# Assessing inorganic arsenic in rice and its health risk to consumers in Ho Chi Minh City, Vietnam

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## Abstract

**Purpose** – The consumption of rice that contains high levels of inorganic arsenic may cause human health risk. This study aims to determine As species concentrations, particularly iAs, in raw rice in Ho Chi Minh (HCM) City and its health risks.

**Design/methodology/approach** – A total of 60 polished raw composite samples of rice were purchased from traditional markets and supermarkets in HCM City. All samples were analyzed by HPLC-ICPMS for As species determination.

**Findings** – Mean concentrations of inorganic arsenic in all samples, which were purchased from supermarket and traditional market, were 88.8 µg/kg and 80.6 µg/kg, respectively. Overall, inorganic arsenic level was 84.7 µg/kg and contributed the highest proportion of arsenic species in rice with 67.7%. The proportion profiles for arsenic species were: As (III) (60 %); dimethylarsinic acid (32.2 %); As (V) (7.7 %) and methylarsonic acid (0.1 %). Inorganic arsenic level in raw rice was below the recommendation of World Health Organization. Using the benchmark dose recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA), all exposure doses were lower than BMDL05. However, as the doses ranged from 3.0 to 8.6 of Margin of Exposure (MOE), the health risk of iAs from rice consumption remains public health concern.

**Originality/value** – The study results report on the surveillance data of the presence of inorganic arsenic in raw rice products, which are available in the supermarkets and traditional markets, and its health risk to consumers in a metropolitan city in Vietnam.

**Keywords** Inorganic arsenic, Raw rice, Health risk, Vietnam

**Paper type** Research paper

## Introduction

Arsenic (As) occurs ubiquitously, is prevalent in the environment and can easily enter the food system through contaminated soil or water. Certainly, inorganic As species are found to be more toxic than the organic form [1, 2]. Inorganic arsenic (iAs) is classified by the



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International Agency for Research on Cancer (IARC) as a class 1 human carcinogen and is associated with skin, lung, liver, kidney and bladder cancers [3]. Different arsenic (As) forms can be found in rice gains. In general, around 50% of the total As in rice grain is in the inorganic form (ranging from 10% to 90%), while the remaining fraction is dimethylarsinic acid (DMA) with trace amounts of methylarsinic acid (MMA), which are considered to be nontoxic [4]. The exposure to iAs is found to cause health risks in various endemic regions in Asia and American with the most important sources being from drinking water and contaminated food [5–7]. In recent years, several published papers have shown that rice produced in some areas was contaminated with arsenic (As), especially inorganic arsenic (iAs), which is a cause for public concern. Rice produced from arsenic-endemic areas in South and Southeast Asian countries is contaminated with iAs about 42–91% [4, 8, 9]. Hence, the consumption of rice among people living in these areas may cause a higher human health risk.

In Vietnam, the Mekong River Delta (MD) and Red River Delta are identified as severe As-contaminated regions. Previous studies have proved that the source of high iAs in MD resulted from natural sources [10–12]. The MD is a crucial rice production region of Vietnam. Thus, the problem arises when using high iAs groundwater for irrigation, iAs will be accumulated in paddy soils and, then, rice grains [13–15]. In the last decade, groundwater has become a significant water supply source in Vietnam [16, 17] and has been supplied mainly for agricultural use at about 769,619 $m^3$/day or 40.01% of the total groundwater abstraction [18]. Huang *et al.* have indicated that the irrigation with iAs-contaminated groundwater in MD had raised the As level in field soil [19]. This may lead to a higher iAs in rice. Other studies have also estimated the As concentration in rice, but most of them estimated iAs indirectly from measured total As (tAs) by using the proportion of iAs in tAs from other studies or rice samples from other countries [20–24]. Few studies are focusing on As species, particularly iAs in rice, which is grown in the MD and their risks to people who consume rice.

Ho Chi Minh (HCM) City is the largest city in Southern Vietnam with a population of more than 8m people [25]. Rice consumed in this city is mainly from MD and some from other regions of Vietnam and countries such as Thailand, Japan and the USA [26]. With a gap about As species concentration in rice from MD and the risk of iAs in rice, this study aims to determine the As species level, particularly iAs, in raw rice, and to assess the health risk of iAs through rice consumption of people in HCM City.

