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ORIGINAL ARTICLE

The distance-to-source trend in vanadium and arsenic exposures for residents living near a petrochemical complex

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Biological monitoring of vanadium (V) and arsenic (As) for residents living near a big petrochemical complex has not been previously studied. This study aims to investigate distance-to-source trends in urinary levels and dispersion-estimated concentrations of V and As in areas surrounding a petrochemical complex in central Taiwan. Our study subjects were 1424 residents living in the townships up to ~40 km from the petrochemical complex, and categorized as near (Zone A), further (Zone B) and furthest (Zone C) from the complex. Urinary and ambient V and As levels were analyzed by inductively coupled plasma mass spectrometry. Two-stage dispersion model was used to estimate V and As concentrations at each study subject's address. Multiple linear regression models were used to study the effects of distance-to-source and estimated air concentrations of V and As on the urinary V and As levels of study subjects. Area-wide levels of both V and As showed a high-to-low trend in urinary levels (μg/g-creatinine) from Zone A (V with 2.86 ± 2.30 and As with 104.6 ± 147.9) to Zone C (V with 0.73 ± 0.72 and As with 73.8 ± 90.8). For study subjects, urinary V and As levels were decreased by 0.09 and 1.17 μg/g-creatinine, respectively, with 1 km away from the emission source of the petrochemical complex, and urinary V levels were significantly elevated by 0.38 μg/g-creatinine with a 1 ng/m^3 increase in estimated ambient V concentrations at their addresses. Our study concludes a distance-to-source gradient in V and As exposures exists for residents living near a petrochemical complex with oil refineries and coal-fired power plants and two-stage dispersion model can predict such a trend for V when inhalation is the major exposure route, but not for As that exposure may be from multiple sources and exposure routes.

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INTRODUCTION

It has been previously reported that combustions of fossil fuels, including oil and coal, are two major air emission sources of vanadium (V) and arsenic (As).[1–3] V is classified as a possible carcinogen (Group 2B), while As is classified as a carcinogen (Group 1) by the International Agency for Research on Cancer.[4] Various toxic effects of these two pollutants have been found in both *in vitro* and *in vivo* studies. Exposing human bronchial epithelial cells to As or V caused changes in gene expression and aberrant growth.[5,6] Pulmonary inflammation and tumor occurrence and promotion were induced by exposing rats and mice to vanadium pentoxide,[7,8] while immunotoxicity was observed in mice exposed to As.[9] Epidemiological studies have also reported adverse health effects of V and As exposure. DNA damage was found in workers who were exposed to vanadium pentoxide during vanadium production.[10] One study showed that airborne As concentrations were associated with elevated standardized mortality ratios of lung cancer in a population living in municipalities in Japan.[11]

As and V were reported to be components of particulate matter (PM) emitted from industrial activities, such as oil refineries and coal-fired plants.[12–14] Previous studies have also shown that environmental concentrations of V in air, soil and vegetables within areas with petrochemical industries were higher than those in areas without petrochemical industries.[15,16] One previous study also showed that airborne As levels measured near coal-fired power plants were higher when compared with those measured at background locations away from the coal-fired power plants.[17] Another study reported that soil As levels were increased in the vicinity of a coal-burning power station in comparison with those measured in areas 5 km away from the power station.[18]

These studies, however, cannot assess the extent of V and As pollutions emitted from a major industrial source, because these studies are generally conducted to compare pollution levels in exposed and nonexposed areas. A study of clear distance-to-source gradient of V and As exposure is needed to clarify the air pollution impact of a major industrial source, such as a petrochemical complex, on its surrounding environment and population. There are even fewer studies on biological monitoring of V and As for residents living in the vicinity of industrial sources. There were no studies on V, but two studies used urinary levels as a biomarker of As exposure to find higher urinary As levels for adults but not for children in the vicinity of a coal-fired power plant.[19,20] The change in urinary As levels, however, was not assessed against airborne As concentrations because environmental monitoring was not conducted in these two studies.

