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Chromium, Cadmium, Lead, and Arsenic Concentrations in Water, Vegetables, and Seafood Consumed in a Coastal Area in Northern Vietnam

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ABSTRACT

BACKGROUND: Heavy metal contamination and related risks for the environment and human health are matters of increasing concern.

METHODS: The levels of 4 heavy metals (Cr, Cd, Pb, and As) were evaluated in 2 water types (surface and well), 4 types of seafood (tiger shrimp, stuffed snail, snake-head fish, and catfish), and 27 types of vegetables (12 leafy vegetables, 4 pea plants, 4 tuber vegetables, and 7 herbs) that are commonly consumed in northern coastal communes located in Vietnam. Atomic absorption spectrometry was employed for quantification.

RESULTS: The mean concentrations of heavy metals detected in water, seafood, and vegetable samples exceeded the national permitted standards and World Health Organization (WHO) recommendation values by at least 2-fold, 2.5-fold, and 5-fold for surface water, vegetables, and well water, respectively. The concentrations of all 4 heavy metals detected in seafood samples were higher than the standards. The levels of heavy metals decreased with increasing distance between the sample collection point and the pollution source.

CONCLUSIONS: This is the first report of heavy metal contamination of common sources of food and water in the northern coastal area of Vietnam. Significantly, the concentrations of heavy metals detected in study samples exceeded the regulatory limits. These results underscore the importance of continued monitoring and the development of intervention measures to ensure that the quality of food and water meets established standards and protects the health of the local population.

KEYWORDS: Heavy metals, pollution, water, food contamination, coastal commune, Northern Vietnam

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Introduction

Environmental pollution is one of the major challenges facing modern human society.[1] One of the major threats to ecological and human health is environmental contamination and pollution by heavy metals.[2,3] Rapid industrialization and urbanization have resulted in contamination of the environment by heavy metals; the rates of mobilization and transport of heavy metals in the environment have greatly accelerated since the 1940s.[4,5] While some metals are essential for human health at trace concentrations due to their role as coenzymes (eg, Fe and Cu), others are toxic at any concentration level (eg, Pb and Cd).[5] Chromium (Cr VI), cadmium (Cd), lead (Pb), and arsenic (As) have been recognized as water, soil, and sediment pollutants in many places in the world.[6,7] As and other heavy metals have even been detected in soil and sediment from estuarine areas in remote locations.[7,8] Metals can cause severe toxicity in humans, depending on the concentration, the exposure pathway, and the duration of exposure. It is thus critical to conduct substantial assessment of heavy metal contamination of the environment and to develop strategies to remediate the contamination and protect human and ecological health.[9]

* These authors contributed equally to this work.

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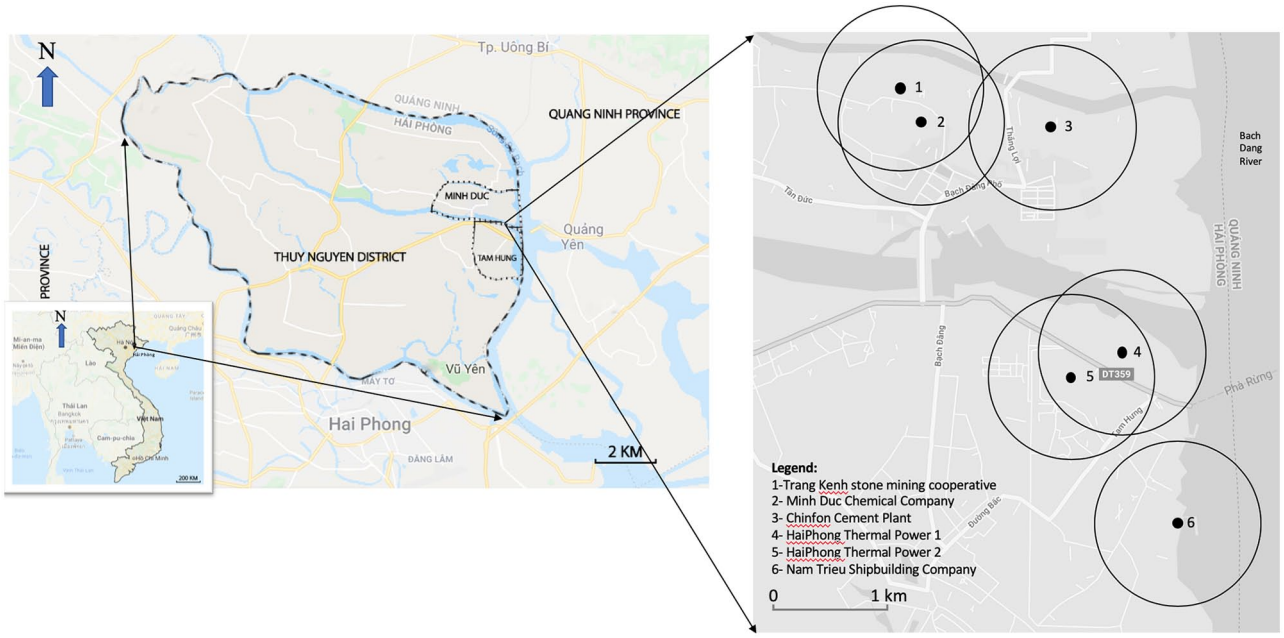


