Assessing the spatial pattern of iron in well water from a small central Florida community

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ABSTRACT

Iron is one of the most common elements in the Earth's crust, which corresponds to it being a common constituent in drinking water supplies. Residents of Bithlo, an unincorporated community in east-central Florida, have observed that their drinking water tastes like metal and stains clothing and teeth. An evaluation of water samples collected from over 200 private drinking water wells revealed iron concentrations that exceeded the US Environmental Protection Agency's (EPA's) secondary standard of 0.3 mg/L. Households with and without point-of-entry treatment were found to have over three times (0.92 mg/L) and ten times (3.86 mg/L) more iron than the EPA's secondary standard, respectively. The human health-based threshold of 4.2 mg/L established by the Centers for Disease Control and Prevention was exceeded in 38.6% of untreated residences. Community-wide statistical and spatial water-quality trends were developed by combining the collected well water quality data with historically available water quality reports. Spatial analyses revealed that greater than 99% of the Bithlo community's private household supplies would exceed the EPA's drinking water secondary standard.

Key words | community-level analysis, drinking water quality, groundwater, iron, medical geography, spatial analysis

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INTRODUCTION

Iron comprises approximately 3.5 to 5% of the Earth's crust, making it one of the most plentiful elements (McMurry & Fay 2003; Lemanceau *et al.* 2009). According to Miller (1997), some forms of iron are present in relatively high concentrations in sandy, organic mucky soils, and also in marine-derived sedimentary layers. Much of peninsular Florida comprises sandy soils and is underlain by marine-derived sedimentary layers (Scott 1993; Miller 1997; Raiswell & Canfield 1998; Hines 2009).

Iron is most commonly found in two oxidative states: ferrous or Fe^{2+} (reduced and water soluble) and ferric or Fe^{3+} (oxidized and much less water soluble). Iron-reducing microbes reduce ferric iron to the ferrous form when water saturates soil and underlying geologic strata, or when oxygen is depleted in other ways (Chapelle 2000). Furthermore, as rainwater percolates through soil and underlying sediments, it dissolves and transports iron into the groundwater supply (Katz *et al.* 2007). Iron solubility is greatly influenced by the biotic, soil, and substrate characteristics of an area (Valcarce & Townsend 2011). Thus, iron concentrations in groundwater vary greatly depending on pH and alkalinity, typically with levels ranging from 0.05 mg/L to 10 mg/L, or even up to 50 mg/L (WHO 1996).

The US Environmental Protection Agency (EPA) has established National Secondary Drinking Water Regulations that set non-mandatory water-quality standards for several contaminants, and were established as guidelines to assist public water systems in managing their drinking

water for aesthetic considerations, such as taste, color, and odor (EPA 2017). Although the EPA does not enforce these secondary maximum contaminant levels (SMCLs), the state of Florida enforces SMCLs per Florida Administrative Code (F.A.C. Chapter 62-550.320). Iron is generally believed not to present a health concern in drinking water, but when iron concentrations reach the EPA secondary threshold of 0.3 mg/L, humans can detect a 'metallic' taste.

There are many alternatives available that can be employed to solve an iron problem in groundwater required for potabilization. Treatment options may include the use of oxidizing filters, cation exchange systems, or water softeners, and depend on water quality and cost. Although the use of a sequesterant chemical (such as polyphosphates) has been employed to treat for iron, this method does not remove iron from the water. Reverse osmosis and pressure aeration/filtration are also recognized as applicable methods to treat for iron; however, these processes are expensive and require continual monitoring, and maintenance). In lieu of treatment, options could include the construction of a new well, eliminating the need for treatment, or (depending on local conditions) the extension of an existing well casing deeper into the aquifer to obtain a better quality water. However, these methods could prove costly for individual residences.

In addition to the staining of fabrics and plumbing fixtures, the staining of teeth by iron has been reported. Stangel *et al.* (1996) reported on the absorption of iron by dentin and its role in discoloration of teeth, indicating that the presence of sulfide can impact the rate of discoloration; in addition, Pushpanjali *et al.* (2004) showed a positive correlation between elevated iron in water and enamel stains in Nepal. It is noted that groundwater in central Florida is known to contain sulfide (He *et al.* 2013). Although a study by Rebelo de Sousa *et al.* (2012) found no association between extrinsic stains on enamel and the level of iron in water, it is noted that the study was based on drinking water where the iron content was less than 0.8 mg/L.

