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Elevated childhood exposure to arsenic despite reduced drinking water concentrations — A longitudinal cohort study in rural Bangladesh

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ABSTRACT

Objectives: The aim of this study was to evaluate the massive efforts to lower water arsenic concentrations in Bangladesh.
Methods: In our large mother–child cohort in rural Matlab, we measured the arsenic concentrations (and other elements) in drinking water and evaluated the actual exposure (urinary arsenic), from early gestation to 10 years of age (n = 1017).
Results: Median drinking water arsenic decreased from 23 (2002–2003) to <2 μg/L (2013), and the fraction of wells exceeding the national standard (50 μg/L) decreased from 58 to 27%. Still, some children had higher water arsenic at 10 years than earlier. Installation of deeper wells (>50 m) explained much of the lower water arsenic concentrations, but increased the manganese concentrations. The highest manganese concentrations (~900 μg/L) appeared in 50–100 m wells. Low arsenic and manganese concentrations (17% of the children) occurred mainly in >100 m wells. The decrease in urinary arsenic concentrations over time was less apparent, from 82 to 58 μg/L, indicating remaining sources of exposure, probably through food (mean 133 μg/kg in rice).
Conclusion: Despite decreased water arsenic concentrations in rural Bangladesh, the children still have elevated exposure, largely from food. Considering the known risks of severe health effects in children, additional mitigation strategies are needed.

1. Introduction

Access to safe drinking water is immensely important for human health and development (Bain et al. 2014; WHO 2011). With the increasing use of ground water for drinking purpose and agricultural needs, naturally occurring toxicants such as arsenic and fluoride have become growing public health concerns (Hunter et al., 2010). Arsenic in rice is a more recently discovered threat to human health (Sohn, 2014). The concern arises from the fact that inorganic arsenic, the main form in both drinking water and rice, is a potent human carcinogen and multi-organ toxicant even at fairly low exposure levels (IARC, 2012; NRC, 2001). Indeed, the challenges are substantial globally. A relevant example is the situation in Bangladesh, where a major part of the millions of tube wells installed since the 1970s were subsequently reported to contain elevated arsenic concentrations (BGS, 2001; Edmunds et al., 2015). Extensive remediation efforts have been undertaken with varying results (Edmunds et al., 2015; Hossain et al., 2014; Ravenscroft et al., 2013). A nation-wide screening of water arsenic in 2009 showed considerable improvement, with only 13.4% of the investigated 14,442 water sources exceeding the Bangladeshi drinking water standard of 50 μg/L of arsenic, however, the concentrations varied substantially across different areas (UNICEF, 2011).

We have repeatedly assessed the exposure to arsenic in children in rural Matlab, about 50 km south-east of Dhaka, where we established a large mother–child cohort more than a decade ago (Vahter et al., 2006). The initial screening (2002–2003) of arsenic in the 13,000 tube-wells in the area, which provide drinking water for most of the 200,000 inhabitants, showed a range from 1 to 3644 μg/L, with more than 60% exceeding the national standard of 50 μg/L and 70% the WHO guideline value (WHO, 2011) of 10 μg/L (Rahman et al., 2006). The wells containing more than 50 μg/L of arsenic were immediately painted red, and people were encouraged to take drinking-water from nearby green-painted wells with lower arsenic concentrations. Additional mitigation options included pond sand filters, home-based "3-pitcher" filters, rainwater harvesting, and deeper wells (Biswas et al., 2012; Hossain et al., 2014; Ravenscroft et al., 2013; von Bromssen et al., 2007).

The exposure assessment has been based on measurements of arsenic concentrations in the drinking water used by the pregnant women

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recruited 2002–2003 and of their children, as well as arsenic in urine, a biomarker of the actual exposure (Gardner et al., 2011; Ljung et al., 2009; Vahter et al., 2006). Still at 5 years of age, the children had only slightly lower urinary arsenic concentrations than their mothers, indicating a low success rate of the arsenic mitigation activities (Gardner et al., 2011). Therefore, the same children have been followed-up again at 10 years of age, at which time arsenic in both drinking-water and urine was measured. Because a strong inverse association was found between the water concentrations of arsenic and manganese (Ljung et al., 2009), an essential element shown to cause neurodevelopmental toxicity at excess exposure (Rodriguez-Barranco et al., 2013), manganese was included in our follow-up analyses. The US life-time health advisory drinking water concentration for manganese is 300 μg/L (EPA, 2012).

2. Methods

2.1. Study design

The study took advantage of a large mother–child cohort, used for evaluation of potential effects of arsenic and other food and water contaminants on pregnancy outcomes and child health and development (Vahter et al., 2006). It was nested in a randomized food and micronutrient supplementation trial (MINIMat; ISRCTN 16581394), recruiting 4436 women in early pregnancy in 2001–2003 in Matlab (Persson et al., 2012). In this pristine rural area, essentially without pollution from industries and traffic, the International Centre for Diarrhoeal Disease Research, Bangladesh (icddr,b) has a Health and Demographic Surveillance System (HDSS) with a central hospital and four smaller regional health care centers, one in each of the administrative areas called blocks A–D. Information on socioeconomic status (SES) for all households was extracted from the HDSS at the time of enrollment in the study. The SES scores were based on household ownership of different consumer items, dwelling characteristics, type of drinking water source, and toilet facilities used, and standardized as described in detail elsewhere (Gwatkin et al., 2000). The SES scores were divided into quintiles where the lowest quintile represents the poorest and the highest quintile the wealthiest. The SES scoring was updated when the children were 10 years of age, showing strong correlation with the previous scoring [Spearman correlation coefficient (r_s) = 0.62, $p < 0.001$], although those at 10 years were generally higher.