Figure 1. Map showing the location of the sampling sites.

The toxicity of heavy metals such as Cd, Pb, Cr(VI), and As to humans is widely recognized and documented.[10,11] For example, the International Agency for Research on Cancer (IARC) classifies As, Cd, and Cr as *Group 1 Carcinogenic to Humans.*[12] Food consumption accounts for up to 80% to 90% of the daily doses of As, Cr, Cd, and Pb to which humans are exposed.[13] Vegetables, which are an important part of the diet in Vietnam, can take up heavy metals when they are grown in contaminated environments; substantial amounts of heavy metals accumulate in the leaves, and therefore, consumption of vegetables may account for a substantial fraction of total exposures to Cd, Pb, and As.[14,15] Likewise, seafood, also an important component of the diet in Vietnam, can accumulate high concentrations of heavy metals when aquaculture is conducted in contaminated areas.[14] This indicates that the consumption of vegetables and seafood may be a major source of exposure to heavy metals. Currently, limited data on the levels of heavy metal contamination in food, especially heavy metal contamination in vegetables and seafood in Vietnam, are available.[16]

Located on the eastern margin of the Indochinese peninsula, bordering the Gulf of Thailand, and the South China Sea, including Gulf of Tonkin, Vietnam has a long coastline of 3260 km, even excluding its 3000 islands that are dispersed along its entire length.[17] A major environmental disaster occurred in the coastal area of Central Vietnam in 2016 when toxic wastewater containing heavy metals was accidentally, or perhaps illegally, discharged to the coast, causing several billions of dollars of damage.[18] The people and the Government in Vietnam are now well aware of the risk of upstream discharges to the coastal areas, including estuarine areas. This study aims to determine the concentrations of Cd, Cr, Pb, and As in water and measure heavy metals in local vegetables and seafood cultivated in 2 coastal communes in Northern Vietnam.

Methods

Study area

The study was conducted in Tam Hung and Minh Duc communes, 2 coastal sites that belong to the Thủy Nguyên district of Hai Phong city, located in the north of Vietnam, 123 km West of Hanoi capital (Figure 1). The study communes cover an area of 23.12 km^2; the population in 2016 was 19 036. The per capita income of 2 communes in 2016 was VND43 000 000 (US$1891).[19,20]

A program for monitoring metal contamination in water and food (vegetables and seafood) was set up in the Minh Duc and Tam Hung communes, Thủy Nguyên district, Hai Phong city in North Vietnam (Figure 1). Industrial production activities that involve wastewater release into the surrounding environment have been operating in this area since the late 1970s. Use of potentially contaminated stream water may enhance the heavy metal concentrations in vegetables and seafood produced near the industrial production areas. Water, vegetable tissue, and seafood samples were taken from crop fields and aquafarms cultivated by local households at 6 industrial sites including site 1—Trang Kenh stone mining cooperative (20°57′42.9″N 106°44′25.3″E), site 2—MinhDuc chemical company (20°57′39.3″N 106°44′27.9″E), site 3—Chinfon cement plant (20°57′36.0″N 106°45′09.2″E), site 4—HaiPhong thermal power 1 plant (20°56′28.5″N 106°45′34.4″E), site 5—HaiPhong thermal power 2 plant (20°56′21.6″N 106°45′21.9″E), and site 6—NamTrieu shipbuilding company (20°55′59.2″N 106°45′42.6″E).

Sampling

Seafood, vegetable, and water samples were collected in areas within a radius of 500 m, 500 to 1000 m, and 1000 to 1500 m

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from factories and enterprises in December 2016. Immediately after the collection, all samples were contained in an airtight insulating box, stored at −4 C, and transported to the Military Hygiene Laboratory, Vietnam Military Academic University for analysis.

