Elevated and variable groundwater iron in rural northwestern Bangladesh

Rebecca D. Merrill, Alain B. Labrique, Abu Ahmed Shamim, Kerry Schulze, Parul Christian, Robert K. Merrill and Keith P. West Jr

ABSTRACT

Over the past 30 years, tubewells have become a ubiquitous source of potable groundwater in South Asia. Considered safer than surface water, groundwater naturally contains minerals that may impact human health; however, few data exist on tubewell water mineral content or its association with human nutritional or health conditions. We surveyed iron concentration in tubewell water across a 435 km^2, contiguous, rural area in northwestern Bangladesh to map and quantify levels of iron in drinking water. One tubewell was randomly sampled from each of 948 adjacent grid cells 675 m^2 in size. Water sampling was standardized and iron concentration measured using a field-based colorimetric kit. The median (interquartile range) concentration of iron in tubewell water was 7.6 (1.6, 17.6) mg l^{-1}. There was high geographic variation (range of 0–46.5 mg l^{-1}), and iron in only 3% of surveyed tubewells fell below the WHO aesthetic cut-off of 0.3 mg l^{-1} suggesting elevated levels of iron throughout the area. Villagers accurately perceived groundwater iron concentration, based on a 4-point ('none', 'a little', 'medium', 'a lot') scale ($p < 0.001$). Water source iron content can be readily quantified in population settings offering the potential to evaluate the health relevance of groundwater iron exposure in rural communities.

Key words | Bangladesh, GIS, groundwater, iron, tubewell

Rebecca D. Merrill
Alain B. Labrique (corresponding author)
Kerry Schulze
Parul Christian
Keith P. West Jr
Department of International Health,
Johns Hopkins Bloomberg School of Public Health,
615 N. Wolfe Street,
Baltimore, MD 21205,
USA
Tel.: +1 410-955-2061
Fax: +1 410-955-0196
E-mail: *alabriqu@jhsph.edu*

Abu Ahmed Shamim
The JiVitA Project,
House 63, Road 3, Karanipara,
Rangpur,
Bangladesh

Robert K. Merrill
Catheart Energy, Inc.,
3811 Hogan Ct,
Sugar Land, TX 77479,
USA

INTRODUCTION

Groundwater use for drinking and cooking has become ubiquitous across much of South Asia, including Bangladesh, since international efforts began during the United Nations (UN) International Decade for Clean Drinking Water of the 1980s. The goal of this global initiative was to provide rural populations with clean water sources free of bacteria such as *Vibrio cholerae*, which causes cholera (Schultzberg 1980). The campaign successfully increased the use of groundwater for daily water consumption by initiating the installation of tens of thousands of hand-pumped tubewells and consequently reducing exposure to waterborne pathogens. With current estimates suggesting that there are over 10 million tubewells in Bangladesh, groundwater provides at least 90% of drinking water and a majority of dry season irrigation water (Ravenscroft 2003). This increased consumption of groundwater, however, has led to long-term population exposure to a wide spectrum of minerals naturally found in aquifers (Kinniburgh & Smedley 2001). For example, in some areas there has been unanticipated exposures to levels of arsenic (Brinkel *et al.* 2009; Rahman *et al.* 2009) and manganese (Wasserman *et al.* 2009) that are consistent with adverse health risks including cancers, cardiovascular disease, diabetes mellitus, hyperkeratosis, pigmentation changes, and kidney, liver, respiratory and neurological disorders (Kapaj *et al.* 2006; Balakumar & Kaur 2009). The government, with help from international and national organizations, is actively working to reduce exposure to arsenic by educating the public about the dangers of consuming toxic levels (defined as water with

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arsenic concentration $>10\,\mu g\ l^{-1}$ or $>50\,\mu g\ l^{-1}$ by the World Health Organization (WHO 2004) and Bangladesh government (Kinniburgh & Smedley 2001), respectively, and providing arsenic-removing filters and alternative water supplies (UNICEF 2009).

