Journal of Hazardous Materials 262 (2013) 1064–1071

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

# Arsenic contamination in the food chain and its risk assessment of populations residing in the Mekong River basin of Cambodia

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## HIGHLIGHTS

- We examined the arsenic exposure from daily food consumption in Cambodia.
- Paddy soil, rice, vegetable and fish were analyzed for total arsenic ($[As]_{tot}$).
- A positive correlation of the $[As]_{tot}$ in paddy soil and paddy rice was found.
- Daily dose of inorganic arsenic ($[As]_i$) from daily food consumption was estimated.
- Kandal residents are at risk of $[As]_i$ intake from their daily food consumption.

## ARTICLE INFO

*Article history:*
Received 7 December 2011
Received in revised form 24 April 2012
Accepted 2 July 2012
Available online 7 July 2012

*Keywords:*
Arsenic
Risk assessment
Daily intake
Foodstuffs
Cambodia

## ABSTRACT

In the present study, we investigated the potential arsenic exposure of Cambodian residents from their daily food consumption. Environmental and ecological samples such as paddy soils, paddy rice (unhusked), staple rice (uncooked and cooked), fish and vegetables were collected from Kandal, Kratie and Kampong Cham provinces in the Mekong River basin of Cambodia. After acid-digestion, digestates were chemically analyzed by inductively coupled plasma mass spectrometry. Results revealed that the means of total arsenic concentration ($[As]_{tot}$) in paddy soils and paddy rice from Kandal were significantly higher than those from Kampong Cham province ($t$-test, $p < 0.05$). Moreover, a significant positive correlation between the $[As]_{tot}$ in paddy soils and paddy rice was found ($r(14) = 0.826$, $p < 0.01$). Calculations of arsenic intake from food consumption indicated that the upper end of the range of the daily dose of inorganic arsenic for Kandal residents (0.089–8.386 $\mu g\, d^{-1}\, kg^{-1}{}_{body\ wt.}$) was greater than the lower limits on the benchmark dose for a 0.5% increased incidence of lung cancer ($BMDL_{0.5}$ is equal to 3.0 $\mu g\, d^{-1}\, kg^{-1}{}_{body\ wt.}$). The present study suggests that the residents in Kandal are at risk of arsenic intake from their daily food consumption. However, the residents in Kratie and Kampong Cham provinces are less likely to be exposed to arsenic through their daily dietary intake. To the best of our knowledge, this is the first report estimating the daily intake and daily dose of inorganic arsenic from food consumption in the Mekong River basin of Cambodia.



## 1. Introduction

Chronic exposure to naturally occurring arsenic-rich groundwater has generally threatened and impaired the well-being of millions of people in South and Southeast Asia. For instance, it is estimated that about 40 million people suffer from arsenic through the groundwater drinking pathway in India and Bangladesh [1]. Similarly, high concentrations of arsenic are also reported in Vietnam [2–4], Lao PDR [5,6] and Cambodia [7–12]. Toxicological studies have shown that chronic exposure to high arsenic concentrations in drinking water has led to dermatological manifestations (raindrop pigmentation, melanosis and hyperkeratosis) and skin cancer [13]. The outcomes of acute arsenic toxicity might include gastrointestinal discomfort, abdominal pain, vomiting, diarrhea, bloody urine, shock, coma and death [14,15]. A dose–response criteria such as no observable adverse effect level (NOAEL), lowest

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http://dx.doi.org/10.1016/j.jhazmat.2012.07.005

observable adverse effect level (LOAEL), and benchmark dose (lowest effect dose for 5% or 1% effect level, based on daily quality, expressed as $LED_{05}$ or $LED_{01}$) have also been observed [15]. One-hundred-twenty milligrams of arsenic trioxide is reported to be the most common minimum lethal dose of humans; however, it can vary from 70 to 180 mg (1–3 mg As $kg^{-1}$) [15]. Likewise, epidemiological studies indicate that ingested and inhaled arsenic can cause skin cancer and lung cancer, respectively. In addition, a number of studies have also shown that arsenic ingestion leads to internal cancer and cancer of the urinary bladder [15]. As a consequence, many approaches have been proposed to assess arsenic risks from drinking water because it has been identified and characterized as a toxin and carcinogen to humans and animals.

Our cross-sectional health risk assessment of inorganic arsenic intake of people residing in the Mekong River basin of Cambodia revealed that 98.65% of residents in the Kandal province study area were confronted with arsenic toxicity. Moreover, cancer risk index was found to average 5 in 1000 exposure [16]. Concurrently, individual variations in arsenic accumulation in human body were observed among the exposed populations while plotting arsenic content in scalp hair, fingernail and toenail with average daily dose (ADD) of arsenic, which was calculated through the groundwater drinking pathway using USEPA health risk assessment models [17]. These variations were explained by several factors. An assumption of high intake of arsenic was that people might ingest an additional amount of arsenic through their daily food consumptions while they were consuming arsenic-rich groundwater [16]. In fact, rice is a dietary staple of Cambodian people, and it is normally eaten three times per day. It was believed that rice grown in the arsenic-contaminated soils had greater arsenic concentration in the grain than that grown the arsenic-free soils. Pot experiments indicate that about 81% of the recovered arsenic in rice is found as inorganic arsenic species [18]. However, a gastrointestinal digestion simulation study revealed that arsenate bioavailability in cooked rice ranged from 63 to 99% [19]. Moreover, arsenic bioavailability in rice is highly dependent on its species in rice, irrigation water and cooking water [20].

