

## Arsenic Pollution in Groundwater of Vietnam and Cambodia : A Review

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### Abstract

Recently, As pollution was reported in groundwater from the Red River delta of Northern Vietnam and the Mekong delta of Southern Vietnam and Cambodia. Although the health of about 10 million people is at risk from the drinking tube well water, little information is available on the health effects of As exposure in the residents of these regions. Also, the countrywide survey on regional distribution of As pollution has not been conducted in these countries. At present, as far as we know, symptoms of chronic As exposure have not yet been reported, probably due to the relative short-term usage of the tube wells in the regions. However, oxidative DNA damage was observed in the residents of Cambodia and so further continuous usage of the tube well might cause severe damage to the health of the residents. In this article, we review literature concerning As pollution of groundwater and its health effects on residents in Vietnam and Cambodia. The mechanisms of As release to the groundwater is also discussed.

**Keywords :** Arsenic, groundwater, human hair, human urine, Vietnam, Cambodia

### 1. Introduction

Consumption of As-polluted groundwater has ad-

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versely affected human health in some areas of the world [1-3]. Particularly, groundwater pollution by naturally occurring As in the Ganges delta poses a significant health risk to about 36 million people who depend on the water [2]. It is known that As exposure causes lung and skin cancer, and also birth defects [4]. In Bangladesh and West Bengal, India, skin, lung, kidney, bladder and liver cancers, and skin lesions appears to be caused by chronic exposure to As through the drinking water [5]. A significant dose-response relationship was also observed between As level in well water and mortality from cancers of bladder, kidney, skin, and lung in residents of southwestern coast of Taiwan [6].

Whereas these areas have been studied comprehen-

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sively for As pollution of groundwater, the human exposure and its epidemiology, there has been little information for the Southeast Asia. Berg et al. [7] and Agusa et al. [8-10] reported the pollution of As in groundwater of the Red River delta in Vietnam. They found elevated As concentrations (up to 3050  $\mu\text{g/l}$ ) in the groundwater samples, some of which contained As over the WHO drinking water guideline (10  $\mu\text{g/l}$ ). Recently, Trang et al. [11] has also reported As pollution in groundwater of Mekong River delta in Southern Vietnam, ranging from < 1 to 850  $\mu\text{g/l}$ . Furthermore, some reports showed that groundwater is polluted also by geogenic As in Cambodia [12-15]. Berg et al. [15] estimated that 10 million people in the Red River delta and 0.5-1 million people in the Mekong delta are at risk of chronic As poisoning. However, very little information is available on the status of As exposure and its related health effects in the residents of Vietnam and Cambodia. This article reviews As pollution in groundwater, its exposure to humans, its potential toxicological risk, and mechanism of the As pollution in groundwater of Cambodia and Vietnam.

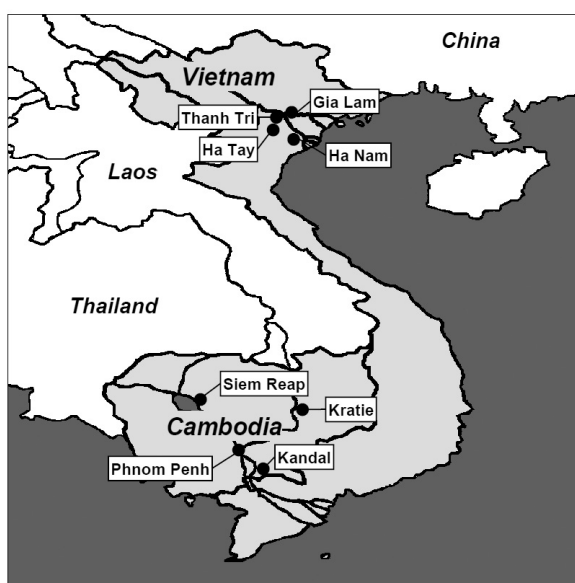
## 2. Arsenic pollution in Vietnam

### 2-1. Arsenic concentration in groundwater

In Vietnam, As pollution of groundwater has been reported for two areas, the Red River delta in the Northern Vietnam and the Mekong River delta in the Southern Vietnam (Fig. 1). The As pollution was first reported for the Red River delta in the Northern Vietnam by Berg et al. [7]. We also collected the groundwater samples from the Gia Lam, Thanh Tri, Ha Nam, and Ha Tay in the Red

River delta (Fig. 1) during 2000-2004 and measured As concentration (Table 1 ; Ref. 9, 10). Concentrations of As in our groundwater samples ranged from < 0.1 - 486  $\mu\text{g/l}$  ( $n = 48$ ). High levels were observed in the Ha Nam (median, 256  $\mu\text{g/l}$  ; max, 486  $\mu\text{g/l}$ ) and Ha Tay (median, 194  $\mu\text{g/l}$  ; max, 344  $\mu\text{g/l}$ ). 36 %, 43 %, 90 % and 100 % of the groundwater samples from the Gia Lam, Thanh Tri, Ha Nam, and Ha Tay, respectively, exceeded the WHO drinking water guideline of 10  $\mu\text{g/l}$  [4]. Much higher concentrations of As were reported for the groundwater of the Gia Lam (2 - 3050  $\mu\text{g/l}$ ) and Thanh Tri (9 - 3010  $\mu\text{g/l}$ ) in the Red River delta [7], although these areas are close to our sampling points. It is well known that As concentrations in groundwater are extremely heterogeneous over small spatial scales in Bangladesh [16] and in the Northern Vietnam [15]. The spatial pattern of groundwater flow path might have cause the difference [16]. Alternatively, the spatial heterogeneity might reflect the rapid change in As level with depth, because the wells would draw water from different depths even though they are close to each other [17]. McArthur et al. [18] reported high stratification gradient of As concentration in groundwater of a well : As level changed from 419  $\mu\text{g/l}$  at 38.3 m to 7  $\mu\text{g/l}$  at 42.8 m in the West Bengal.

