# A Pilot Survey of Potentially Hazardous Trace Elements in the Aquatic Environment Near a Coastal Coal-Fired Power Plant in Taiwan

Victor C Kok[1,2,3], Paul R Winn[4], Yi-Jer Hsieh[3,5], Jien-Wen Chien[3,6,7], Jer-Ming Yang[3] and Guang-Perng Yeh[3,8]

[1]Department of Internal Medicine, Kuang Tien General Hospital, Taichung, Taiwan. [2]Disease Informatics Research Group, Asia University, Taichung, Taiwan. [3]The Scientific Committee, Air Clean Taiwan (ACT) Alliance, Changhua, Taiwan. [4]Waterkeeper Alliance based in Newcastle, Newcastle, NSW, Australia. [5]Department of Anesthesiology, Changhua Christian Medical Foundation Changhua Christian Hospital, Changhua, Taiwan. [6]Institute of Occupational Medicine and Industrial Hygiene, College of Public Health, National Taiwan University, Taipei, Taiwan. [7]Division of Pediatric Nephrology, Changhua Christian Medical Foundation Changhua Christian Children's Hospital, Changhua, Taiwan. [8]Department of Obstetrics and Gynecology, Changhua Christian Medical Foundation Changhua Christian Hospital, Changhua, Taiwan.

## ABSTRACT

**BACKGROUND:** A limited number of potentially hazardous trace elements were quantified in the aquatic environment near the world's second largest coal-fired power plant (CFPP) and the coal combustion residual (CCR) disposition sites in Central Taiwan. We postulated that contamination from specific trace elements would be present in the abovementioned aquatic environments.

**METHODS:** Cross-sectional sampling of trace elements was first performed between September 24, 2017 and October 3, 2017 outside the CFPP, in the effluent sampled from Changhua, a county south of metropolitan Taichung, and at the historical CCR disposal sites, using the intertidal zone surface seawater and the seawater in an oyster farm as controls. Aqueous samples were collected from 12 locations for analysis of 13 trace elements (Al, As, B, Cd, total Cr, Co, Fe, Pb, Mn, Se, Sr, Tl, and V). We used inductively coupled plasma (ICP) optical emission spectrometry to determine B and Fe levels, and ICP mass spectrometry for all other trace elements. The Spearman rank correlation coefficient (Rho) was calculated to examine the pairwise relation among the trace elements.

**RESULTS:** Al (50% of all samples), B (66.7%), Fe (25%), Mn (50%), Sr (8.3%), and V (25%) were identified as being above the Environmental Protection Agency (EPA) regulation limit. The oyster farm seawater had no concerns. Mn ($96.4\,\mu g/L$) in the CFPP drainage effluent was 1.9-fold above the regulation limit. Fe, Mn, and V were detected from the cooling channel at 4379, 625, and $11.3\,\mu g/L$, respectively. The effluent and water from the areas surrounding the 2 CCR dump sites revealed similar magnitudes of trace element contamination. B is highly correlated with Sr (Rho = 0.94, 95% confidence interval [CI], 0.80-0.98). Meanwhile, Fe is highly correlated with Al (Rho = 0.77), Pb (Rho = 0.71), Co (Rho = 0.75), and V (Rho = 0.84).

**CONCLUSIONS:** The EPA must set an explicit regulation limit for aluminum, boron, iron, and strontium in the aquatic environment. This exploratory research will inform policymaking regarding certain trace elements that could potentially have an adverse impact on public health and wildlife.

**KEYWORDS:** Trace elements, coal-fired power plant, coal combustion residuals, aquatic, pollution, boron

## Background

Since May 2016, Taiwanese authorities have launched energy transition and energy industry reforms, aiming at phasing out nuclear energy by 2025 and increasing the share of renewables in electricity generation to 20% to establish a low-carbon and economically efficient energy system that can sustain economic growth.[1,2] Nevertheless, during this transition period, coal-fired power plants (CFPPs) must still be operated to satisfy and maintain economic growth and quality of daily life for residents.

Globally, more than 6600 CFPPs serve an estimated 5 billion people and account for 46% of annual $CO_2$ emissions.[3] Coal combustion produces acid gases, fly ash (the most crucial component of the coal combustion residuals [CCRs]), particulate matter, and potentially hazardous trace metals/elements, which have detrimental effects on humans and wildlife.[4,5] Power plants are currently the dominant emitters of mercury, acid gases, and many toxic metals in the United States.[4] Data from the United States Environmental Protection Agency (EPA) demonstrate that 62% of arsenic, 28% of nickel, 50% of

mercury, 22% of chromium, 13% of $NO_x$, and 60% of $SO_2$ that are toxic air pollutants come from power plants.[6]

Aquatic pollution from a CFPP is also a crucial environmental and public health issue. The coal ash generated from CFPPs can be classified as fly ash and bottom ash; most of it can be reused (in 2014, at least 46 million tons were reused in the United States alone), effectively reducing the environmental burden. Trace metal release from coal ash impoundment can be detrimental to the aquatic environment. In a study conducted to investigate the levels of Se, As, and Hg in the tissue of fish inhabiting water influenced by fly ash, to which the fish have a moderate to high risk of exposure, tissue levels of Se and arsenic were found to be elevated.[7] Coal combustion residuals contain variable amounts of trace metals that can negatively impact the aquatic environment. Coal combustion residual contamination affects mixture pollution as well and CCR can cause multi-element enrichment among a spectrum of trace elements in the water food web.[8] The analysis of metal concentrations and leachability of CCR from 7 CFPPs in Florida, USA, demonstrates that trace element concentrations and the leachability of CCRs depend on soil pH, particularly in the case of As, Ba, Cd, Cr, Pb, and Se.[9]

We performed a literature review on studies examining the aquatic trace metals related to the CCR dumping sites and CFPPs in Taiwan, an island country that has expressed increasing awareness of environmental preservation and environmental hazards to general health. Regrettably, only a few studies have focused on coal-fired power-plant-related pollution of the aquatic or marine environment.

In the past 2 decades, pollution sources such as accidental or deliberate industrial wastewater discharge, or certain cases of heavy metal pollution, eg, cadmium (Cd) or chromium (Cr) pollution on farmland or in certain rivers in Southern Taiwan, have been identified and reported.[10–13] However, the status of trace element identification and quantification in the aquatic environment around the world's second largest CFPP and its CCR dumping sites in Central Taiwan remains unexplored. It is notable that organic chemicals, trace metals, or metalloids, such as As, Cr, Cd, Ni, and Pb, would cause more environmental contamination.[14] Regular monitoring and publicizing of the results of the potentially toxic metal/element concentration in the aquatic environment related to a CFPP are not the usual practice in the region, and no similar research action has yet taken that approach. We postulated that there would exist particular trace element contamination in the abovementioned aquatic environments. To demonstrate this, we performed a cross-sectional survey of the aquatic trace element concentrations in the west coast of Central Taiwan in the CFPP cooling channel and drainage ports and from the CCR dumps, in addition to 2 background sources of seawater for comparison. The data were analyzed to infer the primary sources of the pollutants. The research results will hopefully encourage policymaking in the marine environment protection and elicit more research in this area.

