



Groundwater Constituents and Trace Elements in the Basement Aquifers of Africa and Sedimentary Aquifers of Asia: Medical Hydrogeology of Drinking Water Minerals and Toxicants

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Abstract

The use of groundwater, a major source of potable water, in developing countries has proven to be an invaluable resource for local populations. The ability to safely use this water for drinking, however, depends on its chemical quality, a factor primarily controlled by various aquifer attributes such as geology and geochemistry. On a global scale, groundwater is primarily sourced from either sedimentary or basement aquifers. In this study, we compared the groundwater constituents and trace elements found in these two types of aquifer system in the context of medical hydrogeology, i.e. the status of groundwater mineral nutrients and pollutants, and their complex interaction in relation to human health. The evaluation work used a collated geochemical dataset developed for Bangladesh sedimentary aquifer data ($n=474$), basement aquifer data from Northern Ghana ($n=184$) and Central Tanzania ($n=73$). An assessment of the mineral concentration in regards to dietary needs showed that the sedimentary aquifers found in Bangladesh have almost double the concentration of salubrious minerals such as calcium, magnesium and iron relative to the basement aquifers (Ghana and Tanzania). It should be noted, however, that the groundwater was also found to contain excessive levels of arsenic in the sedimentary aquifers and high levels of fluoride in those countries sourcing water from within basement rock; levels at which both elements pose a serious public health threat. Excessive sodium in drinking water is also an issue as this, combined with the normal dietary sodium level intake, may lead to hypertension and cardio-metabolic diseases. Unfortunately, health-based guideline values for drinking water containing sodium are non-existent or poorly defined, a fact which warrants further consideration at both a national and international level. The use of groundwater for drinking may assist in increasing the level of mineral nutrient uptake in the local population, however, it must also be augmented by a nutritious food supply in order to satisfy normal human dietary requirements.

Keywords Groundwater · Basement aquifer · Bangladesh · Ghana · Tanzania · Medical hydrogeology · Mineral nutrients · Sedimentary aquifer

1 Introduction

In most developing countries, groundwater (usually pathogen-free and distributed resource) is regarded as an invaluable asset for a rural population dependent on it for drinking and other domestic purposes. In addition to its core function of keeping the human body hydrated, previous work (Hoque and Butler 2016a) had identified that minerals found in groundwater in three Asian delta areas supplied part of the recommended daily intake (RDI) for people and, by doing so, supported the observed health status of the local population. Food intake, however, still provided most of the macro- and micro-minerals needed for the body. Studies by Edmunds and Smedley (1996), Chen et al. (1985) and

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Mpenyana-Monyatsi et al. (2012) have highlighted the fact that groundwater quality could also negatively influence the health of local residents. Groundwater pollutants such as arsenic, and the presence of excessive fluoride and sodium have been known to cause widespread morbidity and mortality in many parts of the world (Chouhan and Flora 2010;

Fewtrell 2004; González-Horta et al. 2015; Scheelbeek et al. 2016; Smith et al. 2000).

Groundwater is regarded, in most areas of the world, as a reliable and resilient water resource due to its distributed, subsurface storage and perennial availability. The geological formations which temporarily store this water and allow it to flow, are known as aquifers. Lithologically these can be

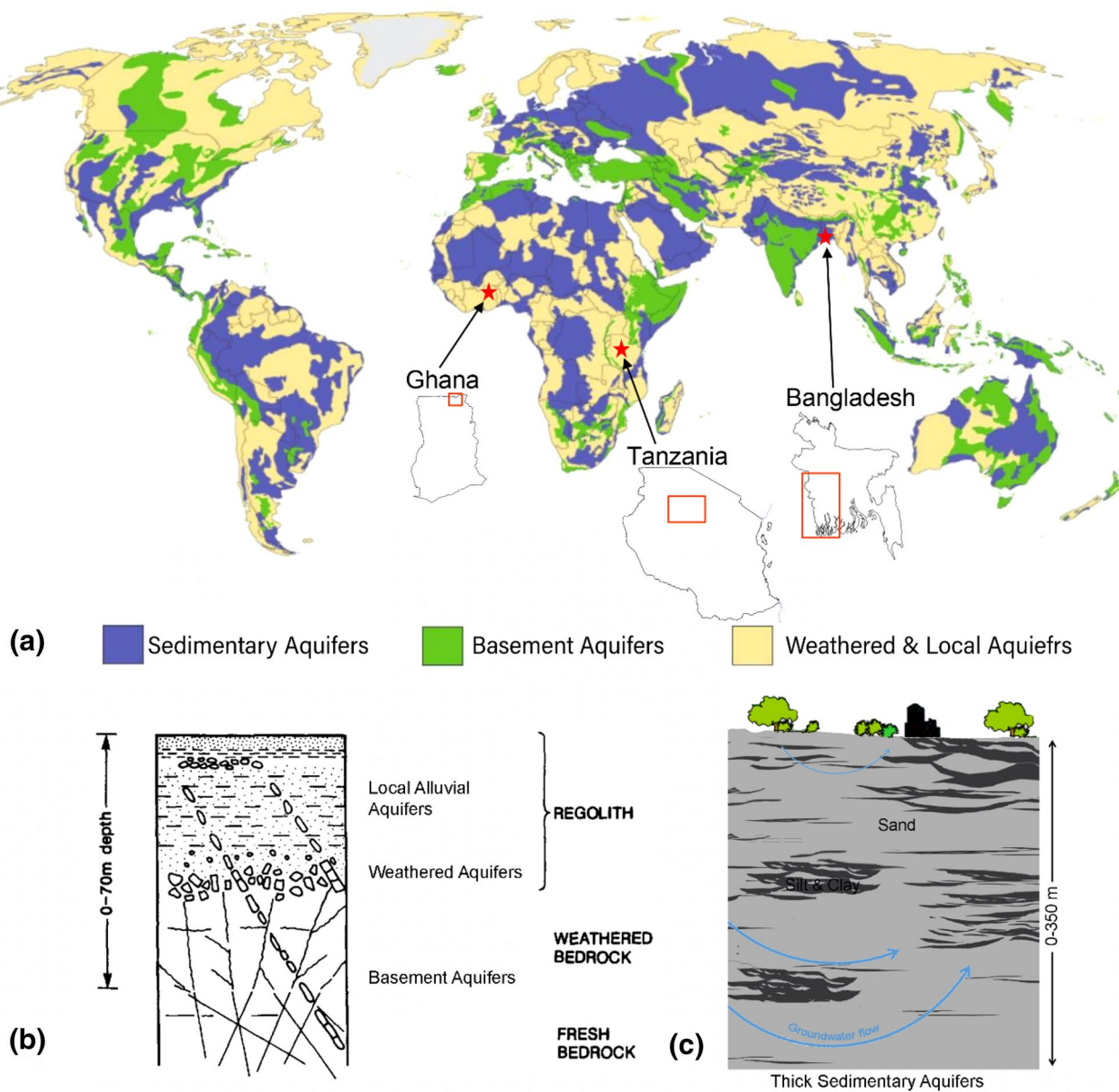


Fig. 1 Aquifer types across the globe. The sites in this study are shown as red rectangles. **a** Globally there are three major aquifer types (BGR and UNESCO 2008); **b** basement aquifers are geologically complex—faulting and fracturing provide the necessary storage and permeability (Wright 1992). In some areas, prolonged weathering of the top few metres of the basement aquifers, often coupled with

fluvial reworking, may lead to local alluvial aquifers but the formation of clay particles reduces the permeability, while underneath basement aquifers of relatively high permeability may occur. On the contrary, **c** large sedimentary aquifers are thick and have multilayer systems that support hierarchy of groundwater flow systems

hosted within either basement (old, crystalline rocks of metamorphic and/or intrusive origin) or sedimentary terrains (Fig. 1). Low primary porosity and permeability are a common characteristic of basement aquifers, and water movement is usually controlled by secondary porosity resulting from chemical weathering, faulting and fracturing (MacDonald et al. 2008; Wright 1992). Weathering and regolith development have an important influence on the storing ability of crystalline rocks and often lead to thin alluvial aquifers of limited extent. Sedimentary formations on the other hand, in particular the sand-clay dominated aquifers which often arranged in thick-layered multi-aquifer structure, have high primary porosity and permeability (Erban et al. 2013; Hoque et al. 2017; Mukherjee et al. 2007). The difference between the basement and sedimentary aquifers in terms of their permeability characteristics and geochemistry can result in significant water quality differences between the two types of systems.