Recently, As pollution of groundwater was also reported for the Mekong River delta in the Southern Vietnam [11, 13]. The As concentration ranged from < 1 to 850  $\mu\text{g/l}$  (Table 1). Although As concentrations as high as 850  $\mu\text{g/l}$  were observed in some wells, the mean concentration (39  $\mu\text{g/l}$  ; Table 1) was relatively low. In these samples, dissolved Mn concentration was reported to be high (mean, 3400  $\mu\text{g/l}$  ; range, < 10 - 34000  $\mu\text{g/l}$  ; Ref. 11) compared to those of the Northern Vietnam. According to Agusa et al. [10], the mean Mn level was 1374  $\mu\text{g/l}$  (range, 2.67 - 5530  $\mu\text{g/l}$ ). Trang et al. [11] also reported that mean Mn levels of 800  $\mu\text{g/l}$ , ranging from < 10 - 2800  $\mu\text{g/l}$  in the Red River delta, the Northern Vietnam. Hence, the reduction of Fe oxyhydroxides may not be enough to release much amount of As to the groundwater in the Southern Vietnam region examined by Trang et al. [11], as discussed later. It was also found that the As levels tend to be higher within a distance of < 10 km from the river (mean, 64  $\mu\text{g/l}$ ), whereas the levels are much lower at a farther distance (mean, 8  $\mu\text{g/l}$ ) [15]. This result might reflect the differences in redox condition of the sediment in these areas because As would be released via reductive dissolution of Fe oxyhydroxides.



**Fig. 1** Map showing sampling locations in Cambodia and Vietnam.

### 2-2. Arsenic concentration in human hair

Human hair may be a useful non-invasive indicator of chronic As exposure because it is easy to collect, transport and preserve [19]. Thus, we analyzed the hair samples collected from the Red River delta in the Northern Vietnam where groundwater was polluted by As. Arsenic concentrations in hair of the residents from the Northern Vietnam ( $n = 59$ ) were 0.09 - 2.77  $\mu\text{g/g}$  dry wt (Table 2). The levels were comparable to or lower than those from other As-polluted areas such as the Bangladesh and West Bengal [20, 21], but were higher than those of residents in non-polluted areas [22]. The As level of 18 % of hair samples from the Thanh Tri exceeded the level of possi-

ble indication of the As toxicity (1  $\mu\text{g/g}$  dry wt ; Ref. 22), although all the values in the Gia Lam were below this threshold level. Similar results were also reported by Berg et al. [15], in which hair As ranged from 0.20 to 2.75  $\mu\text{g/g}$  dry wt in resident of the Northern Vietnam. These results might suggest the potential effects of As on the health of these residents in Northern Vietnam.

A significant positive correlation ( $r = 0.57$ ,  $p < 0.001$ ) between As concentrations in groundwater and hair of residents in the Gia Lam and Thanh Tri, Northern Vietnam was found (Fig. 2). Also, concentrations of As in human hair were positively correlated with cumulative

**Table 1** Concentrations ( $\mu\text{g/l}$ ) of As in groundwater from Vietnam and Cambodia.

Location/ Remark	<i>n</i>	Mean	Min	Max	Median	References
<b>Vietnam</b>						
Red River Delta						
Gia Lam	11	10.8	<0.1	38.2	5.0	[10]
Thanh Tri	14	44.0	<0.1	330	1.5	[10]
Ha Nam	10	209	3.0	486	256	[9]
Ha Tay	13	209	132	344	194	[9]
Dong Anh	48	31	< 1	220		[7]
Gia Lam	55	127	2	3050		[7]
Thanh Tri	45	432	9	3010		[7]
Tu Liem	48	67	1	230		[7]
Luong Yen	6	22.8				[64]
Yen Phu	7	40.5				[64]
Mai Dich	3	1.1				[64]
Ngoc Ha	3	1.6				[64]
Ngoc Si Lien	3	1.4				[64]
Phap Van	5	67.3				[64]
Tuong Mai	4	44.5				[64]
Ha Dinh	5	92.6				[64]
Red River Delta	83	140	1.3	460		[11]
Mekong River Delta	111	39	< 1	850		[11]
<b>Cambodia</b>						
Kandal	8	307	54	720	180	[36]
Kratie	58	88	<1	886	8	[14, 36]
Phnom Penh	2	2	2	3	2	[36]
Countrywide data grouped by regional geology						
Holocene sediments (near Mekong and Bassac)	401	210		1700		[12]
Holocene sediments (other)	346	9.8		390		[12]
Quaternary sediments	217	4.0		270		[12]
Pliocene volcanics	27	0.8		8.7		[12]
Neogene-Quaternary sediments	13	0.9		7.9		[12]
Older units	68	3.4		38		[12]

**Table 2** Concentrations ( $\mu\text{g/g}$  dry wt) of As in hair of residents from Vietnam and Cambodia.

Location	<i>n</i>	Mean (Range)	Median	References
<b>Vietnam</b>				
Red River Delta				
Gia Lam	20	0.40 (0.12 - 0.75)	0.38	[10]
Thanh Tri	39	0.62 (0.09 - 2.77)	0.42	[10]
<b>Cambodia</b>				
Kandal	19	0.26 (0.12 - 1.22)	0.16	[36]
Kandal	11	2.0 (max. 6.5)		[15]
Provinces bordering Kandal	8	0.3		[15]
Kratie	61	2.58 (0.05 - 45.7)	0.79	[14, 36]
Phnom Penh	16	0.15 (0.06 - 0.39)	0.14	[36]

As exposures that were estimated by concentration of As, consumption of groundwater, and record of usage of groundwater [10]. These results imply that residents in the areas have been chronically exposed to As through the consumption of groundwater.

For the Mekong River delta in the Southern Vietnam, hair As level of 0.11 - 2.92  $\mu\text{g/g}$  dry wt was reported [15]. The hair As level in the residents who use groundwater with As level of  $> 50 \mu\text{g/l}$  was significantly higher than those using groundwater with the level of  $< 50 \mu\text{g/l}$  [15], indicating As exposure from the polluted groundwater in this region.

