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Assessing mixed trace elements in groundwater and their health risk of residents living in the Mekong River basin of Cambodia

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ABSTRACT

We investigated the potential contamination of trace elements in shallow Cambodian groundwater. Groundwater and hair samples were collected from three provinces in the Mekong River basin of Cambodia and analyzed by ICP-MS. Groundwater from Kandal ($n = 46$) and Kraite ($n = 12$) were enriched in As, Mn, Ba and Fe whereas none of tube wells in Kampong Cham ($n = 18$) had trace elements higher than Cambodian permissible limits. Risk computations indicated that 98.7% and 12.4% of residents in the study areas of Kandal ($n = 297$) and Kratie ($n = 89$) were at risk of non-carcinogenic effects from exposure to multiple elements, yet none were at risk in Kampong Cham ($n = 184$). Arsenic contributed 99.5%, 60.3% and 84.2% of the aggregate risk in Kandal, Kratie and Kampong Cham, respectively. Sustainable and appropriate treatment technologies must therefore be implemented in order for Cambodian groundwater to be used as potable water.

1. Introduction

Groundwater contamination by trace elements has created substantial concern among environmental health scientists. Although some trace elements are essential for normal growth of humans and animals, high intake and low intake of essential trace elements could lead to toxicity and nutritional deficiency, respectively (Goldhaber, 2003). For instance, trace elements in the drinking water (Muhammad et al., 2011; Kavcar et al., 2009) and food grains grown in contaminated soils (Huang et al., 2008) could pose significant non-carcinogenic effects to their consumers. In general, trace elements in groundwater could be derived from either natural or anthropogenic sources (Ramesh et al., 1995; Chen et al., 2007; Mondal et al., 2010; Banerjee et al., 2012). Ecological communities and living organisms in receiving water are also affected by direct discharge of effluents from various industries into aquatic systems (Krishna et al., 2009). Some of these trace elements in groundwater have been well documented in Vietnam (Agusa et al., 2006), Lao PDR (Chanpiwat et al., 2011), India (Ramesh et al., 1995) and China (Chen et al., 2007). The outcome of acute arsenic toxicity might include gastrointestinal discomfort, abdominal pain, vomiting, diarrhea, bloody urine, shock, coma and death (Hughes, 2002). Chronic exposure to arsenic through groundwater consumption may cause skin lesions (pigmentation, melanosis and keratosis) and development of hard patches of skin on palm of the hand and sole of the feet. Skin cancer, cancer of the bladder, kidney and lung as well as diabetes, high blood pressure and reproductive disorders have also been associated with chronic arsenic exposure (WHO, 2008; ATSDR, 2007a). Humans and animals can acquire Mn, an essential element for metabolism, through many food sources (WHO, 2008). However, an excess or deficiency of Mn might cause adverse health effects. Mn toxicity has been known to occur in particular occupational settings through

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inhalation of Mn-bearing dust and/or fumes (Laohaudomchok et al., 2011). Among all target tissues, the brain was most susceptible to excess Mn; high accumulation of Mn may cause neurodegenerative disorders and neurotoxicity (Wright et al., 2006; Laohaudomchok et al., 2011; ATSDR, 2008). Clinical symptoms of Mn toxicity include movement disorders, psychiatric disturbance, cognitive deficits such as memory loss/impairment, reduced learning capacity, decreased mental flexibility and cognitive slowing and behavioral and/or mood changes (Wright et al., 2006; Laohaudomchok et al., 2011). Neurological disorders resulting from drinking Mn-rich water have been reported in epidemiology studies (WHO, 2008). However, the World Health Organization has recently discontinued its drinking water guideline value of 400 g L^{-1} for Mn because this health-based value was well above Mn concentration normally found in drinking water (WHO, 2011). Although drinking Ba-contaminated water might lead to hypertension, Ba has not shown any evidence of carcinogenic or mutagenic effects (WHO, 2008; ATSDR, 2007b).

A general population census in 2008 revealed that 80% percent of Cambodians live in rural areas (NISC, 2012) and in 2010, 28.5% live under the national poverty—an estimate derived by the Cambodian Government (WFP, 2012). About 16.1% of Cambodian women suffered from malnutrition in 2008, and 29% of Cambodian children were underweight in 2010–2011 (WHO, 2012). It was estimated that 81% and 56% of populations in urban and rural areas had access to improved drinking water sources, which was equivalent to about 61% of the total populations (UNICEF, 2012). Although many populations lived alongside surface waters in Cambodia, shallow groundwater was the main source for drinking water (Phan et al., 2010). Unfortunately, shallow Cambodian groundwater is naturally enriched by arsenic and other trace elements (Polya et al., 2005; Berg et al., 2007; Benner et al., 2008; Kocar et al., 2008; Sthiannopkao et al., 2008; Robinson et al., 2009; Luu et al., 2009; Kim et al., 2011). Arsenic was released from the near-surface, river derived sediments within the Mekong River delta and transported to the underlying aquifer by groundwater flow (Polizzotto et al., 2008). Microbially mediated reductive dissolution coupled with redox cycling in near-surface sediments might play an important role in releasing trace elements into pore water in the Mekong River basin of Cambodia (Phan et al., 2010). Health risk assessment of inorganic arsenic intake through ingestion of contaminated has been investigated (Phan et al., 2010). Furthermore, adverse health impacts from chronic arsenic exposure have also been observed (Sampson et al., 2008; Phan et al., 2011). To date, however, studies on aggregate risk through consumption of mixed trace element intake and the resulting human health impacts have not been conducted. Therefore, the objectives of this study were to (1) investigate the distribution of trace elements in shallow groundwater in the Mekong River basin of Cambodia; (2) assess non-carcinogenic effects from both single and mixed trace elements through groundwater ingestion; (3) determine and compare the concentrations of trace elements in hair samples from local populations; and (4) correlate As, Ba and Mn in hair and groundwater samples as well as their respective average daily dose.