Water samples. A total of 54 surface water samples (3 samples were taken at each designated per distance from each of the 6 industrial sources of pollutions) and 222 drinking (well) water samples were collected. The well water samples were collected from households who responded to a survey (administered to 1010 residents) that they use well water. The samples at each position were mixed in a plastic bucket, and a sample of 1 L was stored in a polyethylene bottle. Water samples were acidified with nitric acid to pH < 2 after collecting and transferred on ice to the laboratory for analysis; 10 mL of each sample were filtered through a 0.45 µm Whatman pore-size disposable capsule filter before elemental determination. Surface water and drinking water samples were collected following the regulations for general guidance on water sampling and quality control from the Ministry of Agriculture and Rural Development, Ministry of Health.[21,22]

Vegetables samples. Samples were collected and processed directly for analysis within 1 to 3 days. Only the edible parts were used for the analysis. Samples included 12 leafy vegetables, 4 pea plants, 4 tuber vegetables, and 7 herbs, which are among the most commonly grown and consumed vegetables in Northern Coastal Vietnam. With 5 replicates per species, a total of 135 samples were collected. Plant samples were collected approximately after 40 to 45 days of sowing.

Seafood samples. A total of 40 seafood samples (10 for each seafood [tiger shrimp, stuffed snails, snake-head fish, and catfish]) were collected at the 2 biggest open markets and the residents' fish ponds in the study area. Immediately after the collection, all samples were placed in an airtight insulating box and transported to the Military Hygiene Laboratory, Military Academic University, Hanoi, Vietnam. Seafood samples were collected by following the regulations on general guidance on food sampling for inspection, quality control, hygiene and safety food published in the Circular No. 14/2011/TT-BYT of the Ministry of Health.[23]

Sample preparation

Sample preparation and digestion methods for vegetables and seafood followed Anh TK Bui et al[16] and P Olmedo[24] with some modification. Only edible parts of each seafood and vegetable type were used for the analysis. Any spoiled or damaged parts were removed before preparation as a simulation of the real food preparation for human consumption. Fresh samples were weighed and then cut into small portions for drying in an oven at 105°C. Complete drying of the samples was achieved within 72 to 96 hours. Samples are considered dry when they attained the stable weight. Dry samples were ground in a household coffee grinder and sieved in a 0.8 mm mesh. The fresh weight (fw) and dry weight (dw) were recorded to calculate moisture contents. Trace element concentrations of vegetables and seafood were determined on a dw basis and converted to a fw basis for comparison with the MLs for contaminants and toxins in foods. The samples were then stored in a pre-labeled polyethylene bag for digestion.

After grinding, vegetable and seafood samples (400 mg per each) were digested with 1 mL H_2O_2 (30%, Puriss p.a.; purchased from Merck, Germany), 1 mL HF (40%, Puriss p.a.; purchased from Merck, Germany), and 6 mL HNO_3 (65%, Puriss p.a.; purchased from Merck, Germany) using the Multiwave PRO (Anton Paar) microwave. The microwave was set to 5 minutes for ramping up the temperature to 170°C and then held at this temperature for 10 minutes. The microwave was then set to 15 minutes ramping up to 180°C and then allowed to cool at room temperature. The digested samples were analyzed for As, Cr, Cd, and Pb using atomic absorption spectrophotometry (AAS/Model: AZ3000, Hitachi, Japan). Prepared reagent blanks and digests of the reference material were analyzed in parallel with each type of water, seafood, and vegetables. The AAS system was calibrated using at least 5 standard solutions for external calibration of each metal. Heavy metal concentrations were expressed as milligram per liter (mg/L) for water samples and as milligram per kilogram fresh weight (mg/kg fw.) for vegetable and fish tissues samples. The standard reference material for heavy metals in spinach leaves (SRM 1570a) and in fish (DORM-4) was obtained from the National Institute of Standard and Technology (NIST), USA and the National Research Council of Canada (NRCC), respectively; and standard reference solutions of 1000 mg/L were used for As, Pb, Cr, and Cd from Merck, Germany.

Quality control of the chemical analyses. The blank method was used to implement the quality control of the chemical analyses, performed through the complete preparation and analytical procedure. The blank spike duplicate (BSD) was performed in duplicate to determine to confirm the procedure was working within established control limits.

Great care was taken during sampling and laboratory analysis to prevent sample contamination and to ensure the reliability, quality, and accuracy of the analysis. We avoided the use of metallic tools whenever possible. All glass and plastic ware was washed with soap and rinsed with Sunfua-cromic (50 g $K_2Cr_2O_7$ mix in 500 mL H_2SO_4 98%) and then rinsed thoroughly with distilled and deionized water before use, to ensure there was no contamination of the laboratory supplies and tools; these procedures were conducted in with the guidelines, standards, and protocols of the National Institute of Standards & Technology (NIST).

