Short Communication

Higher cadmium burden in coastal areas than in inland areas in Korea: implications for seafood intake

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This survey was initiated to examine possible coastal-inland differences in cadmium (Cd) burden in general Korean populations. In total, 268 healthy non-smoking middle-aged women (30 to 49 years; 88 residents in 8 coastal areas and 180 residents in 15 inland areas) participated in the study. They offered peripheral blood and spot urine samples so that cadmium in blood (Cd-B) and urine (Cd-U) were taken as exposure markers. Determination of Cd-B and Cd-U was carried out by graphite furnace atomic absorption spectrophotometry. With regard to Cd burden, geometric means for the coastal and inland residents were 1.70 and 1.72 µg/L for Cd-B, 1.54 and 1.00 µg/L for Cd-U as observed (Cd-U), 2.59 and 1.81 µg/g creatinine for Cd-U as corrected for creatinine (Cd-U_{cr}), respectively. Cd-U and Cd-U_{cr} were higher in the coastal areas than in inland areas. Reasons for higher Cd-U in the coastal areas than in the inland areas were are discussed in relation to major sources of Cd in daily life of the residents. Attention was paid to consumption of fish and shellfish in the coastal areas as major sources of dietary Cd intake. This study shows that Cd burdens were higher in coastal areas than in inland areas in Korea.

Key Words: blood, urine, cadmium, coastal areas, exposure marker

INTRODUCTION

Cadmium (Cd) is well-known toxic metals ubiquitous in the general environment. Our previous survey showed that exposure of general populations in Korea to Cd takes place primarily via foods and drinking water,[1] as to be discussed later in detail. Korea is a country on a peninsula having seas on east, south and west sides with mountainous inland areas in the center. Characteristic differences in dietary habits with regard to consumption of sea-harvested foods have been reported.[2] It is quite conceivable that consumptions of seafood such as fish and shellfish may be higher in the coastal areas than in the inland areas so that the difference may be reflected even in terms of body burden of toxic metals,[3] indicating potential risk to health of local people.[4-6] Nevertheless no reports are available on this possibility of public concern in case of Cd in population of Korea and other countries.

The present study was initiated to investigate possible differences in exposures to Cd between residents in inland and coastal areas in Korea. For this purpose, the Cd levels in blood and urine were determined as representative indicators of exposure.[7-10]

MATERIALS AND METHODS

Ethical issue

The study protocol was approved by the Review Board of Catholic University of Pusan, Pusan, Korea. Each study participant provided informed consent in writing.

Survey sites and populations

In total, 8 fishing villages in coastal areas and 15 rice-producing agricultural villages or residential towns in inland areas were surveyed in April, 2007 to December, 2008. The areas having seashore and the areas without such were defined as coastal and inland areas, respectively. In practice, 268 healthy and non-smoking women aged between 30 to 49 years volunteered to participate in the surveys by offering blood and urine samples, and filling self-administered questionnaires (to be detailed below). Of the 268 women, 88 and 180 women were lived in coastal and inland areas for more than 3 years, respectively. There was no significant difference (p>0.10) in age between the two groups (mean age, 41.1 and 38.6 years in

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coastal and inland participants, respectively). The survey was conducted in the morning, and venous blood and second morning urine sample were collected. For blood sampling, vacuum tubes with heparin for anticoagulation (Vaccutainer, BD, NJ, USA) were employed; the device was previously tested to confirm the absence of metal contamination, including Cd, or leakage.

In parallel with the blood and urine sampling, the participants were requested to answer the questionnaire, for example, height, weight, age, disease record and current status, current administration of medicine, smoking habits (for confirmation of non-smoking) and dietary habits on seafood intake in particular, i.e., if they had seafood in any one of the nine meals in the past three days from survey day. Originally, participants were requested to answer questions if the seafood consumed were finned fish, shellfish or seaweeds, but the answers were incomplete and could not be analyzed further. Women were excluded from the study if they took medicine, had a serious disease, had occupational exposure, or were identified as having environmental exposure to Cd or other hazard materials such as harmful metals, volatile organic compounds, polycyclic aromatic hydrocarbons, PCDD/PCDFs and PCBs.