People living in the rural areas of developing countries often endure low-nutrient, monotonous, diets and may suffer from mineral deficiency and other health-related issues. In such settings access to groundwater supply rich in mineral content can provide a significant amount of the recommended daily mineral requirement. In these cases, access to good quality groundwater has the potential to mitigate some of the possible health issues arising from a nutrient and mineral deficient food diet. Rosborg (2015) noted that medical hydrogeology has traditionally focused on the diseases associated with certain elements (e.g. arsenic, fluoride, manganese, etc.) and has rarely looked into the positive health effects of mineral nutrients (e.g. calcium, magnesium, iron, etc.). Hoque and Butler (2016a) have argued that medical hydrogeology should also consider the presence of beneficial mineral nutrients (including levels of minerals such as calcium) influencing the toxicity of the more harmful elements (e.g. fluoride), and the effect of various physico-chemical characteristics (e.g. bicarbonate, pH, etc.) altering quality of water during storage.

As part of the United Nation's Sustainable Development Goals (SDGs), access to water and sanitation was given

particular attention and was listed as Goal 6 of the SDGs. It was noted that inadequate water supply, poor sanitation and depleting water quality all negatively impact educational opportunities, livelihood choices and the food security of low-income families across the world (Damania et al. 2019). Based on the importance of drinking water in regards to the health status of the local populace in developing countries, and given that groundwater is in many cases the primary source of drinking water, an assessment of the chemical quality of water needs to be considered.

Groundwater constituents are the product of geochemical reactions within an aquifer. These are regulated by residence time, rock type and level of rock-water interaction (Ben Maamar et al. 2015; Elango and Kannan 2007). The basement and sedimentary aquifers vary in terms of geochemical processes due to disparate hydrogeological frameworks and mineral content (Bretzler et al. 2019; Fouché et al. 2019; McArthur et al. 2012). A comparative study should provide an insight into the various groundwater constituents and trace elements, and their significance to human health. This study investigates the disparities in groundwater constituents in relation to medical hydrogeology, i.e. comparing the role and contribution of groundwater to the mineral nutrients (calcium, magnesium, sodium, potassium and iron) along with the status of groundwater toxicants (fluoride and arsenic). The choice of locations was guided by the study aims; southwest Bangladesh having a fluvio-deltaic sedimentary aquifer, while Northern Ghana and Central Tanzania are characterised by a basement aquifer (Fig. 1, Table 1). Although hydrochemical data for all of Bangladesh are available, the study focused on the western section (a typical sedimentary aquifer with groundwater being recharged from the north, composition being affected by saltwater intrusion from the south and along with older regional groundwater at depths) in order to ensure comparability of extents and data density among the chosen case study sites.

Table 1 Summary of the major attributes for the three case study areas

Attributes	Bengal delta	Ghana	Tanzania
Physiography	Within Ganges tidal flood plain and Ganges plain	Between Savannah high plains and Gambaga escarpment	Large plateau
Climate	Tropical wet	Semi-arid sahel	Temperate climate
Vegetation	Cropland, Sundarbans mangrove vegetation	Savannah (guinea) woodland	Groves, bushland and savannah wetlands
Geology	Alluvial and deltaic deposits	Crystalline basement rocks (Precambrian)	Precambrian crystalline basement rocks
Hydrogeology	Alluvial, deltaic and coastal aquifers	Crystalline basement aquifers	Crystalline basement aquifers
Water use	Mainly agricultural and domestic	Livestock, irrigation and domestic	Livestock, irrigation and domestic

2 Physiographical and Hydrogeological Context of the Study Areas

Southwest Bangladesh Khulna Division is a second-order administrative unit located just north of the Bay of Bengal with approximate dimensions of 250 km by 75 km. Physiographically, the area falls within the Ganges delta floodplain and receives most rainfall in summer (around 2000 mm/yr) with little rain in winter. Geologically, the area is composed entirely of fluvio-deltaic sediments (mostly sands and silty clay) of the Quaternary time. The unconsolidated deposits provide a framework for multi-aquifers to an exploitable depth of 350 m. The near-surface aquifers are mostly semi-confined to unconfined (Ahmed 2003; Rahman and Ravenscroft 2003) while the deeper (> 100 m) strata are hydraulically separated by numerous interbedded, laterally discontinuous silty clay layers (Burgess et al. 2010; Hoque et al. 2017).

Northern Ghana The topography of Ghana consists predominantly of gently undulating hills with an elevation nowhere exceeding 500 m above mean sea level. Rainfall generally decreases towards the northern parts of the country. In the far north (around the case study area) annual rainfall is less than 1100 mm, but to the southwest, 1800 mm can be recorded (FAO 2016; Gumma and Pavelic 2013).

Tectonically, northern Ghana occupies the southern part of the West African Craton (Carrier et al. 2008). A regolith horizon, containing in situ chemically weathered material and some transported surface material, is typical of most geological formations in Northern Ghana (Dapaah-Siakwan and Gyau-Boakye 2000). There are two major hydrogeological provinces in this area: Voltaian Basin and Precambrian Basement provinces. The rocks of the Precambrian Basement Province (Carrier et al. 2008; Dapaah-Siakwan and Gyau-Boakye 2000) usually exhibit low primary porosity and permeability. Groundwater occurrence in this province is usually regulated by secondary porosity, resulting from chemical weathering, faulting and fracturing (Gumma and Pavelic 2013). Hydrogeological properties in this area vary mainly due to the anisotropic nature of fracture networks and the different intensity and complexity of weathering processes accountable for regolith development (Dapaah-Siakwan and Gyau-Boakye 2000). In the Voltaian province, sedimentary rocks are characteristically impermeable and well consolidated. Fracturing, faulting and weathering have nonetheless increased the permeability of these rocks on smaller scales (Carrier et al. 2008).

Central Tanzania The centre of Tanzania, which consists largely of an elevated plateau, contains national parks and grasslands to the north and south, and underlying soil which is mostly arable. The mid-plateau region of the country is defined as having a hot arid steppe climate. Two rainy

seasons are found in Tanzania: a short, light season from November to January, and a long, heavy rainfall period stretching from March to May (Rowhani et al. 2011).

Tectonically, the southern and northern highland areas of Tanzania constitute the main East African Rift System, extending to the north through Ethiopia and Kenya (BGS/WaterAid 2000; Grove 1983). The geology of Tanzania consists predominantly of the crystalline basement complex from the Phanerozoic (Upper Palaeozoic, Mesozoic and Cenozoic) and the Precambrian (Archaean, Proterozoic) (BGS/WaterAid 2000). This crystalline complex forms the major basement aquifer in Tanzania (Chacha et al. 2018). Aquifer productivity differs widely depending on type, and varies from one location to the other due to the large variability in geology and existing recharge mechanisms (BGS/WaterAid 2000). In the study area, the crystalline basement of the central plateau is minimally fractured and joints, and therefore, any groundwater flow is limited. Water flow is possibly greater in the uppermost part of the basement complex which has been extremely weathered in places, and hence is highly permeable and friable (Nkotagu 1996).