The evaluation of arsenic exposure and related health effects included pregnant women enrolled in the MINIMat trial during January 2002 to May 2003, and who had donated a urine sample in early pregnancy (about 8 weeks of gestation; GW 8) (Vahter et al., 2006). The drinking water, essentially from tube-wells, used during pregnancy was identified (Rahman et al., 2015) based on the information on lifetime drinking water sources obtained in the parallel screening of well water arsenic carried out in 2002–2003 (Rahman et al., 2006). The main reason for missing maternal water data was that pregnancy occurred after completion of the household water survey, why those women had no information of the drinking water. The children born to the included women were invited to participate in an assessment of exposure and child development at 5 years (2007–2009) (Gardner et al., 2011; Hamadani et al., 2011) and at 10 years (2012–2013). Of the 1265 children with water and urine data at 10 years, 1017 had the corresponding data at both 5 years and during pregnancy and those were included in the present evaluation. Samples of drinking water used by the children were collected at the time of the home visit for collection of data on family characteristics.

The study was approved by the Regional Ethical Committee at the Karolinska Institutet, Sweden, and the Ethical Review Committee at icddr,b, Bangladesh. Informed consent was obtained from the mothers or legal guardians, and all participants were free to refrain from any part of the study.

2.2. Sample collection and analysis

Water samples were collected in 20-mL polyethylene vials after flushing the water with approximately 30 strokes of the pump. The vials contained 30 μL concentrated nitric acid (65% HNO_3, Suprapur, Merck, Germany) to prevent metal precipitation. Initially, the samples were stored at −20 °C at icddr,b and thereafter transported to Karolinska Institutet, Sweden, for analysis of arsenic and other elements. Water concentrations of arsenic, manganese, iron, calcium and magnesium were measured using inductively coupled plasma mass spectrometry (ICPMS; Agilent 7500ce or 7700x; Agilent Technologies, Tokyo, Japan) equipped with an octopole reaction system (Rahman et al., 2013). Before analyses, the samples were diluted 1:10 with 1% HNO_3 (65% Suprapur, Merck, Darmstadt, Germany). The limit of detection (LOD) and results of quality control (NIST 1643e, Trace Elements in Water, National Institute of Standards and Technology, Gaithersburg, MD; USA) for each element are provided in Supplemental Table S1. Thirty water samples ($n = 29$ at 5 years and $n = 1$ at 10 years of age) had an arsenic concentration below LOD of 0.01 μg/L, and they were set to LOD/$\sqrt{2}$.

The actual intake of inorganic arsenic was assessed by measurements of urinary concentrations of inorganic arsenic and its methylated metabolites [methylarsonic acid (MMA), and dimethylarsinic acid (DMA)]. The sum metabolite concentration in urine, hereafter referred to as urinary arsenic, reflects the exposure from both water and food (Vahter, 2002; Vahter et al., 2006). Spot urine samples were collected in the homes or in the health care centers, using disposable, arsenic-free plastic cups, and then transferred to 24-mL polyethylene tubes. Samples were kept at 4 °C until being frozen at −80 °C at the Matlab hospital at the end of each day, awaiting transport by air in cooling boxes to Karolinska Institutet, Sweden, for arsenic analyses. Urinary arsenic was initially (maternal urine) determined by hydride generation atomic absorption spectrophotometry (Vahter et al., 2006), and later on by high-performance liquid chromatography (Agilent 1100 series system, Agilent Technologies, Waldbronn, Germany; Hamilton PRP-X100 column, Reno, NV, USA) on-line with hydride generation and ICPMS (HPLC-HG-ICPMS; Agilent 7500ce, Agilent Technologies, Tokyo, Japan) for the urine samples of the children at 5 years of age (Gardner et al., 2013; Hamadani et al., 2011) and in a sub-set of the children in the present study population at 10 years ($n = 541$). The correlation between HG-AAS and HPLC-HG-ICPMS measurements was strong (Lindberg et al., 2007), and for the above-mentioned sub-set of children, an equally good agreement was observed between the sum of arsenic metabolites measured by HPLC-HG-ICPMS and total urinary arsenic measured by ICPMS ($r_s = 0.98$; $p < 0.001$). Thus, the measurements of child urine at 10 years of age were conducted with ICPMS (Agilent 7700x; Agilent Technologies, Tokyo, Japan). LODs and quality control for the ICPMS analyses are shown in Supplemental Table S1, whereas the corresponding information for the measurements of urinary arsenic of the mothers during pregnancy and the children at 5 years is described in detail elsewhere (Gardner et al., 2013; Hamadani et al., 2011; Vahter et al., 2006).

To compensate for variation in dilution of the spot urine samples, all measured concentrations were adjusted to the average specific gravity of 1.012. Specific gravity was measured by a digital refractometer (EUROMEX RD712 Clinical Refractometer, EUROMEX Holland, Anhem, The Netherlands) (Nermell et al., 2008). It ranged 1.001–1.032 in the mothers' urine and 1.001–1.031 in the children's urine. Specific gravity has been shown to be less affected by age, body size, SES, and arsenic exposure than the more commonly used creatinine adjustment (Nermell et al., 2008).

2.3. Statistical analyses

Statistical analyses were conducted using STATA 12 (STATAcorp LP, College Station, TX, USA). To assess differences in water element