Taichung CFPP is the world's second largest operating power plant with an annual coal consumption of 21 million



**Figure 1.** Locations of the 12 aquatic samples collected are pinpointed in the Google map for trace element contamination survey in Central Taiwan (Map data ©2018 Google), including 3 near the Taichung CFPP, 5 from the Changhua CCR disposal site, 2 from the historic CCR disposal site, and 2 from the background seawater (intertidal zone and Yunlin oyster farm). CCR indicates coal combustion residual; CFPP, coal-fired power plant.

tons; in 2018, this is scheduled to be reduced to 16 million tons. In 2015, coal ash production reached 2007 million tons, of which 1807 million tons were reused in civil construction.[15] Another 10% of CCRs were disposed of at selected sites. Unlike $PM_{2.5}$ in ambient air, the environmental impact of aquatic contamination on resident health and wildlife, and the toxicity profile of the trace metals/elements leached from the CCR disposition sites in Taiwan have not yet been researched well. Our exploratory cross-sectional analysis of trace elements in aquatic samples using a reliable methodology may provide a glimpse into this issue and help stimulate further research.

## Methods

### Study period

We chose the summer as the target research period for the cross-sectional survey because the annual demand for electricity reaches the highest point at this time. This will translate into the highest output of power, and thereby its byproducts, from the Taichung CFPP.[16] During the summer, the Taichung CFPP becomes the primary source of electricity and operates to its maximum capacity.[16] The specimen sampling period was from September 24 to October 3, 2017.

### Sampling

This cross-sectional field study selected 5 major sites, spanning 3 administrative regions, from Taichung City to Changhua County and Yunlin County, for aquatic trace element research:

the CFPP cooling channel and drainage ports; Changhua CCR disposal site; historic CCR disposal site; an intertidal zone 3.5 km southwest of Changhua CCR disposal site; and Yunlin oyster farm.

The Changhua CCR disposal site, which is located ~8 km southeast of the Taichung CFPP, is part of a large reclaimed land site where solid and liquid waste is deposited and is to be used as an expanded industrial site. The disposal site was constructed as a landfill with a human-made material-lined bottom, designated for final CCR disposal. Although leaching of hazardous substances into groundwater is highly suspected, thus far, there is no academic evidence of this occurrence. Solid CCR is delivered to the CCR disposal site by truck, and coal ash slurry (water and ash) is pumped from the power plant to the site via an ~300-mm-diameter steel pipe.

*Sample characteristics* (Supplemental Table S1). In total, 12 unfiltered aqueous samples were collected from 12 locations (refer to Figure 1) using the US EPA Method 1669.[17] Three aquatic samples from Taichung Power Plant (samples 10-BGA, 11-BGC, and 12-AY; Figure 2A), 2 effluent water samples (samples 6-BG and 8-CD), and 3 water samples (samples 4-S2, 5-00, and 7-OD) were collected from the Changhua CCR disposal sites (Figure 2B), located ~8 km southeast of Taichung Power Plant. The effluent water defined in this study was aqueous samples obtained from the steel pipes discharging coal ash slurry (water and ash), pumped from the Taichung CFPP to the CCR disposal site. A 2-L aqueous effluent was allowed to stand for 3 minutes, and the aqueous part was separated for further analysis. Aqueous samples obtained from surface water bodies other than the seawater, river, and the abovementioned effluent water were interchangeably termed water samples in this study. In addition, 1 seawater background sample was collected from the intertidal zone, located ~3.5 km southwest of the Taichung Power CCR disposal site (sample
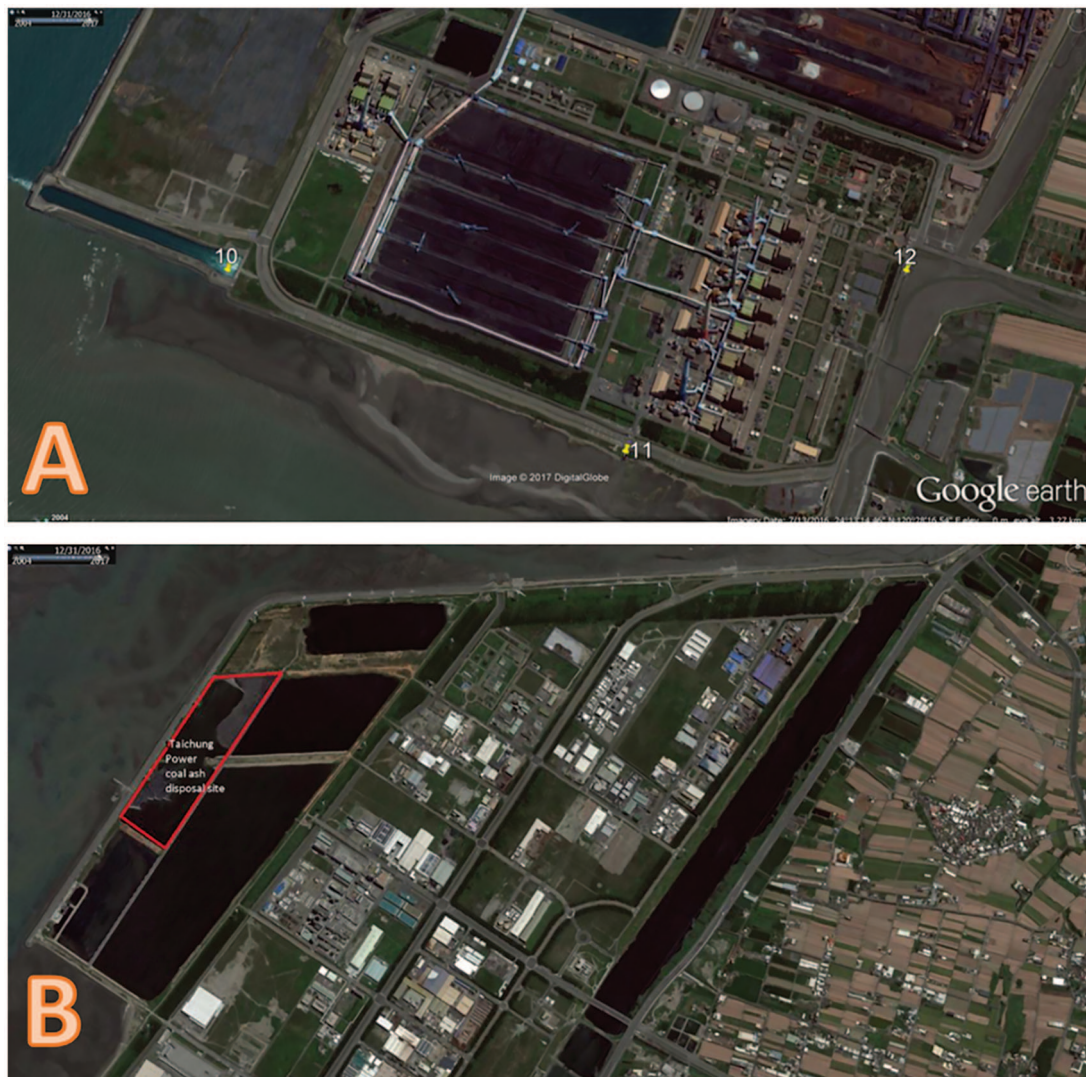


**Figure 2.** Google satellite map (Map data ©2018 Google) showing (A) the Taichung coal-fired power plant where aqueous samples were obtained from No. 10 (Sample 10-BGA), seawater sample from the power plant seawater cooling channel; No. 11 (Sample 11-BGC), a freshwater sample from the power plant southern discharge port; and No. 12 (Sample 12-AY), a freshwater sample from the power plant's eastern discharge port. (B) The location in Changhua considered the coal combustion residual (CCR) disposal site.