Other elements in groundwater may over time provide nutritional benefit. An example is iron, which is known to be present in aquifers in certain areas of Bangladesh varying greatly in concentration from virtually non-existent to levels above $60\,mg\,l^{-1}$ in other areas (Kinniburgh & Smedley 2001). If shown to be a bioavailable source of iron, individuals living within areas of elevated iron in groundwater who routinely consume the iron-rich water through drinking and cooking may be protected from iron deficiency and consequent anaemia. While such a relationship between consuming natural iron in groundwater and human iron status remains speculative, studies have shown that iron found naturally in water at a concentration of approximately $210\,mg\,l^{-1}$ in Gwynedd, UK, is highly bioavailable, reaching absorption rates as high as 40% (Worwood *et al.* 1996; Halksworth *et al.* 2003; McKenna *et al.* 2003). Additionally, one study in Bangladesh revealed a positive association between groundwater iron content categorized as above or below $1\,mg\,l^{-1}$ and weight and height for age of children after controlling for socioeconomic status defined by land and asset ownership and education level of the household head (Briend *et al.* 1990). However, this study did not account for the amount of water consumed.

Dietary studies typically do not consider water as a source of nutriment and, thus, pay little attention to assessing its potential influence on trace mineral status and health of populations. A first step toward investigating the association between dissolved mineral content in drinking and cooking water and nutritional status is to quantify levels of minerals in sources of routinely consumed groundwater in stable populations who rely on tubewells for drinking and/or cooking water.

This report describes a regional survey conducted in 2007 and 2008 by a research project (the JiVitA Project) (West *et al.* 2006) to develop a detailed map of groundwater iron concentration across a large, contiguous, rural area in northwestern Bangladesh for the purpose of serving subsequent studies of tubewell water use, iron exposure through water and maternal nutrition and health. Over 95% of the local population, similar to the rest of Bangladesh, relies on groundwater, sourced from tubewells, to provide drinking and cooking water and a majority of dry season irrigation water (Ravenscroft 2003).

METHODS

This study was conducted from November 2007 to February 2008 in a ~435 km^2 study area, with an estimated population of ~650,000, located in the rural northwestern districts of Gaibandha and southern Rangpur, Bangladesh (Figure 1).

Tubewell selection

Using an established and continuously updated geographic information system (GIS) map of the study area that included landmarks such as roads and tubewells (Sugimoto *et al.* 2007) we overlaid a grid of 675 m^2 quadrangles (25.98 × 25.98 m) onto the entire study area, using the ArcGIS custom overlay grid function. A total of 1,108 quadrangles, or 'cells', were created in the overlay, each containing some part of or touching the boundaries of the study area. Only cells with a centre within the research area ($n = 948$ (86%) cells) were selected to remain in this survey. Two-member teams of field technicians used handheld global positioning system (GPS) units (Garmin E-trex Legend®, Garmin, Kansas) onto which the waypoints, defined by latitude and longitude, for each cell centre had been uploaded and pre-printed paper maps displaying study area boundaries, roads and landmarks (households, government and non-government offices, etc.) for orientation. As seen in Figure 2, the maps depicted the cell centres

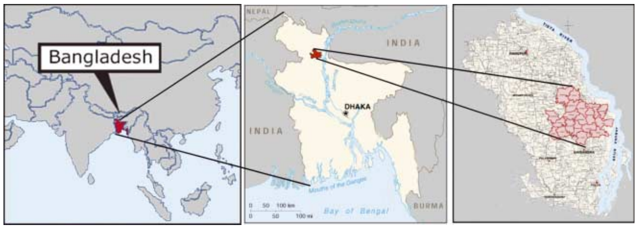

Figure 1 | The JiVitA maternal and child research site in northwest rural Bangladesh, covering 435 km^2 across the districts of Gaibandha and Rangpur.

