This is unavoidable due to current human activities, where surface waters are used for domestic purposes and for waste disposal. Improvements in municipal services and in health and hygiene education are needed. We recommend the application of a groundwater vulnerability mapping approach to inform the choice of measures available for addressing the inorganic and microbiological contamination of shallow groundwater, which is chemically the safest source of drinking water. Once areas for remediation or areas for further groundwater development have been identified, more site-specific risk assessments and testing of water chemistry can be applied in selected locations.

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Glossary

Bedrock: the underground rock that underlies soil. There are three main types that group various rocks: *igneous*, such as charnokite (with quartz), granite and pegmatite (course-grained); *metamorphic*, such as schists and gneisses; and *sedimentary*, such as sandstone and siltstone.

Biotite: ferromagnesian mineral.

$\delta^{18}O$: the ratio of oxygen-18, a natural, stable isotope of oxygen, over oxygen-16, the most abundant oxygen isotope, compared to the Vienna Standard Mean Ocean Water (VSMOW) that has a $\delta^{18}O$ value of zero.

Dental fluorosis: permanent fluoride depositions on the teeth (mottled) that may lead to caries.

Dug well: hand dug shallow well, in Sri Lanka up to 5–10 m in depth.

Escherichia coli (E. coli): bacteria used as an indicator of contamination with faecal material.

Hornblende: group of minerals with calcium–iron–magnesium, aluminium–iron–magnesium, and iron–magnesium silicates.

PHREEQC: a computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations (Parkhurst and Appelo, 1999).

Regolith: weathered rock.

Skeletal fluorosis: permanent fluoride depositions on the bones.

Transmissivity: the rate at which groundwater can flow horizontally through an aquifer.

Tubewell: relatively deep drilled well, in Sri Lanka up to 30–100 m in depth.

WATEQ4F: computer program for processing large numbers of water analyses, developed by the US Geological Survey.