9-BGS), and 2 aquatic samples were taken from a historical CCR disposal site, located ~17 km south of the background sample site (samples 1-BG1 and 2-BG2; Figure 1). Finally, the seawater sample (sample 3-S1) was collected from an oyster farm in Yunlin, located 3.5 km south of the Formosa Plastics oil refinery and naphtha cracking plant (sample 3) (Figure 1). Prior research has identified that the Pacific oyster (*Crassostrea gigas*) can accumulate relatively high levels of copper (Cu), zinc (Zn), and lead (Pb), rendering it a potential biomonitor of these trace elements in aquatic ecosystems.[18] It is the purpose of the research to regard the seawater samples, numbered 9-BGS and 3-S1, as the background control samples.

Sample 10-BGA was collected from the seawater cooling channel of the power plant, which releases discharges into the sea from a submerged port. Sample 11-BGC was collected from an area inside the plant's southern discharge port, which drains into the Dadu River mouth Wildlife Reserve, and sample 12-AY was collected from an area adjacent to the eastern drainage port, which drains into a diversion channel that flows to the Dadu River.

### Chemical analysis

For aqueous samples, both the preparation and analytic methods followed either EPA 200.7 or 200.8 standard.[19] We used inductively coupled plasma optical emission spectrometry (ICP-OES), also referred to as atomic emission spectrometry (AES), in radial and axial modes to determine the boron and iron levels in aquatic samples, in accordance with the US EPA Method 200.7 Version 4.4. A Thermo Scientific iCAP 6500 Duo ICP-OES charge injection device (CID) spectrometer (Thermo Electron Limited, Cambridge, UK) was used for this analysis.[20] In brief, ICP-OES is based on the measurement of atomic emission by an optical spectrometric technique. The aquatic samples were nebulized, and the aerosol produced was directed to the plasma torch where desolvation and excitation took place. Argon gas was used to create the plasma. The atomic line emission was produced by radio-frequency inductively coupled plasma. A grating spectrometer then dispersed the spectra, and line intensities were monitored by a photosensitive CID detector. Photocurrents were processed and controlled by a computer system. To compensate for the variable background contribution during the determination of the trace elements, a background correction technique was applied.

All other trace elements were determined and quantitated using inductively coupled plasma mass spectrometry (ICP-MS) with the Agilent 7700 ICP-MS (Santa Clara, CA, USA), in accordance with the EPA Guidelines Method 200.8 for bioanalytical method validation. Method 200.8 is a well-established method, promulgated by the EPA for the analysis of aquatic samples using ICP-MS.[21] The technology couples the ICP system to mass spectrometry with the ions from the plasma extracted through a differentially pumped vacuum interface into a quadrupole mass spectrometer. The ions are separated by their mass-to-charge ratio, and a Faraday detector receives ion information proportional to the concentration. The concentration of a sample is then determined through calibration and processed by the data handling system.

*Testing*. pH, temperature, and 13 trace metals (limit of detection [LoD]), including aluminum (10 μg/L), arsenic (0.50 μg/L), boron (50.0 μg/L), cadmium (0.080 μg/L), chromium (total; 0.50 μg/L), cobalt (0.50 μg/L), iron (50.0 μg/L), lead (0.10 μg/L), manganese (0.50 μg/L), selenium (0.50 μg/L), strontium (0.50 μg/L), thallium (0.10 μg/L), and vanadium (1.0 μg/L) were tested.

### Sediment sampling and analysis

Sediment samples were collected from 3 sampling sites: two at the Changhua CCR disposal site, approximately 8 km southeast of the Taichung CFPP, and one from a historical CCR disposal site, approximately 17 km south of the CFPP. We collected approximately 100 g of solids from the top 2 cm of bottom sediments using a plastic scoop. Before elemental analysis, we adopted the EPA 3050 guidelines on preparing soils for acid digestion. For reference values, we prioritized the use of the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life developed by the Canadian Council of Ministers of the Environment. For the interpretation of trace element concentration of sediment in the context of aquatic life protection, we adopted the levels from the interim marine sediment quality guidelines (ISQGs), which are equivalent to the threshold effect level (TEL), defined as the threshold level below which adverse biological effects rarely occur.

### Quality control of the chemical analyses

Quality control of the chemical analyses was implemented using the blank method, performed through the complete preparation and analytical procedure. The blank spike duplicate (BSD) was performed in duplicate to determine if the procedure was working within established control limits. The recoveries of the spiked analytes were evaluated to determine accuracy. Comparison of the blank spike to the BSD was made to yield a precise measurement. The matrix spike duplicate (MSD) was analyzed to determine if the procedure was working within established control limits. The recoveries of the spiked analytes were evaluated to determine accuracy in a given matrix. Comparison of the matrix spike to the MSD was performed to estimate precision in a given matrix. Continuing calibration verification was used to determine if the sample analysis was within control limits.

### Statistical analysis

Results of trace element measurements are presented as mean, standard deviation (SD), minimum, median, and maximum

**Table 1.** Summary table of trace metal or element concentration in the aquatic environments including the Taichung coal-fired power plant vicinity, coal combustion residual disposal sites, intertidal zone seawater, and a distant oyster farm.

| TRACE METAL OR ELEMENT | N (%) DETECTED | MIN (MG/L) | MED (MG/L) | MAX (MG/L) | MEAN (MG/L) | SD | N (%) ABOVE EPA |
|---|---|---|---|---|---|---|---|
| Al | 8 (66.7) | 7.1 | 98.1 | 22 200 | 2105.4 | 6350.1 | 4 (33.3) |
| As | 5 (41.7) | 0.35 | 0.35 | 27.3 | 5.0 | 9.1 | 0 (0) |
| B | 10 (83.3) | 35.4 | 2830 | 4550 | 2480.1 | 1821 | 8 (66.7) |
| Cd | 0 (0) | 0.057 | 0.057 | 0.057 | 0.057 | 1.45E−17 | 0 (0) |
| Cr (total) | 1 (8.3) | 0.35 | 0.35 | 42.3 | 3.85 | 12.1 | 0 (0) |
| Co | 3 (25) | 0.35 | 0.35 | 9.7 | 1.7 | 2.9 | 0 (0) |
| Fe | 10 (83.3) | 35.4 | 179.5 | 31 400 | 3373.7 | 8937 | 3 (25) |
| Pb | 3 (25) | 0.07 | 0.07 | 7.9 | 0.7 | 2.26 | 0 (0) |
| Mn | 9 (75) | 0.35 | 42.9 | 774 | 176.8 | 264.3 | 6 (50) |
| Se | 0 (0) | 0.35 | 0.35 | 0.35 | 0.35 | 5.8E−17 | 0 (0) |
| Sr | 12 (100) | 51.2 | 4775 | 7980 | 4423 | 2955.9 | 1 (8.3) |
| Tl | 0 (0) | 0.07 | 0.07 | 0.07 | 0.07 | 1.45E−17 | 0 (0) |
| V | 4 (33.3) | 0.71 | 0.71 | 76.4 | 8.8 | 21.7 | 3 (25) |