Methods of analyses for Cd in blood and urine

The methods of analyses were previously described in detail.[1,7] In short, the biological sample (1 ml urine or heparinized blood) was 10- or 20-fold diluted with a 1:1:1 mixture (a matrix modifier) of 0.2% Triton X-100, 0.2% $(NH_4)_2HPO_4$ and 0.015 % $Mg(NO)_3 \cdot 6H_2O$ in case of Cd analyses. The diluted samples were introduced to the instrumental system, using the standard addition method. The instrument employed was a graphite furnace atomic absorption spectrometer with Zeeman background correction (AA240Z, Varian, Melbourne, Australia), connected with an auto-sampler. The instrumental limit of detection for Cd was 0.1 µg/L. For internal quality control, BioRad Lyphochek whole blood metals (control levels 1 and 2) and urinary metals (control levels 1 and 2) (Bio-Rad Laboratories, Irvine, CA, USA) were analyzed. The ratio of observed values to certified values for Cd in all the control samples were in a range of 85% to 115%. The quality of the analyses was certified also externally by the German Quality Assessment Scheme (G-EQUAS) for Cd in blood and urine (environmental medicine field) in 2007 and 2008.

Creatinine (CR or cr) in urine was measured by conventional colorimetry. In some instances, observed values of Cd in urine (Cd-U) were corrected for CR,[11] and expressed as Cd-U_{cr}, respectively.

Statistical analyses

A log-normal distribution was assumed for both Cd in blood and urine. Accordingly, Cd levels were expressed in terms of geometric means (GM) and geometric standard deviations (GSD). The levels below the instrumental detection limits were assumed to be half the limit. Unpaired t-test was applied to the values after logarithmic conversion. For dichotomous parameters, χ^2 test was employed to detect possible significant difference in distribution.

RESULTS

The results of blood and urine analyses for Cd are summarized in Table 1. Cd-U levels were significantly different (p<0.01) between the two types of areas, ie, 1.54 µg/L for the coastal areas and 1.00 µg/L for the inland areas as geometric means. The difference was maintained even after correction for CR (i.e., 2.59 and 1.81 µg/g cr for the coastal and inland areas, respectively, with p<0.01 for the difference). There was however no significant difference in Cd-B levels between coastal (1.70 µg/L as geometric mean) and inland areas (1.72 µg/L) (p>0.10).

The bottom line in Table 1 shows the prevalence of seafood intake by the women in the coastal and inland areas. A questionnaire survey showed that 77 and 136 women out of 88% and 180 participants in the coastal and inland areas (88% and 76%), respectively, took seafood at least once in nine meals in past three days. The prevalence was significantly (p<0.01) higher for residents in the coastal areas than for those in the inland areas.

DISCUSSION

The dietary route is almost exclusively the main source of Cd intake, accounting for more than 95% of total intake in general population exposure in many Asian countries.[9] Smoking cigarettes is a substantial source of exposure in east Asia where smoking rates are still high, e.g., 47.3% and 6.8% for adult men and women in Korea in 2011.[10,12] In this study, it should be noted that the participants were all non-smokers.

Table 1. Cd levels in blood and urine; comparison between coastal and inland areas

	Unit	Coastal areas		Inland areas		Total		p for difference†
		GM	GSD	GM	GSD	GM	GSD	
Number of cases		88		180		268		
Age‡	years	41.1	4.2	38.6	5.1	39.4	5.0	>0.1
Cd-B	µg/L	1.70	1.58	1.72	1.70	1.71	1.66	0.87
Cd-U	µg/L	1.54	2.11	1.00	2.42	1.15	2.37	<0.01
Cd-U_{cr}	µg/g cr	2.59	2.14	1.81	2.59	2.04	2.48	<0.01
Proportion of seafood intake§		77	(88%)	136	(76%)	213	(80%)	<0.01¶

† p for difference between the coastal and inland areas as examined by unpaired t-test after logarithmic conversion
‡ Arithmetic mean and arithmetic standard deviation are given in place of GM and GSD
§ Intake prevalence of finned fishes, shellfishes and sea weeds in 9 meals in the latest 3 days (percentage in parentheses)
¶ By χ^2 test

Table 2. Mean range and median of dietary Cd levels (μg/g) in each food group[32]

Food group	Median†	Range of means†
Plant-based foods		
Grains and cereals	0.012	0.001-0.023
Potatoes and starch	0.101	0.017-0.185
Sugars and sweets	0.005	0.005
Pulses	0.008	0.004-0.017
Nuts and seeds	0.063	0.063
Vegetables	0.003	0.003-0.016
Fruits	0.002	0.000-0.004
Seaweeds	0.224	0.144-0.304
Beverages	0.002	0.001-0.007
Seasonings	0.009	0.009
Animal-based foods		
Fishes and shellfishes	0.064	0.002-0.559
Meats and poultry	0.03	0.03
Milks and dairy products	0.005	0.005