## Materials and methods

### *Sample collection and preparation of raw rice*

The study was designed at the consumer end, thus, concerning the consumers' purchasing behavior, it was assumed that people would purchase rice with different brands based on price, characteristics, sales promotion and quality. According to the market share, the top five famous brands in each market were also taken into account making the selected scenario more relevant to the real situation. In total, 30 supermarkets and 30 traditional markets in HCM City were selected randomly. In each retail store of the traditional markets and supermarkets, 60 g was taken from each of the five famous rice brands. All samples were kept in plastic zip bags at room temperature before pretreatment.

During pretreatment, regarding the five famous rice brands, the amount of 60 g of each rice brand was mixed to perform a composite sample (300 g). Then, rice grains were homogenized, ground and sieved with a 0.5 mm ring by the Ultra Centrifugal Mill ZM 200 (Retsch, Inc., Haan, Germany) and then oven-dried at 105°C by Memmert Une 500 (Memmert GmbH + Co. KG, Schwabach, Germany) until a constant weight was reached. After that, the powder samples were stored in a room at 4°C.

Finally, about 1 g of the raw rice samples was analyzed by using the high-performance liquid chromatography (HPLC) with the inductively coupled plasma mass spectrometry

(ICPMS) at the Southern Food Testing Centre, Institute of Public Health at Ho Chi Minh City, Vietnam.

*Arsenic determination method and calculation*

Following the US FDA's method [27, 28], about 0.28 mol/L $HNO_3$ solution (EMSURE® Reag. Ph Eur, ISO Merck KGaA, Darmstadt, Germany) was mixed to analyze the samples of rice. Then, all samples were heated at 95°C for 90 min. As species were separated using the HPLC separation with a mobile phase of 10 mmol/L ammonium phosphate dibasic (Sigma-Aldrich, Co., St. Louis, USA) at pH 8.25 (±0.05).

The As species were identified by using a PerkinElmer Flexar™ HPLC system coupled with a PerkinElmer NexION®350X ICP-MS (PerkinElmer, Inc., Massachusetts, USA) and Hamilton® HPLC PRP-X100 (Hamilton Company, Nevada, USA). The conditions were set up with the flow rate of 1 mL/min, RF generator frequency of 40 MHz, a power output of 1350 W, argon flow rate of 15 L/min, auxiliary of 1.2 L/min, carrier of 1 L/min and nebulizer of 0.98 L/min.

A total of 60 composites of raw rice were separated into six batches for analyzing. In each batch, there were 21 samples, including ten composite samples, one method blank, one rice flour Certified Reference Material (CRM), two replications and analyses of a sample, five calibration levels, one calibration check standard and one fortified analytical portion. The CRM used was European Reference Material ERM®-BC211 (Institute for Reference Materials and Measurements of the European Commission's Joint Research Centre, Geel, Belgium) [29].

*Quality assurance (QA)/quality control (QC)*

The largest Analytical Solution Detection Limit (ASDL) and Analytical Solution Quantitation Limit (ASQL) were identified among four measured As species with ASDL (0.1008 µg/kg), ASQL (0.7866 µg/kg), and then, the Limit of Detection (LOD) (5.0509 µg/kg) and Limit of Quantitation (LOQ) (39.4095 µg/kg) were calculated. The results of the calibration curve of As species were 0.99 in all batches. The calibration check standards in line with the mean of iAs, DMA and MMA were 7.44, 3.0 and 3.23 %, respectively. The mean of precision of replicate analytical portions was 2.04% for iAs and 2.94% for DMA while the fortified analytical portion of all batches ranged from 11.95 to 16.07% among As species. Finally, in the ERM-BC211's results, the mean of DMA levels was 123 µg/kg and the mean of iAs was 106 µg/kg.

The level of iAs was calculated by a total of As (III) and As (V) concentrations. The level of tAs was calculated by the sum of the concentrations of As (III), As (V), DMA and MMA.