3 Methods and Materials

3.1 Data

An extensive water quality dataset has been compiled from 731 tubewells (Bangladesh [$n = 474$], Northern Ghana [$n = 184$] and Central Tanzania [$n = 73$]; note that the terms wells and tubewells are used interchangeably in this work) using the published literature (BGS/DPHE 2001; Smedley et al. 2002) available for these three areas (Fig. 1). Most of the wells have recorded a full range of chemical analysis, including trace elements. The range of concentration of minerals in drinking water can be compared to the WHO's RDI values, however this study will be limited to the macro-minerals (calcium, Ca; magnesium, Mg; sodium, Na; potassium, K) and micro-minerals (iron, Fe) and the levels of elements of possible concern [i.e., fluoride (F) and arsenic (As)] commonly found in groundwater in varying concentrations.

3.2 Framework of Data Analysis

The following were selected for this comparative study: calcium (Ca), magnesium (Mg), sodium (Na), potassium (K), iron (Fe), bicarbonate (HCO_3), fluoride (F) and arsenic (As). Details on the analytical procedures conducted, as well as the actual data, can be found in the cited sources (BGS/DPHE 2001; Smedley et al. 2002). The RDI for the different mineral nutrients varies depending on the age group, as

well as for any special groupings (e.g. pregnant women). For simplicity a fixed conservative value, as defined by Hoque and Butler (2016a, b), was used for each mineral nutrient: Ca (800 mg/d), F (4 mg/d), Fe (14 mg/d), K (3000 mg/d), Mg (300 mg/d) and Na (2000 mg/d).

The concentration levels for various minerals and toxicants found in the two aquifer types were compared by aligning the underlying hydrogeochemical processes. In addition, the concentrations of various ions (e.g. Na, Ca, Fe) were compared with the toxicants to determine any possible modulating effects.

For the RDI calculation, it was assumed a person drinks two litres of water from the same water-well on a daily basis

(Hoque and Butler 2016a; WHO 2011), although it should be noted that this figure can vary (Craig et al. 2015; Islam et al. 2011). Inter-aquifer RDIs for various minerals (Ca, Mg, Na, Fe) found in two litres of water were compared by normalising using the following equation.

$$\%R_x = \left(\frac{C_x \times 2}{R_x} \right) \times 100 \tag{1}$$

where $\%R_x$ is percentage RDI (as %) of 'x' (x being Ca, Mg, Na or Fe), C_x is concentration (in mg/l) of x in 1 litre of water, R_x is RDI amount in mg/day and finally multiplied by 100 to have the output in percentage.

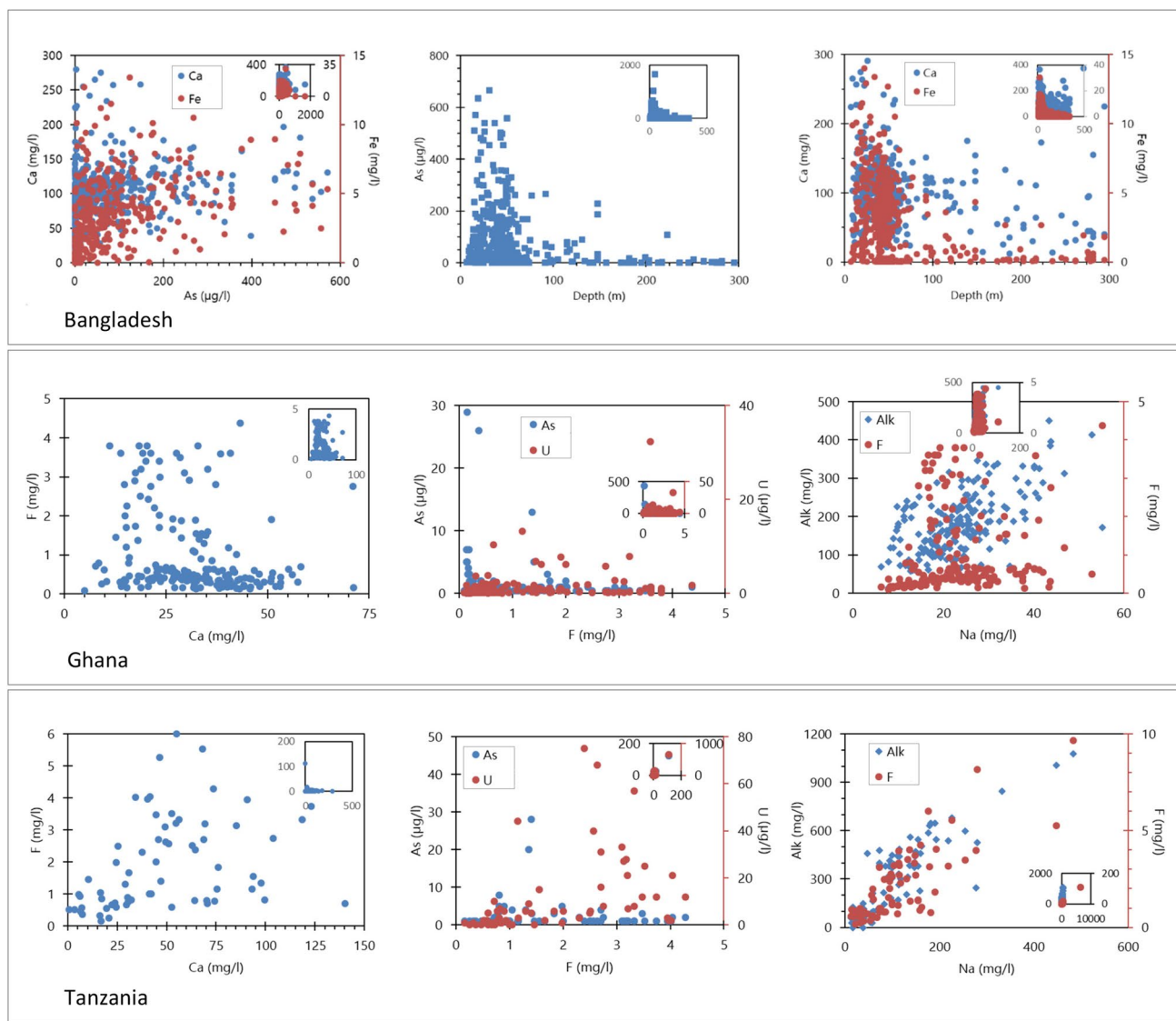


Fig. 2 The co-occurrence of the elements of concern and salubrious minerals. In Bangladesh, wells recording high levels of arsenic also contain a relatively high concentrations of calcium and iron; this tends to be restricted to the shallower depths. In Ghana and Tanzania, high fluoride wells appear to be relatively low in calcium, and arsenic

is not related to fluoride; however, the levels of sodium are relatively higher in the elevated fluoride wells and these same wells also tend to be more alkaline. Full concentration ranges are shown in the inset of the respective plots

Table 2 Summary statistics showing basic properties of groundwater constituents in the three case study areas