### 2-3. Arsenic concentration in human urine

Generally, ingested inorganic As is methylated to monomethyl and dimethyl arsenicals and is excreted through urine in humans [23]. Therefore, As speciation is important for assessing exposure and metabolic capacity

of As in humans. We measured As species including arsenite (As[III]), arsenate (As[V]), monomethylarsonic acid (MMA[V]), dimethylarsinic acid (DMA[V]) and arsenobetaine (AB) in the urine of residents from the Ha Nam and Ha Tay in the Northern Vietnam using a high performance liquid chromatograph - inductively coupled plasma - mass spectrometer (HPLC-ICP-MS) [9].

Concentrations of As species in human urine from the Northern Vietnam are shown in Table 3. Sum of the concentration of As species (S-As ; AB + DMA[V] + MMA[V] + As[III] + As[V]) were 31 - 179  $\mu\text{g/g}$  creatinine. Urinary As composition was similar between the residents of the Ha Nam and Ha Tay in the Northern Vietnam, with DMA [V] being dominant (mean, 63 %). Arsenobetaine (AB), which may be derived from consumption of seafood, was also detected in the urine of residents from the Northern Vietnam (mean, 15 % ; Table 3).

There was a significant positive correlation ( $p < 0.01$ )

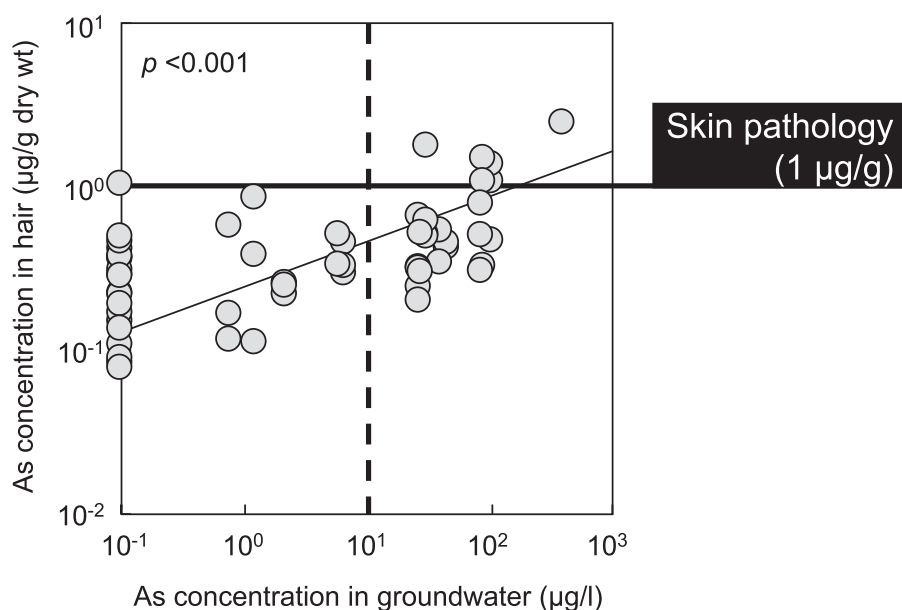
**Table 3** Concentrations ( $\mu\text{g/g}$  creatinine) of As species in urine of residents from Vietnam and Cambodia.

Location	n	DMA		MMA		As[III]		As[V]		As[III] + As[V]		AB		S-As <sup>a</sup>		References
		Mean (Range)	Median	Mean (Range)	Median	Mean (Range)	Median	Mean (Range)	Median	Mean (Range)	Median	Mean (Range)	Median	Mean (Range)	Median	
Vietnam																
Red River Delta																
Ha Nam	56	39 (19 - 64)	35	6 (1 - 18)	4	5 (<1 - 16)	5	3 (<1 - 12)	3	11 (<1 - 58)	10	10 (<1 - 71)	8	63 (31 - 124)	55	[9]
Ha Tay	41	38 (18 - 86)	39	5 (<1 - 20)	5	6 (<1 - 23)	5	5 (<1 - 35)	<1	8 (<1 - 27)	6	9 (<1 - 33)	6	64 (36 - 179)	64	[9]
Cambodia																
Kandal	27	NA <sup>b</sup>	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	62.8 (< 0.5 - 164) <sup>c</sup>	53.4	[36]
Kratie	61	137 (24.5—959)		28.0 (2.2—170)		ND <sup>d</sup>		ND		35.4 (2.9—299)		ND		201 (40.9—1120)		[14]
Kratie	10	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	141 (50.0 - 490) <sup>e</sup>	81.4	[36]
Phnom Penh	10	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	68.0 (10.7 - 123) <sup>f</sup>	73.6	[36]

a: S-As means sum of As species detected.

b: not available.

c: Total As concentration ( $\mu\text{g/l}$ ).



**Fig. 2** Relationship between As concentrations in groundwater and hair of residents from Gia Lam and Thanh Tri, Vietnam [10]. Vertical dashed line indicates the WHO guideline value for drinking water [4]. Samples with As concentrations below the detection limit were plotted as half the value of detection limit.

between concentrations of As in groundwater and S-As in urine for the residents of the Ha Nam and Ha Tay in the Northern Vietnam. Moreover, each As species in urine including DMA[V] ( $r = 0.274$ ,  $p < 0.01$ ), As[III] ( $r = 0.272$ ,  $p < 0.01$ ) and As[V] ( $r = 0.399$ ,  $p = 0.001$ ) also showed positive correlations with As concentrations in groundwater (Fig. 3). In contrast, there was no significant correlation between urinary AB and groundwater As level (Fig. 3). These results suggest that DMA[V], As[III] and As[V] were mainly derived from the groundwater, whereas AB was taken up from the sources other than groundwater (probably from marine fishes) in these residents.

