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Arsenic exposure to drinking water in the Mekong Delta

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HIGHLIGHTS

- Elevated As is found in groundwater used for drinking in the Mekong Delta, Vietnam.
- Arsenic in nails reflects exposure of individuals consuming As-rich groundwater.
- Differential As exposure is observed by the As in nail to As in water ratios.
- Diet and water filtration reduce individual's exposure to As in drinking water.

GRAPHICAL ABSTRACT

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ABSTRACT

Arsenic (As) contamination of groundwater drinking sources was investigated in the Mekong Delta, Vietnam in order to assess the occurrence of As in the groundwater, and the magnitude of As exposure of local residents through measurements of As in toenails of residents consuming groundwater as their major drinking water source. Groundwater ($n = 68$) and toenail ($n = 62$) samples were collected in Dong Thap Province, adjacent to the Mekong River, in southern Vietnam. Fifty-three percent ($n = 36$) of the wells tested had As content above the World Health Organization's (WHO) recommended limit of 10 ppb. Samples were divided into Northern (mean As = 4.0 ppb) and Southern (329.0 ppb) groups; wells from the Southern group were located closer to the Mekong River. Elevated As contents were associated with depth (<200 m), salinity (low salinity), and redox state (reducing conditions) of the study groundwater. In 79% of the wells, As was primarily composed of the reduced As(III) species. Arsenic content in nails collected from local residents was significantly correlated to As in drinking water ($r = 0.49$, $p < 0.001$), and the relationship improved for pairs in which As in drinking water was higher than 1 ppb ($r = 0.56$, $p < 0.001$). Survey data show that the ratio of As in nail to As in water varied among residents, reflecting differential As bioaccumulation in specific exposed sub-populations. The data show that water filtration and diet, particularly increased consumption of animal protein and dairy, and reduced consumption of seafood, were associated with lower ratios of As in nail to As in water and thus could play important roles in mitigating As exposure in areas where As-rich groundwater is the primary drinking water source.

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1. Introduction

The Mekong Delta is a biological diverse and water-rich area, and together with the Red River in northern Vietnam, comprises one of the most productive agricultural regions in Southeast Asia (Berg et al., 2001, 2007). Projected hydropower generation and dam construction on the upstream Mekong River pose risks for water availability in the downstream Mekong Delta. Consequently, groundwater is expected to become a pivotal irrigation and drinking water resource in this region. It is estimated that the demand for groundwater will increase by up to 6.5 times by 2020 (510–520 × 10^9 m^3/year) in comparison to the 1990–2000 period (Van, 2004). In addition to water availability, water quality can limit the sustainability of groundwater in the Mekong Delta, and its potential to substitute for declining surface water resources. In particular, previous studies have highlighted the occurrence of high salinity and As (arsenic) in groundwater that could limit agriculture production and pose human health risks (Buschmann and Berg, 2009; Buschmann et al., 2007, 2008). Understanding the geochemical conditions in which contaminants are mobilized to groundwater and the magnitude of exposure of the local residents to these contaminants is important for evaluating the risks of the expected transition from surface water to groundwater utilization in the Mekong Delta.

Arsenic in Vietnam has been measured in groundwater in both the Red River and the Mekong River Deltas (Berg et al., 2001, 2007, 2008; Buschmann et al., 2007, 2008; Nguyen and Itoi, 2009; Winkel et al., 2011). Elevated As (>10 ppb) levels have been reported to typically occur in shallow groundwater from both the Holocene and Pleistocene aquifers. In the Red River Delta, Winkel et al. (2011) have shown that over-pumping of As-free deep groundwater has resulted in the drawdown of the groundwater levels in the deep aquifers. This has induced the downflow of shallow As-rich groundwater to the deep aquifers and caused contamination of deep groundwater, which has been utilized mainly for drinking water.

Arsenic contamination in groundwater from the Mekong Delta is naturally occurring and caused by chemical and microbial induced reductive dissolution of iron-oxides from the alluvial sediments in the delta (Rowland et al., 2008; Quicksall et al., 2008; Fendorf et al., 2010; Winkel et al., 2011). Following river sediment transport and accumulation in the deltaic reducing conditions, As bound to iron-oxides is released (Berg et al., 2001, 2008; Bissen and Frimmel, 2003; Harvey et al., 2002; Nguyen and Itoi, 2009; Nickson et al., 1998; Polizzotto et al., 2005). The World Health Organization (WHO) recommends As concentrations of up to 10 ppb in drinking water (WHO, 2011), but As concentrations in groundwater over 1300 ppb have been reported in the region (Winkel et al., 2011; Stollenwerk et al., 2007; Buschmann et al., 2008). In addition, other inorganic groundwater contaminants with potential health effects, such as Mn and Ba, have been reported (Buschmann et al., 2007, 2008). Buschmann et al. (2008) reported that high As levels occur selectively in low-saline drinking water wells close to the Mekong River, while groundwater located farther away from the Mekong River is characterized by higher salinity and lower As content.