	Area/Properties	Major cations (mg/l)				Major anions (mg/l)		Other elements		
		Ca	Mg	Na	K	SO ₄	Cl	Fe (mg/l)	F (mg/l)	As (µg/l)
Bangladesh (<i>n</i> = 474)	Minimum	11.9	5.02	4.9	0.6	0.1	–	0.0025	–	0.25
	1st quartile	78.45	22.53	17.43	2.2	0.1	–	0.182	–	1.7
	Median	99	29.6	37.55	3.6	0.1	–	2.48	–	30.75
	Mean	102.8	36.0	142.5	5.1	–	–	3.1	–	84.8
	3rd quartile	120	39.98	193.25	5.8	1.4	–	4.64	–	106
	Maximum	366	193	2460	49.8	181	–	30.4	–	1660
Ghana (<i>n</i> = 184)	Minimum	4.9	1.5	6.3	0.015	0.1	0.015	0.001	0.09	0.2
	1st quartile	21.78	8.3	17.275	1.2	1.58	2.18	0.0025	0.338	0.2
	Median	30.6	11.1	22.75	1.9	2.85	4.3	0.057	0.5	0.2
	Mean	30.9	12.5	23.7	2.3	4.7	8.6	0.2	1.0	4.4
	3rd quartile	38.75	16.15	27.725	2.73	5.75	9.63	0.228	1.41	0.5
	Maximum	71.1	43.1	107.8	15.8	73.1	51.5	2.51	4.37	434
Tanzania (<i>n</i> = 73)	Minimum	0.7	1	13	0.1	1.8	0	0.005	0	0.5
	1st quartile	24.1	6.7	57.5	2.38	11.7	17.63	0.005	0.785	0.5
	Median	44.7	15.95	99.5	4.95	20.85	39.25	0.09	1.505	1
	Mean	52.8	23.0	203.1	6.0	61.3	76.5	0.4	3.9	4.4
	3rd quartile	70.05	29.43	154.75	8.4	55.03	97.7	0.168	3.2075	2
	Maximum	289.8	145	6450	20.8	967	443	11.17	111	123

4 Results

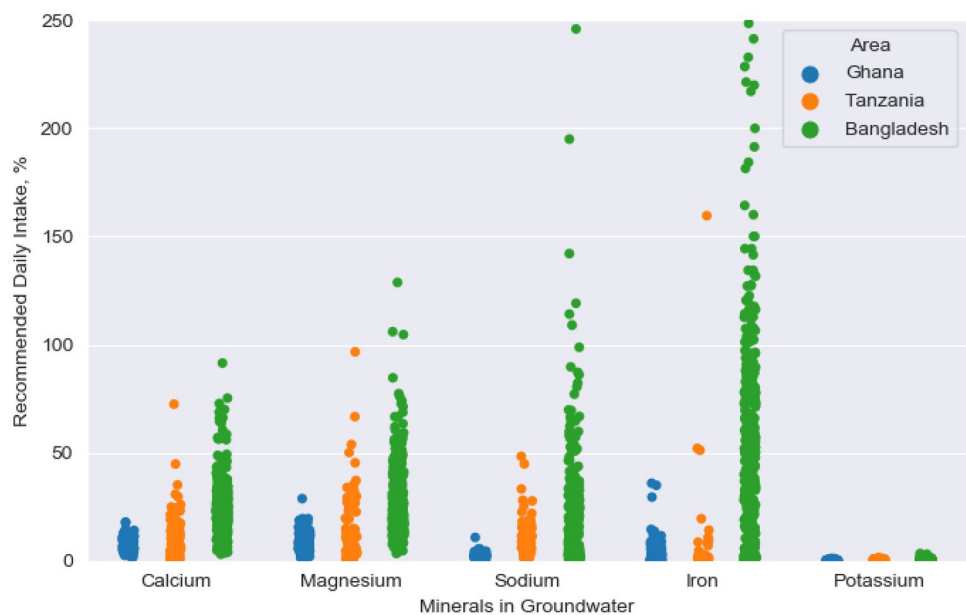
4.1 Macro-minerals

4.1.1 Calcium (Ca)

Calcium was detected in all wells in Bangladesh, but the

actual inter-well variation was substantial. The concentration ranged from 11.9 mg/l to 366 mg/l with an average of 103 mg/l (Fig. 2, Table 2). Elevated concentrations of Ca were found mostly in the shallower (< 100 m) wells (Fig. 2). In Ghana, the concentration ranges from 4.9 to 71.1 mg/l, with an average of 30.9 mg/l (Table 2) and is almost evenly distributed. High F wells also record relatively low Ca (Fig. 2). In Tanzania, the concentration is also highly

Fig. 3 Comparison of the recommended daily intake (RDI) of various mineral nutrients in percentages. It shows that 2 litres of water from the same well could give consumers a significant RDI of certain minerals in Bangladesh as compared to Ghana and Tanzania. If iron and sodium are present in excessive amounts however, water alone can give the full RDI of those minerals with associated detrimental health impacts on individuals



variable and ranges from 0.7 to 289 mg/l, with an average of 52.6 mg/l (Table 2). Wells with high F readings also contain relatively high concentrations of calcium.

About 75% of the wells in Ghana have Ca concentrations higher than 21 mg/l. For a normal adult who drinks two litres of water per day, the mineral uptake would be in the order of 42 mg of calcium. Similarly, in Tanzania and Bangladesh, a normal adult drinking same amount of water would ingest 48 mg and 157 mg, respectively. The recommended RDI value for Ca is 800 mg/day, so people in Bangladesh would get one-fifth of the Ca RDI from the drinking water alone. A percentage RDI plot (Fig. 3) indicates that individual in Bangladesh could get > 50% of Ca RDI from their drinking water, while the citizens of Ghana and Tanzania would rarely get over 25%.

4.1.2 Magnesium (Mg)

In Bangladesh, the concentration of Mg also varies between wells in a similar manner to calcium, ranging from 5.02 to 193 mg/l, with an average of 36 mg/l. In Ghana, the concentration varies from 1.5 to 43.1 mg/l, with an average of 13.0 mg/l (Table 2). In Tanzania, the concentration is highly variable and ranges from 1.0 to 145.0 mg/l, with an average of 22.8 mg/l.

Using an RDI value of 300 mg/day for Mg, people in Bangladesh can get up one-sixth of RDI just from drinking water alone, while this figure can be around one-twelfth for basement aquifer regions in Ghana. A large number of wells in Bangladesh provide > 50% of Mg RDI (similar to Ca), but few are noted in Tanzania and none are seen in Ghana (Fig. 3).

4.1.3 Sodium (Na)

In Bangladesh, the concentration of Na ranges from 4.9 to 2460 mg/l, with an average of 142.3 mg/l. Very high concentrations (> 200 mg/l and above) are restricted to the coastal plains, but it can be as low as 5 mg/l further inland (Fig. 4). The higher concentration of this element is also observed to be associated with shallow (< 100 m) wells. In Ghana, the concentration of Na varies greatly over the study area and ranges from 6.3 to 107.8 mg/l, with an average of 23.7 mg/l (Table 2). Both high and low values of Na were detected in close proximity, even in the same village. In Tanzania, a very wide concentration range characterises Na, i.e. 13–483 mg/l (with the exception of a very high, 6450 mg/l, reading) with an average of 203.8 mg/l (Fig. 4). High Na wells, in both Ghana and Tanzania, also contain high concentrations of F and generally have higher alkalinity (Fig. 2, Fig. 4). In Tanzania, Na has a stronger association with alkalinity and F compared to Ghana.

About 75% of all studied wells have Na concentration higher than ca. 20 mg/l. When compared to 2000 mg/day of Na RDI, at least 15% of the wells contribute over 20% of the RDI, but none of the wells in Ghana fall in this. However, in the coastal areas of Bangladesh and some wells in basement aquifers (particularly in Tanzania), may contribute up to and over 50% of Na RDI just from two litres of drinking water alone (Fig. 3).

4.1.4 Potassium (K)

In Bangladesh, the concentration of K ranges from 0.6 to 49.8 mg/l with a mean of 5.1 mg/l. In Ghana, the concentration ranges are relatively low but the variation is insignificant in basement aquifers, ranging from 0.015 to 15.8 mg/l, with an average of 2.24 mg/l. In Tanzania, it ranges from 0.1 to 20.8 mg/l, with an average of 6.0 mg/l (Table 2). When compared to a RDI value of 3000 mg/day (Fig. 3), it appears that potable water is not a source of any meaningful amount of K in any of the case study areas.