† Medians and ranges of mean values reported in each reference; values are for fresh weight

As for the dietary sources of Cd, rice is regarded as the major source for general Korean populations. For example, Moon and others observed in a field survey including two metropolitan cities of Seoul and Busan, Korea, that rice (as boiled rice) accounted for 24 to 28% of total Cd in food duplicate studies.[1] Kim and Wolt gave a very similar estimate of 25%.[13] Other possible sources of oral Cd intake for Korean populations include dietary supplements (typically for calcium) and oriental medicine, but their contributions are most likely to be limited from quantitative viewpoints.[14,15]

With regard to seafood intake, both total mercury and total arsenic have been taken as indicators of seafood consumption.[16-20] Ahn observed that the level of total mercury in urine samples from school boys was higher (p<0.05) in fishing villages (2.52 μg/g cr) than in farming villages (1.62 μg/g cr).[21] Im estimated that dietary total arsenic intake by adult people was higher in coastal areas (422 μg/day) than in urban and rural areas (308 and 238 μg/day, respectively).[22] Both reports suggest higher seafood intake in coastal areas than in other areas. Regarding seaweed intake in particular, Jeong reported that high school students in a sea coast area had seaweed dishes more frequently than their counterparts in an inland area.[23]

As for studies on increased level of dietary Cd intake in association with seafood intake, the compilation of the results of a literature survey made it clear that seafood is an important source of Cd (Table 2).[32] In agreement, Falco et al reported that daily Cd was high in association with consumption of fish and shellfish in Catalonia, Spain.[24] Marti-Cid et al also observed that seafood was the main contributors to the total dietary intake of Cd.[25] With regard to the seaweed intake, high level of Cd intake was reported in several reports.[26-28] In evaluating the risk associated with seaweed consumption, however, it should be taken into account that the daily consumption of seaweed is quite limited.

Perusal of reports on Cd contents in various foods (Table 2) suggests that Cd contents in dried seaweeds (0.224 μg/g as a median) are 20 times higher than that in grains and cereals (0.012 μg/g).[32] Nevertheless, the dietary habits in Korea are such that people take about 10 g seaweed/day.[29-30] Assuming that about 10% of the seaweed consumed is dry products and 90% is fresh one, that 80% of fresh seaweed weight is attributable to water,[31] and that 10% water will remain in seaweed after drying process, the total seaweed consumption on a dry weight basis is about 2 g/day or less. Thus, less than 0.5 μg Cd/day will come from seaweeds. As Cd from grains (including cereals) and potatoes (including starch) are 3.0 and 3.3 μg Cd/day,[32] the Cd intake from seaweeds is most likely to be less than that from grains or potatoes.

In the present study, it was not possible to clarify separate contribution of finned fish, shellfish and seaweeds intake. Nevertheless, the diet duplicate survey in Busan (a coastal area as defined in the present study; 47 cases), Seoul and Chunan (two inland areas; 53 cases in total) allows us to make dietary Cd estimation as in the previously reported Korean dietary survey.[1] The re-constructed results from Moon et al in terms of seafood types and coastal/inland areas are summarized in Table 3.[1] In the survey,[1] diet samples from participants were 24-hour food duplicate, including snacks and drinks. All participants in the three survey sites took finned fish and shellfish. With regard to seaweed intake, 81% of the participants in Busan (a coastal area) had seaweeds in the food duplicate samples, whereas it was 75% in Seoul+Chunan (the inland areas). Although the prevalence appeared to be higher in the coastal area than in the inland area, the difference was not statistically significant (p>0.10). Interestingly, Cd-U was higher in the coastal area than in the inland areas while there was no difference in Cd-B, in a close agreement with the present observations, shown in Table 1.

In evaluating the burden from seafoods, it should be taken into account that both Cd contents in mussels varied substantially depending on where they were harvested in Korea. Thus, the mussels from the western coast contained substantially more Cd than those from the southern and eastern coasts.[33] It is therefore quite conceivable that seafood from the western coast may carry more Cd than those from other coasts. This factor was however not taken into account in the present analyses. In Korean report of 53 species of fish from the eastern, western and southern coasts of Korea, Cd concentration range was from