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New studies are needed to clarify the impacts of V and As emissions on either exposures or health effects for residents living nearby emission sources. It is particularly important for us to conduct such a study in Taiwan as our previous epidemiological study showed excess mortality of cancers among residents also showed a distance-to-source trend near the No.6 Naphtha Cracking Complex, an industrial complex with oil refineries and coal-fired power plants, in central Taiwan.[21] In that study, residents' standardized mortality rates of lung and liver cancers in Taishi and Mailiao townships (Zone A) were significantly higher than those in their respective comparison townships farther away from the complex, including Baojhong, Sihhu, Dongshih, Lunbei townships (Zone B) and Erlun, Citong, Yuanchang and Huwei townships (Zone C). In a follow-up study since 2010, we observed that residents who lived in Taishi and Mailiao townships had worse lung, liver, and renal functions than those who lived in the other eight townships.[22] Therefore, it is of interest to see whether environmental concentrations and exposure data can link the observed health effects to the industrial emissions of V or As.

This study aims to investigate the distance-to-source trend in air and urinary levels of V and As for residents living near a petrochemical complex with oil refineries and coal-fired power plants by dispersion-based exposure estimation and Euclidean distance.

METHODS

Study Areas

The No.6 Naphtha Cracking Complex, located in Mailiao Township in Yunling County with an area of 2603 Ha, the largest petrochemical complex in Taiwan, is the main source of industrial emissions of our study area. There are 64 plants in this petrochemical complex, including three oil refineries with an oil production capacity of 25 million tons per year, one coal-fired power plant with an electricity generating capacity of 1.8 million kW per year, three co-generation plants with a total electricity capacity of 2.82 million kW and two naphtha cracking plants produced 160 million tons of ethylene, and several petrochemical processing plants.[23] Table 1 list emission sources of PM_{10} from the 249 stacks of the 64 plants in the complex. As indicated in the table, ~ 52.3% of total PM_{10} emissions were from three main stacks at the height of 250 m by coal-fired power plants, 32.3% emissions were from the five main stacks at the heights of 150–250 m by the coal-fired co-generation plants and 5.4% emissions were from two main stacks at the height of 50 m by the oil refinery plants. Residual oil was the main fuel used by the oil refinery plants in this complex.[24]

Our study area includes ten townships with similar social–economic development levels at 0–40 km away from the No.6 Naphtha Cracking Complex. These townships, located at the east to south (90°–180°) of the petrochemical complex, were further classified into three zones based on their administration divisions. The Mailiao and Taishi townships of Zone A are located the nearest by the petrochemical complex. The Baojhong, Sihhu, Dongshih and Lunbei townships of Zone B are further away, located nearby the Zone A in the farther areas of the petrochemical complex. The Erlun, Citong, Yuanchang and Huwei townships of Zone C are the furthest, located in the farther areas of the petrochemical complex than Zone B (Figure 1).

Air Samplings and Metal Analyses

The representative schools of the ten townships in the study area were selected as the air V and As sampling locations, including 3 in Mailiao township of Zone A, 3 in Taishi township of Zone A, 4 in Zone B, and 4 in Zone C (Figure 1) and there were 19 sampling campaigns conducted in the sampling locations from 2009 to 2012. In total, there were 192 ambient air PM_{10} samples (particles with aerodynamic diameter less than 10 μm) collected from these 14 sampling locations, including 64 samples in Zone A, 64 samples in Zone B and 64 samples in Zone C. Each PM_{10} sample was collected for 48 h by using Harvard Impactor (Air Diagnostics and Engineering, Harrison, ME, USA) and SKC pump (Leland Legacy, SKC, PA, USA) operated with a flow rate of 10 l/min. Thirty-seven-mm Teflon filter was selected as sampling medium. The airborne V and As concentrations

Table 1. Major PM_{10} emissions from manufacturing plants of the No. 6 Naphtha Cracking Complex in Taiwan[a].