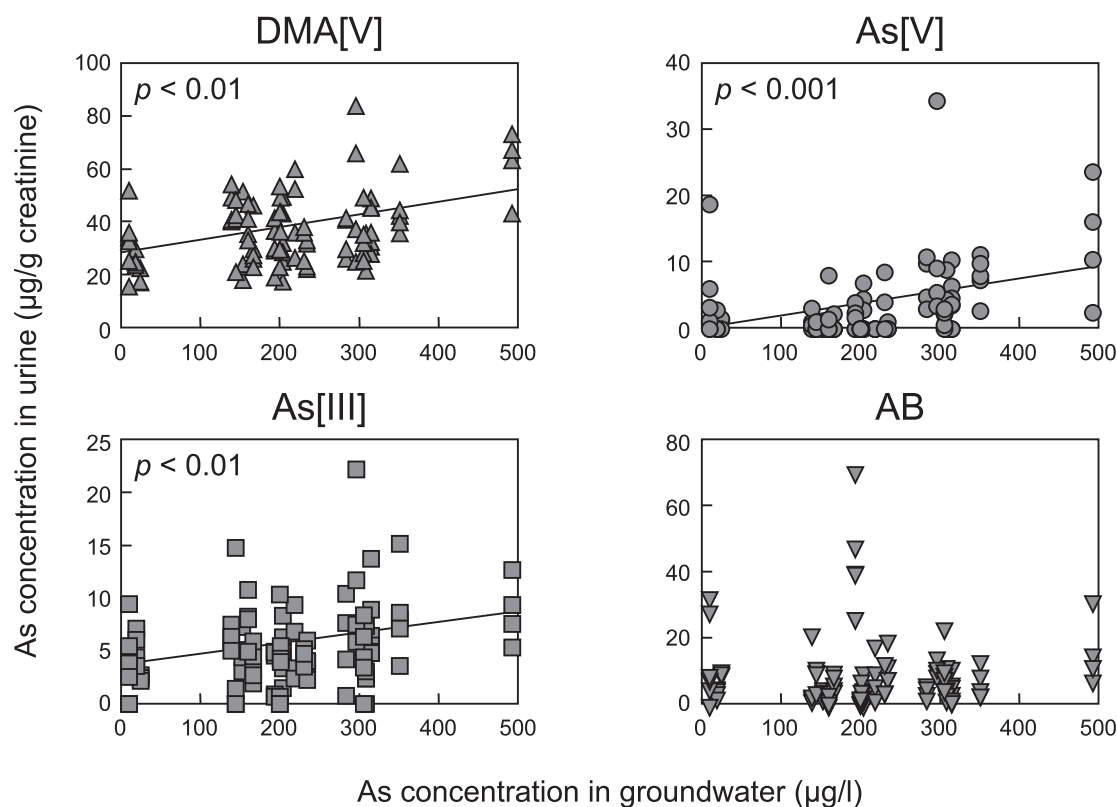
As far as we know, urinary arsenic levels and speciation has not been reported for the residents of the Mekong River delta in the Southern Vietnam.

#### 2-4. Multiple exposure to trace elements in humans from groundwater

Generally, concentrations of Fe and Mn in the groundwater showed the highest values, followed by alkaline earth metals such as Sr and Ba in the Ha Nam and Ha Tay in the Northern Vietnam [10]. In contrast, concentrations of Ag, In, Sn, Sb, Cs, Hg, Tl and Bi were very low

or below the detection limit [10]. Remarkably, concentrations of Mn and Ba in some groundwater samples of the Northern Vietnam exceeded the WHO drinking water guidelines (Mn,  $400 \mu\text{g/l}$ ; Ba,  $700 \mu\text{g/l}$ ; Ref. 4) (Fig. 4). Thus, it is likely that the residents are exposed not only to As but also to Mn and Ba from the groundwater in the Northern Vietnam. Elevated Mn concentrations were also observed in the Mekong River delta in the Southern Vietnam ( $< 10 - 34000 \mu\text{g/l}$ ; Ref. 11), but the data on other elements and the human health effects by Mn exposure have not been reported.

The source of Mn and Ba seems to be geogenic, similar to the case of As. In the groundwater of Northern Vietnam, Mn concentration tends to decrease with Fe levels [18]. Similar relationship was also reported for other As-contaminated areas such as Bangladesh and West Bengal, India [18]. This may reflect the successive reduction processes in the sediments, as proposed by Takai and Kamura [24]. It is generally believed that As is released to groundwater by microbial reduction of As-containing Fe oxyhydroxides in sediments. For such a redox process, reductive dissolution of Mn oxides would occur before the reduction of Fe oxyhydroxides [24-26]. Hence, high Mn levels in groundwater might suggest that reductive



**Fig. 3** Relationship between concentrations of As species in groundwater and urine of residents from Ha Nam and Ha Tay, Vietnam [9]. Samples with concentrations of As species below the detection limit were plotted as half the value of detection limit.

dissolution of Fe oxyhydroxides is not complete yet, and then dissolved As is still low [18]. These results might also suggest that groundwater with low As level contains high Mn level and the groundwater having low Mn level shows high As in these areas. Indeed, in our Northern Vietnam samples examined, the groundwater with low As levels ( $< 10 \mu\text{g/l}$ ) showed higher Mn levels (mean,  $1602 \mu\text{g/l}$ ) than those with high As levels (As,  $> 10 \mu\text{g/l}$ ; mean Mn,  $1025 \mu\text{g/l}$ ), and the groundwater having low Mn levels ( $< 400 \mu\text{g/l}$ ) had higher As levels (mean,  $59 \mu\text{g/l}$ ) than those with high Mn levels (Mn,  $> 400 \mu\text{g/l}$ ; mean As,  $20 \mu\text{g/l}$ ). This relationship should be considered for the supply of drinking water in this region.