Iron is an essential element in human nutrition, as the human body requires approximately 1 to 3 milligrams of iron per day (mg/day), with an average intake of iron approximated at 16 mg/day being provided from food such as green leafy vegetables, red meat, and iron-fortified cereals. Although ferrous and ferric iron are absorbable (Andrews

1999), humans appear to lack a specific iron-excretion pathway, and excess dietary iron can accumulate in tissues, which is associated with a number of chronic health pathologies. Centers for Disease Control and Prevention (CDC) data indicate that chronic iron overload occurs in 1 to 6 people per 100 in the United States (CDC 2014). The average lethal dose of iron is 200 to 250 mg/kg of body weight, although death has occurred following the ingestion of doses as low as 40 mg/kg of body weight (National Research Council 1979). Symptoms of chronic iron overload include fatigue, arthralgia, liver fibrosis and cirrhosis, cardiomyopathy, and an increased risk of liver cancer, among other effects (Andrews 1999; Papanikolaou & Pantopoulos 2005; Shandler & Sazama 2010). According to Shandler & Sazama (2010), accumulating excess iron can result from excessive ingestion of dietary iron and/or supplements, chronic liver disease, chronic transfusion therapy, some hereditary disorders, and drinking water sources. Additionally, up to 5% of the US population exhibits hereditary hemochromatosis, i.e. the genetic inability to restrict iron absorption (Andrews 1999; Papanikolaou & Pantopoulos 2005). This information indicates that a fraction of the population is at risk of chronic iron overload.

Anecdotal evidence of metallic taste and staining issues suggested that iron was a constituent of drinking water in Bithlo, a central-Florida community 24.5 km east of downtown Orlando (Roe 2012). Bithlo relies solely on private water wells constructed at depths no greater than 25 or 30 meters for their household water supply. Many of these shallow wells throughout central Florida also have been found to contain appreciable (0.2 to 2.45 mg/L) sulfide content (Sprinkle 1989). In response to public reports of drinking-water quality, town hall meetings were convened to both inform and hear from Bithlo residents regarding their drinking water concerns (Roe 2012). From these meetings, a number of residents volunteered to have their drinking water tested; each participant would receive a written report of their results as incentive to participate (Roe 2012).

This work aims to be one of the first community-wide spatial scale analyses of groundwater focused on iron concentrations in household private-well supplies in Florida. A major goal of the research was to use a geo-referenced database to conduct community-wide statistical and spatial iron-concentration analyses regarding Bithlo, FL.

METHODS

Study area

Bithlo (28° 33′17″ N; 81° 06′22″ W) is located in eastern Orange County of central Florida, USA (Figure 1). According to the US Census Bureau (US Census 2012), Bithlo is a Census Designated Place of 27.7 km². In 2010, 8,268 people lived in 2,411 households (US Census 2012). Bithlo has existed since the early 20th century as an incorporated town, but in 1929 it ceased to function due to economic hardship. The city was finally unincorporated in 1977, an act which was not finalized until 1982 due to outstanding bonds and legal challenges (Schreyer & Turner 1998). For the years 2007–2011, 21.3% of Bithlo residents were living below poverty level as compared to 14.7% for the entire state of Florida (US Census 2012). Even though Bithlo is near Orlando, Florida, it has never had a public water

supply. Consequently, residences obtain their drinking water from private shallow wells and household wastewater effluent flows into septic systems.