*Exposure assessment and health risk assessment*

An average daily dose exposure (ADD) was estimated using Eqn (1):

$$\mathrm{ADD}_i = \frac{C_i \times \mathrm{IR}_i \times \mathrm{AF} \times \mathrm{EF} \times \mathrm{ED}_i}{\mathrm{BW}_i \times \mathrm{AT}_i} \tag{1}$$

where $Ci$ is the concentration (µg $kg^{-1}$) of As in rice, which was taken from this study; IR is the daily average consumption/ingestion of rice (g/d) (Table 2); BW represents average body weight (kg) (Table 2); EF is exposure frequency (365 d $year^{-1}$) (the US Environmental Protection Agency (U.S.EPA) [30]); ED is exposure duration (49 years from U.S.EPA [30]); AT is the averaging exposure time for noncarcinogenic effects (49 × 365 d $year^{-1}$) from U.S.EPA [30]; AF is bioavailability factor ranging from 70 to 90% from FDA [31].

The information on bodyweight and rice consumption for calculating ADD was obtained from various studies in Vietnam. Based on the National Institute of Nutrition that conducted a nutrition survey in Vietnam during 2009–2010, the rice intake in all regions and the average weight of the 2–5 age group adults aged 20–59 by gender were taken into the calculation, except for the average weight and the rice intake of other groups [32]. For children below 2 years old, their diets and consumption pattern are quite different from adults [33–35]. Thus, the risk of this age group was not assessed in this study. Overall, with the limitations of our data as well as the available evidence, the exposure dose in this study was calculated in limited age groups as detailed in Table 2.

The prediction of health risk related to iAs exposure associated with rice consumption was explained by following equations:

The Margin of Exposure (MOE) was calculated using Eqn (2)

$$MOE_i = \frac{BMDL_{05}}{ADD_i} \tag{2}$$

where ADD from Eqn (1); $BMDL_{05}$ = 3 µg/kg bw/day from JECFA [36]

## Results

### *Characteristics of rice samples in supermarkets and traditional markets*

All raw rice samples purchased in this study were ordinary rice, long-middle grain rice and white (polished) rice. Detailed information about these products was collected from the rice packaging and from interviewing the sellers in traditional markets. In supermarkets, rice samples were collected from 17 companies in Vietnam and one company from Thailand. About 97% of companies were located in the MD in the Southern region of Vietnam. In traditional markets, 91.3% of rice was collected from nonbranded sellers. About 77% of rice was from the MD and 17% was from an unknown location.

### *Level of arsenic species in rice sample*

In this study, since the process capacity analysis in the laboratory was limited, only As (III), As (V), DMA and MMA were able to be determined. The concentration of AsB was not included in the study. As (III), As (V) and DMA were detected in all 60 raw rice samples and were reported in dry-weight based (µg/kg). The mean ± SD concentration of tAs and iAs were 125.5 ± 14.7 µg/kg and 84.7 ± 7.4 µg/kg, respectively. As the MMA concentration was 98.3 % below LOD, the organic form was mainly from DMA and was found to be lower than those inorganic forms about 50%. The As (III) concentration was found to be the highest level (mean ± SD: 74.9 ± 6.2 µg/kg), followed by DMA levels (mean ± SD: 40.8 ± 9.0 µg/kg) and As (V) (mean ± SD: 9.8 ± 3.8 µg/kg). The concentration of As species in rice samples from supermarkets was significantly higher than those from traditional markets. (Table 1)

The iAs is the main component found in these samples with a mean proportion of 67.7% (ranged from 57 to 72.9%). The proportion profiles for the As species were: As (III) (60 %),

| | Concentrations in µg/kg (mean ± SD) | | | | | |
|---|---|---|---|---|---|---|
| Source | iAs | As(III) | As(V) | DMA | MMA | tAs* |
| Super market ($n$ = 30) | 88.8 ± 6.7[a] | 77.0 ± 6.2[a] | 11.7 ± 4.5[b] | 43 ± 9.6[b] | 0 ± 0 | 131.8 ± 14.1[b] |
| Traditional market ($n$ = 30) | 80.6 ± 5.6 | 72.8 ± 5.5 | 7.8 ± 1.3 | 38.5 ± 8 | 0.24 ± 1.3 | 119.3 ± 12.6 |
| Total ($n$ = 60) | 84.7 ± 7.4 | 74.9 ± 6.2 | 9.8 ± 3.8 | 40.8 ± 9 | 0.12 ± 0.92 | 125.5 ± 14.7 |

**Note(s)**: a: significance difference in mean ($p$ < 0.05) by $t$-test
b: significance difference in median ($p$ < 0.05) by Mann–Whitney $U$ test
*Calculated as the summation of As (III), As (V), DMA and MMA