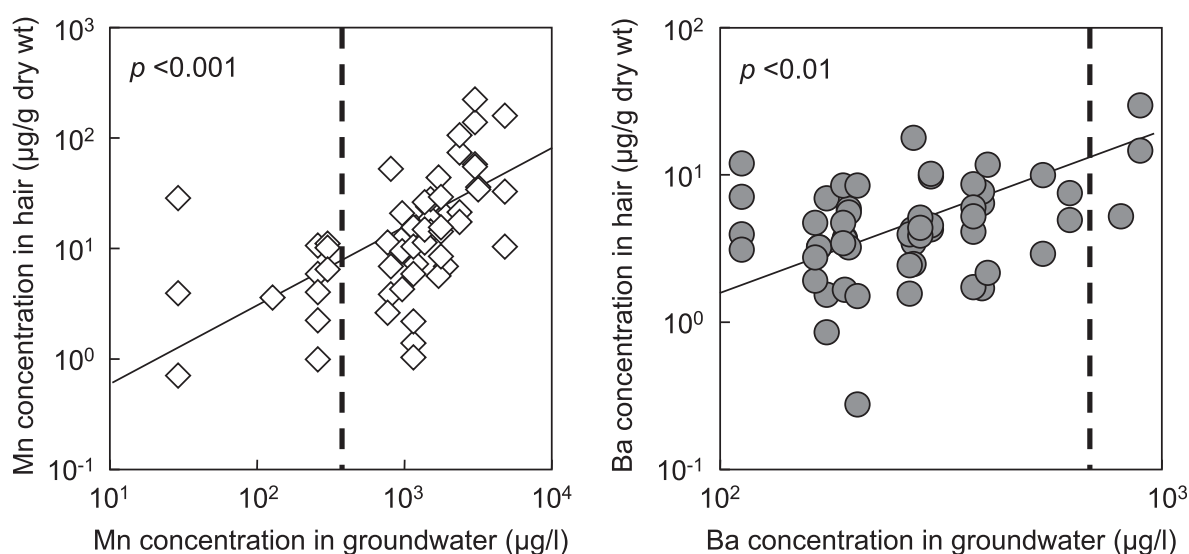
High Ba concentrations in the groundwater of the Northern Vietnam (Fig. 4) may be due to the dissolution of carbonates, as suggested for the groundwater of Bangladesh [27]. Calcium, derived from the carbonates, exhibits similar distribution pattern with As in the groundwater of Bangladesh [28]. In our groundwater samples, Ba showed a significant positive correlation with As [10], although Ca was not determined. This can be attributed to the fact that all these elements are released to groundwater by redox processes. The redox condition of the sediments is caused mainly by the microbial activity using the organic matter. The  $\text{CO}_2$  produced by the microbial respiration is known to induce dissolution of carbonates [29]. Under such a redox condition, As-containing Fe oxyhydroxides is also dissolved.

To evaluate multiple exposure by trace elements, concentrations of trace elements (V, Cr, Mn, Co, Cu, Zn, Rb,

Sr, Mo, Ag, Cd, In, Sn, Sb, Cs, Ba, Tl, Hg, Pb and Bi) other than As were also determined in human hair from the Gia Lam and Thanh Tri, the Northern Vietnam. Hair concentrations of V (median,  $0.119 \mu\text{g/g}$  dry wt) and Mn (median,  $16.7 \mu\text{g/g}$  dry wt) in the residents of the Gia Lam and Thanh Tri were relatively high in comparison with those of V ( $0.029 \mu\text{g/g}$  dry wt) and Mn ( $0.42 \mu\text{g/g}$  dry wt) in Japanese [30]. Also, Ba concentrations in hair of both Gia Lam and Thanh Tri ( $0.3 - 34 \mu\text{g/g}$  dry wt) were higher than the levels of general population from non-polluted areas ( $0.6 - 5.6 \mu\text{g/g}$  dry wt; Ref. 31).

The high hair concentrations of Mn and Ba in the residents of the Gia Lam and Thanh Tri were consistent with the results observed for the groundwater. Also, concentrations of Mn ( $p < 0.001$ ), Co ( $p < 0.001$ ), Ba ( $p < 0.05$ ), Mo ( $p < 0.05$ ) and Sr ( $p < 0.05$ ) in human hair were positively correlated with those in groundwater collected from the wells that had been utilized by the residents whose hair samples were collected [10]. Thus, this relationship strongly indicates that consumption of the groundwater is a main source of these elements for the residents.

While Mn is known as an essential element, teratogenicity and neurotoxic effects are caused at the high dose [32]. Abnormal neurological scores were observed in older persons ( $n = 77$ ) who had consumed Mn-polluted drinking water (range,  $1800 - 2300 \mu\text{g/l}$ ) in northwest Peloponnesos, Greece, whose mean Mn concentration in hair was  $11 \mu\text{g/g}$  dry wt [33]. Recently, Woolf et al. [34] reported that a child who had received



**Fig. 4** Relationships between concentrations in groundwater and hair of residents from Gia Lam and Thanh Tri, Vietnam for Mn and Ba [10]. Vertical dashed line indicates the WHO guideline values for drinking water [4].

Mn loading for 5 years thorough drinking water had striking difficulties in both visual and verbal memory, which are typical toxic effects of Mn. Manganese concentrations in the drinking water and hair of the child were 1210  $\mu\text{g/l}$  and 3.09  $\mu\text{g/g}$  dry wt, respectively [34]. Mean Mn values of groundwater and human hair from both the Gia Lam (1520  $\mu\text{g/l}$  in groundwater and 15.5  $\mu\text{g/g}$  dry wt in hair) and Thanh Tri (1260  $\mu\text{g/l}$  in groundwater and 38.9  $\mu\text{g/g}$  dry wt in hair) were comparable to or higher than the Mn levels reported by Woolf et al. [34].

It is known that Ba exerts toxic effects associated with hypokalemia and electrocardiographic changes [35], but little attention has been paid on the association of concentration in hair with its health effects. Further studies are necessary to assess the risk of these elements and also their mixed toxicity on the residents.