Plant	*Major products*	*Pollution control technologies*	*Fuel*	*No. of plants*	*No. of stacks*	*Stack height (m)*	*Stack Diameter (m)*	*Stack exit gas temp. (°C)*	*Stack exit gas velocity (m/s)*	*PM_{10} emissions*	
										Tons/year	Percentage (%)
Coal power plant	Electricity	Electric precipitator wet flue gas desulfurization selective catalytic reduction	Bituminous coal	1	3	250	6.2	33	12.6	488.42	52.3
Cogeneration plants	Electricity, steam, pure water	Electric Precipitator	Bituminous coal	3	2	250	6.2	30–210	13.8–21.0	229.68	24.6
		Wet flue gas desulfurization Selective catalytic reduction	Pulverized coal		3	150	3.3	52–53	16.8–18.5	71.55	7.7
Oil refinery plants	Naphtha, liquefied petroleum gas gasoline, diesel, kerosene	Selective catalytic reduction Wet flue gas desulfurization	Residual oil Natural gas	3	2	50	4.6	62	5.2–15.2	50.06	5.4
Other downstream plants	Petrochemicals and raw plastics	Wet scrubber	Natural gas	57	239	20–37	0.3–1.8	26–168	2.4–14.8	63.26	6.8
Non stack releases[b]					62					31.60	3.4
Total				64	249					934.57	100.0

Abbreviation: PM, particulate matter. [a]The emission data is from Taiwan Emission Data System (TEDS 7.0) of Taiwan Environmental Protection Administration (TEPA, 2007). [b]These facilities are fugitive emission source without any stacks.

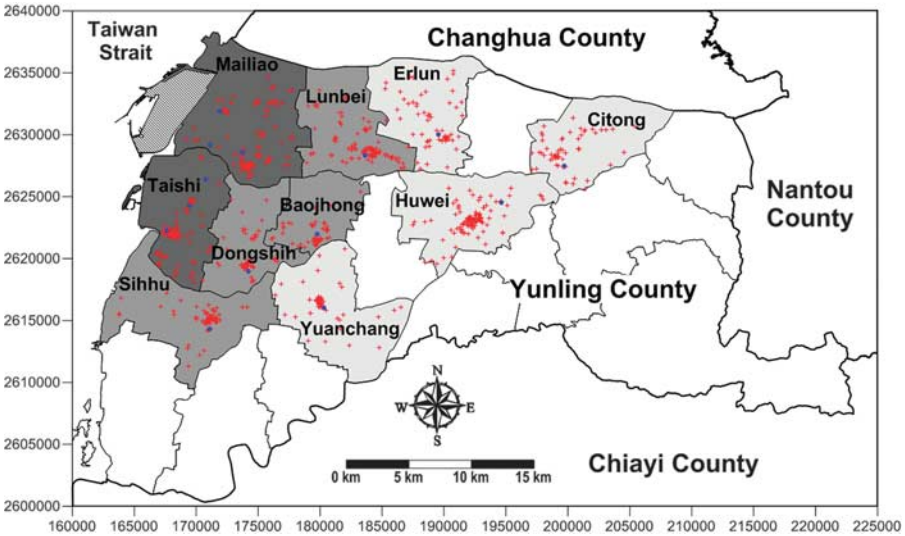

Figure 1. The map of the No.6 Naphtha Cracking Complex, study areas, air sampling sites and locations of study subjects in Yunling County in Taiwan, and the wind rose for PM_{10} sampling period from the Taishi air-quality monitoring station from 2009–2012. Note: The area of the No.6 Naphtha Cracking Complex (▨); the locations of air sampling sites (●); the addresses of 1424 study subjects (┼)

were determined by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500 s, Agilent, Santa Clara, TX, USA).

Study Subjects

There were 29 recruiting activities were held in the downtowns of the ten townships in the study area during 2009 and 2012, and there were total of 3307 local residents recruited to participate in our study. At the time of recruitment, the residents have conducted questionnaire survey, health examination and urine collection. We established a cohort of 3230 residents who have lived more than 5 years in these townships and completed all the data collection. Because of the outward migration of young people in the study areas, the disproportionate age distribution of only 25% of our recruited residents aged below 35 years was found. Finally, we selected 1424 adult residents of 276 in Zone A, 418 in Zone B and 730 in Zone C, who were aged above 35 years, from the established cohort as the study subjects in the present study, and the distribution of residential locations of the study subjects was shown in Figure 1. This study was approved by the Research Ethics Committee of National Taiwan University Hospital, and informed consent was given to us by each participant.