**Table 1.**
As species concentration in raw rice in Ho Chi Minh City ($n$ = 60)

| Age | Gender | Wt (kg) | Rice intake (g/d) | ADD (μg/kg/d) (AF 70–90%) | MOE |
|---|---|---|---|---|---|
| 2–5 | Boy | 16.4 [37] | 204.7 [32] | 0.74–0.95 | 3.2–4.1 |
| | Girl | 15.2 [37] | | 0.80–1.02 | 3.0–3.8 |
| 20–24 | Male | 54.19 | 330.3 [32] | 0.36–0.47 | 6.4–8.3 |
| | Female | 45.98 | | 0.43–0.55 | 5.5–7.0 |
| 25–29 | Male | 55.6 | | 0.35–0.45 | 6.7–8.6 |
| | Female | 47.03 | | 0.42–0.54 | 5.6–7.1 |
| 30–34 | Male | 55.68 | | 0.35–0.45 | 6.7–8.6 |
| | Female | 48.15 | | 0.41–0.52 | 5.8–7.3 |
| 35–39 | Male | 55.73 | | 0.35–0.45 | 6.7–8.6 |
| | Female | 48.15 | | 0.41–0.52 | 5.8–7.3 |
| 40–44 | Male | 55.58 | | 0.35–0.45 | 6.7–8.6 |
| | Female | 49.66 | | 0.39–0.51 | 5.9–7.7 |
| 45–49 | Male | 55.67 | | 0.35–0.45 | 6.7–8.6 |
| | Female | 49.52 | | 0.40–0.51 | 5.9–7.5 |
| 50–54 | Male | 54.94 | | 0.36–0.46 | 6.5–8.3 |
| | Female | 50.72 | | 0.39–0.50 | 6.0–7.7 |
| 55–59 | Male | 54.42 | | 0.36–0.46 | 6.5–8.3 |
| | Female | 49.62 | | 0.40–0.51 | 5.9–7.5 |

**Note(s)**: MOE (Margin of Exposure) = $BMDL_{05}/ADD_i$ ; $BMDL_{05}$ = 3 μg/kg bw/day [36]

**Table 2.** Individual exposure dose of iAs from consuming rice by age groups and gender

DMA (32.2 %), As (V) (7.7 %) and MMA (0.1 %). The As (III) was the main species, which ranged from 47.6 to 67.6%) of tAs while there was a small proportion of MMA. (Data was not shown)

The exposure dose and health risk associated with the consumption of rice with iAs

Given the exposure factors described in Eqn (1), the daily exposure doses of iAs from consuming rice among adults ranged from 0.35 to 0.55 μg/kg bw/day. The exposure doses among females were found to be higher than those found among males. The results also showed that the ADD among children aged 2–5 was two times higher than that in adults. All exposure doses were below the $BMDL_{05}$. The MOE for children aged between 2 and 5 years was 3.0–4.1, while the range for the adult groups was 5.5–8.6 (Table 2). The MOE of children was found to be lower than that of the adult groups.

## Discussion

The results of QA and QC complied with the recommendation of the FDA's instruction. The recovery was compared to the CRMs value of iAs at 85%. This value had fallen between 80 and 110%, which was acceptable in accordance with the Association of Official Analytical Chemists Standard Method Guidance [38]. The ERM-BC211 for As species determination was used and compared to the Certified CRM [29] and other studies [35, 39–44]. In this study, tAs was calculated as the summation of As (III), As (V), DMA and MMA; thus, the tAs levels were 239 ± 8 (μg/kg). This level was lower than those of other studies that used LC-ICP-QQQ, HG-AAS, SS-HG-CT-AAS and LC-ICPMS methods [35, 39–42]. However, it is found that this level was close to those of other studies that used the same method [43, 44]. The As (III) in this study was about 104 ± 3 (μg/kg) and those in other studies that used the same method of HPLC-IPMS ranged from 104 to 106 (μg/kg). Our results were similar to other studies with respect to the level and standard deviation of iAs levels [43, 44].