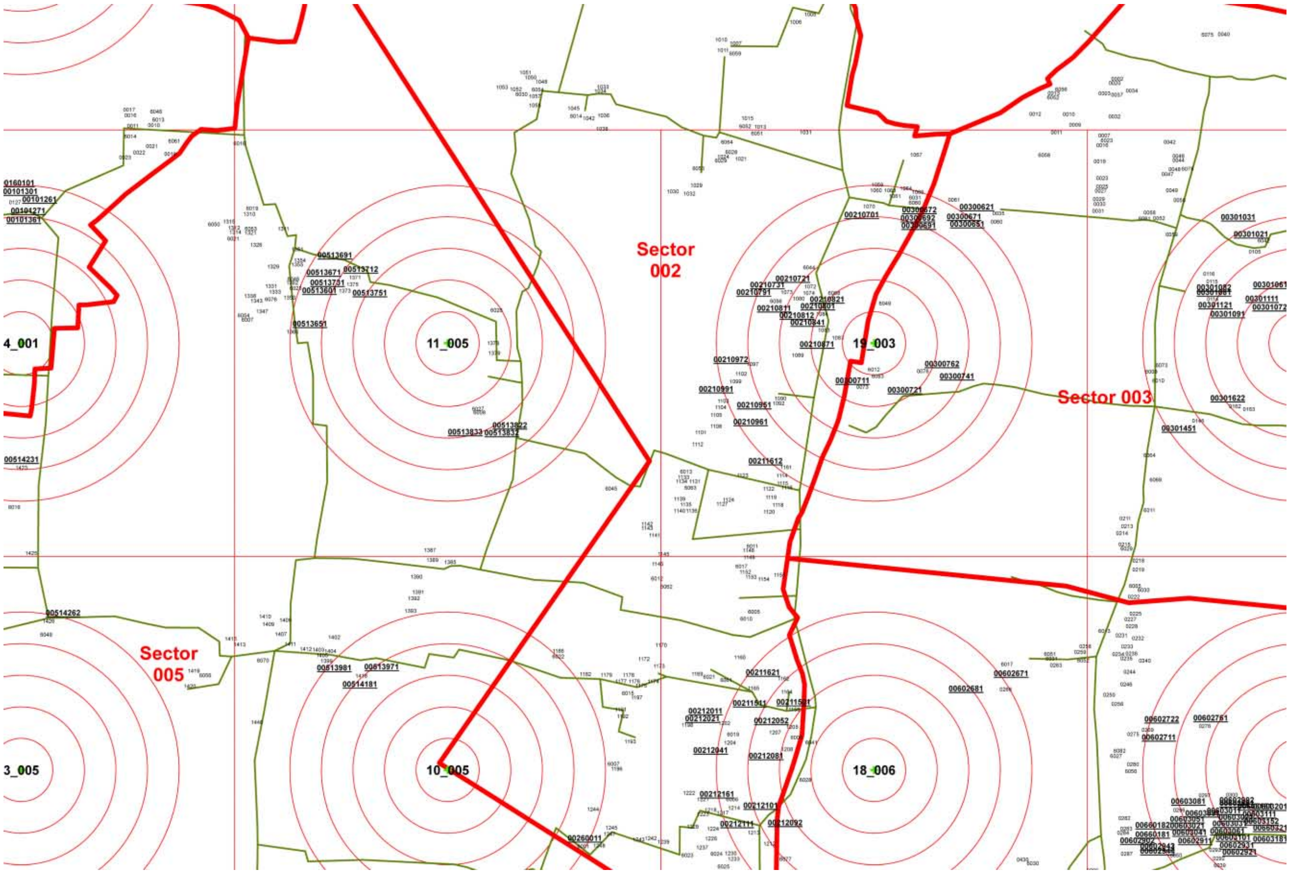

Figure 2 | Example of grid and landmark map used by field technicians to assist with selecting one tubewell per cell as close as possible to the grid centre. Bold lines represent study-defined administrative boundary; number in the grid cell centre represents the grid cell number and study-defined administrative unit in which is it found; 50 m concentric 'buffer zones' displayed within each individual 675 m^2 cell; households and other landmarks displayed using appropriate field codes and identifiers.

surrounded by five concentric 'buffer zones', each representing a 50 m increase in radius over the previous, smaller buffer. The objective of these maps was to facilitate the selection of one tubewell per grid cell as close to the centre of that cell as possible.