Abbreviation: EPA, Environmental Protection Agency.

values. In addition, the number detected and number beyond the regulation limits are displayed. A summary table of trace metal or element concentrations in aquatic environments, including the Taichung CFPP vicinity, coal combustion residual disposal sites, intertidal zone seawater, and a remote oyster farm, is displayed. When the analyte concentrations (data points) obtained were below the LoD of the method used, those "missing" data points were treated according to the recommendations by the US EPA Data Quality Assessment.[22] Spearman rank correlation coefficient (Rho) was adopted with a 95% confidence interval (CI) (Fisher *Z* transformed), measuring the strength of the relationship between pairwise trace elements determined from all aquatic sampling sites. A Rho value > 0.7 indicates a high correlation.

*Availability of data and materials*

The dataset supporting the conclusions of this article is included within the article and its additional file.

## Results

Out of the 13 trace elements examined, 7 were within the national limit or the environmental quality standards of other advanced countries. These were As, Cd, total Cr, Co, Pb, Se, and Tl, in which Cd, Se, and Tl were undetectable in all 12 aquatic samples (Table 1). In total, 6, 8, 3, 6, 1, and 3 samples were identified to be above the EPA limit for Al, B, Fe, Mn, Sr, and V, respectively (Tables 1 and 2 and Supplemental Table S1). The EPA limit values are listed in Table 2.

*Conditions of seawater trace elements in the 2 control regions*

The seawater sampled from the Yunlin oyster farm was tested for the 13 trace elements, among which only the level of boron (4550 μg/L) was identified to be above the Japanese environmental quality standards for human health (4.5-fold) but was still considered normal by the Canadian Water Quality Guidelines for the Protection of Aquatic Life (5000 μg/L; Tables 1 and 2). In the seawater from the intertidal zone (sample 9-BGS), only aluminum (217 μg/L) and boron (2680 μg/L) were slightly higher than the referenced standards (Table 2). It is noteworthy that the literature reports boron level measured in *seawater* to be between 4500 and 5000 μg/L.[23] Hence, from the available data, it is likely that these 2 control water samples do not identify any apparent influence from the nearby CFPP or the CCR disposal sites.

*Status of trace elements in the aquatic samples obtained from the periphery of the Taichung CFPP compared with the control surface seawaters*

Although the levels of aluminum, arsenic, boron, cadmium, total chromium, cobalt, lead, selenium, strontium, and thallium were within the regulation limit in all the aqueous samples obtained from the CFPP, the level of manganese was 96.4 μg/L in the effluent obtained from the eastern drainage port, which was 1.9-fold above the regulation limit, set at 50 μg/L (Figure 2A and Supplemental Table S1). Iron, manganese, and vanadium were detected at 4379, 625, and 11.3 μg/L, respectively,

**Table 2.** The quality standards on specific trace metals in the aquatic environment compared in different regions.

| TRACE METAL IN SURFACE WATER | TAIWAN NATIONAL LIMIT VALUE (μG/L) | UNECE STANDARD STATISTICAL CLASSIFICATION OF SURFACE FRESHWATER QUALITY FOR MAINTAINING AQUATIC LIFE (CLASS IV[a]) (μG/L) | CANADIAN WATER QUALITY GUIDELINES FOR THE PROTECTION OF AQUATIC LIFE (μG/L) | ENVIRONMENTAL QUALITY STANDARDS FOR HUMAN HEALTH, JAPAN (μG/L) | US SALT WATER CRITERION MAXIMUM CONCENTRATION (μG/L) | US FRESH WATER CRITERION MAXIMUM CONCENTRATION (μG/L) |
|---|---|---|---|---|---|---|
| Aluminum | N.A. | 5-75 | 100 | N.A. | N.A. | N.A. |
| Arsenic | 50 | 190-360 | 50 | ≤10 | 69 | 360 |
| Boron[b] | N.A. | N.A. | 5000 | ≤1000 | N.A. | N.A. |
| Cadmium | 10 | 1.1-3.9 | 1.8 | ≤10 | 42 | 3.7 |
| Chromium (total) | N.A. | 11-16 | 20 | ≤5 (hexavalent) | 1100 (hexavalent) | 565 (III + IV) |
| Cobalt | | | WHO: marine environment at 20 μg/L (for the protection of 99% of marine species with 50% confidence) and for the freshwater environment at 8 μg/L (for the protection of 95% of freshwater species with 50% confidence) (Kim et al. 2006 Geneva: World Health Organization, p. 93.) | | | |
| Iron | N.A. | N.A. | 300 | N.A. | N.A. | N.A. |
| Lead | 100 | 3.2-82 | 7 | ≤10 | 210 | 65 |
| Manganese | 50 | N.A. | 430 (long-term guideline) | ≤200 | N.A. | N.A. |
| Selenium | 50 | N.A. | 1 | ≤10 | 290 | 20 |
| Strontium | | A widely accepted upper limit of normal for strontium in seawater samples from both the cold North Atlantic area and the warm subtropical Caribbean area is <7800 μg/L (Angino et al. *Chemical Geology* 1966;1:145-153.) | | | | |
| Thallium | N.A. | N.A. | N.A. | N.A. | 1.7 (water and organisms) | N.A. |
| Vanadium | | | The Dutch environmental quality standard for vanadium in water is proposed at 1.2 μg/L for long-term exposure and 3.0 μg/L for short-term concentration peaks (Smit CE, 2012) | | | |

Abbreviations: N.A., not available; UNECE, United Nations Economic Commission for Europe; WHO, World health organization.
[a]Class IV—the hazardous substance concentrations may exceed the chronic values occasionally but should not lead to chronically toxic conditions, concerning either concentration, duration, or frequency.
[b]The measured boron level in the *seawater* has been reported to range between 4500 and 5000 μg/L.[23]

from the seawater cooling channel (Figure 2A and Supplemental Table S1).

*Results of trace elements in the aqueous samples from the CCR disposal site in Changhua compared with that from the historic CCR disposal site*

The effluents and water from the surrounding areas of these 2 main targeted research sites disclosed similar patterns of the potentially hazardous trace element contamination. In both sites, 6 trace elements (As, Cd, Cr, Pb, Se, and Tl) were of no concern. In an analysis of the effluents sampled from these 2 CCR disposal sites, marked increases in the levels of other trace elements were readily noted: aluminum—up to 22 200 µg/L; boron—up to 4550 µg/L; cobalt—9.7 µg/L (mildly elevated in one of the historic disposal sites); iron—up to 31 400 µg/L; manganese—up to 774 µg/L; strontium—up to 7980 µg/L; and vanadium—up to 76.4 µg/L. All trace elements with the highest concentration were detected in the historic CCR disposal sites except for manganese (Supplemental Table S1).

