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Dietary intake and urinary metals among pregnant women in the Pacific Northwest☆

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Pregnancy is a period when the mother and her offspring are susceptible to the toxic effects of metals. We investigated associations of intake of frequently consumed foods with urinary metals concentrations among pregnant women in the Pacific Northwest. We measured urinary cadmium (U-Cd), arsenic (U-As) and molybdenum (U-Mo) concentrations from spot urine samples in early pregnancy (15 weeks of gestation, on average) among 558 women from Seattle and Tacoma, Washington. We assessed periconceptional dietary intake using a semi-quantitative food frequency questionnaire (FFQ). We also determined early pregnancy zinc concentrations in serum. Statistical analyses involved multivariable linear regression models, adjusted for smoking status, age, race/ethnicity, multivitamin and supplement use, education, estimated total energy intake, and gravidity. The geometric mean and range in μg/g creatinine for U-Cd, U-As and U-Mo were 0.29 (0.1–8.2), 18.95 (3–550), and 72.1 (15–467), respectively. U-Cd was positively associated with dietary zinc intake (P-value = 0.004) and serum zinc (P-value<0.001) while it was negatively associated with coffee intake (P-value = 0.03). U-As was positively associated with dietary fish [(Lean fish, fatty fish, shellfish and non-fried fish) (P-values<0.01)], selenium (P-value = 0.004), zinc (P-value = 0.017), vegetables (P-value = 0.004), and low-fat yogurt (P-value = 0.03). Women who reported higher intake of dietary magnesium (Mg)(P-value = 0.04), insoluble fiber (P-value = 0.03), and low-fat yogurt (P-value = 0.04) had higher U-Mo concentrations. Our study suggests that vegetables, fish, fiber and yogurt might be significant dietary sources of metals. Future studies aimed at investigating the risk of exposure to metals from other various food sources among reproductive-age and pregnant women are needed.

1. Introduction

Maternal diet influences the health of the mother and the developing fetus (Emmett et al., 2015). The American College of Obstetricians and Gynecologists recommends that pregnant women include five food groups (i.e., grains, fruits, vegetables, protein foods and dairy) in their daily diet (American College of Gynecologist, 2015). Food sources of micronutrients, vitamins and minerals, however, might also contain metals and lead to dietary exposures to metals (Kones, 1990; Satarug et al., 2010; Basu et al., 2011). For instance, vegetables and grains might be a source of cadmium (Cd), as vegetables may accumulate the heavy metal from soils naturally rich in Cd as well as from the use of Cd-containing fertilizers and pesticides (Cabrera et al., 1998). Cd is also found in meats, particularly liver and kidney, and in certain areas, in shellfish (Järup and Akesson, 2009). Seafood consumption can also lead to exposure to organic arsenic, a non-toxic form of arsenic (Fort et al., 2014). Rice and vegetables might provide inorganic arsenic (As)-considered more toxic than organic arsenic (Ramirez-Andreotta et al., 2013; Signes-Pastor et al., 2016). A recent study among Canadian pregnant women has shown that inorganic arsenic metabolite, dimethylarsinic acid (DMA), is the main inorganic

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arsenic species detected in maternal urine (Ettinger et al., 2017). Findings from some epidemiologic studies (Ettinger et al., 2009; Romano et al., 2015; Kile et al., 2016; Yang et al., 2016), but not all (Guo et al., 2017), indicate associations of As or Cd exposure with adverse maternal and fetal outcomes. For example, environmental exposure to As and Cd has been associated with low birth weight and a reduction in head circumference (Kippler et al., 2012; Davis et al., 2015; Kile et al., 2016). Previous studies have also reported higher risk of gestational diabetes and impaired glucose tolerance in pregnant women environmentally exposed to Cd or As, respectively (Ettinger et al., 2009; Romano et al., 2015).

Diet might also be a source of molybdenum (Mo), a ubiquitous trace element found in food and multivitamin/multimineral supplements (IOM, 2001; NHANES III, www.cdc.gov). Dietary sources of Mo include legumes (beans, lentils, and peas), grain products and nuts (IOM, 2001). Mo content in animal products, fruit and many vegetables is generally low (IOM, 2001). In the United States, dietary Mo intake is well above the recommended dietary allowances (RDA) for Mo (45 μg/day for adults) among women (76 μg/day) and men (109 μg/day) (IOM, 2001). Few epidemiologic studies have examined the health effects related to Mo intake. For example, prenatal Mo exposure (45.6–54.0 μg/g of creatinine) was associated with low psychomotor index in Mexican children (Vázquez-Salas et al., 2014). Several studies have evaluated metal exposure through food sources in men and non-pregnant women, yet much remains unknown about the extent of dietary exposure to metals among pregnant women. This study aimed to determine the dietary factors associated with urinary Cd, As and Mo concentrations in pregnant women from Seattle and Tacoma, Washington.