Before visiting a cell, teams used the grid cell maps to assess the likelihood of locating a tubewell within 50 m of the cell centre. The ubiquitous use of tubewells in this population (100% of JiVitA participants reported collecting household drinking water from tubewells; unpublished data) allowed the field technicians to assume that where a landmark or household was present there would be a high probability of finding a tubewell nearby. If there was no landmark or tubewell depicted on the map within 50 m of the grid centre, the team travelled to the landmark nearest to the grid centre. Once at the cell centre or landmark nearest the cell centre, the team selected the closest visible tubewell. If there were no or multiple tubewells in sight, the team spun an empty glass bottle on the ground to select a random direction to walk until a working tubewell was found and selected. Using this methodology, one working tubewell was selected from each of 948 grid cells.

On-site tubewell analysis

Exact tubewell location was recorded using the GPS handheld units. If present and available, the tubewell owner or a regular user of the selected tubewell was asked about the year of installation and well depth (in feet). Data collectors used relevant events, such as weddings or birth of children, to assist the respondent with year of installation. As the cost of tubewell installation is directly related to the number of pipes used, which have standard lengths, depth was calculated by asking the number of iron (5 feet), filter (6 or 10 feet), and/or plastic (15 feet) sections. A question about the perceived amount of iron in the tubewell water on a 4 point scale (e.g. 'none', 'a little', 'a medium amount' and 'a lot') was asked to assess the validity of using a simple question based on perception to assess magnitude of iron concentration. Subjects were readily able to answer this question without explanation most likely because of the organoleptic qualities associated with iron.

Initially, tubewells were purged of any residual minerals on the inside of the pipe and the aquifer source was reached by pumping a steady stream of water for 5 min. Subsequently, a water sample was collected directly from the tubewell water flow in a 1 l plastic container, previously rinsed three times using that tubewell's water. Temperature to the nearest 0.1°C (Portable Thermometer, Model No: ST-9269A/B/C, Winning Technology Ltd, Hong Kong) and pH to the nearest 0.1 pH unit (ADWA AD100 pH Electronic Meter, Adwa Instruments, Belgium) were recorded. After filling the 1-l bucket with a fresh sample of water directly from the tubewell flow, iron concentration to the nearest 0.1 $mg\,l^{-1}$ was measured using a previously validated field-based colorimetric iron test kit (HACH Iron Test Kit, Model IR-18B) following methods described in detail elsewhere (Merrill *et al.* 2009).

Quality control

From a random 10% of cells, water samples were collected, acid-preserved to $pH < 2.0$ to prevent iron precipitation, and stored the same day in a cool dark room until analysis within 1 month by atomic absorption spectrophotometry (AAS), an accepted standard for assessment of water iron content. Cells from which quality-control water samples were collected ($n = 109$) were pre-selected using GIS technology to ensure they were equally distributed across the study area and consequently collected periodically throughout the survey period because cells were visited from north to south. Field technicians were trained and standardized on form completion and field water analysis techniques and randomly observed by the study supervisor.

Statistical analysis

Variable distributions were explored and log transformed when necessary to achieve a normal distribution (groundwater iron). Correlations between continuous water characteristics (year of installation, depth (ft), pH, temperature (Celsius), iron concentration ($\ln(mg\,l^{-1})$)) were assessed using the Pearson correlation coefficient. Distributions across categorical variables (perceived iron concentration, categorized iron concentration) were compared using Fisher exact tests and non-parametric Kruskal–Wallis one-way analysis of variance.

Four categories were defined to describe tubewell groundwater iron concentration based on established recommendations for iron in water and daily dietary iron intake. Tubewells were defined as having 'no or minimal' iron if they were below the WHO aesthetic limit for iron in water defined at 0.3 $mg\,l^{-1}$ (WHO 2006). Where iron concentration was above 0.3 $mg\,l^{-1}$ but below the 2.0 $mg\,l^{-1}$ Joint FAO/WHO Expert Committee on Food Additives (JECFA) defined limit of iron in water, the tubewell was defined as having 'elevated' iron. The JECFA limit was established by allocating 10% provisional maximum tolerable daily intake (PMTDI) for iron, defined as 0.8 mg iron/kg of body weight assuming average body weight of 60 kg, to water assuming an average daily water consumption of 2 l (WHO 1984, 2004). Tubewells were defined as having 'high' levels of iron if the iron