The As species in rice that other studies identified were As (III), As (V), Arsenobetaine (AsB), DMA and MMA. However, AsB, which was one of the types of organic arsenic and was the main species in fish or seafood, was found to be very low and rarely detected in rice grains [22, 45–48]. Besides, it was found that there was no significant difference between the direct determination of tAs and the summation of As species. Agusa *et al.* calculated tAs in rice as the sum of As species including As (III), As (V), AsB, DMA and MMA [22]. Sofuoglu *et al.* also calculated tAs as the sum of As (III), As (V), DMA and MMA [49].

When comparing tAs in this study to other studies, the results were in line or higher than those found in Iran (120 µg/kg), Ghana (110 µg/kg), Japan (95 µg/kg), India (46–80 µg/kg), the Hunan Province of China (129 µg/kg) and Taiwan (116.6 µg/kg) [23, 50–53]. However, they were lower than those found in Turkey (202 µg/kg), Brazil (229 µg/kg), France (280 µg/kg), Spain (200 µg/kg), USA (250 µg/kg) and Finland (250 µg/kg) [9, 46, 47, 49]. The tAs concentrations in this study were lower than the Chinese and Hungarian tAs recommendation [54, 55]. However, the concentrations were in the global normal range (82–202 µg/kg) of rice grains as reported by Zavala and Duxbury [56] with the assumption of 10% moisture content [57].

Likewise, compared to a previous study of Vietnamese rice, the tAs concentrations were similar to those from previous studies in the local markets in Thailand, Ghana and Cambodia [21, 23, 24]. However, these results were lower than those from As-contaminated areas in Vietnam including An Giang province in the Mekong Delta and Ha Nam province in the Red River Delta [22, 58].

The high level of tAs in rice grown in the Mekong Delta and Ha Nam province may involve the contaminated groundwater used for irrigation. Farmers used a huge amount of groundwater for the irrigation of paddy rice fields due to the shortage of water during the dry season or even in monsoon season. This process would increase the As deposition in soil over time [5, 59, 60]. Pal *et al.* showed a significant correlation between the level of As in rice, irrigation water and soil [61]. Concerning the area contaminated with As, the As concentrations in the affected area of Vietnam were lower than those found in Bengal and India [61, 62]. It would be the influence of the amount of irrigation water and the duration of using groundwater, which was shorter than the duration in Bengal. By comparing the results of tAs in rice in markets in Ho Chi Minh City and contaminated areas in Vietnam as well as the earlier discussion, the raw rice distributed in the markets may come from not only the MD but also other areas.

As species concentrations in rice were compared between samples collected from supermarkets and traditional markets. Although the main products distributed in Ho Chi Minh city were not from the contaminated areas, this could be from various factors such as soils, types or cultivars. In addition, all of these samples were polished rice, the type of rice that has the lowest iAs level due to the removal of the bran. Also, Narukawa *et al.* noted that tAs and iAs decreased with the increase of the degree of polishing the rice [63].

The iAs was the main component in these rice samples; however, the concentrations of iAs were lower than the recommendation of WHO (200 µg/kg) [64]. Previous studies from various countries in the world as well as in this study showed that the iAs form was found to be the major part of As species in rice with the percentage ranging between 44 and 100% [9, 52, 65, 66]. The As species including As (III), As (V), DMA and MMA were also similar to those of other studies from Vietnam [21, 22] as well as other Asian countries [9, 21, 50, 53]. Besides, there was a wide variation of As species levels in rice since the As levels in soils and other factors could result in large coefficients in As species results [53, 67]. In this study, due to the sample preparation as a composite and replication strategy, the variations would be reduced.

The daily exposure doses of iAs from rice consumption were similar to those of a study in Pakistan (0.3 µg/kg bw/day), Finland (0.38–0.46 µg/kg bw/day), EU (0.22 µg/kg bw/day), Spain (0.41 µg/kg bw/day), Thailand (0.37 µg/kg bw/day) and China (0.4 µg/kg bw/day) [21, 47, 68–71]. However, the results were lower than those found in other studies in Vietnam and other countries [22, 58, 72–74], as well as being higher than those from USA (0.02 µg/kg bw/day), Turkey (0.1 µg/kg bw/day) and Iran (0.09 µg/kg bw/day) [49, 51, 75].