2. Methods

2.1. Study population

Participants were women attending prenatal care clinics affiliated with the Swedish Medical Center and Tacoma General Hospital in Seattle and Tacoma, WA (Enquobahrie et al., 2005; Qiu et al., 2011). Eligibility criteria included: beginning prenatal care before 20 weeks' gestation, ability to consent and to communicate in English, age 18 years or older and planned to deliver at either of the two hospitals.

Participants completed an interviewer-administrated questionnaire during early pregnancy. They also completed a 121-item semi-quantitative food frequency questionnaire (FFQ) (Patterson et al., 1999). From medical records, we abstracted information on pregnancy outcome information. All procedures and study protocols were reviewed and approved by the institutional review boards of the study hospitals. All participants provided written informed consent.

The study population was derived from participants enrolled in the Omega Study between 1996 and 2008. During this period, 5825 eligible women were approached and 4602 (approximately 79%) agreed to participate. We selected a sub-cohort of 732 women randomly drawn from the full study cohort. Women found to have multi-fetal pregnancies (n = 28), physician diagnosed type 2 diabetes (n = 17), renal disease (n = 7), chronic hypertension (n = 26), iron deficiency anemia (n = 15), or unknown anemia status (n = 9) were excluded. Also excluded were women who reported extreme levels of daily total energy intake (<500 calories/day [n = 5] or >3500 calories/day [n = 67]). A cohort of 558 women remained for analyses.

2.2. Data collection

We collected information from structured-questionnaires and medical records on covariates including maternal age, smoking, pre-pregnancy weight and height, reproductive and medical histories. Maternal smoking was classified as never smokers, former smokers or current smokers.

Self-reported pre-pregnancy weight and height were used to calculate pre-pregnancy body mass index (BMI): weight in kilograms divided by height in meters squared. Participants completed a self-administered, validated, semi-quantitative food frequency questionnaire (FFQ) (Patterson et al., 1999) at a mean gestational age of 15 weeks to provide information on periconceptional (i.e., three months before conception and up to three months' post conception) dietary intake. The questionnaire, Women's Health Initiative (WHI) FFQ, allowed for assessment of intake, portion size, and food additives. Participants were provided clear instructions including photos of portion sizes. The WHI FFQ has documented reliability of accurately recording intake over an extended period of observation (Patterson et al., 1999). Micronutrient intake from dietary sources was estimated using food composition tables from the University of Minnesota Nutrition Coding Center nutrient database (Nutrition Coordinating Center, Minneapolis, MN). We also identified the major sources of dietary calcium in our population originating from high fat and low-fat dairy products, and whole grains as previously described (Osorio-Yañez et al., 2016).

Seafood was categorized into shell, lean, or fatty fish groups (Mohanty et al., 2015). Fried fish was described as "fried fish, fish sandwich and fried shellfish (shrimp and oysters). The snack chips category included "potato chips, corn chips, cheese crackers and tortilla chips" (Patterson et al., 1999; Stott-Miller et al., 2013).

2.3. Urinary metals

Urinary metals (Cd, As and Mo) were assessed in early pregnancy at 15 weeks of gestation, on average (standard deviation = 2.9, interquartile range = 13–17 gestational weeks). A clean-catch spot urine was collected in polyethylene containers, promptly separated into 2 mL aliquots, and stored at −80 °C until analysis. Urine concentrations were quantified using a validated method of inductively coupled mass-spectrometry (ICP-MS) and published protocols (Heitland and Koster, 2004) at Metametrix Clinical Laboratory, a Clinical Laboratory Improvement Amendments (CLIA) certified facility in Duluth, Georgia. ICP-MS calibration was performed by employing calibration curves generated by assaying a series of standards with known concentrations.

Briefly, urine samples were shaken and 1 mL was acidified with 1% HNO_3 (100 μL). An internal standard solution containing scandium, rhodium, and germanium (500 μL) was added. Samples were diluted to 5 mL with deionized water. Polyatomic interferences were minimized by utilizing ICP-MS with a dynamic reaction cell (DRC) (PerkinElmer SCIEX Elan DRC II with ESI SC-4, FAST Autosampler). Oxygen (0_2) as a reaction gas was used in combination with the appropriate bandpass settings (0_2 = 2.0 mL/min; RPq = 0.75) to avoid overlapping between molybdenum oxide (MoO) and Cd isotopes. The accuracy of ICP-MS was checked by conducting proficiency testing using a urine reference material (New York Toxic/Trace Elements in Urine Event 3#1 2012; A1209200170). The mean concentrations of As, Cd and Mo in the urinary reference material were 27.2, 5.3 and 84 μg/L, respectively. The mean concentration of metals was within the range reported for each metal in the urinary reference material [As (21.1–33.2), Cd (4.3–6.3) and Mo (58.8–109.2) μg/L]. Urine creatinine (Cr) concentration was assessed using a commercially available kit (Genzyme Diagnostics, Catalogue #221–30/#221–50) with improved Jaffe Reaction. The concentrations of Cd and Mo in maternal samples were all above the limits of detection of 0.10 μg/g Cr and 1.84 μg/L, respectively. Only one sample was below the limit of