One calibration blank (HNO_3 1%) was used to establish the analytical curve. Method blanks were used to check possible contamination from the sample preparation procedure. The blank and the standard samples were analyzed. The spike

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Table 1. Heavy metal concentrations (mg/L) in water samples (n = 276).

METAL	SURFACE WATER (N = 54)		WELL WATER (N = 222)	
	$\bar{X}$ (MIN, MAX)	TIMES EXCEEDED PERMISSIBLE LIMIT (%)	$\bar{X}$ (MIN, MAX)	TIMES EXCEEDED PERMISSIBLE LIMIT (%)
Cd	0.02 (0.00-0.03)	2.00 (64.81)	0.03 (0.00-0.15)	10 (84.68)
Pb	0.17 (0.03-0.39)	3.40 (87.04)	0.12 (0.01-0.42)	12 (94.59)
As	0.19 (0.02-0.42)	3.80 (68.52)	0.06 (0.01-0.48)	6 (83.33)
Cr	2.56 (0.32-4.32)	5.12 (98.15)	0.25 (0.02-0.82)	5 (74.77)

Abbreviation: WHO, World Health Organization. Allowable limits of Cd, Pb, As, and Cr in surface water recommended by the WHO guideline 2008 and Vietnam Standards 08:2008/BTNMT are 0.01, 0.05, 0.05, and 0.5 mg/L, respectively. Allowable limits of Cd, Pb, As, and Cr in well water recommended by the WHO guideline 2004[25] and Vietnam Standards 01:2009/BYT are 0.003, 0.01, 0.01, and 0.05 mg/L, respectively.

method was done in each analysis. Cadmium, lead, arsenic, and chromium levels in the blank samples were lower than limit of detection (LOD). The LOD was calculated by multiplying the SD of minimum values from 5 times repetition running by 3.

Continuing calibration verification was used to determine whether the sample analysis was within control limits (a close linear correlation between the concentration of the standard solution and the absorbance, with a correlation coefficient $r = 0.9990$).

The recovery rates were 93.08% to 93.52%, 88.81% to 89.19%, 83.62% to 84.38%, and 93.82% to 94.18% for Cd, Pb, As, and Cr, respectively.

Statistical analysis. Descriptive statistics including mean and standard deviation (SD) were calculated. Differences in the concentration of heavy metals among places were assessed using a one-way analysis of variance (one-way analysis of variance [ANOVA]) with Tukey honestly significant difference (HSD) post hoc test. The statistical significance was set as $P < .05$. All the statistical analyses were done with the IBM SPSS statistics (version 19.0).

Ethical considerations. The protocol of this study was approved by the Scientific and Ethical Committee in Biomedical Research, Hai Phong University of Medicine and Pharmacy. Local commune authorities were asked for their permission and all households were asked for their consent before colleting samples.

Results

Levels of Cd, Pb, As, and Cr in water samples

The levels of Cd, Pb, As, and Cr in the surface water and well water detected in the current study are presented in Table 1. The average concentrations of Cd, Pb, As, and Cr detected in surface water collected from 2 coastal communes in Northern Vietnam ranged between 0.0 and 0.03, 0.03 and 0.39, 0.02 and 0.42, and 0.32 and 4.32 mg/L, respectively, while the concentration of Pb, Cd, As, and Cr in well water ranged from 0.01 to 0.42, 0.0 to 0.15, 0.01 to 0.48, and 0.02 to 0.82 mg/L, respectively (Table 1). In brief, Cr was detected at the highest concentrations, followed by Pb, and then Cd.

The highest mean concentrations of metals detected in both surface and well water were for Cr and the lowest concentrations were observed for Cd. The concentration of As was higher than Pb in surface water, while the adverse result was found in drinking water (well water). The maximum limits (MLs) for Cd, Pb, As, and Cr in surface water according to the Vietnam standards (QCVN 08:2008 BTNMT) are 0.01, 0.05, 0.05, and 0.50 mg/L, respectively. These results indicate that the concentrations of all 4 heavy metals in surface water samples were 2- to 5-fold higher than national standards. The concentration of all 4 heavy metals in well water samples was 5- to 12-fold higher than WHO guidelines and national standards, as the MLs of Cd, Pb, As, and Cr in well water according to the Vietnam standards (QCVN 01:2009 BYT) are 0.003, 0.01, 0.01, and 0.05 mg/L, respectively.

The heavy metal concentrations in water by distance between the sampling sites and the pollution sources are shown in Figure 2. The concentrations of Pb, As, and Cr in surface water samples were statistically reduced in accordance to longer distances between the sampling sites and the pollutants sources ($P < .01$). Cd concentrations in surface water >1000 m from pollution source were statistically lower than the samples taken closer to the pollution source. No difference in Cd concentrations was found in surface water <500 m from the location and 500 to 1000 m distant from pollution sources ($P = .092$).