4.2 Micro-minerals

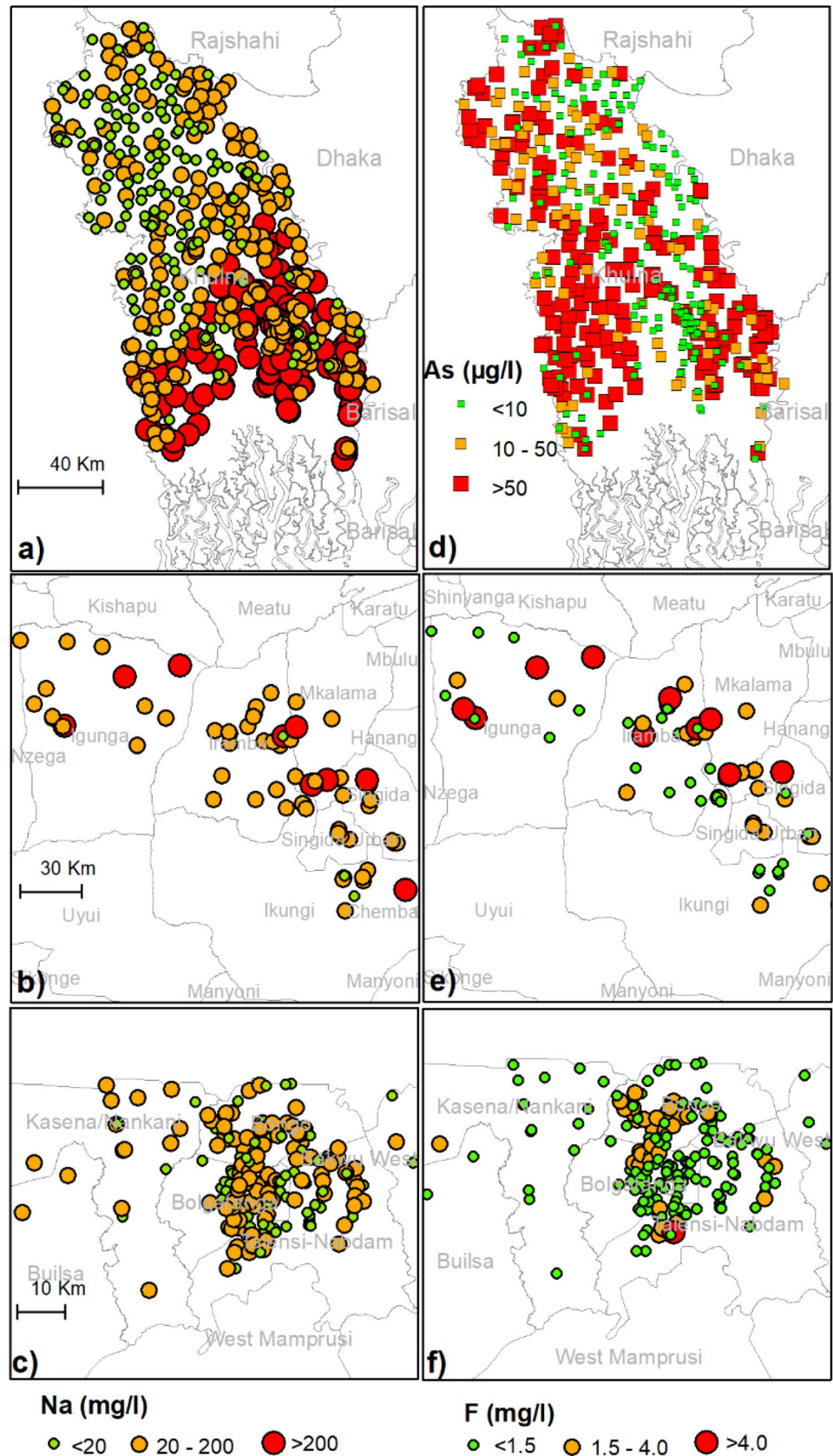
4.2.1 Iron (Fe)

In Bangladesh, Iron concentration ranges from 0.0025 to 30.40 mg/l with an average of 3.1 mg/l (Table 2). A high degree of variability was observed, with some wells having very low concentration, particularly those wells with a depth > 100 m. They are also low in Ca content (Fig. 2). For Ghana, the Fe concentration ranges from 0.001 to 2.51 mg/l, with an average of 0.18 mg/l. Iron concentrations in Tanzania are similar to Ghana, ranging from 0.005 to 11.17 mg/l; note the third quartile is 0.168 mg/l. When compared to an RDI value of 14 mg/day (Fig. 3), some wells, particularly in Bangladesh, provide a significant part of the daily Fe intake.

4.2.2 Fluoride (F)

The Bangladesh dataset does not include information about the amounts of F found in wells, but other studies reveal that the concentration is insignificant (Hoque et al. 2003). In Ghana, however, it ranges from 0.09 to 4.37 mg/l, with an average of 1.0 mg/l (Table 2). The high concentration of this element in the study area is associated with the central areas (around Bolgatanga), where the aquifer is mainly characterised by granitic, basement rocks. In Tanzania, the concentration ranges from zero to 111.0 mg/l, with an average of 3.94 mg/l (Table 2, Fig. 2); the figure of 111 mg/l was recorded in one of the wells. All other wells recorded a concentration of < 17.5 mg/l, with an average of 2.5 mg/l, an amount which is still higher than the WHO guideline

Fig. 4 Spatial concentration variation of elements of concern. Sodium (a–c: Bangladesh, Tanzania, Ghana) varies spatially within a region across the countries, but the concentration is significantly higher in sedimentary coastal aquifers. The fluoride in Bangladesh groundwater is not a problem but arsenic is a serious threat to public health (d), excessive fluoride is a widespread issue in basements aquifers (e [Tanzania] and f [Ghana]), and often, high fluoride wells have high sodium (b, c)



of 1.5 mg/l, although lower than the Tanzanian standard of 4 mg/l.

Most wells in Ghana and Tanzania provide a significant portion of the daily intake of F. Around 25% of the studied wells in Tanzania, and some in Ghana, do contain excessive levels of F, often contributing to the entire dietary RDI for F from just drinking 1 litre of water a day (Fig. 2).

4.3 Toxicants

4.3.1 Arsenic (As)

The presence and concentration of arsenic in groundwater is one of the most studied topics in Bangladesh. The concentration of As in groundwater is highly variable and ranges from 0.25 to 1660 µg/l with an average of 84.7 µg/l (Table 2). Extreme As values are rare, but these are real not outliers, and are likely to be associated with penecontemporaneous reduction of iron and manganese oxides (which reached to their maximum adsorption capacity). These are normally found at geological boundaries where grey, reduced sands are in lateral contact with brown, oxidised sands with the groundwater flowing from the former to the latter areas (e.g. Hoque et al. 2014). As shown in Fig. 2, the observed high concentrations are mainly associated with shallow wells which record high values of Ca and Fe. Table 2 presents a summary of the results from the three locations indicating that As is not widespread in the hardrock, basement aquifers.

5 Discussion

5.1 Geology and Water Quality

Comparison of the results from the three study sites demonstrates a clear difference in the chemical composition of the water. The chemical constituents in groundwater are normally derived from dissolution of the rocks when they come in contact with water during recharge, and subsequently while flowing through the respective geological medium. Laboratory studies and field observations show that Ca, Mg, Na and K dissolved more rapidly from the more highly reactive carbonate and sedimentary rocks (limestone, dolomite, sandstone, mudstone and siltstone), as compared with the relatively unreactive igneous and metamorphic rocks (Brantley 2008; Saxena and Ahmed 2001). Regions with the crystalline basement aquifers (such as Central Tanzania and Northern Ghana) are characterised by granitic and basaltic aquifers with fractures and joints. These rocks are often slow to weather, and rainwater infiltrates into fractures and remains very dilute and depleted in terms of mineral content. A factor that can influence the water chemistry,

however, is the water retention time within the fractures, as with increasing time the water can be influenced by the rock chemistry. This rock–water interaction is shown in a case study conducted in an area in Central Tanzania where high concentrations of water Na were derived from sodic alkaline rocks and volcanic activity (BGS/WaterAid 2000).