Urinary V and As Determination

We collected one morning spot urine sample from each study subject in a 15-ml BD tube (Vacutainer). Each sample was stored at −80 °C until analysis. V and As in urine were analyzed by an ICP-MS method as explained in the following steps. A 1.0-ml urine sample was first diluted with 2.0 ml 2% nitric acid and filtered with a 0.45-μm filter. The filtrate was then transferred to an ICP-MS instrument for analysis (Agilent 7500c, Agilent, Santa Clara, TX, USA). The urinary V and As levels of standard reference materials (SERO, Billingstad, Norway) analyzed by our method were all within acceptable ranges provided by the standard reference materials. The relative error of the ten spiked samples for each batch of the experiment was below 10% for V and As. The method-detection limits of urinary V and As were determined to be 0.026 and 3.325 μg/l, respectively. Creatinine analysis was conducted on all urine samples to adjust the measured urinary V and As levels. Urinary creatinine concentrations below 30 or above 300 mg/dl were used as criteria to exclude abnormal urine samples of unknown reasons for further data analysis. One-half of the method-detection limit was used to represent urinary V and As levels for samples below the method-detection limits.

Questionnaire Survey

Well-trained interviewers administered face-to-face questionnaire surveys to each study subject to collect exposure-related demographic and lifestyle data including age, gender, education level, addresses, smoking, alcohol drinking habits, betel nut chewing, sources of drinking water (including tap water, groundwater, spring water and other specific water) and fish consumption (including none, seldom and how many dishes on the previous 3 days).

Euclidean Distance

The geographic information system map of study subjects' residential locations was generated by using Google Earth 6.1 and Quantum GIS 1.74 according to the addresses with Universal Transverse Mercator (UTM) coordinate system where provided from study subjects.[25–27] On the basis of our understanding, the most contributions of V and As emissions of the No.6 Naphtha Cracking Complex were from its oil refineries and coal-fired power plant; two representative stacks were selected to be the emission source of V of the oil refineries (UTM-E: 167883, UTM-N: 2633516) and As of the coal-fired power plant (UTM-E: 167247, UTM-N: 2632285) in the No.6 Naphtha Cracking Complex. The distances of 1424 study subjects' addresses to these representative stacks of V and As emission sources in the No.6 Naphtha Cracking Complex were calculated by Euclidean distance formula.

Dispersion-based Exposure Estimation

Two-stage dispersion modeling approach developed in our previous study was used to estimate air concentrations of V and As at the addresses of the study subjects.[28] Briefly, the emission stacks from two main emission sources, that is, oil refineries for V and coal-fired power plants for As, were used to represent all V and As emission sources from the No.6 Naphtha Cracking Complex in the two-stage dispersion model (Table 1). At the first stage of modeling, a backward fitting approach with measured V and As concentrations from 192 ambient air PM_{10} samples monitored at 14 sampling locations across Zone A, B and C in the ten study townships from 2009 to 2012 was used to estimate V and As emissions (g/s) from both oil refineries and coal-fired power plants. The prevailing wind direction for overall air sampling campaigns was northeast (Figure 1). At the second stage of modeling, model-derived V and As emission rates and meteorological data were fed to the Industrial Source Complex, version 3 models to estimate the spatial distributions of V and As concentrations on the days when the subjects' urine samples were collected. The prevailing wind direction for overall study subjects recruited days was northeast. Kriging interpolation method and shift weighting averages were then applied to estimate the V and As concentrations of ambient air at each subjects' geocoded address from spatial distributions of V and As derived from Industrial Source Complex, version 3 models in the simulation domain of 50 km × 40 km.

Statistical Analysis

Data analysis started with descriptive statistics for study-subject variables. According to the WHO definition of elderly, study subjects were dichotomized as middle aged (35–65-years old) and elderly (>65-years old). Education level was classified as primary (below elementary school), middle (above junior high school and below university) and high (above university). The sources of drinking water were categorized as tap water and others (including spring water, groundwater and others). To compare the differences among study subjects in the three Zones, X^2-testing was employed for the analysis of discrete variables. To analyze the differences in distance of address-to-source, urinary metal levels and dispersion-based estimated air-metal concentrations of the study subjects in the three zones, we conducted analysis of covariance test for urinary metal levels by adjusting age, gender, education level, smoking, alcohol consumption, nut chewing, fish consumption and source of drinking water with the post comparison Scheffe test. After adjusting important potential confounders, including age, gender, education level, smoking, alcohol consumption, nut chewing, fish consumption and source of drinking water, a multiple linear regression model was conducted to assess the associations between urinary metal levels, distance of address-to-source and estimated personal air-metal concentrations at the home addresses. All statistical analyses were performed by using SAS 9.2 for Windows. The difference was considered significant when $P < 0.05$.

