Exposure levels to various arsenic species and their associated factors in Korean adults

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ABSTRACT: Arsenic (As) has been known a human carcinogen, which is widely distributed in the environment. This study was performed to evaluate the exposure level to arsenic and to determine contributing factors to human exposure of arsenic in Korean adults. Diet intake and urine sample were obtained from 2,044 study subjects older than 19 years of age. Human exposure to the arsenic was assessed by using the concentrations of various arsenic species such as inorganic As (As(V), As(III)), MMA, DMA, arsenobetaine in urine. The concentration of arsenic species in urine was analyzed by HPLC-ICP-MS method. In the arsenic speciation analysis, the geometric mean concentrations were 41.36 $\mu\text{g L}^{-1}$ of arsenobetaine, 24.72 $\mu\text{g L}^{-1}$ of DMA, 1.42 $\mu\text{g L}^{-1}$ of MMA, 2.96 $\mu\text{g L}^{-1}$ of inorganic As. The levels of various arsenic species in urine were influenced generally by age, smoking, inhabitant area and seafood intake. The consumption of seafood was positively correlated with some of arsenic species such as inorganic As, DMA and arsenobetaine. These findings suggest that the seafood might be a one of major source to the arsenic exposure in Korean adults, and overconsumption of seafood could increase human exposure to inorganic arsenic as well as organic.

1 INTRODUCTION

Arsenic (As) has been known one of major environmental pollutants which is ubiquitously distributed in the environment. Arsenic occurred from anthropogenic source such as mining, industry, agricultural uses as well as from naturally in soil, rock and volcanic eruption (ATSDR, 2007). There are several forms of arsenic. The toxicities of arsenic are quite different from their chemical form and valence, inorganic is much toxic than organic and trivalent is more toxic compared to pentavalent. Inorganic arsenic, arsenate (As(V)) and arsenite (As(III)), are metabolized in the body to the much less toxic metabolites, monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA), through reduction and methylation process, however, which are more toxic than organoarsenics such as arsenobetaine (AsB), arsenocholine (AC) and arsenosugars (AS). Organic arsenics are rapidly excreted in urine without metabolism in the body, which are essentially non-toxic. Human exposure to arsenic can be estimated from based on arsenic levels in blood, hair, nails, and urine (Choi *et al.*, 2010; Meliker *et al.*, 2006). Arsenic concentration in urine is being used as the most valuable biomarker which reflects the exposure to arsenic within the last several days (Navas-Acien *et al.*, 2011; Saoudi *et al.*, 2012). However, several forms of arsenic such as organoarsenic as well as inorganic arsenic and their metabolites, are excreted in urine. So, it is necessary to determine various arsenic species respectively for the risk assessment in human arsenic exposure. Several studies reported that seafood consumption increased DMA excretion in urine and suggested that kind of organoarsenic in seafood could be metabolized to

DMA in the body. Accordingly, it is questionable yet whether overconsumption of seafood could be hazardous to human health or not. Seafood is one of favorable food in Korean. However, there are very limited data for arsenic species analyses in the urine of Korean.

In this study, we performed speciation analysis of arsenic in urine and analyzed the relations between seafood consumption and various arsenic species in urine, which will be useful to do the risk assessment for arsenic in human exposure.

2 MATERIAL AND METHODS

Total of 2,044 study subjects, 888 males and 1,156 females were recruited, who were 19 years old or older. They had not been exposed to As occupationally. We sampled study subjects by probability sampling methods stratified by sex and age from the 102 sampling sites in Korea. Written informed consent was obtained from all study subjects participated in this study. We had conducted 1:1 interviews to investigate demographic characteristics and diet information during the last 24 h. Diet study was performed by the 24-h recall method. Also, we asked whether seafood intake within 72 hours before this study. Then, urine samples were collected and stored at -70°C until As analysis. The speciation analysis of As, such as As(V), As(III), MMA, DMA, arsenobetaine in urine was performed by HPLC-ICP-MS (HPLC pump PerkinElmer Series 200, PerkinElmer NEXION 300S). The levels of various arsenic in urine were presented as arithmetic mean, geometric mean, median and value at 95 percentiles. The concentrations of arsenic in urine were distributed log-normally rather than normal distribution, which were log-transformed for the statistical analyses. And

statistical analyses were performed with SAS version 9.2 (SAS institute Inc., Cary, NC, USA). The level of statistical significance was set at $p < 0.05$.

3 RESULTS AND DISCUSSION

The concentrations of geometric mean of various arsenic species in urine were 2.96 $\mu\text{g L}^{-1}$ of InAs (As(III) + As(V)), 1.42 $\mu\text{g L}^{-1}$ of MMA, 24.72 $\mu\text{g L}^{-1}$ of DMA, 41.36 $\mu\text{g L}^{-1}$ of AsB, and also were 30.90 $\mu\text{g L}^{-1}$ of TmetAs (As(III) + As(V) + MMA + DMA) and 84.67 $\mu\text{g L}^{-1}$ of TsumAs (As(III) + As(V) + MMA + DMA + AB) in Korean adults (Table 1).

Urinary As level in Korean adult is similar or less than Taiwan, Japan, China, while is higher compare to United State, France, Germany and United Kingdom (Fig. 1).

In general, contributing factors to the concentrations of arsenic species in urine were determined as age, coastal area inhabitant, and seafood consumption (Table 2).

Table 1. Mean concentrations of various As species in urine ($\mu\text{g L}^{-1}$).