The geology across most of east-central Florida is a thick sequence of sedimentary rocks of Ocala and dolomitic limestone (O'Reilly *et al.* 2002). Water-bearing sediments in this area are primarily limestone, dolomite, shell, clay, and sand. There are three aquifers underlying Bithlo (O'Reilly *et al.* 2002; Adamski & German 2004; USGS 2013). The surface aquifer is a non-artesian system comprising quaternary undifferentiated sands, clayey sands, clays, marls and peats beginning just under the soil and averaging 5 to 20 meters thick and is confined below by the intermediate confining unit, itself ~30 m thick (O'Reilly *et al.* 2002; Adamski & German 2004). Below this formation is the Upper Floridan Aquifer system ~90 to 200 m below surface, with the deepest aquifer being the Lower Floridan Aquifer, ~300 to 700 m deep (O'Reilly *et al.* 2002; Adamski & German 2004; USGS 2013). Additionally, Bithlo is in the Econlockhatchee River basin, which is itself part of a federally monitored water district for the St. John's River (O'Reilly *et al.* 2002).
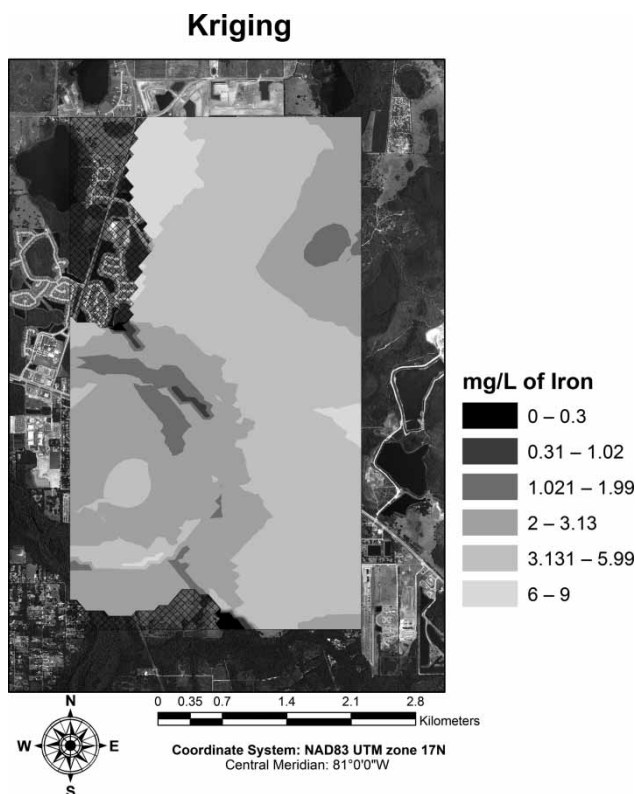
Water quality and study design

The Orange County Environmental Protection Division collected 202 water samples from 196 residential locations between January and June 2012 in Bithlo, Florida (Roe 2012). The aqueous samples were subsequently delivered to the Orange County Utilities (OCU) Central Laboratory for chemical analysis using EPA methods 200.8 and 200.9 for iron. The results of these water-quality tests were individually mailed to participants. Publicly funded agencies collected and analyzed the water samples, thus the water quality reports are available to the public (Florida Statutes 2010). In this work, a convenience historical (samples collected in 2012), cross-sectional (each water supply was visited once across the study area) design was relied on to assess overall statistical and spatial iron-concentration trends.

Whereas household water-quality results were in separate files, the individual records acquired were combined into a single geo-referenced database. This collection of water-quality reports formed the basis of an analytical, community-wide, and cross-sectional spatial layer for statistical

Kriging

mg/L of Iron

- 0 – 0.3
- 0.31 – 1.02
- 1.021 – 1.99
- 2 – 3.13
- 3.131 – 5.99
- 6 – 9

0   0.35 0.7   1.4   2.1   2.8
Kilometers

Coordinate System: NAD83 UTM zone 17N
Central Meridian: 81°0'0"W

Figure 1 | Map showing location of the study area in central Florida. The geographic center of the study area is 28° 34′ N and 81° 06′ 11″ W and is approximately 25 km east of downtown Orlando, Florida. All water samples were collected from within the outlined area in the figure.

and spatial analyses. It is noted that complaints or reports of metallic taste and staining issues do not necessarily indicate a systemic iron problem throughout Bithlo, as these issues could also be the result of corroded well housings due to age and environmental conditions.

Treated vs untreated water samples

Treated refers to collected water samples that had first passed through some type of filtering or purification system, which residents had in their homes. Untreated refers to collected water samples that had not passed through some type of filtering or purification system in the household where data had been collected and relied on in this work.

Geodatabase and geocoding

A single geo-referenced database was created using individual water-quality reports acquired in digital form from Orange County EPD and transcribed into a single database. This database included iron-concentration records from both untreated and treated samples in conjunction with the household address associated with each sample. Then, each location from which iron was tested was geocoded by address (ArcGIS 10.1 geocode tool, centroids along roads); specific points are not displayed for security and privacy purposes. These geocoded locations served as proxies for well locations. It was estimated that these proxy locations averaged less than 60 m from the wellheads, and this was reasonable because the average distance between geocoded locations was over 120 m.

Statistical analyses and surface interpolation

To assess community-wide patterns of groundwater, iron concentration averages, medians, standard deviations, 95% confidence intervals and ranges for treated, untreated and combined treated and untreated samples were determined. Furthermore, spatial autocorrelation analysis (Incremental autocorrelation) on the untreated sample concentrations and geographic coordinates were also conducted.

One of the analytical benefits of having a geocoded, statistically based dataset is that geographic point data can be interpolated into surfaces (Butler 2003; Jones 1997). Surface interpolation from point data generalize information from specific locations into broader spatial trends (Jones 1997).