RESULTS

The recruited 1424 adult study subjects aged above 35 years accounted for 0.8% of the population in the ten townships, and were proportional to the respective populations of the three zones by 276 (19.4%) in Zone A, 418 (29.3%) in Zone B and 730 (51.3%) in Zone C. Basic characteristics of the 1424 study subjects among the three study zones in the vicinity of the petrochemical complex are shown in Table 2. Our study subjects were comparable among the three study zones as there were no differences in gender, smoking, alcohol consumption, betel nut chewing and fish consumption among them. There were, however, differences in age, education and drinking water sources among the three zones. Study subjects were the youngest in Zone B, had the lowest education level in Zone A, and used tap water the least in Zone C.

Comparisons of address-to-source distances, urinary levels and measured and estimated air concentrations of V and As for the study subjects among the three study zones of the petrochemical complex are listed in Table 3. There was a distance-to-source trend of urinary V and As levels in our study subjects living in the vicinity of the petrochemical complex: their levels increased from Zone C (V with 0.73 ± 0.72 μg/g-creatinine and As with 73.8 ± 90.8 μg/g-creatinine) to Zone A (V with 2.86 ± 2.30 μg/g-creatinine and As with 104.6 ± 147.9 μg/g-creatinine). The measured ambient V concentrations of 14 sampling locations in the study area were 8.45 ± 2.97 ng/m^3, 7.19 ± 1.43 ng/m^3 and 6.86 ± 1.17 ng/m^3 in Zones A, B and C, respectively, and the measured ambient As concentrations were 2.05 ± 1.19 ng/m^3, 2.38 ± 1.27 ng/m^3 and 2.26 ± 1.21 ng/m^3 in Zones A, B and C, respectively. It showed a significantly distance-to-source trend of dispersion-based estimated ambient V concentrations at the subjects' addresses from Zone A (8.50 ± 1.71 ng/m^3) to Zone C (5.34 ± 0.87 ng/m^3), but the reverse outcome was observed in the dispersion-based estimated ambient As concentrations from Zone A (2.16 ± 0.37 ng/m^3) to Zone C (2.40 ± 0.4 ng/m^3).

Tables 4 and 5 shows the relation between urinary V and As levels (μg/g-creatinine) and the distances from sources (km) for 1424 subjects estimated by multiple regression models. After adjusting for age, gender, education level, smoking, alcohol consumption, nut chewing, fish consumption and source of drinking water, it showed that the study subjects' urinary V levels were decreased by 0.09 μg/g-creatinine with 1 km away from the V emission source of the petrochemical complex, and it also found that elderly people had significantly higher urinary V levels when compared with middle-aged people, and subjects with higher educational levels had significantly lower urinary V levels than subjects with lower education levels. Nevertheless, our model showed no effect on urinary V levels by gender, smoking, alcohol consumption, betel nut chewing, fish consumption, or the source of drinking water. For As, the model demonstrated the study subjects' urinary As levels were significantly decreased by 1.17 μg/g-creatinine with a 1 km far from the As emission source of the petrochemical complex with no effect by other confounding factors.

The associations between urinary levels (μg/g-creatinine) and dispersion-based estimated ambient concentrations at study subject's addresses (ng/m^3) for V and As are shown in Tables 4 and 5. Results showed that the study subjects' urinary V levels were significantly elevated by 0.38 μg/g-creatinine with a 1 ng/m^3 increase in estimated ambient V concentrations at their addresses after adjusting potential confounding factors, and elderly people and subjects with higher educational levels had significantly higher urinary V levels. By contrast, no such association has been found between estimated airborne As concentrations at home addresses and individual urinary As levels among study subjects.