*Correlation between trace metals examined using Spearman rank correlation coefficient (Rho)*

When the arbitrary cut-off point estimate of Spearman Rho used to indicate high correlation is set to >0.7, boron is highly correlated with strontium (Rho = 0.94, 95% CI, 0.80-0.98). Iron is highly correlated with aluminum (Rho = 0.77, 95% CI, 0.35-0.93), lead (Rho = 0.71, 95% CI, 0.23-0.91), cobalt (Rho = 0.75, 95% CI, 0.32-0.93), and vanadium (Rho = 0.84, 95% CI, 0.52-0.95). In addition to iron, vanadium is highly correlated with lead (Rho = 0.84, 95% CI, 0.52-0.96) and cobalt (Rho = 0.90, 95% CI, 0.66-0.97). Arsenic is highly correlated with manganese (Rho = 0.78, 95% CI, 0.36-0.93) and cobalt (Rho = 0.76, 95% CI, 0.34-0.93). Manganese is highly correlated with cobalt (Rho = 0.74, 95% CI, 0.29-0.92; Table 3).

The results of the trace element concentration in the sediment retrieved from 3 CCR disposal sites are presented in Table 4. The levels of cadmium, chromium, copper, lead, and zinc from the sediment were all below ISQG levels, indicating no significant ecological risk. Although the levels of arsenic were above the ISQG (TEL), they were still well below the probable effect level (PEL) above which (41.6 mg kg$^{-1}$) adverse effects frequently occur.

**Discussion**

We have discovered that the levels of the 13 selected trace elements of the background surface seawater, sampled from the intertidal zone and oyster farm, are within the national regulation limits or the quality standard established by the EPA of at least one of the advanced countries. Unfortunately, there are some concerns regarding the unchecked effluents from either the historical or Changhua CCR disposal sites. Al, Fe, Mn, and V are particularly noticeable. Regrettably, Taiwan EPA has not

**Table 3.** Spearman rank correlation coefficient (Rho) with its 95% confidence interval (CI) (Fisher Z transformed) measuring the strength of the relation between 2 trace elements determined from pairwise regression analyses.

| SPEARMAN RHO (95% CI) | SR | MNa | B | V | AL | CR | FE | PB | CO |
|---|---|---|---|---|---|---|---|---|---|
| B | **0.94 (0.80 to 0.98)** P=.3 | −0.58 (−0.87 to 0.008) P=.14 | | | | | | | |
| V | 0.70 (0.21 to 0.91) P=.3 | P=.1 | P=.4 | | | | | | |
| Al | P=.3 | P=.17 | P=.4 | P=.06 | | | | | |
| Cr | P=.06 | P=.25 | 0.58 (−0.003 to 0.86) | 0.57 (−0.003 to 0.86) P=.05 | | | | | |
| Fe | 0.58 (0.007 to 0.87) P=.47 | P=.35 | | **0.84 (0.52 to 0.95)** | **0.77 (0.35 to 0.93)** P=.05 | P=.06 | | | |
| Pb | P=.44 | P=.08 | | **0.84 (0.52 to 0.96)** P=.2 | P=.2 | 0.63 (0.09 to 0.89) | **0.71 (0.23 to 0.91)** | | |
| Co | P=.44 | **0.74 (0.29 to 0.92)** | | **0.90 (0.66 to 0.97)** P=.3 | 0.59 (0.02 to 0.87) | 0.63 (0.09 to 0.88) | **0.75 (0.32 to 0.93)** | 0.60 (0.04 to 0.87) | |
| As | P=.3 | **0.78 (0.36 to 0.93)** P=.18 | | 0.61 (0.06 to 0.88) P=.18 | P=.1 | P=.08 | 0.57 (−0.0007 to 0.86) P=.18 | P=.18 | **0.76 (0.34 to 0.93)** P=.18 |

Bold values represent the point estimate of Rho value > 0.7 indicating a high correlation. Cd, Se, and Tl were not included due to being under the limit of detection.
aNo significant correlation between Mn and Sr (P=.1).

**Table 4.** Trace element concentration in the sediment retrieved from 3 coal combustion residual (CCR) disposal sites.

| SITE | CHANGHUA CCR DISPOSAL SITE | CHANGHUA CCR DISPOSAL SITE | HISTORIC CCR DISPOSAL SITE | REFERENCE VALUE FOR SEDIMENTS |
|---|---|---|---|---|
| Sample ID | B | E | 1 | |
| Corresponding to the aquatic sample in Supplemental Table S1 | 5-00 | 8-CD | 1-BG1 | |
| Date | 10/03/17 | 10/03/17 | 9/24/17 | |
| Time | 10:15 | 11:00 | 10:00 | |
| pH | 8.3 | 9.1 | – | |
| Temperature (°C) | 32.7 | 32.9 | – | |
| Matrix | Solid | Solid | Solid | |
| Percent moisture | 43.7 | 46.7 | 60.8 | |
| Latitude (decimal degree) | 24.15425 | 24.15127778 | 23.98128889 | |
| Longitude (decimal degree) | 120.4269444 | 120.4247222 | 120.3441667 | |
| Unit[a] | mg/kg av.6010/6020 | mg/kg av.6010/6020 | mg/kg av. MET 6010/6020 | |
| Aluminum | 9580 | 20 700 | 15 700 | No guideline[b] |
| Arsenic (11-33 mg/kg)[c] | 13.9 | 18.3 | 16.8 | 7.24 |
| Boron | 59.0 | 538.0 | 53.0 | No guideline |
| Cadmium (0.65-2.49 mg/kg)[c] | <0.12 (undetectable) | 0.28 | <0.16 (undetectable) | 0.7 |
| Chromium (total) (76-233 mg/kg)[c] | 23.7 | 24.8 | 24.3 | 52.3 |
| Cobalt | 5.8 | 16.3 | 11.2 | 20[d] |
| Copper (50-157 mg/kg)[c] | 16.3 | 23.2 | 15.0 | 18.7 |
| Iron | 18 600 | 15 000 | 27 200 | No guideline |
| Lead (48-161 mg/kg)[c] | 16.4 | 14.1 | 19.4 | 30.2 |
| Manganese | 196 | 186 | 410 | 152–1152 (East China Sea)[e] |
| Selenium | 4.4 | 6.2 | 2.0 | No guideline |
| Strontium | 73.8 | 501.0 | 119 | No guideline |
| Thallium | <1.2 (undetectable) | Not measured | <1.6 (undetectable) | No guideline |
| Titanium | 99.8 | 711 | 133 | 4160[d] |
| Vanadium | 29.5 | 75.5 | 24.7 | 170[d] |
| Zinc (140-384 mg/kg)[c] | 71.0 | Not measured | 78.3 | 124 |