Ordinary kriging is the surface-modeling technique to apply when the study area exhibits a high degree of geological and topographical isotropy (Isaaks & Srivastava 1989; Goovaerts 1997). Bithlo is flat; topography varies by only a few meters, and the underlying surficial geology extends throughout much of central Florida. Due to this we assume very minimal to no spatial bias or directionality of iron concentrations in the groundwater.

In this work, a surface was created using ordinary kriging from the untreated point locations. First, untreated water samples ($N = 79$) reasonably represent groundwater iron levels since the supply was being withdrawn from the shallow alluvial aquifer, often through metallic-cased wells. Second, each geocoded location is coupled to a geographic coordinate system, and because each point location contains a continuous variable (iron concentration) connected to it, these coordinates and attribute values are well suited for spatial interpolation via kriging (Goovaerts 1997).

An inherent condition in spatial analyses is that local means can vary significantly across a study area (Goovaerts 1997). The amount of iron detected at different well sites across our study area varied by almost four orders of magnitude. Thus, to better capture and model this variation ArcGIS provides cross-validation statistics and user-defined parameters. In this way, the user can analytically compare different interpolation results to produce a best-fit interpolated surface. Our best-fit, ordinary kriging interpolation model used nearest-neighbor points (number of untreated wells) to be no less than 20 and no more than 40 because of the wide range of iron-concentration values, over three orders of magnitude, found across the study area; a nearest-neighbor geometry of a circle with four sectors of 45° each, major/minor semiaxes of 952 m, 12 lags of 97 m (half the average nearest-neighbors distance, ArcGIS 10.1), anisotropy setting of 'no' and the 'Stable' model parameters (ArcGIS 10.1). Furthermore, we employed incremental autocorrelation analysis, 25 m increments, to assess spatial autocorrelation of the untreated, well water locations.

RESULTS

The overall iron concentrations ranged from below detectable limits (<0.009 mg/L) found for 12 treated samples to one sample measured at 36 mg/L. The highest value detected was almost twice as large as the next highest (19 mg/L). To assess whether this result was an outlier we performed Grubbs' test for outliers (Grubbs 1969). The calculated Grubbs' statistic for the 36 mg/L value was 6.12 ($P < 0.01$), indicating strong support for outlier status; consequently, this value was removed when calculating statistics and creating the surface interpolations. Nevertheless, there is no indication that this high iron-concentration value is artefactual, but represents an actual, extreme, iron concentration in a Bithlo groundwater supply for household use.

In general, untreated water contained the highest overall iron concentrations, treated water the lowest; and when these two sets of samples were combined, the results exhibit intermediate values between the other two (Table 1). All averages ranged well above the EPA secondary standard (Table 1). All median values were less than their mean counterparts (Table 1). However, both the treated and combined median values were less than half their corresponding mean value while the untreated samples' median was approximately 35% lower (Table 1). There was almost a three-fold greater likelihood of an untreated sample having an iron concentration greater than the EPA secondary standard as compared to treated samples and, overall, there was almost a 60% chance of any sample having a higher iron concentration than the EPA recommendation (Table 1).

The results of the statistical analyses also suggest several distinct patterns. First, the greatest iron concentrations were found in the untreated water samples, which averaged over 11 times greater than the EPA secondary standard (Table 1). Most (91%) contained an iron concentration greater than 0.3 mg/L. This result is reflected in that by the 8th percentile of ranked, untreated samples, iron concentrations started to exceed 0.3 mg/L (Table 1). Following this trend of a large proportion of untreated samples exceeding 0.3 mg/L with many of the samples containing iron concentrations many times higher than the EPA secondary standards, 50% of the untreated samples exceeded 2.39 mg/L or eight times greater than the EPA secondary standard. Second, the average iron concentration detected in treated water was 73% lower as compared to the untreated samples' average (Table 1). Nevertheless, almost 40% of treated samples exceeded the EPA secondary standard (Table 1). Third, the range of iron concentrations detected in the untreated samples varied by three orders of magnitude (Table 1).

The results of the incremental autocorrelation analysis indicate that untreated well-water iron concentrations were not spatially autocorrelated as no 25 m increment P-value result was less than 0.142 (20 bands, starting at 150 m, ArcGIS 10.1). We started at 150 m because the average distance between wells was over 120 m. The best-fit modeling resulted in a relatively broad 'neighborhood' of nearest neighbors. This is because of (1) the relatively large number of nearest neighbors needed to capture the great variation of iron concentrations found across the study area and (2) the geocoded well locations were not evenly distributed.