Cambodia is an agriculture-based country, and most Cambodians live in rural areas alongside watersheds. They totally relied on their extensive farming methods, rain-fed flooding of paddies, for their rice productions in the past. However, some households in the Mekong River basin have recently gained access to shallow groundwater through the inexpensive and easily drilled boreholes. In addition to using this secured source of water for daily drinking water, some households have changed their traditional farming to an intensive one using shallow groundwater irrigation systems. However, a number of studies have reported that shallow groundwater in the Mekong River basin of Cambodia is highly contaminated with arsenic and other toxic trace elements [7,16,21,22]. The irrigation of paddy fields with arsenic-rich shallow groundwater may lead to accumulation of arsenic in paddy soils and potentially affect the production yield and rice quality. Although the mechanisms of arsenic transfer from arsenic-contaminated irrigation water to paddy soils and the transfer from arsenic-contaminated paddy soils to rice remained unclear [23], the study of arsenic contamination in the food chain may provide additional evidence and explanations for the variations in individual arsenic accumulations. Therefore, the objectives of the present study were to (1) investigate arsenic distributions and correlations between paddy soils and paddy rice in the Mekong River basin of Cambodia, (2) determine and compare a distribution of the total arsenic concentration ($[As]_{tot}$) among the three staple foodstuffs, namely rice, vegetable and fish, and (3) estimate the daily intake and daily dose of inorganic arsenic from daily food consumption of arsenic exposed populations.

## 2. Materials and methods

### 2.1. Study area

The design of the present project was a cross-sectional study. Sampling was carried out in three provinces in the Mekong River basin of Cambodia. Kratie (Sambok village, Sambok commune, Kracheh district) and Kampong Cham (Veal Sbov village, Ampil commune, Kampong Siem district) are located along the Mekong River upstream of Phnom Penh whereas Kandal (Preak Russey village, Kampong Kong commune, Koh Thom district) is located between the Mekong River and Bassac River, downstream of Phnom Penh (Fig. 1).

### 2.2. Field sampling and sample preparation

Sampling was conducted twice for the present study. The first batch of sampling was carried out in February 2009, when paddy soils and paddy rice were collected from households where groundwater was used to irrigate their paddy fields in Kandal (paddy soils $n = 8$; paddy rice $n = 8$) and Kampong Cham (paddy soils $n = 8$; paddy rice $n = 8$). Paddy soils and paddy rice were separately packed in paper bags in the field and transported to a laboratory in the Department of Chemistry, Royal University of Phnom Penh, Cambodia. Paddy soil samples were dried in the open air under diffused sunlight at room temperature for 48 h, and sieved (10 mesh) to separate debris. Paddy rice was manually ground with mortar and winnowed to separate the grain from its husk. The grain was then ground to fine powder. The second batch of sampling was carried out in March 2011, when staple rice (uncooked rice $n = 10$; cooked rice $n = 10$) and five types of popular fruit vegetables (cucumber $n = 3$; gourd $n = 3$; papaya $n = 3$; pumpkin $n = 3$; tomato $n = 3$) were collected from the visited households and family gardens, respectively. Concurrently, two kinds of popular fish (snakehead fish $n = 5$; catfish $n = 5$) were collected from the natural habitats where people usually fish in the study areas of Kampong Cham and Kandal provinces. In addition, sampling was extended to Kratie (uncooked rice $n = 10$; cooked rice $n = 10$; snakehead fish $n = 5$; catfish $n = 5$; gourd $n = 3$; papaya $n = 3$; pumpkin $n = 3$), which is known as a moderately arsenic-contaminated area [16]. However, cucumber and tomato were not available over there at the time of sampling. All rice, fruit vegetable and fish samples were kept separately in labeled plastic ziplock bags, placed in a cold box in the field, and transferred into a refrigerator where they were stored at 4 °C until further treatment. After thawing at room temperature for several hours, fruit vegetables were peeled and cut into small pieces with a quartz knife. Fish was dissected; viscera were removed and only the edible parts were taken. All rice, vegetable and fish samples were washed with deionized water and dried in the open air under diffused sunlight for 24 h; next, samples were oven-dried at 50 °C for several days to achieve complete dryness, and they were subsequently manually ground to a fine powder with a mortar and passed through 35-mesh sieve. Each sample was repacked in a labeled plastic ziplock bag and shipped to GIST, Korea for chemical analyses.

### 2.3. Sample analyses

Paddy soil samples were digested using a modified Aqua Regia method. Briefly, 0.50 g of the dried paddy soil was accurately weighed into a 15 mL polyethylene tube. Exactly 3.75 mL of HCl (36%) and 1.25 mL of $HNO_3$ (65%) were added and the mixture was allowed to stand overnight. In the next morning, the mixture was heated at $96 \pm 3$ °C for an hour using a heating block. After cooling down at room temperature, 5.00 mL of deionized water was added, after which it was centrifuged at $1824 \times g$-force (3000 rpm, rotor