



Trends in urinary arsenic among the U.S. population by drinking water source: Results from the National Health and Nutritional Examinations Survey 2003–2014



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ABSTRACT

Background: In 2001, the United States revised the arsenic maximum contaminant level for public drinking water systems from 50 µg/L to 10 µg/L. This study aimed to examine temporal trends in urinary arsenic concentrations in the U.S. population from 2003 to 2014 by drinking water source among individuals aged 12 years and older who had no detectable arsenobetaine - a biomarker of arsenic exposure from seafood intake.

Methods: We examined data from 6 consecutive cycles of the National Health and Nutrition Examination Survey (2003–2014; N = 5848). Total urinary arsenic (TUA) was calculated by subtracting arsenobetaine's limit of detection and detectable arsenocholine from total arsenic. Additional sensitivity analyses were conducted using a second total urinary arsenic index (TUA2, calculated by adding arsenite, arsenate, monomethylarsonic acid, dimethylarsinic acid). We classified drinking water source using 24-h dietary questionnaire data as community supply (n = 3427), well or rain cistern (n = 506), and did not drink tap water (n = 1060).

Results: Geometric means (GM) of survey cycles were calculated from multivariate regression models adjusting for age, gender, race/ethnicity, BMI, income, creatinine, water source, type of water consumed, recent smoking, and consumption of seafood, rice, poultry, and juice. Compared to 2003–2004, adjusted TUA was 35.5% lower in 2013–2014 among the general U.S. population. Stratified analysis by smoking status indicated that the trend in lower TUA was only consistent among non-smokers. Compared to 2003–2004, lower adjusted TUA was observed in 2013–2014 among non-smoking participants who used community water supplies (1.98 vs 1.16 µg/L, $p < 0.001$), well or rain cistern users (1.54 vs 1.28 µg/L, $p < 0.001$) and who did not drink tap water (2.24 vs 1.53 µg/L, $p < 0.001$). Sensitivity analyses showed consistent results for participants who used a community water supplier and to a lesser extent those who did not drink tap water. However, the sensitivity analysis showed overall exposure stayed the same or was higher among well or rain cistern users. Finally, the greatest decrease in TUA was among participants within the highest exposure percentiles (e.g. 95th percentile had 34% lower TUA in 2013/2014 vs 2003/2004, $p < 0.001$).

Conclusions: Overall, urinary arsenic levels in the U.S. population declined over a 12-year period that encompassed the adoption of the revised Arsenic Rule. The most consistent trends in declining exposure were observed among non-smoking individuals using public community water systems. These results suggest regulation and prevention strategies to reduce arsenic exposures in the U.S. may be succeeding.

1. Introduction

Arsenic is a public health concern worldwide including in the United States. Chronic exposure to inorganic arsenic (iAs) is associated with adverse health effects such as various cancers, skin disorders,

cardiovascular disease, and immunotoxicity (ATSDR, 2007; Cardenas et al., 2015, 2016; IARC, 2012; Naujokas et al., 2013). A naturally occurring element, there are numerous anthropogenic and natural sources of iAs in the United States. Exposures to iAs can come from contaminated soils or dust, emissions from industrial smelting processes

Abbreviations: iAs, inorganic arsenic; As^{III}, arsenite; As^V, arsenate; MMA, monomethylarsonic acid; DMA, dimethylarsenic acid; AsB, arsenobetaine; AsC, arsenocholine; TUA, Total urinary arsenic minus AsB and AsC; TUA2, Sum of As^{III}, As^V, MMA, and DMA; SDWA, Safe Drinking Water Act; EPA, United States Environmental Protection Agency; BMI, body mass index (kg/m²); PIR, family poverty-income ratio

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or specialized glass manufacturers, mining effluents, or household pesticides and chemicals (ATSDR, 2007). Inorganic As is also a common drinking water contaminant. Elevated concentrations of iAs in groundwater occurs throughout the United States although it is more prevalent in the Northeast, Midwest, and Western regions of the country (ATSDR, 2007; Frost et al., 2003; Nielsen et al., 2010). While drinking water contaminated with iAs is a major route of exposure in the United States (ATSDR, 2007; Naujokas et al., 2013), people can also be exposed to iAs from eating rice or rice-based products (Davis et al., 2012; Navas-Acien et al., 2011). Other dietary sources of arsenic include seafood, grains, fruits, and various juice products (Davis et al., 2012; deCastro et al., 2014; Navas-Acien et al., 2011). People can also be exposed to iAs from cigarette smoke (Caruso et al., 2013; Pappas, 2011).

Arsenic has been regulated in drinking water in the United States since 1942 when the United States Public Health Service set a standard of 50 $\mu\text{g/L}$ (USPHS, 1943). Amendments to the 1996 Safe Drinking Water Act (PL 1040182) required the United States Environmental Protection Agency (EPA) to issue a primary drinking water regulation for arsenic based on additional evidence of its health effects, occurrence, and treatment costs at low concentrations in drinking water. In 2001, the EPA adopted the revised Arsenic Rule, which reduced the maximum contaminant level (MCL) to 10 $\mu\text{g/L}$ for public water systems. This rule became enforceable in January 2006, but many small or highly affected water systems were provided renewable 3-year waivers to reach compliance (EPA, 2001). The EPA estimated that the revised arsenic MCL would affect more than 4000 water systems serving at least 12.7 million people (EPA, 2001). Water systems required to comply with the revised MCL include community water systems serving at least 25 people year-round (e.g. most cities and towns) or with at least 15 connections and non-transient, non-community water systems that serve at least 25 of the same people for at least 6 months per year (e.g. schools, churches, and businesses). The EPA does not regulate or monitor water sources considered private, which typically includes domestic wells serving a single or a limited number of homes (Nielsen et al., 2010). Thus, approximately 12% of the U.S. population who are served by domestic wells were not required to comply with the revised MCL even though it is estimated that 11–19% of private wells contain arsenic in excess of 10 $\mu\text{g/L}$ (Focazio et al., 2006; Kumar et al., 2010; Montgomery et al., 2003). It is likely, therefore, that the revised Arsenic Rule would reduce arsenic exposure only among people who receive their drinking water from a community water source.