Abbreviation: EPA, Environmental Protection Agency.
[a]Results are reported on a "dry weight" basis and adjusted for percent moisture, sample size, and any dilutions.
[b]For the reference values, we prioritize the use of the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life developed by the Canadian Council of Ministers of the Environment. In the context of aquatic life protection, we adopt the interim marine sediment quality guidelines (ISQGs) level which is equivalent to the threshold effect level (TEL) defined as the threshold level below which adverse biological effects rarely occur.
[c]Taiwan EPA established reference values of various metal concentrations in the sediments to represent the sediment quality.
[d]Value is 'World's average suspended sediments' from the publication by Alagarsamy R and Zhang J. Geochemical characterization of major and trace elements in the coastal sediments of India. *Environ Monit Assess*. 2010;161:161-176.
[e]Derived from the paper by Fang TH et al. Distribution and contamination of trace metals in surface sediments of the East China Sea. *Marine Environ Res* 2009;68:178-187.

set any regulation limit yet for trace elements such as aluminum, iron, cobalt, and vanadium in the aquatic environment. Background values of these trace elements determined mainly by local geochemical factors should be established first. Risks

of contamination of air, soil, and aquatic ecosystems associated with trace element leachates or other forms of contamination are closely linked to biogeochemical conditions, material leachability, and contaminant mobility. In evaluating the possible

impact of different biogeochemical conditions on the mobility of hazardous trace elements, one should consider the maintenance of acid and oxidative conditions, the sustainability of biogeochemical processes affecting element transport, leachate composition, material leachability, metal solubilization, geochemical characteristics of sediment, and basic soil geochemistry (eg, pH, Fe, Mn).[24-29] Leaching simulations performed in the laboratory identified that the pH and ash/water ratio determine the CCR contaminant abundance and geochemical composition of the impacted water.[25] In a study of fly ash from Koradi and Khaperkheda, thermal power plants located in India, disposal is performed in an area surrounded by many small villages whose residents use the groundwater for drinking and domestic purposes. The investigators discovered relatively high concentrations of Mo, As, B, and F in circulating waters, associated with the leaching from fly ash. This has a direct spatial correlation between the concentration of fluorides in the groundwater and their closeness to the ash ponds.[30]

High concentrations of Al and Fe (up to 222-fold and 104.7-fold, respectively, above the quality standard set by the Canadian Water Quality Guidelines), Mn (up to 6-fold above the national regulation limit), and V (up to 25-fold above the Dutch quality standard proposed for short-term concentration peaks) in the effluent water collected from one of the CCR disposition sites indicate that contamination of the surrounding aquatic environment via metal leachability could be an issue.

Most of the manganese metals enter bottom and fly ash in the combustion process at CFPPs.[31,32] Manganese overexposure could lead to several neurologic disorders, such as Parkinson disease, as Mn is deposited in the basal ganglia causing neurodegeneration.[33] The Canadian Water Quality Guidelines for the Protection of Aquatic Life set a long-term exposure limit value at $430\,\mu g/L$ for dissolved Mn. The highest abnormal result of the 3 abnormal aqueous samples was $774\,\mu g/L$, detected at a Changhua CCR disposal site. We think it is necessary to conduct further study to control the release of Mn into the wildlife environment.

Whereas the boron concentration in the background seawater (sample IDs: 9-BGS and 3-S1; Supplemental Table S1) may reflect geogenic sources, namely, seawater, the high concentration in other effluent aqueous samples from the CCR dumping can only be explained by anthropogenic activities, particularly the byproducts from the CFPP nearby. Boron and strontium exhibited a very strong correlation coefficient (Spearman Rho = 0.94). The potential toxicities to human after significant exposure to boron (from boric acid [$H_3BO_3$ containing 17.48% boron] and sodium borates [$B_4Na_2O_7$ containing 11.34%-14.85% boron]) have been under investigation, mainly regarding reproductive toxicity and potential non-inheritable adverse effects on the development of offspring.[23,34] Coal combustion residuals comprise fly ash, bottom ash, flue gas desulfurization products, and boiler slag, which are typically enriched in toxic elements such as As, B, Se, Hg, Mn, and

Sr.[35] It has been documented that the ecosystems surrounding rivers are sensitive to fly ash spilled into rivers, with higher concentrations of arsenic, iron, manganese, and selenium found in turtles from rivers affected by spills.[36] Nonetheless, no research report examining the implications of aluminum, boron, iron, manganese, and vanadium in Taiwan has been reported.

Strontium (Sr) is an alkaline metal; once in the environment, it is mostly present as the divalent cation $Sr^{2+}$ or various complexes, depending on the physicochemical characteristics of the environment.[37] In the United States, CCRs are the most abundant industrial waste; they are mainly stored in surface impoundments and landfills.[38] Harkness et al,[38] in their recent work on evaluating possible leaks from coal ash ponds on the geochemistry of seeps and surface water from 15 sites in 5 US states (Tennessee, Kentucky, Georgia, Virginia, and North Carolina), identified that strontium and boron exceeded the background values of 100 and $150\,\mu g/L$, respectively, at all sites.

There are several limitations in this study. Serial monitoring over time and more extensive sampling of different aquatic sources surrounding the CFPP were not available due to resource limitations. Moreover, although the impact on public health is potentially recognizable, this pilot screening survey is not feasible for model building.

Contamination of aquatic systems with trace metals such as those in this report is a significant public health problem worldwide.[39–41] Coastal trace metal contamination has been a long-standing environmental protection issue, and the long-term consumption of seafood contaminated with metals has been reported as an important route of human exposure to abnormally high trace metals in Taiwan, where fish is consumed on a large scale.[42,43] The elevated concentrations of trace metals identified indicate the introduction of pollution into the wetland from human activities such as agricultural non-point sources,[39] urban wastewater treatment, and other possible sources such as a CFPP. The Taichung Power Plant wastewater discharge ports should be the focus of continued monitoring and water sampling to determine the frequency and magnitude of trace metal contamination over time. To inform environmental health policymaking in a time-sensitive context, the results of this aquatic pollution pilot survey can raise the awareness of certain trace elements that could potentially have an adverse impact on human health and wildlife through aquatic contamination.

The potential health impact from toxic-level trace element exposure is one of the primary rationales for our research. In recent years, there have been many studies examining the association between certain trace metals and type 2 diabetes, including metabolic syndrome.[44–47] Although the exact mechanisms are yet to be revealed, human overexposure to trace metals is still believed to induce oxidative stress, damage, and inflammation in human tissues. A report on the Chinese residents of Wuhan, China has identified that urinary aluminum, titanium, cobalt,

nickel, copper, zinc, selenium, rubidium, strontium, molybdenum, cadmium, antimony, barium, tungsten, and lead were significantly associated with increased risk for altered fasting plasma glucose, impaired fasting glucose, and diabetes, after adjusting for potential confounders.[47] This Wuhan study suggests that human metabolism, such as glucose homeostasis, could be altered by certain trace metal overexposure.