		InAs ^a	MMA ^b	DMA ^c	AsB ^d	TmetAs ^e	TsumAs ^f
Total	AM $\pm$	6.02 $\pm$	2.22 $\pm$	34.11 $\pm$	98.15 $\pm$	42.35 $\pm$	140.49 $\pm$
(n = 2044)	SD	13.75	2.12	39.35	208.37	50.99	221.62
	GM	2.96	1.42	24.72	41.36	30.90	84.67
	(GSD)	(3.33)	(3.47)	(2.25)	(4.20)	(2.18)	(2.63)
	Median	2.95	1.73	25.29	43.82	31.88	82.85
	P95	19.1	5.62	83.11	337.96	100.85	407.07
	PTsum	4.20%	1.60%	24.30%	70.00%	30.00%	100.00%

*a: As(III) + As(V), b: monomethylarsinic acid, c: dimethylarsinic acid, d: arsenobetaine, e: As(III) + As(V) + MMA + DMA, f: As(III) + As(V) + MMA + DMA + AsB, PTsum: percentage in relation to Tsum, AM: arithmetic mean, SD: standard deviation, GM: geometric mean, GSD: geometric standard deviation, P95: value at 95 percentile. * TmetAs: As(III) + As(V) + MMA + DMA, TsumAs: As(III) + As(V) + MMA + DMA + AsB.

Figure 1. International comparisons of arsenic concentration in urine ($\mu\text{g/L}$) (TmetAs, TsumAs).

Table 2. Mean concentrations of various As species in urine by demographic characteristics ($\mu\text{g/L}$).

		N	InAs ^a	MMA ^b	DMA ^c	AsB ^d	TmetAs ^e	TsumAs ^f
Gender	Male	888	2.97 (3.19)	1.50 (3.59)	24.24 (2.21)	45.55 (3.87)	30.37 (2.16)	87.68 (2.59)
	Female	1156	2.95 (3.45)	1.36 (3.38)	25.10 (2.28)	38.40 (4.45)	31.31 (2.21)	82.4 (2.66)
Age (yrs)	19–29	355	2.76 (3.25)	1.45 (3.10)	20.61 (2.39)	29.10 (4.84)	26.56 (2.26)	66.42 (2.74)
	30–39	360	3.10 (3.07)	1.31 (3.78)	23.72 (2.34)	37.83 (4.03)	30.09 (2.17)	78.67 (2.57)
	40–49	450	3.20 (3.13)	1.55 (3.25)	26.82 (2.17)	47.08 (3.99)	33.19 (2.14)	92.89 (2.56)
	50–59	475	2.78 (3.33)	1.48 (3.28)	26.22 (2.08)	46.65 (4.01)	32.29 (2.07)	92.17 (2.61)
	60–	404	2.97 (3.88)	1.30 (4.02)	25.64 (2.27)	45.83 (4.07)	31.68 (2.29)	91.35 (2.62)
Inhabitant area	Inland	1541	2.77 (3.31)	1.46 (3.34)	23.88 (2.21)	35.24 (4.13)	29.86 (2.13)	76.13 (2.50)
	Costal	503	3.63(3.33)	1.30 (3.88)	27.49 (2.36)	67.54 (3.97)	34.30(2.33)	117.30 (2.86)
Seafood intake[†]	No	734	2.61 (3.40)	1.34 (3.83)	20.97 (2.28)	29.18(4.46)	26.58 (2.20)	65.62 (2.60)
	Yes	1295	3.17 (3.26)	1.46 (3.29)	27.15 (2.20)	50.71 (3.90)	33.65 (2.15)	98.12 (2.57)

[†]: seafood intake during the last 3 days before this study. Data are presented as geometric mean and geometric standard deviation, a: As(III) + As(V), b: monomethylarsinic acid, c: dimethylarsinic acid, d: arsenobetaine, e: As(III) + As(V) + MMA + DMA, f: As(III) + As(V) + MMA + DMA + AsB.

No relations were observed between total food consumption and arsenic levels in urine. However, the consumption of fish/shellfish was positively correlated with InAs, DMA, arsenobetaine, TmetAs and TsumAs. The consumption of seaweeds was also correlated positively with InAs, DMA, TmetAs and TsumAs, but not arsenobetaine.

4 CONCLUSIONS

Urinary As level in Korean adult is similar or less than Taiwan, Japan, China, while is higher compare to United State, France, Germany and United Kingdom. Our data suggests that seafood was primary cause of arsenic exposure among Korean adults, and overconsumption of seafood could increase human exposure level to the inorganic As as well as organic As.

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REFERENCES

Choi, B.S., Choi, S.J., Kim, D.W., Huang, M., Kim, N.Y., Park, K.S., Kim, C.Y., Lee, H.M., Yum, Y.N., Han, E.S., Kang, T.S., Yu, I.J. & Park, J.D. 2010. Effects of repeated seafood consumption on urinary excretion of arsenic species by volunteers. *Arch. Environ. Contam. Toxicol.* 58(1): 222–229.

Meliker, J.R., Franzblau, A., Slotnick, M.J. & Nriagu, J.O. 2006. Major contributors to inorganic arsenic intake in southeastern Michigan. *Int. J. Hyg. Environ. Health* 209(5): 399–411.

Navas-Acien, A., Francesconi, K.A., Silbergeld, E.K. & Guallar, E. 2011. Seafood intake and urine concentrations of total arsenic, dimethylarsinate and arsenobetaine in the US population. *Environ. Res.* 111(1): 110–118.

Saoudi, A., Zeghnoun, A., Bidondo, M.L., Garnier, R., Cirimele, V., Persoons, R. & Fréry, N. 2012. Urinary arsenic levels in the French adult population: the French National Nutrition and Health Study, 2006–2007. *Sci. Total Environ.* 433: 206–215.