### 3. Arsenic pollution in Cambodia

#### 3-1. Arsenic concentration in groundwater

We determined As concentrations in the groundwater from Phnom Penh, Kratie and Kandal (Fig. 1) in the Mekong River Basin, Cambodia [14, 36]. The levels ranged from  $< 1$  - 886  $\mu\text{g/l}$  in our samples ( $n = 58$ ) (Table 1). The levels in groundwater from the Kandal (median, 180  $\mu\text{g/l}$ ; max, 720  $\mu\text{g/l}$ ) were highest. About 100 %, 45 %, and 0 % of groundwater samples from the Kandal, Kratie, and Phnom Penh exceeded 10  $\mu\text{g/l}$ , as WHO drinking water guideline for As, respectively [4].

Polya et al. [12] summarized groundwater As data of more than 1000 wells in Cambodia; these concentrations were determined by various organizations. In their analysis, it was found that the hot spot of As pollution is immediately the south and southeast of Phnom Penh. Also, the groundwater As levels were related to the geology: the highest concentration was observed in the Holocene sediments near the Mekong and Bassac Rivers (mean, 210  $\mu\text{g/l}$ ), whereas the Pliocene-Pleistocene volcanics (mean, 0.8  $\mu\text{g/l}$ ) and the Neogene-Quaternary sediments (0.9  $\mu\text{g/l}$ ) showed the lowest levels (Table 1).

#### 3-2. Arsenic concentration in human hair

Arsenic concentrations in hair of the residents from Cambodia ( $n = 96$ ) were 0.05 - 45.7  $\mu\text{g/g}$  dry wt (Table 2; Ref. 14, 36). Among the locations examined including those of Northern Vietnam, the highest concentration of As in hair was observed in the residents of Kratie (mean, 2.58  $\mu\text{g/g}$  dry wt), and also an extraordinarily high concentration, 45.7  $\mu\text{g/g}$  dry wt, was found from a donor of this site (Table 2). Also, 5 %, 43 %, and 0 % of

the subjects from the Kandal, Kratie, and Phnom Penh, respectively, exceeded the level of As in hair that may be related to skin pathology [22].

Recently, Berg et al. [15] also reported the hair As concentrations in Cambodia: the mean levels were 2.0  $\mu\text{g/g}$  dry wt in the Kandal and 0.3  $\mu\text{g/g}$  dry wt in the provinces bordering Kandal.

#### 3-3. Arsenic concentration in human urine

Total As concentrations were determined in the urine of residents from the Kandal, Kratie, and Phnom Penh, Cambodia (Table 3; Ref. 14, 36). Although concentrations of As in groundwater from the Kandal (median, 180  $\mu\text{g/l}$ ) were much higher than those from Phnom Penh (median, 2  $\mu\text{g/l}$ ) (Table 1), there was no significant difference in urinary As concentrations among locations (Table 3). However, the maximum concentration (490  $\mu\text{g/l}$ ) of total As in urine was found in a donor from Kratie (Table 3). Also, arsenic speciation was conducted for the residents of Kratie [14]. Percentages of urinary DMA[V], MMA[V] and inorganic As were 68 %, 14 % and 19 % (mean), respectively, for the residents (Table 3). Whereas AB was detected in the urine of the residents of Northern Vietnam as described above, this arsenical was not observed in the residents of Kratie, Cambodia (Table 3), suggesting that intakes of marine products may be small in the residents of Kratie. It should be noted that urinary As concentrations were significantly correlated with those of hair in the residents of Kratie, which also implies that these residents have been chronically exposed to As from the groundwater [14].

#### 3-4. Oxidative DNA damage in humans

Oxidative stress is recognized as one of the most plausible modes of action for As carcinogenesis [37-40]. 8-hydroxy-2'-deoxyguanosine (8-OHdG), an oxidative DNA damage product, is one of the predominant forms of radical-induced lesions in DNA, and is known to be useful for assessments of the carcinogenic risk of oxygen radical-forming chemicals [41, 42]. Elevated 8-OHdG levels were reported in the urine of patients with acute As poisoning [43] and of inhabitants chronically exposed to As through drinking water in Inner Mongolia, China [44], As-related skin neoplasms and keratosis of Bowen's diseases [45], and As-related human skin tumors of inhabitants in As polluted area in Gejiu, China [39]. Thus, the elevated 8-OHdG levels are considered to be an early symptom for the carcinogenic process of As [43].

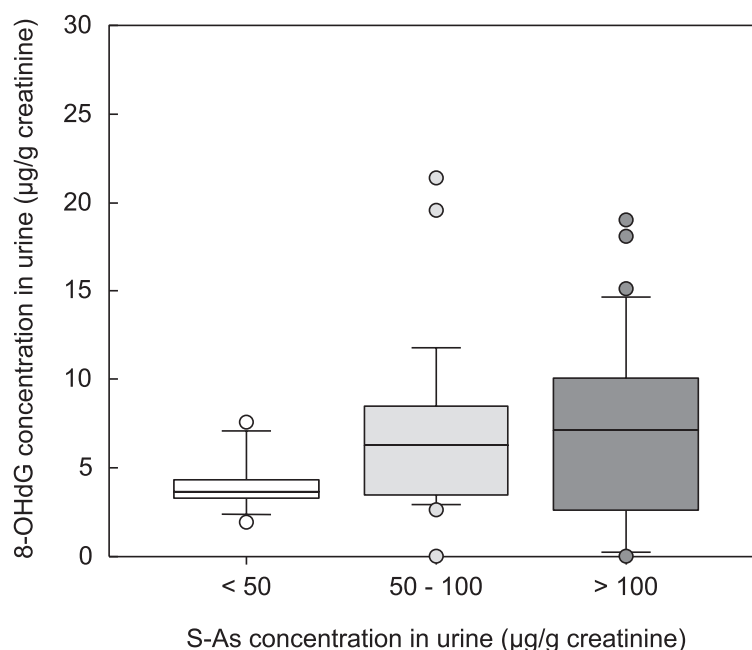


We determined the concentration of urinary 8-OHdG in the residents from Kratie, Cambodia to evaluate the oxidative stress by As exposure from groundwater [14]. Although there is no significant correlation ( $p > 0.05$ ) between concentrations of 8-OHdG and As in the urine, 8-OHdG level tended to increase with an increase in urinary As level (Fig. 5). Furthermore, urinary 8-OHdG level was significantly higher in the residents with hair As level of  $\geq 1 \mu\text{g/g}$  dry wt, the level of possible indication of As toxicity [22] than those of residents with  $< 1 \mu\text{g/g}$  dry wt (Fig. 6).