Table 1 | Tubewell and water characteristics

	N	Mean (SD)	Median (IQR)	Range
Year of installation	942	2000.0 (7.0)	2002 (1997, 2005)	1938, 2008
Depth (ft)	815	46.4 (18.8)	45 (35, 55)	24, 185
pH	864	6.6 (0.8)	6.8 (6.3, 7.1)	3.8, 8.6
Temperature (Celsius)	879	25.9 (0.8)	25.8 (25.4, 26.4)	19.4, 36.2
Iron ($mg\,l^{-1}$)	948	11.0 (10.5)	7.6 (1.6, 17.6)	0.0, 46.5

Table 2 | Distribution of 948 tubewells across water iron concentration ($\mathrm{mg\,l^{-1}}$) categories based on current WHO and JECFA water-related and IOM dietary daily iron intake recommendations

Category	**N (%)**	**Mean (SD)**	**Median (IQR)**
Minimal 0– $< 0.3^{*}\,\mathrm{mg\,l^{-1}}$	25 (3)	0.2 (0.1)	0.2 (0.2, 0.2)
Elevated 0.3–$2.0^{\dagger}\,\mathrm{mg\,l^{-1}}$	234 (25)	1.0 (0.4)	1.0 (0.7, 1.3)
High >2.0–$22.5^{\ddagger}\,\mathrm{mg\,l^{-1}}$	518 (55)	10.1 (5.7)	9.5 (4.7, 14.8)
Very high $> 22.5\,\mathrm{mg\,l^{-1}}$	171 (18)	29.0 (5.3)	27.3 (25.0, 31.3)

*WHO (2006) aesthetic cut-off.

†JECFA provisional maximum tolerable daily intake for iron in water (WHO 1984, 2004).

‡Per litre equivalent of the IOM recommended tolerable upper intake level of 45 mg iron/day for daily iron intake for adults (excluding iron supplements) assuming 2 l/day water consumption (Otten *et al.* 2006; WHO 2006).

concentration was greater than $2.0\,\mathrm{mg\,l^{-1}}$ but less than or equal to $22.5\,\mathrm{mg\,l^{-1}}$. This limit represents the daily per liter equivalent, again assuming average daily water intake of 2 l, of the Institute of Medicine (IOM) recommended daily tolerable upper intake level for iron of 45 mg for men and women aged 9 years and older (Otten *et al.* 2006; WHO 2006). Finally, tubewells with an iron concentration greater than $22.5\,\mathrm{mg\,l^{-1}}$ were defined as having 'very high' iron. Categories were used to explore the distribution of iron concentration across the study area.

Ordinary kriging was used to develop a smoothed, three-dimensional display of groundwater iron concentration across the study area (Hill *et al.* 2009). This geostatistical method is used to estimate values, in this case groundwater iron concentration, in non-sampled areas using data from sampled points and calculated spatial correlation, rather than absolute distance, to apply weights during estimation.

RESULTS

From each of the 948 cells one tubewell was successfully identified and selected. Tubewell and water characteristics are presented in Table 1. A majority of tubewells ($n = 726$, 77%) were installed within 10 years of the survey (median (interquartile range, IQR) year of installation: 2002 (1997, 2005)). Median (IQR) depth (feet) ($n = 815$) was 45 (35, 55); 98% of tubewells were less than 100 feet deep. Depth was positively associated with iron in water ($\ln \mathrm{mg\,l^{-1}}$) ($r = 0.18$, $p < 0.001$). Median (IQR) temperature (degrees Celsius) ($n = 879$) and pH ($n = 864$) were 25.8 (25.4, 26.4) and 6.8 (6.3, 7.1), respectively. Temperature and pH were not significantly associated with iron in water ($\ln \mathrm{mg\,l^{-1}}$) ($r = -0.06$, $p = 0.09$; $r = 0.05$, $p = 0.17$, respectively) or each other ($r = -0.03$, $p = 0.42$). Tubewell iron concentration was high (Table 2) with a median (IQR) iron concentration of 7.6 (1.6, 17.6) $\mathrm{mg\,l^{-1}}$ (Table 1, Figure 3) and showed a wide range (0.0 to $46.5\,\mathrm{mg\,l^{-1}}$). Field-based results did not differ from gold standard AAS results ($n = 109$, mean difference: $0.52\,\mathrm{mg\,l^{-1}}$, $p > 0.25$ by Student's t test).