A statistically significantly lower concentration in As was detected in well water from sampling sites located at longer distances from the pollution source ($P < .001$). The concentration of Pb, Cd, and Cr in well water samples from locations near the pollution source (<500 m) was higher than locations that were 500 to 1000 m from the pollution source ($P < .001$); however, there was no difference in the 3 heavy metals in wells located at 500 to 1000 m and >1000 m from the pollution source ($P = .928$; $P = .998$; $P = .866$ respectively; Figure 2).

Heavy metal in seafood from aquaculture in sampling sites in Thuy Nguyen district, Hai Phong city

Heavy metal concentrations detected in seafood samples are shown in Table 2. The concentration of Cd was higher than

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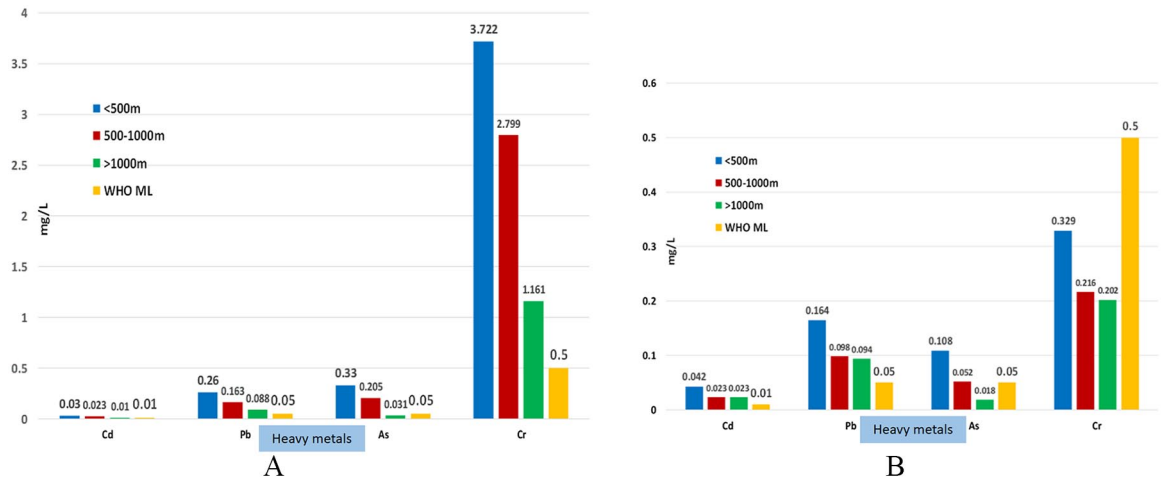


Figure 2. Heavy metal concentration in water by distance between the sampling sites and the pollution sources (n = 276). (A) Surface water and (B) Well water. WHO ML indicates World Health Organization maximum limit.

Table 2. Heavy metal concentration (Mean ± SD, mg/kg) in seafood samples.

SEAFOOD	PB	CD	CR	AS
Tiger shrimp	1.24 ± 0.46[a]	1.62 ± 0.24[a]	1.46 ± 0.28	0.80 ± 0.01
Stuffed snails	2.13 ± 0.34[a]	3.26 ± 0.27[a]	1.69 ± 0.40	1.20 ± 0.01
Snake-head fish	0.08 ± 0.01	2.30 ± 0.26	2.12 ± 0.13	1.18 ± 0.003
Catfish	0.10 ± 0.02	1.06 ± 0.09	2.25 ± 0.15	1.66 ± 0.08

Abbreviation: WHO, World Health Organization; FAO, Food and Agriculture Organization. Allowable limits of Pb, Cd, Cr, and As in seafood recommended by the WHO/FAO, Codex and Vietnam National Technical Regulation are 0.5 mg/kg for Pb and Cd in tiger shrimp; 1.0 and 2.0 mg/kg for stuffed snails, respectively.
[a]The heavy metal concentrations are higher than permitted standards.

those of the other 3 metals in seafood samples except catfish, in which the concentration of Cr exceeded that of the other 3 metals. The concentrations of metals detected in tiger shrimp and stuffed snail samples were the highest for Cd, followed by Pb and Cr.

In contrast to the other types of seafood, in snake-head fish, the order of the heavy metal concentrations was Cd > Cr > As > Pb. The concentrations of Pb and Cd in bottom layer seafood (shrimp and snail) were detected at levels higher than National permissible values and Food and Agriculture Organization (FAO)/(WHO)[26] tolerable concentrations, indicating that consuming these seafood products may pose a risk to human health.