The cross-validation results of the best-fit kriged surface were determined to be a mean prediction error of -0.0002, a standardized root mean square of 1.008, and an average standard error of 3.84 (ArcGIS 10.1 cross-validation comparison). These results indicate a good fit. The results of kriging in ArcGIS produce a geo-referenced, probability surface map. The surface is displayed as a series of isopleths of different attribute values ranging across a study area. Each contour represents a statistically significant probability that the attribute of interest would be expected to occur with the calculated (from the point data) magnitude.

Table 1 │ Descriptive statistics of iron concentrations recorded in Bithlo

	Untreated	Treated	Total
N	79	123	202
Mean	3.45 mg/L	0.92 mg/L	1.91 mg/L
95% CI	±0.84	±0.3	±0.41
Median	2.4 mg/L	0.14 mg/L	0.58 mg/L
St. dev.	3.83	1.68	2.99
Range	0.028–19 mg/L	<0.009–7.7 mg/L	<0.009–19 mg/L
Percentile ≥0.3 mg/L[a]	8th	64th	43rd
Probability of ≥0.3 mg/L	0.91	0.37	0.58

[a]EPA secondary level for iron.

In this work, ordinary kriging results reveal that the range of iron concentrations across the study area geostatistically clustered into six, iron-concentration categories (Figure 2). The study area encompassed approximately 1.2×10^7 m^2, of which less than 0.1% (5,000 m^2) was predicted to have well-water iron concentrations that could be less than 0.3 mg/L. This result was confined to two small areas: a tiny cluster on the mid-western edge of the study area and a narrow strip just south-east of the preceding cluster (Figure 2). Therefore, the surface-analysis results indicate it is very unlikely that untreated well water from any site in our study area would contain iron concentrations less than the EPA secondary standard. Highest iron-concentrations in the study area were located in the north-western corner and west-central portions, with smaller patches distributed south and west (Figure 2). Not only was it found that there is a large number of household untreated water supplies containing high iron, as revealed by the statistical analyses, but also that these locations indicate that high iron

concentrations are expected to occur throughout the study area. Furthermore, the resulting lack of spatial autocorrelation in conjunction with the resulting broad-scale patterns of high iron concentrations across the study area indicate that geocoding to centroids along streets would not be a factor affecting results. This suggests that Bithlo community households reliant on shallow wells will continue to be exposed to levels that exceed the EPA's SCML, especially those without treatment.

DISCUSSION

There do not appear to be any systematic geospatial assessments correlating iron concentrations in shallow alluvial drinking water supplies within a rural water community. Our analyses and results indicate that Bithlo, Florida exhibits community-wide high iron concentrations in household drinking water supplies. The results of the statistical analysis indicated the propensity for untreated, household water supplies to contain relatively high iron concentrations. However, more information was extracted by utilizing spatial statistics. Kriging, like other spatial statistical techniques, assesses more than an attribute's magnitude, but also includes length, location, proximity, direction, area and/or orientation in the calculations. In doing so, spatial statistics explain more than just magnitude of a phenomenon, but also place and space of a phenomenon.

The surface analysis indicates that broad swaths of Bithlo are predicted to have high iron in household drinking water drawn from the shallow, alluvial aquifer underlying this community; in many cases five- to ten-fold higher than the EPA's SMCLs. This study strongly suggests that Bithlo citizens' complaints regarding taste and staining of their home drinking water is not likely to be a function of a few, unsettled residents. Rather, the negative aesthetic characteristics of staining and taste associated with household water high iron concentrations would likely be found throughout the community. Some of the drinking water issues could be a result of corroding well casings and household plumbing. However, the fact that virtually all untreated water samples exhibit iron concentrations higher than EPA SCMLs, and that these samples were distributed across the study area, indicates that a more general condition, like
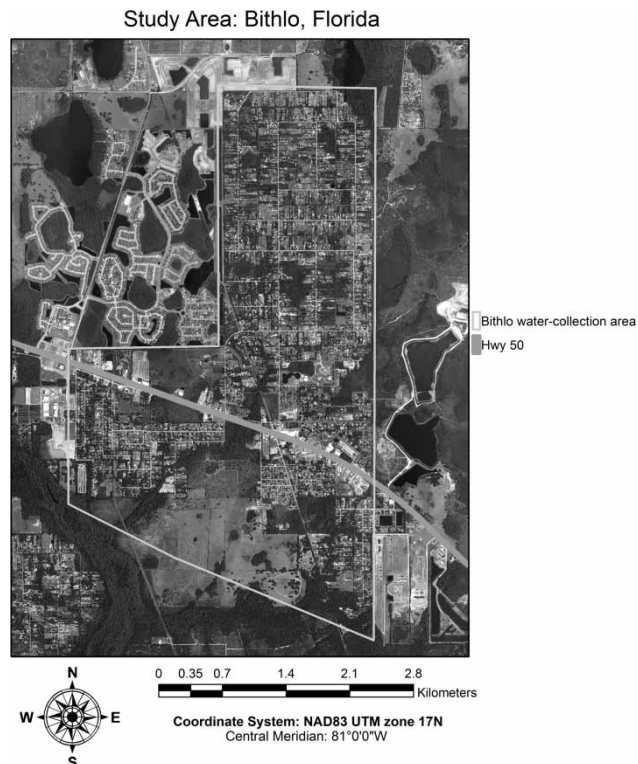