Only 3% of tubewells had 'no or minimal' iron and a majority of the tubewells (73%) had iron concentrations defined as 'high' or 'very high'. Tubewells with extreme iron concentration values were distributed across the study area (Figure 4). However, after interpolating iron concentration values using ordinary kriging, general areas with 'very high'

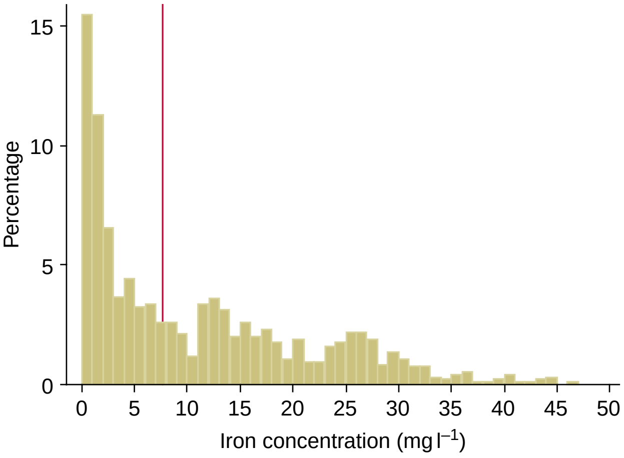

Figure 3 | Histogram of tubewell water iron concentration ($\mathrm{mg\,l^{-1}}$) ($n = 948$) across the survey area including line at median value ($7.6\,\mathrm{mg\,l^{-1}}$).

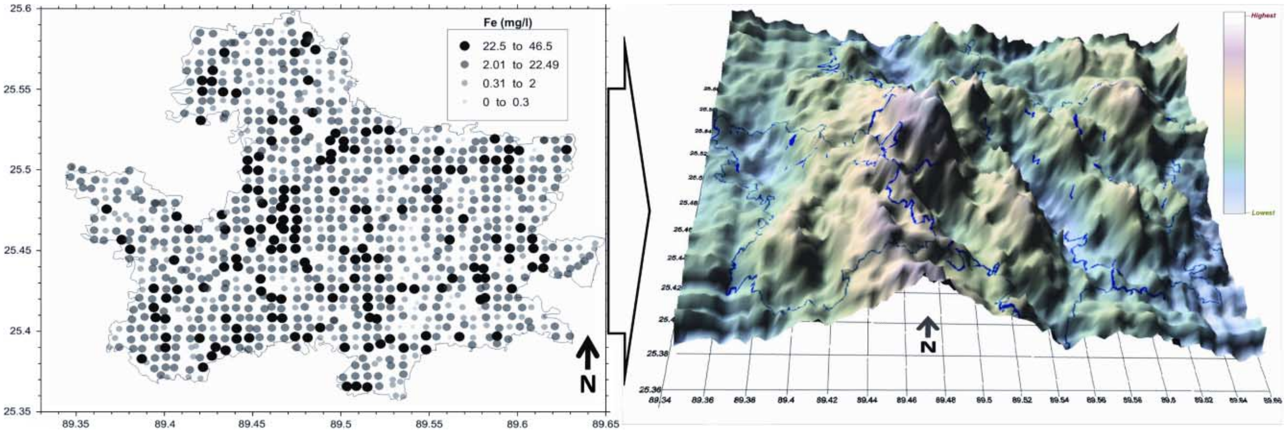

Figure 4 | Distribution of tubewell ($n = 937$) iron concentration across JiVitA study area (elevation in conjunction with colour depicts the estimated levels and variation in groundwater iron concentration).

iron concentration values were exposed. Respondents' report of perceived iron concentration was positively associated with measured iron concentration (Figure 5, $p < 0.001$). Median (IQR) iron concentrations by increasing perception score were 1.0 (0.6, 2.1), 3.0 (1.1, 6.4), 11.7 (6.2, 17.7), and 18.9 (13.5, 26.6) ($n = 214$, 243, 149 and 340, respectively).