Several studies also represented DNA damage and oxidative stress in residents chronically exposed to As in drinking water in Inner Mongolia. Feng et al. [46] measured DNA fragmentation in residents exposed to As via drinking water in Inner Mongolia (mean,  $527.5 \mu\text{g/l}$ ). Elevated serum level of lipid peroxides, a marker of oxidative stress, was also observed in residents (mean As level was  $1.8 \mu\text{g/g}$  dry wt in hair) chronically exposed to As in drinking water (mean,  $410 \mu\text{g/l}$ ) in Inner Mongolia, China [47]. In West Bengal, India, elevated DNA damage in peripheral blood lymphocytes was recorded by comet assay in As-exposed participants (mean As levels in groundwater,  $247.1 \mu\text{g/l}$ ) with  $2.06 \mu\text{g/g}$  dry wt of mean hair level [48]. Although the As levels in the groundwater in the studies of Inner Mongolia [46, 47]

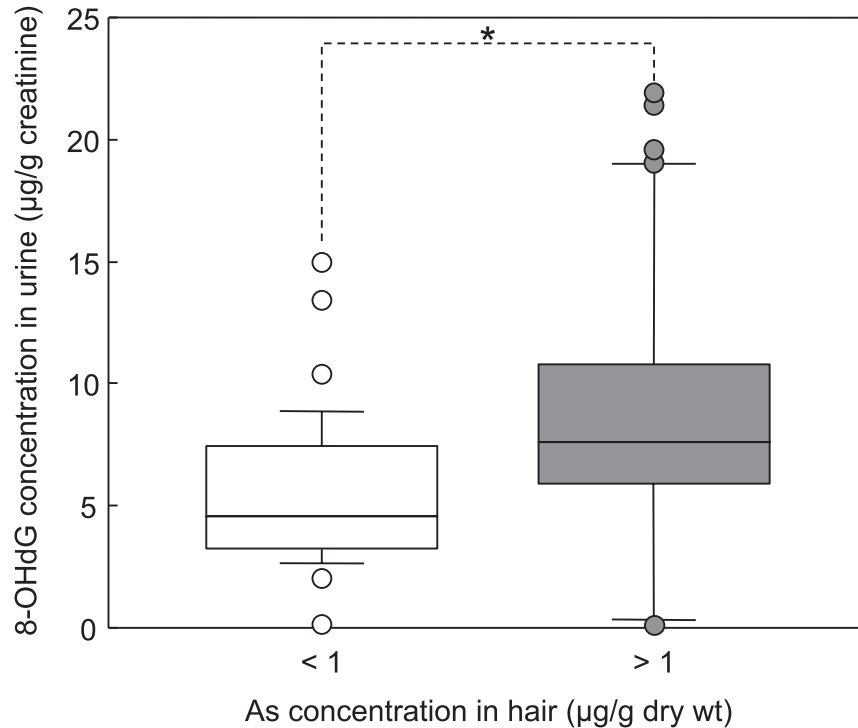
and West Bengal [48] were higher than those in the present study (Table 1), the mean hair As level in Kratie ( $1.77 \mu\text{g/g}$  dry wt, when one outlier,  $45.7 \mu\text{g/g}$  dry wt was excluded) was comparable to those in Inner Mongolia and West Bengal. Furthermore, Schwerdtle et al. [49] suggested that As in drinking water at the level of  $100 \mu\text{g/l}$  may cause DNA damage in humans based on the studies on DNA strand breaks and lesions in cultured human cells. In a village in Wuyuan Prefecture, Inner Mongolia, associations of As (mean,  $158 \mu\text{g/l}$ ) in well water with elevated skin abnormalities and urinary 8-OHdG concentrations in 61 of the 109 subjects have also been demonstrated [44]. These As levels in drinking water were comparable to those in Kratie, Cambodia (Table 1). Therefore, it seems likely that the oxidative DNA damage was caused by the chronic As exposure from groundwater in the residents of Kratie.

To our knowledge, symptoms of chronic As exposure from groundwater have not yet been reported for the residents in the Vietnam and Cambodia. This is probably due to the fact that the occurrence of cancer is caused by long-term exposure to As (the latency period for human carcinogenesis is thought to be 30-50 years; Ref. 43) and that most of the tube wells were built in the past decade in Vietnam and Cambodia [15]. However, further continuous consumption of the As-polluted tube well



**Fig. 5** Relationship between concentrations of S-As and 8-OHdG in the urine of residents from Kratie, Cambodia [14]. For the box and whisker plots, the horizontal bars indicate median, the vertical bars correspond to the range of 10-90%, and the horizontal boundaries of the boxes represent the range of 25-75%.





**Fig. 6** Relationship between concentrations of As in hair and 8-OHdG in urine of residents from Kratie, Cambodia [14]. For the box and whisker plots, the horizontal bars indicate median, the vertical bars correspond to the range of 10-90%, and the horizontal boundaries of the boxes represent the range of 25-75%. \*  $p < 0.05$ .

water might induce carcinogenesis in the future. Therefore, a larger and comprehensive epidemiological study is needed for the accurate risk assessment of As in residents of Mekong Basin in Cambodia and Vietnam and Red River delta in Vietnam.