Given the adoption of the Arsenic Rule, we hypothesized that there would be a population level decrease in iAs exposure following its implementation. We examined urinary arsenic levels among the general U.S. population to evaluate these trends in exposure among different water users by using 6 consecutive cycles of the National Health and Nutrition Examination Survey (NHANES) spanning from 2003 to 2004–2013–2014. NHANES collects biological monitoring data that is used to evaluate trends in population-level exposure to chemicals and NHANES urinary arsenic measurements have been used by multiple studies to improve our understanding of the health effects of iAs exposure (Cardenas et al., 2015; Jones et al., 2011; Navas-Acien et al., 2011) and sources of iAs exposure (Davis et al., 2012; deCastro et al., 2014; Mantha et al., 2017; Xue et al., 2010). Here we examined temporal urinary arsenic trends among people who receive their drinking water from a community supply, wells or rain cisterns, or who did not drink tap water. We hypothesized that decreased arsenic exposure would be greatest for individuals using public water systems (impacted by the revised Arsenic Rule) compared to individuals using wells (not impacted by the rule change).

2. Materials and methods

2.1. Study design

NHANES data is collected annually and publicly released in two-year cycles by the National Center for Health Statistics (NCHS) of the Centers for Disease Control and Prevention (CDC). The survey uses a complex multistage probability sample design to select a representative sample of the civilian, non-institutionalized U.S. population (CDC, 2015). Each cycle includes multiple survey stages that include questionnaires, physical exam, biospecimen collection and a variety of laboratory tests (Zipf et al., 2013). All participants provided informed consent and study protocols were approved by the NCHS research ethics review board (CDC, 2012).

We used publicly available data from six NHANES cycles: 2003–2004 ($n=2554$), 2005–2006 ($n=2568$), 2007–2008 ($n=2545$), 2009–2010 ($n=2855$), 2011–2012 ($n=2501$), and 2013–2014 ($n=2640$) cycles. This analysis was restricted to participants who had data on urinary arsenic speciation, which was first collected by NHANES in the 2003–2004 cycle (Caldwell et al., 2009). Urinary arsenic concentrations were measured in a random one-third subsample of participant's ≥ 6 years of age ($N=15,663$ from the 6 pooled NHANES survey cycles) (Caldwell et al., 2009). We further restricted our analysis to individuals who had urinary arsenobetaine concentrations below the limit of detection. Arsenobetaine (AsB) is the dominant form of organic arsenic found in seafood with 90% of the ingested AsB excreted in urine within 66 h (Molin et al., 2015; Schmeisser et al., 2006). Thus, by restricting to individuals without detectable AsB levels in the urine we are greatly reducing the potential for seafood intake to confound our analyses. Finally, to account for the potential confounding by smoking status we further restricted the sample to individuals who completed a smoking history questionnaire. This questionnaire was only asked to participants ≥ 12 years old. Thus, our final sample size was ($N=5848$ from the 6 pooled NHANES survey cycles). We compared the selected socio-demographic and exposure characteristics between the unrestricted and restricted analysis and observed no difference in underlying population characteristics (data not shown).

2.2. Urinary arsenic analysis

The methods describing urinary arsenic collection and measurement have been described previously (Caldwell et al., 2009; CDC, 2014). Briefly, spot urine samples were collected during the survey physical examination and shipped to the CDC's National Center for Environmental Health where the samples were analyzed within three weeks of collection by high-performance liquid chromatography and inductively coupled-plasma dynamic reaction cell-mass spectrometry. NHANES provides the following urinary arsenic measurements: total arsenic, arsenite (As^{III}), arsenate (As^{V}), arsenobetaine (AsB), arsenocholine (AsC), monomethylarsonic acid (MMA), dimethylarsenic acid (DMA), and trimethylarsine oxide (CDC, 2014). The arsenic metabolites AsB and AsC are commonly found in seafood and understood to be relatively nontoxic (Caldwell et al., 2009; Choi et al., 2010; deCastro et al., 2014). Thus, we defined total urinary arsenic (TUA) as total arsenic minus AsB and AsC. We also constructed a second total urinary arsenic composite (TUA2) by the sum of As^{III} , As^{V} , MMA, and DMA and re-ran all models as a sensitivity analysis (Supplemental Tables S4–S6, Fig. S1). These composite measures of urinary arsenic have been used by the CDC and in previous studies to represent iAs (Cardenas et al., 2015; CDC, 2017).

The limits of detection (LOD) for the three arsenic species used to compute TUA varied by survey cycle (Table S1). To enable unbiased comparisons of TUA across cycles, we assumed the maximal LOD for each arsenic species observed in one cycle (i.e. Total As = 0.88 $\mu\text{g/L}$, AsB = 0.84 $\mu\text{g/L}$, AsC = 0.42 $\mu\text{g/L}$, As^{III} = 0.85 $\mu\text{g/L}$, As^{V} = 0.71 $\mu\text{g/L}$, MMA = 1.35 $\mu\text{g/L}$, DMA = $\mu\text{g/L}$) and applied it to each cycle. Then, any samples below the maximal LOD were assigned the maximal LOD

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