In spite of the extensive literature on the overall toxic effects of As, it is difficult to establish a direct link between health affects and As exposure from drinking water in a given population due to the long latency period between the window of exposure and the development of health outcomes. Keratin, such as in hair and nails, has been shown to be the preferred method to monitor long-term exposure to As in drinking water. While blood and urine are useful biomarkers for smaller exposure windows, the nails reflect an integrated exposure time ranging from 3 months to a year (Schroeder and Balassa, 1966; Slotnick and Nriagu, 2006; Yoshida et al., 2004). Toenails are thought to be better than fingernails at capturing As exposure due to the fact that their slower growth rate provides greater As levels per mass compared to fingernails. Both are thought to be better than hair because an individual's hair growth rates vary more across populations relative to individual's nail growth rates (Karagas et al., 1996, 2000; Slotnick and Nriagu, 2006).

Although elevated As in drinking water sources of the Mekong and Red River Deltas in Vietnam have been identified as a major health concern, no exposure study through the monitoring of As in nails has been conducted in the Mekong Delta Region. Nguyen et al. (2009) found a correlation between As concentrations in groundwater drinking wells from the Red River Delta and As concentrations in women's hair, while Berg et al. (2007) found a correlation between As concentrations in drinking water and As concentrations in hair in the Mekong Delta (in both Vietnam and Cambodia), and also in the Red River Delta. This paper aims to fill the literature gap, focusing on As occurrence and human exposure in the Mekong Delta by measuring As concentrations in drinking water and in nails of local residents that consume groundwater as their major drinking water source.

The objectives of this study are (1) to evaluate As occurrence in the Mekong Delta groundwater; and (2) to assess its bioaccumulation in populations exposed to As in their drinking water. We collected samples measuring As in groundwater and nails from the Dong Thap Province in southern Vietnam and compared the As data to previous studies. By understanding the extent of As distribution in groundwater and its accumulation in the local populations in the Mekong Delta this paper provides the foundation for evaluating the health risks associated with the increased utilization of groundwater, which will likely result from the projected reduction of the Mekong River flow.

2. Methods

2.1. Field sampling

IRB approval was obtained from Duke University and Ho Chi Minh Science University and the Department of Natural Resource and Environment of Dong Thap Province. The study site spans approximately 70 km north to south in the Dong Thap province of Vietnam. Wells were selected based on government approval, as well as consent from individual well owners. In total, 68 groundwater wells were tested as well as 5 surface water samples from the Mekong River.

2.2. Groundwater sampling and analytic techniques

Groundwater from private and monitoring wells, as well as surface waters from the Mekong River were collected following USGS protocols (USGS, 2011). Samples to be analyzed for trace metals were filtered at the site location using 0.45 μm syringe filters, preserved using nitric acid, and then shipped to Duke University for analysis. The samples were analyzed for major elements using an ARL SpectraSpan 7 (Thermo Fisher Scientific, Inc.) direct current plasma optical emission spectrometry (DCP-OES), anions by an ICS 2100 (Dionex) ion chromatography (IC), and trace metals by a VG PlasmaQuad-3 (Thermo Fisher Scientific, Inc.) inductively coupled plasma mass spectrometry (ICP-MS) at Duke University. While more elements were analyzed, for this paper we report only As (detection limit = 0.05 ppb) and Cl (detection limit = 0.2 ppm). Speciation of As was performed in the field and preserved according to methods in Bednar et al. (2002) by using EDTA and anion exchange to isolate the uncharged As(III) species. Parameters collected in the field include pH, temperature, and oxidation–reduction potential (ORP) (YSI pH100A pH/ORP), conductivity (EC) (YSI EC300A), and dissolved oxygen (DO) (YSI DO200A). Meter calibration was performed prior to sampling. More detailed field and laboratory analytical methods can be found in Ruhl et al. (2010).

2.3. Nail sampling and analytic techniques

Toenails were collected from individuals whose water had been sampled. Researchers approached participants, explained the study, and obtained consent. The individuals were then surveyed to collect basic demographic information as well as water consumption patterns and basic health issues. Toenails were then clipped using new clean