Figure 2 | Kriged surface result. We used geometric classification (ArcGIS 10.1) process for the legend and display. This classification system divides classes into similar statistical and frequency categories based on geometric means of the classes.

the underlying hydrogeology, would more likely be the cause for such a community-wide effect.

Chronic iron overload occurs in up to 6% of the US population (CDC 2014). A contributing factor to this percentage could be high iron fractions in groundwater sources used for human consumption (Shandler & Sazama 2010). Humans do not have a specific iron-excretion pathway and research suggests that too much ingested iron over a long period of time could have repercussions for human health (Andrews 1999; Papanikolaou & Pantopoulos 2005; Shandler & Sazama 2010). The underlying geology and hydrology of the study area extends throughout a large portion of peninsular Florida (Scott 1993; Raiswell & Canfield 1998; Hines 2009). Thus, households reliant on private wells drawing from surficial alluvial groundwater supplies in other parts of peninsular Florida could also be at risk of elevated iron concentrations in their drinking water.

It is assumed that wells in the study area penetrate into the surficial, non-artesian alluvial aquifer, which is not deeper than 30 m (O'Reilly *et al.* 2002; Adamski & German 2004). Anecdotal evidence suggests that higher iron fractions could be expected in even shallower wells. According to a report via a local newspaper (Stokes 2012), the 36 mg/L iron sample came from a well only 3 m deep. For comparison, a second well was drilled at the site to 20 m depth and the recorded iron concentration was 6.1 mg/L. Nevertheless, iron concentrations could be related to well depth. Thus, ascertaining well depth could help refine future water quality analyses of the area.

Bithlo is underlain by hydrogeological features that extend throughout central Florida, so the local geology of the study area is part of larger geological trends. Too much iron could be a human health concern (Papanikolaou & Pantopoulos 2005). Thus, not only could residents of the Bithlo area be at risk of chronic iron overload, so could other communities in central Florida. Communities typically avoid a risk posed by chronic iron overload as a result of poor hydrogeologic conditions by employing centralized treatment plants that provide disinfected water to residences and businesses through a series of distribution lines. It would stand to reason that the residents living within the Bithlo community would benefit from a centralized approach to water treatment to reduce the risk of iron overload to its consuming population. An extension of a pipeline

two miles from the closest water purveyor, OCU, could offer a permanent solution to Bithlo's iron exposure. However, a preliminary opinion of probable construction cost to extend OCU's closest drinking water supply main and associated residential supply lines and meters to Bithlo residences ranges between $5.25 million and $6.25 million (University of Central Florida 2013). Under current conditions, it remains unlikely that the Bithlo community could afford to tie into the nearest municipal water supply to resolve exposure to elevated iron in drinking water at this time.

CONCLUSION

Individual households located within Bithlo, Florida report that untreated drinking water contains iron concentrations that often greatly exceed the EPA secondary standard of 0.3 mg/L. The amount of iron in untreated samples averaged an order of magnitude greater than the EPA's secondary standard. A third (38.6%) of untreated samples contained iron concentrations greater than the human consumption health-based threshold set by the CDC (4.2 mg/L). Even treated water averaged approximately three times (0.92 mg/L) higher than the EPA secondary standard for iron.

This work is the result of collaboration between different universities, local governments, health institutions, and advocacy groups. We recognize the wealth of research that has been successfully conducted by such interorganizational partnerships (Varda *et al.* 2012) and suggest the results of this work buttress interorganizational collaboration as an effective means to assessing, analyzing, and evaluating human health-related drinking-water issues.

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