Sedimentary aquifers formed from materials deposited by geologically recent rivers or glaciers can also be highly permeable. Wells drilled into this type of aquifer typically vary from few tens of metres to few hundred metres with variable water quality, and often contain highly elevated levels of mineral nutrients compared to basement aquifers. Hoque and Butler (2016a) identified a number of processes at work when defining water composition at various depths in sedimentary aquifers: recharge, geochemical processes and groundwater flow. It was concluded that most mineralisation of infiltrating low-mineral rainwater occurs during recharge when carbonate dissolution and silicate weathering occurs, leading to increased levels of Ca, Mg, Na and K.

Dissolution of redox-sensitive constituents often leads to excessive amounts of some minerals in groundwater (Brantley 2008). For instance, oxidative dissolution of sulphide minerals, if and where available, i.e. in mineralised zones and in basement aquifers (generally unconfined and oxic) leads to As pollution (Ahoulé et al. 2015; Bretzler et al. 2017, 2019). On the other hand, in the semi-confined, anoxic aquifers found in Bangladesh, reductive dissolution of iron minerals (containing adsorbed As) causes widespread As pollution (Fendorf et al. 2010; McArthur et al. 2001; Nickson et al. 1998; Ravenscroft et al. 2009). Although both aquifers, through dissolutions gaining large quantity of Fe(II) into solution, often exceeding 0.3 mg/l (WHO aesthetic guideline value for drinking water), anoxic sedimentary aquifer has a relatively large quantity of dissolved Fe. In basement aquifers, long residing groundwater with high pH and alkalinity dissolves fluoride where apatite, biotite and hornblende minerals are present (Jacks et al. 2005; Saxena and Ahmed 2001; Smedley et al. 2002), and often also liberates uranium (> 30 ppb—WHO guidelines for drinking water). The water enriched with F also contains high Na (Fig. 2) most likely resulting from ion exchange (Rama and Keshav 2018). The local topography can influence the amount of groundwater flow divides, and local flow systems can lead to shorter residence times and intense flushing and, therefore, low content of minerals in groundwater (Bhattacharya et al. 2012). In contrast, coastal sedimentary aquifers often contain excessive Na, as found in Bangladesh (Figs. 3 and 4), because of seawater incursion into storage areas of remnant connate water (Worland et al. 2015), episodic recharge from storm surges or from ponding of saline water (Rahman et al. 2018). Fine-grained sedimentary deposits such as clay or silts are often interbedded within the sand layers in sedimentary aquifers, known as aquitards,

providing hydraulic basis for hierarchical flow systems (Hoque et al. 2017). Often the deeper flow systems (made of regional groundwater bodies of relatively homogeneous water quality) contains low amounts of As and Na, and can provide superior drinking water (Burgess et al. 2010; Hoque et al. 2017; Ravenscroft et al. 2018).

5.2 Efficacy of Nutritional Value and Toxicity

In general, the mineral constituents in drinking water can be healthy, harmful or toxic depending on the respective concentrations (i.e. dosage). The simple ionic form of minerals found in water allows easy absorption within the human gut compared to the more complex minerals found in food (WHO 2009), however current protocols used in assessments conducted to estimate the amount of minerals found in local diets does not consider drinking water (Sharma 2011). The minerals in the water of a region have previously been linked to health outcomes of local population in some Asian deltas (Ahmed et al. 2018; Chase et al. 2019; Hoque and Butler 2016a; Naser et al. 2019). In this current study, groundwater quality from three case areas was compared with respect to recorded values of Ca, Mg, Na, K and Fe (Fig. 3). It was found that drinking water provides variable amount of Ca, Mg and Fe in all areas, but the amount is significantly higher (almost double) in the sedimentary aquifers found in Bangladesh.

Calcium insufficiency (Bromage et al. 2016) and Fe deficiency (Kamruzzaman et al. 2015) are prevalent in Bangladesh; therefore, adventitious contribution by drinking water (Hoque and Butler 2016a) would be compromised when a person switched to required As-safe deep wells which are generally low in Ca, Mg and Fe (Fig. 2), and appropriate intervention through food choices needs to be made. The prevalence of anaemia in much of Bangladesh population decreased around 2001 when most of the population switched from using surface water to shallow groundwater so as to avoid various pathogenic gastrointestinal diseases associated with the surface water. The shallow groundwater was found to contain excessive iron, which inadvertently reduced anaemia, but ironically the same water was found to contain excessive arsenic. The drilling of deeper wells to reduce the concentration of arsenic, which also resulted in lower iron levels, may have resulted in a deterioration of the anaemia situation. It has been observed that arsenic-induced skin lesions in Bangladeshi women are three times higher in those who are anaemic (Kile et al. 2016), but many high arsenic wells also have significant levels of Fe which may mask the skin manifestation of arsenic poisoning.

People relying on shallow groundwater for drinking may be getting a significant amount of Ca, Mg and Fe but that often may be associated with the carcinogen As. There

is currently no understanding of how the coexistence of these various minerals moderate the toxicity of As (Hoque and Butler 2016a). It is likely that the harm caused by As outweighs the benefits of the salubrious minerals. At the same time some of the same water contains excessive Na which, with dietary intake, may result in hypertension (Khan et al. 2011, 2014; Scheelbeek et al. 2017, 2016).

Fluoride is commonly found in the basement aquifers (Kimambo et al. 2019), though in some places, including Tanzania and some wells in Ghana the concentration can be excessively high (Smedley et al. 2002). In these cases it can provide double the RDI value, i.e. over 8 mg/d from two litres of drinking water. Locals get F from food, which is also the case for Na. It is well known that individuals with nutrient deficiencies, particularly those with insufficient Ca intake, are more susceptible to F toxicity, including the effects on bone health (Marier and Rose 1977; Shankar et al. 2013). Ironically, high F wells generally contain lower levels of Ca (Fig. 2). In addition, high F wells are also seen to have relatively high amounts of Na (particularly in Tanzania) contributing up to 50% of RDI (Fig. 3) and excessive uranium (occasionally over the WHO guideline value of 30 µg/l) (Fig. 2). Hypertension associated with high Na contributions from fluoride-polluted drinking water is poorly understood, although Na derived from drinking water was found to be associated with hypertension in Bangladesh (Scheelbeek et al. 2017). The wells recording high Na and F readings also contain proportionally higher levels of alkalinity (Fig. 2) which were found to counter the impact of excess Na in some settings (Santos et al. 2010; Schoppen et al. 2004); however this effect has not been studied in fluoride-affected areas.