The disposal of CCR is generally conducted in surface impoundment or landfill ponds, after it is mixed in certain appropriate proportions of water to form a slurry. Accidental leaching of CCR effluent into aquatic ecosystems would cause deleterious environmental effects impacting human health, either through direct exposure to contaminated water or indirectly via exposure of CCR in the food web.[48] Some of the abundant metals and trace elements present in CCR are readily taken up at high concentrations by aquatic organisms. A previous study using biota dosimetry, rather than water dosimetry, to evaluate heavy metal contamination status in the aquatic ecosystems, was performed along the inshore area of Changhua.[18] The study identified that the soldier crab (*Mictyris brevidactylus*) in aquatic ecosystems accumulated relatively high levels of Cu, Zn, Pb, and Ni. The relevance of this study is that the data may be extrapolated to a spectrum of potentially hazardous trace elements derived from CCR, which may be taken up into aquatic food web and ultimately consumed by humans. Coal combustion residual exposure may also lead to numerous histopathological, behavioral, and physiological (reproductive, energetic, and endocrinological) effects in certain vertebrates and invertebrates.[48,49]

Moreover, in regions with dense populations, CCR is often stored in landfills and slurry ponds located near residential areas, indicating a high potential for increased health risks. An interesting community-based study involving 4 neighborhoods adjacent to a large coal ash storage facility in the United States was conducted by the University of Louisville School of Public Health, using focus groups to identify themes regarding children's exposure to coal ash and resultant children's health.[50] Most of the parents (85%) self-reported that their children suffered from respiratory, emotional, and behavioral disorders and strongly implied that exposure to coal ash from the landfill was the cause of their children's health issues.[50] Studies such as these are lacking for the Taichung CFPP's CCR disposition in Taiwan. The authors believe that there is an excellent rationale for human and health research to gather local evidence in this regard.

## Conclusions

In this pilot screening survey, we researched the trace element contamination associated with the vicinity of the world's second largest CFPP and combustion residual residue disposition sites. The effluent from one of the drainage ports from the CFPP was identified with an increase in Mn concentration, nearly 2-fold above the national limit. The levels of Al, B, Fe, Mn, Sr, and V in the effluents from the CCR dumping sites, particularly the old historic disposition site, were a matter of some concern as well. Boron levels were markedly correlated with Sr levels in our analysis. The leachability issue is evident in this exploratory cross-sectional study on the effluent water obtained from the CCR dumping sites. This exploratory research will elicit further national coordinated efforts to establish a guideline for the effective control of point-source trace metal pollution and eagerly awaited regulation limits for those trace metals neglected in the past.

## Author Contributions

PRW, J-MY, and G-PY conceptualized and determined the sampling sites and timing of research. VCK, PRW, and J-MY analyzed and interpreted the data regarding the chemical analysis of trace elements. VCK was a major contributor in writing the manuscript. All authors read, made revisions, and approved the final manuscript.

## Availability of Data and Materials

All chemical analytical results are presented as a supplement document in Supplemental Table S1.

## ORCID iD

Victor C Kok (iD) https://orcid.org/0000-0003-3440-8154

## Supplemental Material

Supplemental material for this article is available online.

### REFERENCES

1. Lu W-C. Greenhouse gas emissions, energy consumption and economic growth: a panel cointegration analysis for 16 Asian countries. *Int J Environ Res Public Health*. 2017;14:1436.
2. Fell H-J. *Taiwan's New Energy Policy: Climate Protection and the Phasing-Out of Nuclear Power by Expanding the Scope of Renewable Energy* (Business & Diplomacy). Berlin, Germany: Diplomat Media; 2017.
3. Olivier JGJ, Peters JAHW, Janssens-Maenhout G, Muntean M. *Trends in Global CO2 Emissions (Report)*. Den Haag, The Netherlands; Ispra, Italy: PBL Netherlands Environmental Assessment Agency; Institute for Environment and Sustainability (IES), Joint Research Centre (JRC); 2013.
4. Kittner N, Fadadu RP, Buckley HL, Schwarzman MR, Kammen DM. Trace metal content of coal exacerbates air-pollution-related health risks: the case of lignite coal in Kosovo. *Environ Sci Technol*. 2018;52:2359–2367.
5. Arumugam S, Ramaiah S. Concentrations of toxic metals (Pb, Cd, Cr) in the tissues and their effects on diversification of *Devario aequipinnatus* populations. *Int J Environ Health Res*. 2018;28:379–390.
6. US Environmental Protection Agency. Cleaner power plants. https://www.epa.gov/mats/cleaner-power-plants. Up-dated 2019. Accessed March 5, 2019.
7. Reash RJ. Selenium, arsenic, and mercury in fish inhabiting a fly ash exposure gradient: interspecific bioaccumulation patterns and elemental associations. *Environ Toxicol Chem*. 2012;31:739–747.

8.  Brandt JE, Simonin M, Di Giulio RT, Bernhardt ES. Beyond selenium: coal combustion residuals lead to multielement enrichment in receiving lake food webs. *Environ Sci Technol*. 2019;53:4119–4127.

9.  da Silva EB, Li S, de Oliveira LM, et al. Metal leachability from coal combustion residuals under different pHs and liquid/solid ratios. *J Hazard Mater*. 2018;341:66–74.

10. Lu LT, Chang IC, Hsiao TY, Yu YH, Ma HW. Identification of pollution source of cadmium in soil: application of material flow analysis and a case study in Taiwan. *Environ Sci Pollut Res Int*. 2007;14:49–59.

11. Tsai LJ, Yu KC, Ho ST. Cadmium distribution in sediment profiles of the six main rivers in southern Taiwan. *J Hazard Mater*. 2007;148:630–639.

12. Dong CD, Chen CW, Chen CF. Distribution and contamination status of chromium in surface sediments of northern Kaohsiung Harbor, Taiwan. *J Environ Sci (China)*. 2013;25:1450–1457.

13. Lin YC, Chang-Chien GP, Chiang PC, Chen WH, Lin YC. Multivariate analysis of heavy metal contaminations in seawater and sediments from a heavily industrialized harbor in Southern Taiwan. *Mar Pollut Bull*. 2013;76:266–275.

14. Barbieri M, Sappa G, Nigro A. Soil pollution: anthropogenic versus geogenic contributions over large areas of the Lazio region. *J Geochem Explor*. 2018;195: 78–86.

15. Taiwan Power Company. *2016 Taiwan Power Company Sustainability Report*. Taipei, Taiwan: Taiwan Power Company; 2016.

16. Fang GC, Xiao YF, Zhuang YJ, Cho MH, Huang CY, Tsai KH. PM2.5 particulates and metallic elements (Ni, Cu, Zn, Cd and Pb) study in a mixed area of summer season in Shalu, Taiwan. *Environ Geochem Health*. 2017;39:791–802.

17. Office of Water, US Environmental Protection Agency. *Method 1669: Sampling Ambient Water for Trace Metals at EPA Water Quality Criteria Levels: Draft*. Washington, DC: Office of Water, US Environmental Protection Agency; 1996.

18. Yeh HC, Chen IM, Chen P, Wang WH. Heavy metal concentrations of the soldier crab (*Mictyris brevidactylus*) along the inshore area of Changhua, Taiwan. *Environ Monit Assess*. 2009;153:103–109.