The difference in exposure dose among gender could be explained by a difference in body weight. Other studies showed that males consumed a higher rice intake than females in Asian countries such as Singapore, Korea, China and Cambodia [76–79]. Besides, with similar food consumption patterns among Asian countries [80, 81], the assumption of similar rice intake by gender in Vietnam could be applied. With this scenario, either the overestimate or the lower estimate for the ADD of both genders is likely to happen. A study in Taiwan that focused on the iAs risk assessment from rice consumption showed that women were exposed to iAs with a lower ADD than men [50].

The finding indicated that higher ADD among children aged 2–5 years compared with adults was similar to other countries such as Finland, Taiwan, Sweden and the USA [82]. In addition, the EU's study about As contamination in food stated that the ADD of iAs reduced with an increase in age [82]. Compared to other countries, ADD in children aged 2–5 from rice consumption in this study was higher than those in Sweden in those aged 4 years (0.185 μg/kgd) [83], in USA in those aged 0–6 (0.543 μg/kgd) [31] and Finland among the 1–6 age group (0.57–0.67 μg/kgd) [47].

The results of the exposure dose of iAs through rice consumption varied across age groups, gender and countries. This could be explained by the difference in rice intake and body weight. With the same mean of iAs level in rice, although females consumed more iAs than males in this study, the females could get a lower level of ADD than males due to their lower amount of rice intake. The children consumed higher ADD than adults due to the higher ratio of rice intake/bodyweight. From the perspective of countries, the iAs level in this study was lower than or equal to other countries but the exposure dose was relatively equal to or higher than those populations due to high rice intake per capita among the population [21, 47, 49, 51, 68–71, 75].

The results of the MOE are also similar to other studies [47, 84, 85] and were found to be lower than that in a study in Belgium where people consumed less rice and followed different consumption patterns [86]. At present, there are no international guidelines for MOE criteria. However, if the MOE is higher, then the public health concern is lower. In a worst-case scenario, consumption of rice contaminated with iAs, which is higher than $BMDL_{05}$, may lead to a 0.5% increased incidence of lung and bladder cancers.

Besides, studies about iAs in food showed that although rice is the major food of iAs exposure, other food also contributed the amount of iAs into humans [8,69–71, 85]. Thus, the actual risk of iAs intake from all food might be higher in this population. This study focused on the health risk of iAs from rice consumption only; thus, lower estimation is likely to occur. Further study on inorganic As in other food items should be considered for characterizing the significant sources of exposure.

The limitation of this study is that the origin of the rice cannot be traced back due to the complicated production system. This does not reflect the variation of geography as well as the cultivar and the farm. Another uncertainty is the lack of rice consumption level specific to the study area. The data related to rice intake including age and gender groups were taken from the National Survey and other studies. Another factor is the bioavailability of As with a wide range of 55–71% [87] and 70–90% [88]. With respect to the worst-scenario approach of risk assessment, the range of 70–90% of bioavailability was used for assessing the risk. The overestimated risk would be generated.

## Conclusions

The results of this study show that most people in HCM City consumed rice with the mean of iAs in raw rice 84.7 μg/kg in dry weight. The As speciation results indicate that an iAs form in rice and their distribution were similar to other Asian rice profiles. Although the iAs concentrations found in rice grains being sold in the local market in HCM City are likely to be safe for consumers according to WHO recommendation, based on JECFA's criteria for chronic and cancer risk, the health risk could not be excluded. A risk communication of iAs in rice needs to be raised for public health awareness about the high exposure from rice consumption and chronic iAs disease. People should change their routine preparation and rice cooking methods. More importantly, people should promptly view the new information on how to reduce contamination, for instance, following appropriate washing procedures, choosing the rice already approved by the health sectors, avoiding using arsenic-contaminated water for irrigation or having a balanced and varied diet.

A monitoring program for iAs contamination in other food items should be considered. Although the use of groundwater is occasional, the risk of accumulated iAs in rice and foods grown in contaminated areas would occur over a long-term period, especially among young children. Besides, the Mekong delta, although an important region for rice production in Vietnam, is thought to be an As-contaminated area. Thus, a monitoring system for As in rice, the original production of rice, As in groundwater as well as As in soil over time should be established. In addition, rice consumption should be investigated for specific regions, gender and age groups for a more accurate risk assessment. Finally, further study on dietary intake of iAs should be carried out as the overall risk of iAs from foods in HCM City would benefit from further research.

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