Level of heavy metals in vegetables grown in sampling sites in Thuy Nguyen district, Hai Phong city

The concentrations of Pb, Cr, Cd, and As (mg/kg fw) in local vegetables collected from the 6 study sites ranged between 0.11 and 1.96, 0.02 and 1.57, 0.0 and 3.27, 0.17 and 1.70 mg/kg fw, respectively (Table 3). The concentrations of Pb, Cr, and Cd detected in 100% of the vegetable samples were higher than permitted standards. The concentrations of As in the vegetables samples were lower than the permitted standard except some species of bean (lady's fingers, green bean, and French bean), red amaranth, and some herb species (rau dang, Laksa leaves, Perilla, and Lettuce). The concentrations of heavy metals across Brassica vegetable samples were the highest for Pb, followed by Cd, As, and then Cr, while the order of heavy metal concentrations in bean vegetable samples was As > Pb > Cr > Cd except lady's fingers, for which the levels of Cd were higher than Cr. The order of concentration of heavy metals in amaranth was Cd > As > Pb > Cr but in Den tieu Pb was the highest, followed by As and Cd and then Cr.

The highest concentrations of Pb and Cd (mg/kg fw) were found in mustard greens (1.20 and 1.25, respectively; Table 3). The highest concentrations of As (mg/kg fw) were identified in green beans and French beans (1.26).

Figure 3 shows that the average concentrations of As and Pb in vegetables grown further from the sources of pollutant release were statistically lower than vegetables grown nearer to the sources of pollution ($P < .001$). Cd concentrations in samples from >1000 m distance from the pollution sources were lower than those from <500 m distance from the pollution sources ($P = .024$). The Cr concentrations in vegetable grown at <500 m from pollution sources were statistically different from vegetables grown farther away, 500 to 1000 m ($P = .006$)

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Table 3. Concentration of heavy metal (mg/kg) in grown vegetables (n = 135, 5 samples/species).

LOCAL NAME	ENGLISH NAME	LATIN NAME	PB	CR	CD	AS
Mean (Min-Max)			0.80* (0.11-1.96)	0.51* (0.02-1.57)	0.82[a] (0.00-3.27)	0.87 (0.17-1.70)
Cải bẹ dung			0.79[a]	0.09	0.13[a]	0.72
Cải bẹ xanh	Mustard green	*Brassica juncea*	1.20[a]	0.75[a]	1.25[a]	0.88[a]
Cải ngọt	Kale	*Brassica integrifolia*	0.50[a]	0.19[a]	0.57[a]	0.49
Cải xanh	Brown mustard	*Brassica juncea*	0.80[a]	0.32[a]	0.24[a]	0.89
Dền đỏ	Red amaranth	*Amaranthus gangeticus*	0.84[a]	0.50[a]	1.21[a]	1.07[a]
Dền tiểu	Amaranth		0.70[a]	0.48[a]	0.65[a]	0.67
Dền xanh	Green amaranth	*Amaranthus viridis*	0.66[a]	0.38[a]	0.82[a]	0.71
Mồng tơi	Vine spinach	*Basella alba*	0.75[a]	0.40[a]	0.91[a]	0.77
Rau đắng			1.00[a]	0.47[a]	0.90[a]	1.07[a]
Rau đay	Jute mallow	*Corchorus olitorius*	0.82[a]	0.53[a]	0.88[a]	1.02[a]
Rau lang	Sweet potato leaf	*Ipomoea batatas*	0.58[a]	0.51[a]	1.01[a]	0.78
Rau muống	Water spinach	*Ipomoea aquatic*	0.65[a]	0.50[a]	1.21[a]	0.90
Đậu bắp	Lady's fingers	*Abelmoschus esculentus*	0.97[a]	0.58[a]	0.96[a]	1.05[a]
Đậu cô ve	Green bean and French bean	*Phaseolus vulgaris*	0.96[a]	0.80[a]	0.66[a]	1.24[a]
Đậu đũa	asparagus bean	*Vigna unguiculata*	0.73[a]	0.73[a]	0.47[a]	0.90
Đậu rồng	Dragon bean	*Psophocarpus tetragonolobus*	0.83[a]	0.84[a]	0.50[a]	0.88
Mướp	Sponge gourd	*Luffa aegyptiaca*	0.63[a]	0.40[a]	1.22[a]	0.69
Cà tím	Long purple eggplants	*Solanum melongena*	0.70[a]	0.11[a]	0.09	0.67
Dưa leo	Cucumber	*Cucumis sativus*	0.86[a]	0.67[a]	1.06[a]	0.85
Khổ qua	Bitter melon	*Momordica charantia*	0.83[a]	0.77[a]	0.87[a]	0.86
Diếp cá	Fish mint	*Houttuynia cordata*	0.67[a]	0.46[a]	0.74[a]	0.75
Húng cây	Indian borage	*Plectranthus amboinicus*	0.57[a]	0.44[a]	1.02[a]	0.60
Húng quế	Basil	*Ocimum basilicum*	0.84[a]	0.42[a]	0.88[a]	0.79
Lá lốt	Lolot pepper	*Piper lolot*	0.72[a]	0.51[a]	0.74[a]	0.80
Rau răm	Laksa leaves	*Persicaria odorata*	0.87[a]	0.50[a]	1.18[a]	1.02[a]
Tía tô	Perilla	*Perilla frutescens*	1.04[a]	0.80[a]	0.97[a]	1.11[a]
Xà lách	Lettuce	*Lactuca sativa*	1.10[a]	0.60[a]	0.98[a]	1.16[a]

