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Dietary Intake of Total and Inorganic Arsenic by Adults in Arsenic-Contaminated Dan Chang District, Thailand, Using Duplicate Food Approach

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Abstract Dan Chang district, approximately 100 km west of Bangkok, was a site of tin mines operated almost 40 years ago. Mining operations caused arsenic contamination in soil, surface water, and groundwater within the district. The specific aim of this study was to estimate the dietary intakes of total and inorganic arsenic in 60 adults (30 males and 30 females) residing in Dan Chang district, using a duplicate food approach. The daily intake rates of inorganic arsenic ranged from 0.496 to 1.817 µg/kg BW for males and 0.342 to 1.778 µg/kg BW for females.

Keywords Total arsenic · Inorganic arsenic · Duplicate food approach · Dan Chang district

Dan Chang district is located in Suphan Buri province approximately 100 km west of Bangkok. Dan Chang district was a site of several former tin mines operated almost 40 years ago. As a result of mining operation, arsenic contaminated the soil, surface water, and groundwater (DEQP 2010; Charoenchaisri 2008). Concentrations of arsenic in contaminated areas have been reported to be 5.27–380 µg/g in soil, 17–9,965 µg/L in surface water, and 4–41 µg/L in groundwater (DEQP 2010; Charoenchaisri 2008). Although the mines are no longer in operation, concern still remains over the public health problems caused by arsenic exposure through the consumption of contaminated food. At present, people residing in the contaminated areas use rainwater and commercial water for consumption and cooking. The concentrations of arsenic in rainwater were 0.51–1.12 µg/L (DEQP 2010), which were far below the limit of 10 µg/L for drinking water recommended by the World Health Organization.

It has long been known that food is a significant source of arsenic exposure in humans (WHO 2001). Arsenic in food is present in several forms that have different toxicities. Inorganic arsenic [As(III) and As(V)] is the most toxic form of arsenic. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) has established the provisional tolerable weekly intake (PTWI) for inorganic arsenic, but not total arsenic (WHO 1989).

The specific aim of this study was to estimate daily intake of total and inorganic arsenic by adults residing in arsenic-contaminated Dan Chang district using a duplicate food approach for three consecutive days.

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Materials and Methods

Standard Reference Material (SRM) 2977 (mussel tissue, organic contaminants and trace elements) was purchased from the National Institute of Standards and Technology (NIST), USA. SRM 1566a (oyster tissue, NIST) was a gift from Dr. Chanchai Jaengsawang of the Thailand Department of Medical Sciences. SRM 1568a (rice flour) was a gift from Mr. Panompon Wongsapan of the Environmental Research and Training Centre, Thailand.

All participants in this study had lived and worked in Dan Chang district for at least 10 years. The participants consisted of 30 males and 30 females with an age range of 20–48 years. None of participants had gastrointestinal disorders or drank alcoholic beverages. No participants were vegetarians or were on special diets. All participants were paid for duplicate food samples and for participating in this project. Weights of participants were recorded at Day 1 (being 49–85 kg for males and 45–72 kg for females). Duplicate samples of food and beverages of each participant were collected in polypropylene containers from the breakfast of Day 1 through the evening meal of Day 3. Food samples were sent daily to the laboratory in Bangkok. In the laboratory, the daily food and beverage samples of individual participants were pooled, weighed, homogenized, freeze-dried, again weighed, and kept at 4°C until analysis. Food samples were collected in August 2009.

Sample preparation for determination of total and inorganic arsenic was described by Munoz et al. (1999). An atomic absorption spectrometer (Perkin–Elmer AAnalyst 300) equipped with an AS90 autosampler and flow injection system (Finorganical Arsenic 400) was used to determine total and inorganic arsenic concentrations in the final solutions. The atomic absorption spectrophotometric conditions were previously described by Ruangwises and Saipan (2010).