Table 3. Intake weight and frequencies of seafood by diet duplicate method

Cd exposure parameter	Coastal (Busan)	Inland (Seoul + Chunan)	Total	p
No. of participants	47	53	100	
Food analysis†				
Intake by weight (GM: g/day)				
Seaweeds	0.58	0.10	0.23	0.282‡
Fishes and shellfishes	55.3	26.7	37.6	0.002‡
Prevalence				
Seaweeds (%)	38 (81)	40 (75)	78 (78)	0.517§
Fishes and shellfishes (%)	47 (100)	53 (100)	100 (100)	>0.10§
Cd in blood and urine (GM)†				
Cd-B (μg/L)	1.26	1.11	1.17	0.252‡
Cd-U (μg/L)	1.73	1.26	1.46	0.059‡
$Cd\text{-}U_{sg}$ (μg/L)	1.73	1.27	1.47	0.072‡
$Cd\text{-}U_{cr}$ (μg/g cr)	2.09	1.65	1.85	0.048‡

†Recalculated from Moon et al. 1995[1]. Cd-B is for Cd in blood, and Cd-U is for Cd in urine; Cd-U, $Cd\text{-}U_{sg}$ and $Cd\text{-}U_{cr}$ are Cd-U as observed, Cd-U after correction for a specific gravity of urine of 1.016, and afrer correction for creatinine concentration, respectively.
‡ Comparison by unpaired t-test.
§ Comparison by χ^2 test.

<0.001 to 0.091 μg/g. Small fishes, such as ancovy (*Engraulis japonica*) and sand lance (*Ammondytes personatus*) had low Cd concentrations (<0.001 μg/g and <0.001 μg/g to 0.014 μg/g, respectively). The fish with the highest Cd concentrations were cloudy catshark (*Scyliorhinus torazame*; 0.083 μg/g), conger eel (*Conger myriaster* ; 0.039 μg/g), and Brown sole (*Limanda herzensteini*; 0.053 μg/g).[35] Fish contained a comparatively lower Cd concentration than shellfish (0.232-0.559 μg/g in range) in a report from coastal regions of Korea.[36]

The reason for the lack of a difference in Cd-B in costal vs. inland participants, despite such a difference being present in Cd-U (Table 1) is not clear from the present study, although similar results were obtained in a previous study.[1] One possible explanation is the difference in biological half-life, ie, the half -life is much longer for Cd-U (10-30 years) than for Cd-B (75-130 days for the rapid phase and 7.4-16 years for the slow phase).[34] As in many other countries, it is quite conceivable that, in the past, people depended more on locally harvested foods such as seafood in case of the residents in coastal areas. This implies that Cd exposure was more intense for coastal people than for inland people because of larger consumption of Cd-rich seafood. Higher accumulation of Cd in the body, typically in the kidney, in the past may be reflected in higher Cd-U in coastal residents, whereas recent development of nation-wide food supply system is likely to have made the dietary Cd exposure more even throughout the country so that no significant differences would be detected in Cd-B. Such a working hypothesis should deserve further study for verification and validation.

Cd-U and $Cd\text{-}U_{cr}$ as long-term Cd exposure markers were higher in the coastal areas than in the inland areas, suggesting that the body burden is higher in residents in coastal areas than those in inland areas. Cd-B in short-term Cd exposure markers however did not show a significant difference between the two areas. The main influential source was estimated to be a higher intake of fish and shellfish in coastal areas than in inland areas.

AUTHOR DISCLOSURES

The authors declare that they have no conflicts of interest. This study was supported by Research Grant 2011 from Catholic University of Pusan, Korea.

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韩国沿海地区镉负担高于内陆：海产品摄入的启示

此调查旨在研究韩国沿海人群和内陆人群体内镉(Cd)含量的差别。此项调查共有 268 位不吸烟的健康中年女性参加(年龄 30-49 岁，其中有 88 位来自 8 个沿海区域，180 位来自 15 个内陆区域)。对她们采集外周血样和随机尿样，从而了解血液镉(Cd-B)和尿液镉(Cd-U)的含量。采用石墨炉原子吸收分光光度法测定 Cd-B 和 Cd-U。结果显示，内陆和沿海居民 Cd 含量的几何平均值 Cd-B 分别为 1.7 μg/L 和 1.72 μg/L；Cd-U(Cd-Uob)分别为 1.54 μg/L 和 1.00 μg/L；肌酐矫正值(Cd-Ucr)为 2.59 μg/g 肌酐和 1.81 μg/g 肌酐。沿海居民 Cd-Uob 和 Cd-Ucr 均高于内陆居民，其原因可能是沿海居民生活接触大量镉有关。值得注意的是，沿海地区鱼类和贝类的摄入可能是其体内 Cd 的主要来源。本研究表明，韩国沿海地区居民镉负担要高于内陆居民。

关键词：血液、尿液、镉、沿海地区、暴露标志物