Anderson County Board of Commissioners Intergovernmental Committee Agenda

January 13, 2020

Anderson County Courthouse Room 312 at 5:00 p.m.

- 1. Call to order
- 2. Bull Run potential health hazards for residents in close proximity of stored coal ash

Unfinished Business

New Business

Adjournment

Name - Email Address

1, BRADMCZ Ogmail.com

). SBTOODE PRODECY. NET

3. julie bledsoc Ome. com
p tabathabreeden a gmail. com

S. Kminault @gmail.com

6. wells. 37716@gmail.com

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Article

Respiratory Health in Adults Residing Near a Coal-Burning Power Plant with Coal Ash Storage Facilities: A Cross-Sectional Epidemiological Study

Department of Epidemiology and Population Health, University of Louisville, Louisville, KY 40292, USA

Department of Epidemiology, Brown University, Providence, RI 02912, USA

Department of Environmental Health Sciences, School of Public Health, University of Alabama at Birmingham, Birmingham, AL 35294, USA

Author to whom correspondence should be addressed.

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Abstract: Coal ash, the byproduct of burning coal made up of small particles, including heavy metals and radioactive elements, is discarded in openair landfills where it can be emitted into the air, contributing to air pollution in the surrounding community. Few regulations exist regarding the storage, disposal, and transport of coal ash. There is limited research on the health impacts of coal ash exposure on communities. The purpose of this study was to examine the prevalence of respiratory symptoms among adults exposed to coal ash and non-exposed adults. A cross-sectional epidemiological study was conducted among two populations: one exposed to coal ash and one not exposed to coal ash. Perception of health (p-Value < 0.0001), cough (Adjusted Odds Ratio (AOR) = 5.30, 95% Confidence Intervals (CI) = 2.60–11), shortness of breath (AOR = 2.59, 95% CI = 1.56–4.31), hoarseness (AOR = 4.02, 95% CI = 2.45–6.60), respiratory infections (AOR = 1.82, 95% CI = 1.14–2.89), and mean overall respiratory health score (p-Value < 0.0001) were all statistically significantly greater in exposed adults (N = 231) when compared to non-exposed adults (N = 170). Adults residing near the coal ash facility were more likely to report respiratory symptoms than the non-exposed population. More research on the health impact of coal ash and storage regulations needs to be conducted.

Keywords: coal ash; fly ash; coal combustion residuals; coal-burning power plants; respiratory health; air pollution; environmental health

1. Introduction

Coal is a combustible sedimentary rock that is largely formed from plant debris [1]. It is made up of carbon, hydrogen, oxygen, nitrogen, and sulfur, as well as small amounts of various metals and radioactive materials [1,2]. Coal is mined to meet the needs of the world's increasing demand for fuel.

Despite efforts to increase natural gas production, coal combustion power plants continue to play a major role in electricity production [1,3]. Coal-burning power plants generate approximately 27% of the electricity in the United States [4].

Coal combustion residuals (CCR), the byproduct of coal combustion, consist of small particles that contain naturally occurring radioactive materials, polycyclic aromatic hydrocarbons, and a variety of heavy metals, including aluminum, arsenic, iron, lead, and mercury [5,6,7,8,9,10,11,12]. In 2017, the United States produced nearly 111 million tons of CCR, more commonly known as coal ash [13]. Coal ash is not coal dust, which is the term used to refer to a type of pollution generated from the mining of coal. The composition and particle size of coal ash are different compared to coal dust.

Coal ash is an overarching term that can include flue gas desulfurization solids, boiler slag, bottom ash, and fly ash [3]. During the coal combustion process, small spherical particles measuring ≤10 µm in diameter ascend up the stack and are collected on filters. These small particles are known as fly ash and account for nearly 40%–70% of coal ash product [3,14]. Although some effort is being made to reuse coal ash, in 2017, only 64% of coal ash was recycled and used in products such as cement [13]. The coal ash that is not reused gets stored in designated landfills and ash ponds where it becomes a likely source of pollution [15]. Fugitive fly ash emissions from coal ash storage facilities may be a significant contributor to the concentration of ambient air particulate matter (PM) [16,17].

Although studies on coal ash exposure and human health are lacking, several researchers have investigated the occupational health hazards of coal ash. Two researchers found that coal ash workers exposed to high arsenic levels had higher malignancy-caused death and higher rates of cancer mortality when compared to coal ash workers exposed to "normal" levels of arsenic [18,19]. More recent studies sought to examine occupational health hazards of working at a fly ash treatment plant compared to a bottom ash recovery plant. One researcher found that when compared to bottom ash workers, workers in the fly ash treatment plant had significantly higher plasma malondialdehyde [20]. Another researcher found that fly ash treatment plant workers had more DNA damage when compared to their bottom ash counterparts [21]. Although a few occupational health studies have been conducted, epidemiologic studies investigating the impact coal ash has on the surrounding community are limited [22,23]. Since coal ash landfills and ponds are maintained in residential communities, it is important to understand community exposure.