19. Gibbs RJ, Guerra C. Metals of the bottom muds in Belize City Harbor, Belize. *Environ Pollut*. 1997;98:135–138.

20. Pancras JP, Norris GA, Landis MS, Kovalcik KD, McGee JK, Kamal AS. Application of ICP-OES for evaluating energy extraction and production wastewater discharge impacts on surface waters in Western Pennsylvania. *Sci Total Environ*. 2015;529:21–29.

21. Dehouck P, Cordeiro F, Snell J, de la Calle B. State of the art in the determination of trace elements in seawater: a worldwide proficiency test. *Anal Bioanal Chem*. 2016;408:3223–3232.

22. US Environmental Protection Agency. *Data Quality Assessment: Statistical Methods for Practitioner* (EPA QA/G-9S). Washington, DC: US Environmental Protection Agency; 2006:1–198.

23. Nigro A, Sappa G, Barbieri M. Application of boron and tritium isotopes for tracing landfill contamination in groundwater. *J Geochem Explor*. 2017;172: 101–108.

24. Uddh Soderberg T, Berggren Kleja D, Astrom M, et al. Metal solubility and transport at a contaminated landfill site—from the source zone into the groundwater. *Sci Total Environ*. 2019;668:1064–1076.

25. Ruhl L, Vengosh A, Dwyer GS, Hsu-Kim H, Deonarine A. Environmental impacts of the coal ash spill in Kingston, Tennessee: an 18-month survey. *Environ Sci Technol*. 2010;44:9272–9278.

26. Mavakala BK, Le Faucheur S, Mulaji CK, et al. Leachates draining from controlled municipal solid waste landfill: detailed geochemical characterization and toxicity tests. *Waste Manag*. 2016;55:238–248.

27. Ghayaza M, Le Forestier L, Muller F, Tournassat C, Beny JM. Pb(II) and Zn(II) adsorption onto Na- and Ca-montmorillonites in acetic acid/acetate medium: experimental approach and geochemical modeling. *J Colloid Interface Sci*. 2011;361:238–246.

28. Fonti V, Dell'Anno A, Beolchini F. Influence of biogeochemical interactions on metal bioleaching performance in contaminated marine sediment. *Water Res*. 2013;47:5139–5152.

29. de Souza Machado AA, Spencer KL, Zarfl C, O'Shea FT. Unravelling metal mobility under complex contaminant signatures. *Sci Total Environ*. 2018;622–623: 373-384.

30. Spadoni M, Voltaggio M, Sacchi E, et al. Impact of the disposal and re-use of fly ash on water quality: the case of the Koradi and Khaperkheda thermal power plants (Maharashtra, India). *Sci Total Environ*. 2014;479–480:159-170.

31. Deng S, Shi Y, Liu Y, et al. Emission characteristics of Cd, Pb and Mn from coal combustion: field study at coal-fired power plants in China. *Fuel Process Technol*. 2014;126:469–475.

32. Zhao S, Duan Y, Chen L, et al. Study on emission of hazardous trace elements in a 350 MW coal-fired power plant. Part 2. Arsenic, chromium, barium, manganese, lead. *Environ Pollut*. 2017;226:404–411.

33. Chen P, Culbreth M, Aschner M. Exposure, epidemiology, and mechanism of the environmental toxicant manganese. *Environ Sci Pollut Res Int*. 2016;23: 13802–13810.

34. Duydu Y, Basaran N, Ustundag A, et al. Is boric acid toxic to reproduction in humans? Assessment of the animal reproductive toxicity data and epidemiological study results. *Curr Drug Deliv*. 2016;13:324–329.

35. Ruhl LS, Dwyer GS, Hsu-Kim H, Hower JC, Vengosh A. Boron and strontium isotopic characterization of coal combustion residuals: validation of new environmental tracers. *Environ Sci Technol*. 2014;48:14790–14798.

36. Van Dyke JU, Jachowski CM, Steen DA, Jackson BP, Hopkins WA. Spatial differences in trace element bioaccumulation in turtles exposed to a partially remediated coal fly ash spill. *Environ Toxicol Chem*. 2017;36:201–211.

37. Boyer A, Ning P, Killey D, et al. Strontium adsorption and desorption in wetlands: role of organic matter functional groups and environmental implications. *Water Res*. 2018;133:27–36.

38. Harkness JS, Sulkin B, Vengosh A. Evidence for coal ash ponds leaking in the southeastern United States. *Environ Sci Technol*. 2016;50:6583–6592.

39. Jimenez-Ballesta R, Garcia-Navarro FJ, Bravo S, Amoros JA, Perez-de-Los-Reyes C, Mejias M. Environmental assessment of potential toxic trace element contents in the inundated floodplain area of Tablas de Daimiel wetland (Spain). *Environ Geochem Health*. 2017;39:1159–1177.

40. Mudila H, Prasher P, Kumar M, et al. An insight into Cadmium poisoning and its removal from aqueous sources by graphene adsorbents. *Int J Environ Health Res*. 2019;29:1–21.

41. Huang X, Hu J, Li C, Deng J, Long J, Qin F. Heavy-metal pollution and potential ecological risk assessment of sediments from Baihua Lake, Guizhou, P.R. China. *Int J Environ Health Res*. 2009;19:405–419.

42. Lin YJ, Ling MP, Chen SC, et al. Mixture risk assessment due to ingestion of arsenic, copper, and zinc from milkfish farmed in contaminated coastal areas. *Environ Sci Pollut Res Int*. 2017;24:14616–14626.

43. Ling MP, Hsu HT, Shie RH, Wu CC, Hong YS. Health risk of consuming heavy metals in farmed tilapia in Central Taiwan. *Bull Environ Contam Toxicol*. 2009;83:558–564.

44. Yang A, Liu S, Cheng N, et al. Multiple metals exposure, elevated blood glucose and dysglycemia among Chinese occupational workers. *J Diabetes Complications*. 2017;31:101–107.

45. Planchart A, Green A, Hoyo C, Mattingly CJ. Heavy metal exposure and metabolic syndrome: evidence from human and model system studies. *Curr Environ Health Rep*. 2018;5:110–124.

46. Li XT, Yu PF, Gao Y, et al. Association between plasma metal levels and diabetes risk: a case-control study in China. *Biomed Environ Sci*. 2017;30:482–491.

47. Feng W, Cui X, Liu B, et al. Association of urinary metal profiles with altered glucose levels and diabetes risk: a population-based study in China. *PLoS ONE*. 2015;10:e0123742.

48. Rowe CL, Hopkins WA, Congdon JD. Ecotoxicological implications of aquatic disposal of coal combustion residues in the United States: a review. *Environ Monit Assess*. 2002;80:207–276.

49. Kravchenko J, Lyerly HK. The impact of coal-powered electrical plants and coal ash impoundments on the health of residential communities. *N C Med J*. 2018;79:289–300.

50. Zierold KM, Sears CG. Community views about the health and exposure of children living near a coal ash storage site. *J Community Health*. 2015;40: 357–363.