Abbreviation: WHO, World Health Organization; FAO, Food and Agriculture Organization. Allowable limits of Pb, Cd, Cr, and As in vegetables recommended by the WHO/FAO, Codex, and Vietnam National Technical Regulation are 0.3, 0.1, 0.1, and 1.0 mg/kg in fw, respectively.
[a]The heavy metal concentrations are higher than permitted standards.

and >1000 m ($P < .001$); however, no difference was seen in vegetables grown 500 to 1000 m and >1000 m from the source of pollution ($P = .545$). The concentrations of Cr, Cd, and Pb in vegetables were 2-to 10-fold higher than MLs within a 1.5 km radius from pollution sources. Except <500 m distance from pollution source, As was detected below the ML.

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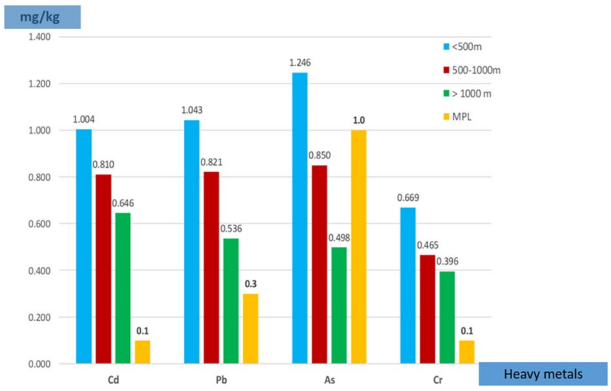


Figure 3. Heavy metal contaminations in vegetable samples by distance between sampling sites and pollution sources (n = 135).

Discussion

Peiravi et al[27] reported that industrial activities, especially plastic, chemical industries, and metal smelting, are major sources of heavy metals in water. These results led us to measure the levels of 4 heavy metals in water, seafood, and vegetables at a coastal site in North Vietnam where 6 industrial plants are located. Our results indicate that the industrial plants may affect the drinking water quality in this region. Our results also suggest that the stream used as irrigation water for vegetable crop production and seafood aquaculture was contaminated by wastewater from industrial production activities, resulting in elevated heavy metal concentrations in vegetables and seafood from the receiving areas. Altogether, our results indicate that suitable sanitation improvement programs should be used to protect the health of local residents.

The levels of heavy metals in water samples

The detection of heavy metals in water samples from Vietnam has some similarities and differences to the results from studies of heavy metals conducted in other areas. For example, the findings from Table 1 show that in surface water, Cr was detected at the highest concentration, followed by As, and then, Pb and Cd are similar to recent results in Taiwan, which was conducted in surface water in environment near a coastal coal-fired power plant.[28] Like surface water, in well water, Cr was detected as the highest concentration and Cd was detected at the lowest concentration. However, the concentration of Pd in well water was higher than the concentration of As. These results are consistent with results of a previous study conducted in the southeast coast of India, which reported a similar trend with Pb and Cd, although the concentrations of Cd and Pb measured in the present study were much higher than those reported in the previous study both pre-monsoon and postmonsoon.[29] Likewise, studies conducted in Iran and China also found that Cr was present in the highest concentration, although the concentration of Cr in water samples from Vietnam are higher.[30,31] By contrast, studies conducted in Thailand, India, Malaysia, and Bangladesh detected Pb at higher concentrations than Cr drinking water.[32-35] The As concentration detected in this study is higher than that detected in HaNam province—a hot spot of arsenic in underground water in Vietnam, which ranged from 8 to 579 ppb (mean 301 ppb), equivalent to 0.008 to 0.579 mg/L, mean 0.301 mg/L.[36]