The purpose of this study was to assess the prevalence of respiratory symptoms and illness among adults living in neighborhoods surrounding a coal-burning power plant with coal ash storage facilities compared to adults living in a non-exposed environment. The exposed population lives in Louisville, Kentucky, an area that is consistently ranked by the American Lung Association in the top 25 cities in the United States for the most particulate matter pollution in the country. We hypothesized that the adults residing in neighborhoods near the coal-burning power plant with coal ash storage facilities would have poorer respiratory health and more respiratory symptoms than adults who are not exposed to coal ash. By characterizing respiratory symptoms in the populations, our analysis begins to fill the gap in the research literature regarding the potential health effects of coal ash among adults.

2. Methods

In this analysis, we used a cross-sectional questionnaire which was developed by the research team in conjunction with community members living near the coal-burning power plant. This questionnaire was used to identify respiratory symptoms and health in an exposed and non-exposed comparison population. This study was a community-based mixed methods study (focus groups and a cross-sectional epidemiological study) that took place in four neighborhoods surrounding a coal-burning power plant with coal ash storage facilities in Southwest Louisville, Kentucky [22,23]. The exposed neighborhoods that were included in this study were all within 1/4 to 1/2 miles of the storage facility. The storage facility, located just east of the Ohio River, is home to one large coal ash landfill and five ponds, two of which are known to store coal ash. The landfill was last estimated to have an elevation of over 500 feet and a surface area of 110 acres [24]. The main coal ash storage pond, which is located just 1200 feet from the Ohio River, has a surface area of approximately 40 acres [25]. To compare the prevalence of respiratory symptoms and illness in the coal ash exposed population, a non-exposed population without coal-burning power plants or coal ash storage facilities located in Orange County, Indiana, was chosen.

All methods were approved by the University of Louisville Institutional Review Board (IRB#: 09.0141). Participants were provided a preamble prior to completing the questionnaires. Adults who finished the questionnaire and submitted it to the research team were considered to have consented.

2.1. Study Populations

2.1.1. Exposed Group

Exposure was defined as residing in one of the four neighborhoods adjacent to the coal ash facility. The study encompassed a community-based research design in which residents were recruited from four neighborhoods surrounding a large coal-burning power plant. The four neighborhoods included in the study span two zip codes. According to the U.S. Census in 2010, the first zip code region was home to 40,746 people; 63% were white, 76.3% were 18 years or older, 14.3% were 65 years or older. Additionally, the average household size was 2.41. According to the 2010–2014 American Community Survey 5-Year Estimates, 37.9% of those 25 years and older were at least high school graduates; the poverty level among these same individuals was 15.7%.

The 2010 U.S. Census reported that the second zip code region was made up of 26,465 people; 86.6% were white, 75.1% were 18 years or older, 13.6% were 65 years or older, and the average household size was 2.54. According to the 2010-2014 American Community Survey 5-Year Estimates, 44.1% of persons 25 years and older were at least high school graduates, 7.5% were living in poverty.

2.1.2. Non-Exposed Comparison Group

A non-exposed comparison population was chosen from Orange County, Indiana, approximately 60 miles from Louisville, Kentucky. Orange County was chosen because its population has similar demographics and it is rurally situated, without close proximity to a coal-burning power plant or coal ash storage facilities. In 2010, the U.S. Census reported that of the 19,840 residents of Orange County, Indiana, 98.2% were white, 75.4% were 18 years or older, and 15.8% were 65 years or older. Additionally, the average household size was 2.49. Of the people 25 years and older, 45.1% were at least high school graduates and 12.2% were living in poverty.

2.2. Collection of Data

Data were collected from a cross-sectional survey of exposed and non-exposed adults. The questionnaire for the survey was developed by the research team based on results from five focus groups that were held with 26 adult community members that lived near a coal-burning power plant with coal ash storage facilities [22,23]. In addition to the health conditions and symptoms that resulted from the focus group discussions, health conditions and symptoms that were known to be associated with exposure to particulate matter were included in the questionnaire. Although the reliability and validity of the questions were not assessed, the questionnaire underwent pilot testing with a group of community leaders who provided feedback on the length, time, and appropriateness of the questions. After the feedback was incorporated, the questionnaire was used for the survey. In order to recruit community members to participate from each of the four neighborhoods, the research team distributed informational flyers to residents in the neighborhoods. The

flyers informed potential participants about the study and invited them to participate by filling out the questionnaire. A central location in each neighborhood was chosen and written questionnaires were available on on multiple days for participants to complete on their own. English-speaking adults 18 years and older were included in this study.