#### 4. Mechanism of arsenic release into groundwater

Very few studies are available for the mechanisms explaining As release to groundwater in the Red River delta of Northern Vietnam and the Mekong River Basin of Southern Vietnam and Cambodia. Hence, we discussed this subject primarily based on the results of West Bengal and Bangladesh, because all these areas have a variety of common features such as river drainage from the rapidly weathering Himalayas, rapidly buried organic matter-bearing, relatively young sediments, very low hydraulic gradients, anoxic conditions, and high concentration of  $\text{Fe}^{2+}$  in groundwater [50].

The high As concentration in the groundwater in the Ganges, Red River and Mekong River deltas are not related to the As concentrations in their sediments [7, 27, 51]. Therefore, more complicated processes should be involved in the elevated groundwater As levels. It is generally accepted that vulnerable areas for arsenic pollution

are typically Holocene deltaic and alluvial sediments comprising highly reducing aquifers. It has been reported that groundwater As levels are high in aquifers beneath the Holocene floodplains [27, 51]. Also, bioavailable organic matter should be present to maintain the anoxic condition in the subsurface. Indeed, the areal distribution of As pollution is known to correspond closely to the areal distribution of buried peat in the Ganges delta [27].

The original source of the As may be pyrite from the granite and metamorphic source region of the Himalayas [18]. During Pleistocene, erosion of mountain belts was enhanced by glaciers [52]. Further chemical weathering released As to surface waters during Holocene, leading to the fluvial transport and sedimentation of As-enriched Fe oxyhydroxides [52]. Also, warm Holocene climate induced the high biological productivity, leading to the codeposition of organic matter and As adsorbed to the Fe oxyhydroxides [53]. Red River and Mekong River deltas also have young sedimentary deposits of Holocene age [15] and rapidly buried organic matter [50].

It is widely accepted that As is derived from the reductive dissolution of As-containing Fe oxyhydroxides in sediments from West Bengal and Bangladesh [27, 28, 54] and the Red River delta of Northern Vietnam and the

Mekong River Basin of Southern Vietnam and Cambodia [7, 10, 15]. Recently, it was revealed that this reductive processes are conducted by anaerobic bacteria [55-57]. As shown in Fig. 7, Fe-reducing bacteria reduce Fe(III) to Fe(II), leading to the release of As(V). Then, the As(V) will be reduced to As(III) by As-reducing bacteria [58]. It should be noted that released As, especially As(V), can be readsorbed on the Fe oxyhydroxides if the surface sites are not saturated. Therefore, the As levels in groundwater will be controlled by the balance between activities of Fe- and As-reducing bacteria depending on the quantity and bioavailability of organic matter, and the content of Fe oxyhydroxides available to sorb As [18]. In the Red River delta of Northern Vietnam, Berg et al. [7] found a positive correlation between As and Fe in sediments, and Agusa et al. [10] observed a similar correlation in the groundwater. These results are consistent with the hypothesis that the reductive dissolution of As-containing Fe oxyhydroxides causes mobilization of As in the sediments of Red River delta.

## 5. Conclusions

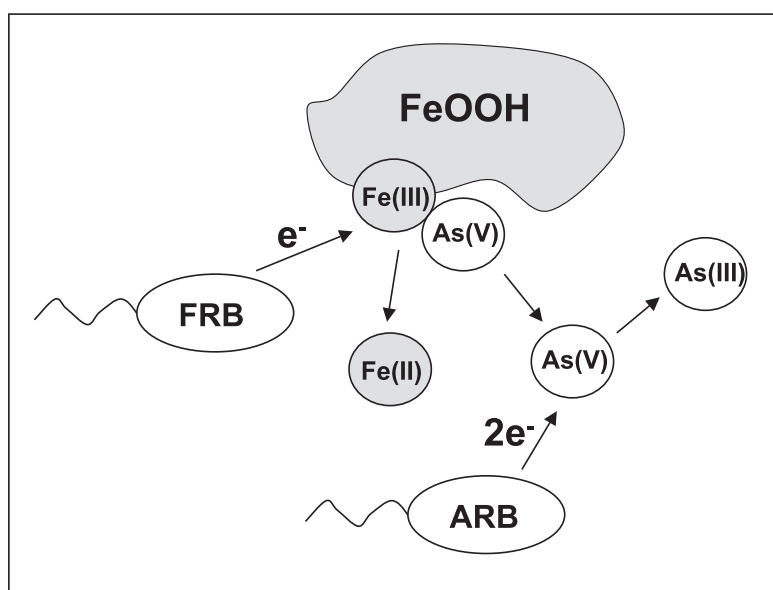
Elevated As concentrations were recently observed in groundwater of Vietnam and Cambodia, which were comparable to or lower than those from other As contaminated regions such as Bangladesh, West Bengal and Taiwan [6, 20, 21, 59]. Also, hair As concentrations of the residents of Vietnam and Cambodia were comparable

to or lower than those from other As-polluted areas such as Bangladesh and West Bengal [20, 21], but higher than those of residents in non-polluted areas [22]. The finding of As pollution in these areas might suggest that As pollution is much more widely distributed in the Asian countries. According to Stanger [60], As pollution of groundwater might also occur at river basins of Ayeyarwady, Chindwin, Salween, Jinsha Yangtze, and Black Da in the Southeast Asia. Hence, further studies are needed to address this issue.

Also, development of As removing techniques for polluted groundwater is required. The technique should be suited for these regions, especially for rural areas. Promising techniques include the household sand filters [61] and the three-pitcher method [62]. International technical and financial assistance would help promote the spread of such a mitigating action.

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**Fig. 7** Mechanism of As release from Fe oxyhydroxides (FeOOH) to groundwater by Fe- and As-reducing bacteria. FRB and ARB correspond to Fe- and As-reducing bacteria, respectively. Modified from Oremland and Stolz [63].

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