Limits of quantification (LOQ) for total and inorganic arsenic were determined using the Q2B method of the USFDA (1996). For determination of the LOQ for total arsenic, food samples (0.25 g) were fortified with SRM 2977 (mussel tissue) equivalent to total arsenic at concentrations of 1, 2.5, 5, and 10 µg/g; blank samples were not fortified with arsenic. Concentrations of total arsenic in arsenic-fortified samples and blank samples were quantified as performed for determination of total arsenic. Calibration curves (n = 6) were obtained by the least-square linear regression analysis of the residual peak heights of standard arsenic versus fortified total arsenic concentrations. For determination of the LOQ for inorganic arsenic, food samples (0.5 g) were fortified with an inorganic arsenic mixture [As(III):As(V) 1:1 w/w] at concentrations of 0.25, 0.5, 0.75, and 1.0 µg/g; blank samples were not fortified with inorganic arsenic. Concentrations of inorganic arsenic in arsenic-fortified samples and blank samples were quantified as performed for determination of inorganic arsenic. Calibration curves (n = 6) were obtained by the least-square linear regression analysis of the residual peak heights of standard arsenic versus fortified total arsenic concentrations.

The accuracy of determination of total arsenic was assessed by analyzing NIST SRM 1568a (rice flour) and SRM 2977 (mussel tissue). Since no commercial standard reference materials for inorganic arsenic are available, the amount of inorganic arsenic in SRM 1566a (oyster tissue) and 1568a (rice flour) were determined and compared with the previously reported values.

The differences of each parameter between males and females were analyzed using Student's t test. Statistical significance was considered when $p < 0.05$. SPSS Statistics version 17.0 for Windows was used for the statistical analysis.

Results and Discussion

The LOQ calculation was based on the standard deviation of y-intercepts of linear regression analysis (σ) and the slope (S) using the equation LOQ = 10 σ/S (USFDA 1996). The LOQs for total and inorganic arsenic were 0.044 and 0.024 µg/g dry weight (dw), respectively. Concentrations of total arsenic found in SRM 1568a (rice flour) and SRM 2977 (mussel tissue) were 0.286 ± 0.012 µg/g (n = 5; reference value 0.29 ± 0.03 µg/g) and 8.70 ± 0.43 µg/g (n = 6; reference value 9.2 ± 0.5 µg/g), respectively. The levels of inorganic arsenic found in SRM 1566a (oyster tissue) and SRM 1568a (rice flour) were 0.601 ± 0.037 µg/g (n = 4) and 0.103 ± 0.017 µg/g (n = 6), which agreed well with the previously reported values of 0.647 ± 0.027 µg/g and 0.110 ± 0.027 µg/g, respectively (Munoz et al. 1999, 2002).

Daily weights of lyophilized duplicate food samples ranged from 259 to 561 g for males and 248 to 467 g for females (Table 1). Statistical analysis showed that the average daily amounts of food consumed by males were higher than those consumed by females. Concentrations of total arsenic in daily food samples for males ranged from 0.475 to 1.063 µg/g; these values were comparable to those for females (0.289–1.730 µg/g). The average concentration of inorganic arsenic in food consumed by males (0.180 ± 0.046 µg/g) was significantly higher than that in food consumed by females (0.163 ± 0.050 µg/g). The percentages of inorganic arsenic with respect to total arsenic in food consumed by males and females were 11.1–47.5 and 10.9–51.4, respectively.

The daily amounts of total and inorganic arsenic consumed by males were significantly higher than those by females. The amounts of total and inorganic arsenic consumed by individual males ranged (respectively) from 158 to 493 µg/day and 27.8 to 120 µg/day; while the values for individual females were 92.2–561 and 20.2–114 µg/day, respectively. When the body weights (BW) of individual participants were used to calculate the daily intake rates of total and inorganic arsenic, the average daily intake rates of total and inorganic arsenic in males were also higher than those in females. The daily intake rate of inorganic arsenic

Table 1 Daily food intake, dietary arsenic concentration, daily arsenic intake, and daily arsenic intake rate by 60 participants for three consecutive days

Participant	n	Food intake (g/day, dw)	Arsenic concentration (µg/g, dw)		% Inorganic arsenic in food	Daily intake (µg/day)		Daily intake rate (µg/kg BW/day)	
			Total	Inorganic		Total	Inorganic	Total	Inorganic
Male	90	398 ± 69.0 (259–561)	0.745 ± 0.160 (0.475–1.063)	0.180 ± 0.046 (0.090–0.294)	25.2 ± 8.1 (11.1–47.5)	295 ± 76.0 (158–493)	71.0 ± 19.7 (27.8–120)	4.662 ± 1.108 (2.808–7.048)	1.122 ± 0.289 (0.496–1.817)
Female	90	325 ± 45.7* (248–467)	0.702 ± 0.252 (0.289–1.730)	0.163 ± 0.050* (0.063–0.291)	24.9 ± 8.6 (10.9–51.4)	230 ± 94.0* (92.2–561)	52.9 ± 18.2* (20.2–114)	3.906 ± 1.428* (1.601—9.073)	0.905 ± 0.284* (0.342–1.778)
Overall	180	362 ± 69.1 (248–561)	0.723 ± 0.211 (0.289–1.730)	0.172 ± 0.049 (0.063–0.294)	25.1 ± 8.4 (10.9–51.4)	262 ± 91.3 (92.2–561)	62.0 ± 21.0 (20.2–120)	4.284 ± 1.330 (1.601–9.073)	1.014 ± 0.306 (0.342–1.817)