Table 2. Comparisons of basic characteristics of 1424 study subjects among three study zones in the vicinity of the petrochemical complex.

Areas / *Variables*	*Zone A* (n = *276*)	*Zone B* (n = *418*)	*Zone C* (n = *730*)	P-*value*[a]
Age, n *(%)*				
≤65 years	181 (65.6)	303 (72.5)	455 (62.3)	0.0022
>65 years	95 (34.4)	115 (27.5)	275 (37.7)	
Gender, n *(%)*				
Male	86 (31.2)	150 (35.9)	281 (38.5)	0.0952
Female	190 (68.8)	268 (64.1)	449 (61.5)	
Education level, n *(%)*				
Primary	189 (68.5)	198 (47.4)	331 (45.3)	< 0.0001
Middle	79 (28.6)	191 (45.7)	331 (45.3)	
High	8 (2.9)	29 (6.9)	68 (9.3)	
Smoking, n *(%)*				
No	254 (92.0)	370 (88.5)	654 (89.6)	0.3215
Yes	22 (8.0)	48 (11.5)	76 (10.4)	
Alcohol intake, n *(%)*				
No	251 (90.9)	359 (85.9)	651 (89.2)	0.0921
Yes	25 (9.1)	59 (14.1)	79 (10.8)	
Nut intake, n *(%)*				
No	263 (95.3)	393 (94.0)	701 (96.0)	0.3026
Yes	13 (4.7)	25 (6.0)	29 (4.0)	
Fish consumption, n *(%)*[b]				
No	213 (77.2)	336 (80.4)	599 (82.0)	0.2150
Yes	63 (22.8)	82 (19.6)	131 (18.0)	
Water source, n *(%)*[c]				
Tap water	252 (91.3)	374 (89.5)	553 (75.7)	< 0.0001
Other	24 (8.7)	44 (10.5)	177 (24.3)	

[a]Comparing means by X^2-test. [b]The "yes" including seldom and how many dishes they had on the previous 3 days. [c]The "other" including groundwater, spring water and other specific water.

DISCUSSION

This is the first study to report a distance-to-source gradient in urinary V concentrations for residents living in the vicinity of a petrochemical industry complex at an individual as well as a township level. Urinary V is an effective biological marker for

Table 3. Comparisons of distance of address-to-source, urinary levels, measured and dispersion-based estimated air concentrations of V and As in study subjects among study zones of the petrochemical complex.

Areas metals[a]	*Zone A (276)*	*Zone B (418)*	*Zone C (730)*	P-*value*[b]	*Scheffe*
Distance of address-to-source, km					
V	9.65 ± 2.42	16.7 ± 2.29	25.5 ± 4.97	< 0.0001	C > B > A
As	9.13 ± 2.00	16.3 ± 2.17	25.6 ± 5.20	< 0.0001	C > B > A
Urinary levels, μg/g-creatinine					
V	2.86 ± 2.30	0.85 ± 1.02	0.73 ± 0.72	< 0.0001	A > B > C
As	104.6 ± 147.9	79.6 ± 77.9	73.8 ± 90.8	< 0.0001	A > B > C
Measured ambient concentrations in air, ng/m³ [c]					
V	8.45 ± 6.57	7.19 ± 4.40	6.86 ± 4.17	0.1878	
As	2.05 ± 1.19	2.38 ± 1.27	2.26 ± 1.21	0.2993	
Dispersion-based estimated air concentrations at subjects' addresses, ng/m³					
V	8.50 ± 1.71	6.80 ± 1.46	5.34 ± 0.87	< 0.0001	A > B > C
As	2.16 ± 0.37	2.27 ± 0.32	2.40 ± 0.40	< 0.0001	C > B > A

Abbreviation: ANCOVA, analysis of covariance. [a]All data are presented as the mean ± SD. [b]Comparison among three zones by ANCOVA test adjusting age, gender, education level, smoking, alcohol consumption, nut chewing, fish consumption, and source of drinking water with a post comparison by Scheffe test. [c]The average concentrations of collected air samples in study area with 64 PM_{10} samples for each Zone.