5.3 WHO Guideline Versus Regional Standards: 'Political Exposure'

The approach of the WHO in dealing with the issue of drinking water in developing countries has been concentrated more on accessibility rather than quality (Zimmerman et al. 2008). This has led to a debate regarding the quality versus quantity of drinking water, with an emphasis on chemical quality gaining more attention. WHO have established health-based guideline values in drinking water for F (1.5 mg/l) and As (10 µg/l) (WHO 2011). The value for Fe has been set at 0.3 mg/l, as above this level water may cause staining and be aesthetically poor. The limits for Cl and Na are set at 250 and 200 mg/l, respectively, as concentrations above these levels make the water taste salty. In regards regional drinking water standards, the case study areas have their own standards which have been compared to the WHO guidelines (Fig. 5). Except for the gold mining belt in Ghana, there is seemingly no problem associated with As in groundwater both in Ghana and Tanzania

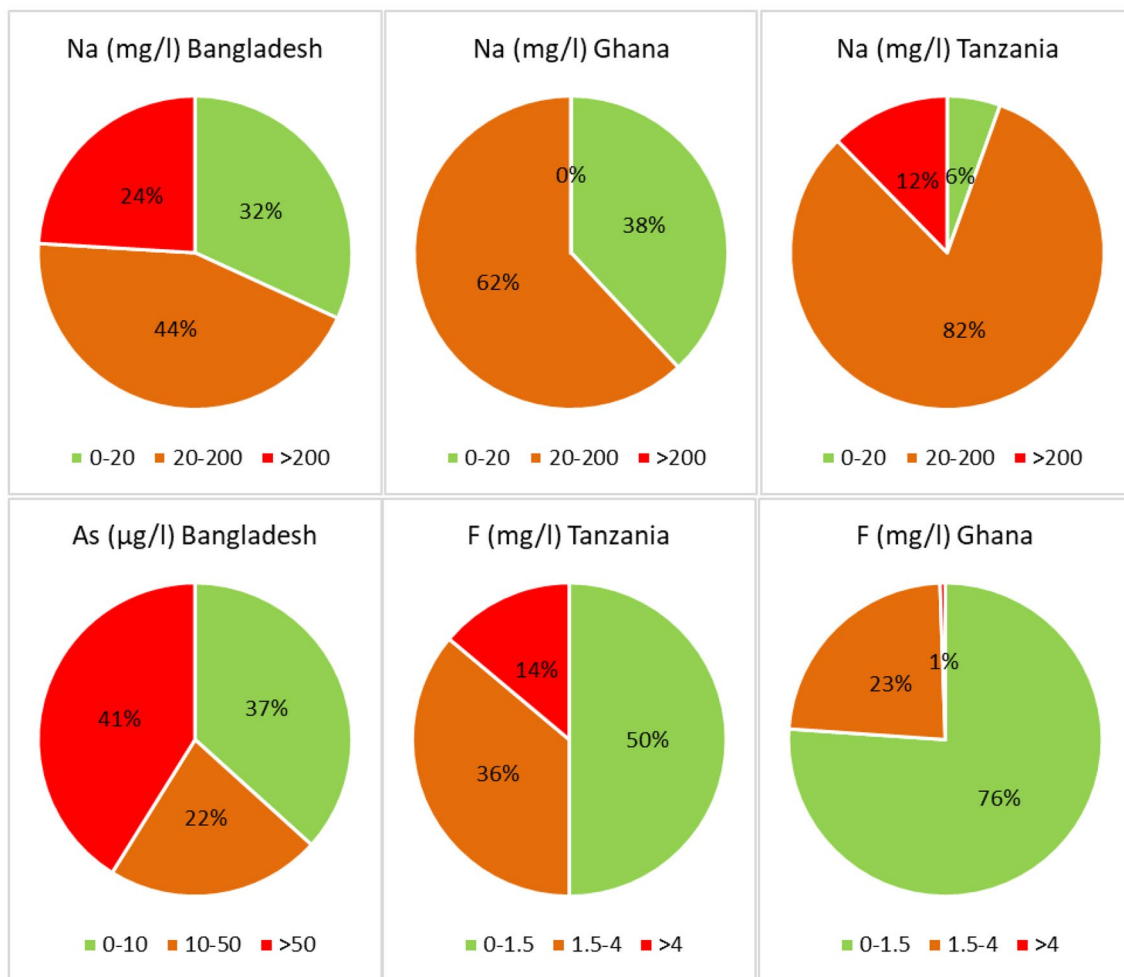


Fig. 5 Percentage of wells in relation to local standards, acceptability limit and international guidelines. Currently, sodium in drinking water has no WHO guidelines (except 200 mg/l as a taste threshold, above this value water may taste salty), while WHO states 20 mg/l from drinking water in the daily total dietary intake in the calculation

of sodium (WHO 2003). Arsenic has a WHO guideline of 10 µg/l while most developing countries (including Bangladesh) use 50 µg/l as the local standard. The fluoride in groundwater is excessive in Ghana. In Tanzania, groundwater often exceeds the WHO acceptable limit of 1.5 mg/l while the local standard for fluoride is 4 mg/l

(Ahoulé et al. 2015). However, in Bangladesh, As is particularly problematic, where around 60% of wells exceed the WHO guideline (10 µg/l) while about 40% exceed the Bangladesh standard of 50 µg/l (Fig. 5). The current study has used an old dataset for the analysis, therefore more recent data may show lower exposure levels, however as of BBS/UNICEF (BBS/UNICEF 2018) >40% wells have As level over WHO guideline values in that particular area of Bangladesh. The results show there are no issues with Fe in Ghana and Tanzania, as about 75% of the wells analysed in both areas have concentrations lower than the WHO guideline value of 0.3 mg/l. In Bangladesh, more than 75% of the wells analysed have concentrations exceeding the WHO guideline value for Fe, a situation diametrically opposed to that found in both Tanzania and Ghana. However, as most people in rural areas quickly drink the raw water, rarely confronted

with the aesthetic aspects of this excessive Fe, and indeed tend to benefit from the reduction in anaemia that it confers (Briend et al. 1990; Hoque and Butler 2016b; Merrill et al. 2011).

In Tanzania, the national standard for F is 4.0 mg/l, while Ghana has adopted the WHO value of 1.5 mg/l (Malago et al. 2017). Fluoride concentrations above 1.5 mg/l in drinking water can result in dental fluorosis and cognitive impairment with concentrations above 4 mg/l leading to skeletal fluorosis (Peckham and Awofeso 2014). Evidence of dental fluorosis has been reported in some places in Tanzania (Smedley et al. 2002).

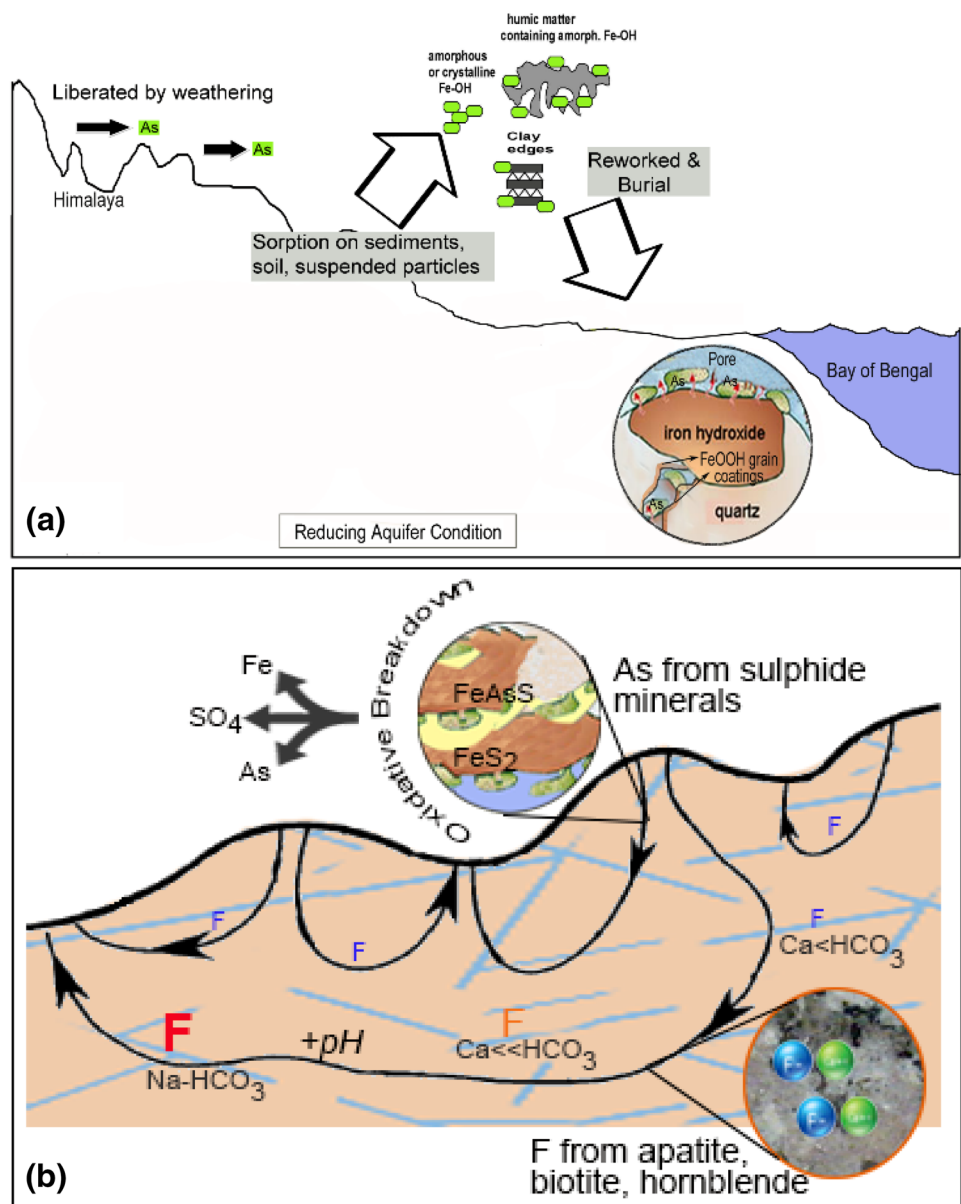
Arsenicosis and fluorosis, two endemic diseases known to result from (low) concentrations in groundwater over long-term exposure timeframe, have been major public health concerns for several decades (Chouhan and Flora