Heavy metal in seafood from aquaculture in sampling sites

Heavy metals, which have adverse effects on human health, can accumulate in fish and shrimp tissues, which are generally found in the last zone of the aquatic food chain. Metals are usually taken up from food and water in fish and shrimp, distributed by circulation and eventually accumulate in target organs.[37] The lower concentration of Cr detected in shrimp in this study is similar to that observed in the research of Batvari et al who conducted a study in southeast coast of India, whereas the concentrations of Cd was higher than the concentration of Pb in this study; the inverse was observed in studies of other coastal regions in Asia, India, Turkey, and Yemen.[33,37,38] The As levels in seafood in this study were higher than those presented in other studies, which reported levels ranging from 0.003 to 0.08 µg/g and from 0.021 to 0.048 µg/g in common carp.[39]

The trend of As levels being higher than Pb levels in fish tissues in our study is consistent with results reported from Bangladesh, which showed the mean concentration of As and Pb to be 1.59 and 1.13 mg/kg in summer and 1.81 and 1.45 mg/kg in winter, respectively.[40] Interestingly, Peshut et al[41] found that arsenic in marine organisms or seafood is mainly in various organic forms, such as arsenobetaine, arsenoribosides, and arsenocholine, which are effectively nontoxic.

The present study results, which found the order of heavy metal concentrations to be Cr > As > Pb > Cd, are different from the findings from Le Quang Dung et al,[42] which reported the order of heavy metal concentration in oysters along the Hai Phong-Ha Long coast was As > Cd > Pb > Cr with concentrations of 10.10 to 19.33, 3.53 to 12.74, 0.79 to 6.20, and 0.81 to 4.47 mg/kg dry wt, respectively.

Heavy metal in vegetables in sampling sites

Heavy metal contamination in vegetable samples is presented in Table 3. Levels of Pb in mustard greens in this study were higher than in other studies.[11,15,16,43,44] Pb-Zn mining nearby results in contamination of soil and irrigation water.[16] Fertilizer and other agrochemicals, atmospheric deposition, and irrigation with contaminated water have also been implicated in Pb contamination of crops.[44]

Similarly high Cd concentrations were found in leafy vegetables sampled at Dabaoshan mine[45] and at Bac Kan.[16] Other studies have found lower[6,46] and higher[47,48] Cd concentrations in vegetables compared to those found in our study.

The highest concentrations of As (mg/kg fw) were identified in green beans and French beans (1.26). There have been very few studies that explored As content in vegetables, and our

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results were much higher than those reported in another study conducted in Pearl River Delta, South China[43] but much lower than those found in the study carried out in Bac Kan, Vietnam.[16]

Figure 3 shows the trace element contamination in vegetable samples by distance between sampling sites and pollution sources. Only the As concentration in vegetables grown at a distance >500 m from the pollution source is lower than ML. It is important to consider the health risks from heavy metal contamination of vegetables to the local community because these vegetables are the most favorite foods consumed in Vietnam, especially in the North.

Limitations of the study

To the best of our knowledge, this is the first study to measure heavy metal contamination in water, seafood, and vegetables collected from coastal communes in Northern Vietnam. However, the current data represented only a cross-sectional snapshot. We did not consider the change of daily weather during sampling period to avoid the possible bias in the analysis. Moreover, this present study did not include a health risk assessment to predict the population health problems related to environmental factors. Further studies are needed to investigate the impact of the heavy metals in water, vegetables, and seafood on human health.

Conclusions

In summary, our results indicate that water sources (*surface, drinking*) and food (*vegetables, seafood*) are contaminated with heavy metals (*arsenic, lead, cadmium, chromium*). The concentrations of heavy metal in water and food samples decreased with the distance between where the samples were collected and the pollution sources, especially for arsenic and lead. The research results show that the use of water and food grown near the pollution sources may increase public health risks due to heavy metals.

Recommendations

Considering all available evidence, we do not recommend field cultivation and consumption of vegetables grown 1 km from the sources of pollution. Further studies need to be conducted not only to calculate the risks, especially the cumulative risk resulting from consumption of contaminated water, food, and seafood, but also to apply appropriate measures to ensure the quality of food and water sources to protect the health of the local community.

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Author Contributions

NTMN, NVC, PVH, HAS, and PVT conceived and designed the study, agreed with the results and conclusions, and came up with arguments for this manuscript. NTMN, NVC, HCS, NQD, and NBT collected data and samples in the field. NTTThao, NTTB, NTTTrang, and NQD analyzed the data. NTMN, NVC, NTTThao, NQD, EVW, and HN wrote the first draft of the paper. All the authors read, made revisions, and agreed on the final version of manuscript. NTMN, NTTThao, and PVT reviewed and approved the final manuscript for submission.

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