Table 4. Multiple regression models for predictors of urinary V levels (μg/g-creatinine) of study subjects ($N = 1,424$).

	Model I		*Model II*	
Variables	*β (95% CI)*	P-*value*	*β(95% CI)*	P-*value*
Intercept	2.74 (2.35, 3.12)	< 0.0001	− 1.44 (−1.88, − 1.00)	< 0.0001
Distance of address-to-source (km)	− 0.09 (−0.10, − 0.08)	< 0.0001	—	—
Estimated air level at address (ng/m³)	—	—	0.38 (0.34, 0.42)	< 0.0001
Age (ref: ≤65 years)	0.19 (0.02, 0.35)	0.0261	0.23 (0.06, 0.39)	0.0066
Gender (ref: male)	0.17 (−0.01, 0.34)	0.0580	0.11 (−0.06, 0.28)	0.2232
Education level (ref: primary)				
Middle	− 0.37 (−0.54, − 0.20)	< 0.0001	− 0.40 (−0.56, − 0.23)	< 0.0001
High	− 0.53 (−0.82, − 0.23)	0.0005	− 0.71 (−1.00, − 0.43)	< 0.0001
Smoking (ref: none)	− 0.02 (−0.29, 0.25)	0.8849	− 0.13 (−0.39, 0.14)	0.3448
Alcohol intake (ref: none)	− 0.02 (−0.27, 0.22)	0.8506	− 0.01 (−0.25, 0.23)	0.9105
Nut intake (ref: none)	− 0.06 (−0.42, 0.30)	0.7394	0.05 (−0.30, 0.40)	0.7844
Fish consumption (ref: none)	− 0.04 (−0.22, 0.14)	0.6496	− 0.05 (−0.22, 0.13)	0.5954
Water source (ref: tap water)	0.09 (−0.10, 0.27)	0.3695	0.04(−0.14, 0.23)	0.6567

Abbreviation: CI, confidence interval. Note: The adjusted *R*-square of regression for Models I and II were 0.23 and 0.25, separately.

exposure because urinary excretion is the main metabolic pathway of V exposure.[1] V has been indicated as a good tracer for environmental pollution from the petrochemical industry, as V is commonly emitted from the burning of residual oil and coal by the refineries and power plants such as an industrial complex.[29] Our study found that township-average urinary V levels of study subjects living in Zone A were about four-fold higher than those living Zone C (Table 3), and the individualized distance-to-source trend in urinary V levels of study subjects also be observed after controlled by potential confounders (Table 4). Urinary V concentrations of this study, 0.73–2.86 μg/g-creatinine, were higher than nonoccupational exposure levels, 0.052 μg/g-creatinine, reported in Germany,[30] and comparable to occupational exposure levels, 0.8–1.4 μg/g-creatinine, in USA.[31] In addition, it should be noted that elderly persons and residents with a relatively low level of education have higher urinary V levels than their counterparts in our study (Table 4). Such results are in agreement with findings of previous studies.[32–34] The effects of age and education (a surrogate indicator of socioeconomic status) on urinary V levels may be explained by differences in exposure and metabolic rate associated with these factors. Such differences can be attributed to different time-activity patterns between study subjects. Time span indoors is believed to be longer for elderly people and highly educated adults with office work in the present study. This may reduce their V exposure from ambient air. The excess V burdens borne by residents living near the petrochemical complex may pose potential health risks, as V has been shown to have various toxicological effects, such as irritation of the respiratory tract, gene expression changes and DNA damage.[3,5,10] In our previous studies, the higher standardized mortality rates of lung and liver cancers and the worse lung, liver and renal functions were found in residents living near the petrochemical complex.[21,22]

This is also the first study to show a good correlation between the internal exposure of urinary levels and the external exposure of estimated ambient air concentrations for assessing V exposure among people (Table 4). Our two-stage dispersion models implies that the petrochemical complex is the major exposure source of air V in the area (Table 3), which then contributes to resident's V exposures via the inhalation route.

Table 5. Multiple regression models for predictors of urinary As levels (μg/g-creatinine) of study subjects ($N = 1424$).