Numbers in parentheses are ranges

* Significantly different from males ($p < 0.05$)

were between 0.496 and 1.817 µg/kg BW for males, and 0.342–1.778 µg/kg BW for females. In this study, daily intake rates of inorganic arsenic in both males and females did not exceed the provisional tolerable daily intake (PTDI) of 2.1 µg/kg BW recommended by the JECFA (WHO 1989).

Table 2 shows a comparison of daily arsenic intakes in various countries studied by the duplicate food approach. Wilhelm et al. (2003) reported dietary intakes of total arsenic by 14 German children living on a North Sea island, Amrum, ranging from 0.60 to 98.0 µg/day. Jorhem et al. (1998) studied the dietary intake of total arsenic by 15 Swedish adults residing in Stockholm and reported an average value of 60.0 ± 0.04 µg/day. Mohri et al. (1990) estimated dietary intake of total and inorganic arsenic by four Japanese adults living in Fukuoka being 27.0–376 and 1.80–22.6 µg/day, respectively. When compared to the values reported from these three countries, the daily amounts of total and inorganic arsenic consumed by the 60 adults residing in Dan Chang district were relatively high. This can be explained by the fact that all of the studies in those countries were conducted in uncontaminated areas.

Ruangwises and Saipan (2010) reported the daily intake of total and inorganic arsenic by 20 adults living in an arsenic-contaminated area of Ron Phibun district as 68.2–564 and 15.8–146 µg/day, respectively. Ron Phibun district, located in Nakhon Sri Thammarat province approximately 800 km south of Bangkok, was a site of several tin mines operated almost 100 years ago. Mining operations also caused arsenic contamination in soil, surface water, and groundwater within the district. Concentrations of arsenic in contaminated areas were 150–14,200 µg/g in soil, 35.8–583 µg/L in surface water, and 1.25–5,114 µg/L in groundwater (Visoottiviseth et al. 2002; Williams et al. 1996). Arsenic concentrations in soil and groundwater in Ron Phibun district were relatively higher than those found in Dan Chang district, but the arsenic level in surface water was lower. The daily intake rates of total and inorganic arsenic by adults residing in Ron Phibun district were 4.97 ± 1.36 and 1.43 ± 0.43 µg/kg BW/day, respectively, which were comparable to those by adults in Dan Chang district.

Like the Ron Phibun district case, high intake rates of total and inorganic arsenic by adults living in Dan Chang district may be partly due to possible uses of contaminated well water and surface water for cooking and the consumption of vegetables and fruits locally grown in the contaminated soil. It has been shown that several edible plants grown in contaminated soils accumulated high levels of arsenic (Abedin et al. 2002; Baroni et al. 2004). Further investigation is being conducted to estimate the concentrations of total and inorganic arsenic in individual vegetables and fruits grown in Dan Chang district.

Table 2 Daily arsenic intakes in various countries studied by the duplicate food approach

Country	Number of participants	Study days	Daily arsenic intake (μg/day)		Reference
			Total	Inorganic	
North Sea island, Amrum, Germany	14	7	6.90 ± 12.4 (0.60–98.0)	ND	Wilhelm et al. (2003)
Stockholm, Sweden	15	7	60.0 ± 0.04 (<50–180)	ND	Jorhem et al. (1998)
Fukuoka, Japan	4	7	182 ± 114 (27.0–376)	10.3 ± 5.50 (1.80–22.6)	Mohri et al. (1990)
Ron Phibun district, Thailand	20	7	287 ± 97.7 (68.2–564)	82.4 ± 27.8 (15.8–146)	Ruangwises and Saipan (2010)
Dan Chang district, Thailand	60	3	262 ± 91.3 (92.2–561)	62.0 ± 21.0 (20.2–120)	Present study

ND = not determined

Numbers in parentheses are ranges

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