2. Materials and methods

2.1. Study area

The Mekong River originates in Tibet and flows 4600 km across six countries including China, Myanmar, Thailand, Lao PDR, Cambodia and Vietnam before discharging into the South China Sea. The area of the Mekong River basin is about 800,000 km^2 with 470 km^3 of annual water and 160 million tons of sediment deposition (Tamura et al., 2007; MRC, 2010). The furthest upstream reaches of the Mekong delta is defined close to the capital city of Phnom Penh; the delta expands downstream of Phnom Penh and covers an area of 62,520 km^2 with two major distributary channels, including the Mekong and Bassac Rivers (MRC, 2010). The present study was conducted in three provinces in the Mekong River basin of Cambodia. Kratie (Preak Samrong I and II villages, Khsarch Andaet commune, Chhloung district) and Kampong Cham (Andoung Chros and Veal Sbov villages, Ampil commune, Kampong Siem district) were located along the Mekong River upstream of Phnom Penh, whereas Kandal (Preak Russey village, Kampong Kong commune, Koh Thom district) was located between the Mekong and the Bassac Rivers, downstream of Phnom Penh.

2.2. Field work

Our field work was conducted after our research proposal was approved by the National Ethics Committee for Health Research (Reference No. 131NECHR, 12/12/2008) under the Cambodian Ministry of Health and informed consent was obtained. Groundwater samples were randomly collected from the study areas of Kandal ($n = 46$) and Kampong Cham ($n = 18$) in February 2009 and Kraite ($n = 12$) in August 2009. Sampling was conducted based upon the accessibilities to tube wells, the willingness of respondents to provide hair samples and respondent claims of tube well use for an extended period of time. Each groundwater sample was collected from a tube well after 5–10 min of flushing in order to remove any standing water from the tube. Groundwater sample was filtered (0.45 μm) into a polyethylene bottle and acidified with concentrated nitric acid in order to prevent the precipitation of Fe, Mn and As and adsorption of other trace elements to the bottle surface during field storage. Simultaneously, on-site measurements of pH and redox potential were taken using HORIBA pH/Cond meter D-54. The collected samples were kept in an ice box and then transferred to a fridge where they were stored at 4 °C until delivery to GIST, Republic of Korea for analyses. Concurrently, hair samples were collected using stainless steel scissors from the nape of the head as near as possible to the scalp of several members of the volunteered families in the Kandal ($n = 270$), Kratie ($n = 84$) and Kampong Cham ($n = 173$) province study areas, who claimed to routinely use a tube well. Individual demographic information was also collected to calculate the average daily dose of each trace element to individuals in the study populations. Age, gender, ingestion rates and exposure duration were collected using a structured questionnaire. Body weight was measured using a bathroom scale which was calibrated to zero prior to each measurement. The collected hair samples were separately kept in a labeled plastic ziplock bags and stored in darkness until analyses.

2.3. Sample preparation and analyses

Groundwater samples from Kandal and Kraite were diluted (1:25) with 2% nitric acid (prepared by 18.2 MΩ *MilliQ* deionized water and 70% nitric acid) to measure the concentrations of As, Mn, Ba and Fe. Samples from Kampong Cham did not require dilution, because the concentrations of As, Mn, Ba and Fe fell into the standard calibration curves. Concurrently, Ag, Al, Cd, Co, Cr, Cu, Ga, Ni, Pb, Se, U and Zn were determined using the original samples. Hair was cut into small pieces (3 mm) and alternately washed with acetone and deionized water as described in our previous report (Phan et al., 2010). Washed hair was dried at 60 °C overnight before digestion. Acid-digestion was performed following our previously used method (Phan et al., 2010). Chemical measurements of all groundwater and digestate of hair samples were employed by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500ce) using an external calibration method. Calibration standard solutions (0.1 μg L^{-1}, 1 μg L^{-1}, 5 μg L^{-1}, 10 μg L^{-1}, 20 μg L^{-1}, 50 μg L^{-1} and 100 μg L^{-1}) were prepared from a stock solution (Multi element 2A), also with 2% nitric acid. Two percent nitric acid was also used as an analytical blank. Standard reference material (Trace Element in Water, SRM 1643e) was analyzed to check the ICP-MS accuracy. If the recovery rate become out of the recommended range (90–110%), samples were reanalyzed with a new calibration curve. Digestions of two replicated hair samples were conducted to verify a validity of acid digestion method. Three replicates of human hair standard reference materials (GBW07601) were also treated in the same manner as the sample to check the accuracy of the digestion methods. The recovery rates of Ag (108.74%), As (94.80%), B (82.90%), Ba (87.29%), Cd (108.55%), Cu (101.00%), Mn (93.91%), Mo (83.68%), Ni (77.98%), Pb (93.94%) were in good agreements with the certified values.

2.4. Health risk assessment

Health risk assessment procedures from the USEPA (1989) were applied to calculate non-carcinogenic effects of single and mixed trace elements. The average daily dose of single element is calculated from the following Equation (1).

$$\mathrm{ADD} = \frac{C_w \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \quad (1)$$

where ADD is the average daily dose from ingestion (mg kg^{-1} d^{-1}); C_w is the trace element concentration in groundwater (mg L^{-1}); IR is the ingestion rate of groundwater (L d^{-1}); EF is the exposure frequency (d y^{-1}); ED is the exposure duration (y); BW is the body weight (kg) and AT is the averaging time (d). Field surveys (Table 1) showed that the residents in the study areas of Kandal ($n = 297$) and Kratie ($n = 89$) have consumed groundwater 9 months per year (EF = 270 d y^{-1})