	Model I		*Model II*	
Variables	β (95% CI)	P-*value*	*β (95% CI)*	P-*value*
Intercept	90.3 (61.0, 120)	< 0.0001	82.2 (40.5, 124)	0.0001
Distance of address-to-source (km)	−1.17 (−1.89, −0.45)	0.0015	—	—
Estimated air level at address (ng/m^3)	—	—	−5.27 (−19.3, 8.74)	0.4604
Age (ref: ≤65 years)	8.52 (−4.18, 21.2)	0.1882	9.02 (3.72, 21.8)	0.1651
Gender (ref: male)	2.89 (−10.4, 16.2)	0.6688	0.36 (−12.9, 13.6)	0.9574
Education level (ref: primary)				
Middle	−2.84 (−15.6, 9.96)	0.6635	−6.21 (−18.9, 6.45)	0.3361
High	−5.77 (−28.3, 16.8)	0.6160	10.1 (−32.7, 12.5)	0.3803
Smoking (ref: none)	9.30 (−11.0, 29.6)	0.3697	8.15 (−12.3, 28.6)	0.4333
Alcohol intake (ref: none)	1.96 (−16.7, 20.6)	0.8373	1.79 (−16.9, 20.5)	0.8513
Nut intake (ref: none)	7.62 (−19.7, 35.0)	0.5846	7.71 (−19.8, 35.2)	0.5822
Fish consumption (ref: none)	3.68 (−10.0, 17.4)	0.5983	4.07 (−9.69, 17.8)	0.5618
Water source (ref: tap water)	−8.03 (−22.4, 6.36)	0.2737	−13.1 (−27.2, 1.03)	0.0693

Note: The adjusted *R*-square of regression for Models I and II were 0.06 and 0.01, separately.

Our finding also shows a distance-to-source gradient in urinary As levels evidenced by individual subject's Euclidean distance to the petrochemical complex (Table 5). This is in agreement with previous studies on populations living near industrial sources of petrochemical industries and coal-fired power plants.[19,20] Urinary As concentrations of the present study, 73.8–104.6 μg/g-creatinine, were higher than nonoccupational exposure levels, 12 μg/g-creatinine, reported in Germany,[30] The failure of using dispersion-based estimated exposure to associate with urinary As levels in our study indicates that inhalation may not the only route for As exposure for residents in the study area. Urinary As levels in the present study may represent various environmental exposures of As from sources of food, water and soil through ingestion and dermal contact, in addition to inhalation of As from the polluted air.

Some of limitations of the present study should be mentioned. First, dietary intake is one of the important metal exposure sources for the general population, but only the source of drinking water and fish consumption were controlled in this study. It is particularly important for As in this study because total urinary As is used as an exposure indicator. The primary form of As found in urine is organic As,[2] while the dominant arsenic from fossil fuel and industrial processes is inorganic As. Further investigation into determining the important environmental exposure sources of As in this study area and determining urinary arsenic speciation for the residents should be conducted in order to clarify the role of these influential factors on the distance-to-source trend of urinary As levels found in the current study. Further complete survey on the residents' dietary habits and the measurements of metal levels in food and drinking water are suggested for clarifying the contribution of dietary intake on urinary metal levels of residents living in the vicinity of an industrial complex. Second, the urinary metal levels with short biological half-lives might be influenced by the recent exposure before the urine collection of study subjects. Nevertheless, it should not affect the direction of the present results because of continuous long-term emission from the industrial complex since its start of operation in 1999 and because all of the study subjects lived in this area for at least 5 years. Third, the meteorological conditions of measured data, such as wind speed, wind direction, temperature and humidity at the air sampling locations during air samplings, may not fully reflect dispersion scenarios of the areas over the entire modeling period. More air monitoring is needed to verify the distribution of the dispersion-based air pollution at study subject's addresses.

CONCLUSIONS

The residents' urinary V and As levels are proportional to their proximity to the industrial sources of oil refineries and coal-fired power plants, and would be appropriate biomarkers to explore the environmental exposure for the residents living nearby. Two-stage dispersion models can predict such a trend in V exposure well when inhalation is the major exposure route, and project potential multi-route of As exposures for residents living in the area.

CONFLICT OF INTEREST

The authors declare no conflict of interest

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