DISCUSSION

We found in a rural northwestern area of Bangladesh that only 3% of the 948 tubewells sampled had an iron concentration below the WHO aesthetic limit ($< 0.3 \, \mathrm{mg\,l^{-1}}$) and 72% had high to very high levels of iron ($> 2.0 \, \mathrm{mg\,l^{-1}}$). Using ordinary kriging to interpolate the entire 435 $\mathrm{km^2}$ study area based on the field-based water analysis results, we showed that the area has elevated iron concentration in groundwater. However, iron concentration in the study area may be slightly higher than our ordinary kriging methods estimated based on findings from previous studies showing that this method, when predicting groundwater arsenic levels, may underestimate mean concentrations in non-sampled areas (Hossain *et al.* 2007; Hill *et al.* 2009). The findings from our study suggesting elevated and variable levels of iron in groundwater are in agreement with results from the less densely sampled nationwide British Geologic Survey (BGS) survey published in 2001 (Kinniburgh & Smedley 2001). We also detected considerable variation in the concentration of iron in the region. For example, tubewells less than 50 m apart were found to have a 25-fold difference in groundwater iron concentration. By combining the densely sampled data using a 625 $\mathrm{m^2}$ grid system sampling scheme with ordinary kriging to interpolate the non-sampled areas, we assured, with a high degree of resolution, geographic balance and representation across a large survey area. Additionally, iron concentration was measured on the spot using a field kit previously validated against gold standard atomic absorption spectrophotometry (Merrill *et al.* 2009).

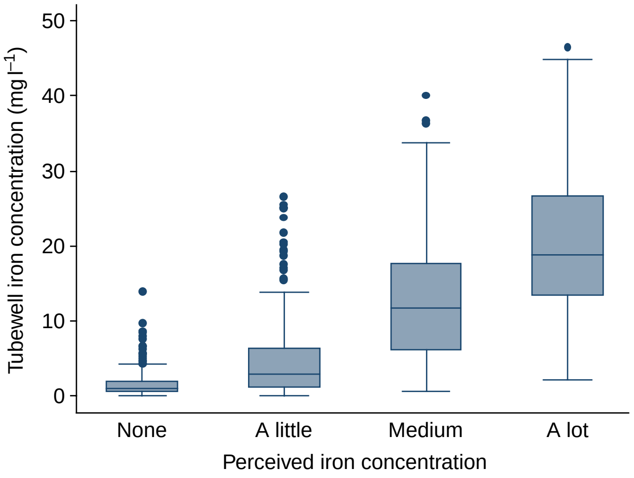

Figure 5 | Iron concentration in tubewell water ($\mathrm{mg\,l^{-1}}$) as determined by field analysis, stratified by category of respondent-perceived iron, on a 4 point scale (based on the question 'How much iron does this tubewell contain?'; significant difference in iron concentration by perception score, $p < 0.001$ by Kruskal Wallis test).

It is noteworthy that the local population was capable of estimating, using a 4 point scale, the level of iron in their tubewell, as shown by the significant increase in median iron concentration with reported perception score from 1.0, 3.0, 11.7 and 18.9 mg l^{-1} for 'none', 'a little', 'a medium amount' and 'a lot', respectively. Their ability to perceive the magnitude of iron present is most likely due to the increase in negative organoleptic properties, such as a rust-like smell and taste and a red discoloration, associated with increased iron concentration. Using this simple question, 'How much iron do you think is in the water that you pump from this tubewell?', to estimate the magnitude and variability of iron exposure through groundwater, a population health and dietary survey carried out in areas with variable groundwater iron concentration may be able to obtain an initial, semi-quantifiable impression of the iron distribution in water sources and consumption levels.

CONCLUSION

This survey has documented the elevated levels of iron exposure through groundwater used for drinking and cooking in a typical, northern, rural area of Bangladesh. Iron concentration was quantified on the spot with a validated, easy to use, field kit as well as by magnitude with a short survey question. Both of these methods would be easy to incorporate into a large, field-based survey. The consequences, if any, either positive or negative, of chronic exposure to this iron are unknown at this time and deserve additional attention.

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