2010). Excessive Na in drinking water is now on the list, as this may lead to hypertension and cardio-metabolic diseases when added to any Na acquired from a normal diet. Although there is a health-based guideline value for As and F, no such guideline exists for Na. The WHO argue that drinking water does not contain excessive level of Na (WHO 2011) and implied that it is 20 mg/l (WHO 2003) but this value is not be correct for many water wells situated in deltaic aquifers (Hoque and Butler 2016a) and many wells with excessive F drilled into basement aquifers (Fig. 5). The implied concentration in drinking water is 20 mg/l, as WHO assumed drinking water containing 20 mg of Na per litre would lead to a daily intake of about 40 mg of Na (WHO 2003). If a person takes 20 mg/l as

a cut-off, then ca. > 60% wells in Bangladesh and Ghana and > 80% wells in Tanzania contain excessive levels of Na (Fig. 5). Therefore, the WHO should look into their recommendation for drinking water Na concentration, which has not been revised since 1993, if they want to reduce the total daily intake of Na for better cardiovascular health outcomes.

The WHO's 1993 guideline values were adopted as a national standard by most developing countries where excessive As, F and Na in groundwater is highly prevalent. It is well known that individuals with nutrient deficiencies are more susceptible to arsenicosis or fluorosis, and therefore, developing countries should adopt newer, usually lower, concentration thresholds rather than older, higher ones. It

Fig. 6 Conceptual medical hydrogeology models. **a** Arsenic is released into in reducing aquifer in sedimentary terrain because of iron reduction which also facilitates release of other trace metals in water, while b) oxidative environment may allow breakdown of some minerals and rock-water interaction leading to fluoride and others minerals including arsenic. In the basement aquifer, with suitable mineral suit, fluoride is generally associated with long residence time, high pH and excessive bicarbonate and ion exchange along the flow line leading to sodium enrichment. Arsenic is sporadic in the basement aquifers as the sulphide minerals which hosts are patchy



should be noted that the adoption of a national standard is effectively voluntary, as compliance is not enforced. It has limited statutory value as in most cases a consumer cannot make anybody liable for breach of standard limits (Meharg and Raab 2010; Sharma 2017). Higher threshold values assist governments politically by providing theoretically 'safe water' to its citizens. In reality, 22% of the population in the Bangladesh study area (Fig. 5) are, according to the national standard for As, sourcing water from a presumed As-safe supply. Many people may have sourced their water from a safe-well (in light of WHO's guideline) if they knew their principal water source was not really As-safe. The situation is similar for As where 36% in Tanzania (Fig. 5) could have sourced drinking water from alternative low-F wells if they were not given the 'false' threshold. In many rural settings, people are willing to pay for their water (Whittington et al. 1990; Witt 2019), and therefore, setting national standard carefully, or replacing the so-call (non-legal) standard with lower unenforced recommended limit, may help many to avoid this unnecessary 'political exposure'. Food is the main source of daily exposure to Na, therefore, its content in water needs to be communicated to people in similar way by both international and national policy instruments. This may lead to a reduction in number of preventable cardiovascular cases globally.

5.4 Heterogeneous Water Quality, Medical Hydrogeology and Water Supply Framework

Groundwater quality is heterogeneous, varying from one area to another at any given scale (from a continental to a village, and even between wells in a village, which makes it a challenge to maintain water quality standards, especially in rural settings. A distributed and individually-managed service delivery has many benefits to overall efficiency and sustainability of water supplies, however problems arise when complying with specific water quality throughout an area. The investigative analysis of water quality at various spatial scales under a 'medical hydrogeology' framework may help in regionalised water quality (Fig. 6). Medical (hydro)geology traditionally focuses on toxicants in groundwater (Selinus 2013); some apparently harmless looking content in water can be harmful if considered in a holistic manner (Hoque and Butler 2016a, b). For instance, Na in water added to Na from the normal diet, may lead to a total ingested amount over the relevant health threshold, which may lead to hypertension and other cardio-metabolic diseases. Conversely, the toxicity of some toxic substances (e.g. As, F) may be moderated by the presence of specific minerals in the same water or in dietary intake. Therefore, medical hydrogeology should focus on holistic water quality and their interactions with other coexisting ions in terms of both interference and synergy. Regionalised water quality and

an understanding of medical hydrogeology of specific local aquifer/zones may be used for rural water supplies. Furthermore, health risk assessment for ingestion of minerals and toxicants of concern via drinking water (e.g. Kavcar et al. 2009) may aid the decision-making process. In this case, there would be a need for a licensing system to be developed, as opposed to the current practice in most developing countries, where money is invested and equipment installed without needing any permission. A licencing system would help government organisations to guide individuals and provide the structure and resourcing to effectively assess and monitor water quality.

6 Conclusions

We compared groundwater constituents and trace elements between the basement and sedimentary aquifers in the context of medical hydrogeology, i.e. status of groundwater mineral nutrients and pollutants, and their complex interaction in relation to human health. The three study areas (two from basement aquifers in Africa [Ghana and Tanzania] and one from sedimentary aquifer in Asia [Bangladesh]) were analysed in order to understand the relationship between aquifer properties and groundwater mineral nutrients. From this study, the following conclusions can be drawn:

- Geology plays an important role in determining types of minerals in groundwater. The sedimentary aquifers, as opposed to the basement aquifers, have a higher concentration of calcium, magnesium and iron, leading to a higher RDI. This must, however, be augmented by adequate food intake to meet the dietary needs of the local population;
- Regional standards in most developing countries are not current and do not provide adequate reduction in exposure. This is observed with both fluoride (in Tanzania) and arsenic (in Bangladesh). Careful consideration when setting standards and developing guidelines may reduce exposure risks;
- Current sodium guideline values developed by WHO for potable water needs revision. Regional water directives (industrial and developing countries alike) should also look into the contribution of drinking water derived sodium to total daily dietary intake of respective population.

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Compliance with Ethical Standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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