The questionnaire used to sample the non-exposed comparison population was adapted in order to survey the comparison group in Orange County, Indiana. All demographic, smoking, health, and symptom questions remained the same, but the questions about coal ash exposure and behaviors related to coal ash were altered into questions about outdoor health and wellness. Research team members traveled to a comprehensive health clinic in the community on multiple occasions to administer the questionnaire. English-speaking adults, 18 years or older, that were visiting the clinic were invited to complete the questionnaire. This study assessed adults because adulthood exposure to particulate matter can contribute to respiratory illness and exacerbate symptoms of chronic respiratory diseases. For example, air pollution exposure increases adulthood risk for both Chronic Obstructive Pulmonary Disease (COPD) and emphysema. Respiratory symptoms can greatly disrupt the quality of life of adults and put them at risk of other health concerns.

2.3. Respiratory Symptoms Questions

The questionnaire that was utilized in this study assessed several respiratory symptoms and health conditions. To determine the symptoms that exposed and non-exposed participants reported, we asked "How frequently do you experience: cough, shortness of breath, hoarseness, and respiratory infections". Responses consisted of never, sometimes, and frequently. For this analysis the symptoms were dichotomized as "never" and "sometimes/frequently". To determine whether a participant had asthma, allergies, or other lung diseases, we asked "Have you ever been told by a doctor or health care provider that you have (circle Y if Yes)". Participants who were told by their health care professional that they had these health conditions circled a "Y". For this analysis, health conditions were coded as "1" for yes and "0" for no.

2.4. Data Analysis

Data were statistically analyzed using SAS 9.4 (SAS Institue Inc., Cary, NC, USA). The data analysis included a total of 401 questionnaires from participants; 231 completed questionnaires from the exposed population and 170 questionnaires from the non-exposed comparison population. SAS 9.4 was used to examine the prevalence of demographic characteristics and general behaviors of the study populations. The median age of participants in the exposed and non-exposed groups was compared using a Wilcoxon median two-sample test. For categorical demographic variables, Pearson's chi-squared test was used to compare the proportion of the selected variable between the exposed group and non-exposed comparison group. Differences in the prevalence of self-reported respiratory symptoms and health conditions, including cough, shortness of breath, hoarseness, respiratory infections, asthma, allergies, and other lung conditions, were assessed between the exposed and non-exposed groups using Fisher's exact test or chi-square tests. Logistic regression was utilized to calculate odds ratios. First, non-adjusted odds ratios and 95% confidence intervals from simple models with just the symptoms and exposure were determined. Then, final models that were adjusted for gender, age, and years of smoking, were used to report adjusted odds ratios, and 95% confidence intervals. The Cochran-Armitage two-sided trend test was used to assess if participants who spent more time outside were more likely to experience respiratory infections.

An overall respiratory health score was created based on self-report of all respiratory symptoms: cough, shortness of breath, hoarseness, respiratory infections, and asthma, and other lung diseases. Variables were dichotomized as "0" for never responses and "1" for sometimes or frequently responses. The overall respiratory health score reflects a numeric score for the number of positive responses of respiratory symptoms. The greater the reported respiratory symptoms the higher the overall respiratory health score. A Wilcoxon Rank-Sum test was used to compare the mean score for the exposed versus non-exposed comparison group.

3. Results

In the exposed population, 49% of participants lived in their home for >20 years, 30% lived in their homes for 5-20 years, and 20% lived in their home for <5 years. In the non-exposed population, 23% lived in their homes for >20 years, 36% lived in their homes for 5-20 years, and 42% lived in their homes for <5 years. Table 1 describes the demographics and general behaviors compared between the exposed and non-exposed populations. Age, gender, and years smoked were all statistically significantly different (p-value < 0.05) between the exposed and non-exposed groups.

Table 1. Characteristics among the non-exposed and exposed populations.

The exposed population had a more even ratio of female to male participants, while nearly three-fourths of the non-exposed population was female. Additionally, the exposed population was slightly older, median age was 52 years compared to 43 years for the non-exposed population. While the exposed population reported having smoked longer compared to the non-exposed population, other general behaviors like current smoker status and time spent outside were similar between the two populations.

The population exposed to coal ash was more likely to report having a poorer perception of health. For example, 46.8% of the exposed population perceived their health as poor or fair (16.2% and 30.6%, respectively), compared with only 28.8% of the non-exposed population (10.2% and 18.6%, respectively). The non-exposed population was more likely to perceive their health as very good or excellent 37.7% compared to 11.4% of the exposed population.

In addition to a poorer perception of health, the exposed group also more frequently experienced several respiratory irritations and illnesses (Table 2).

Table 2. Prevalence of reported respiratory symptoms.

The exposed population was more likely to report frequent coughing compared to the non-exposed comparison group (36.8% vs. 9.2%, p-Value < 0.0001). The exposed group was also more likely to report frequent shortness of breath (33.6% vs. 9.1%, p-value < 0.0001), hoarseness (15.9% vs. 1.2%, p-value < 0.0001), and respiratory infections (16.8% vs. 2.42%, p-value < 0.0001). On average, participants in the exposed group reported more respiratory symptoms than the non-exposed comparison group (mean overall respiratory health score: 3.87 vs. 2.82, p-value < 0.0001).

Table 3 reports the results from the logistic regression modeling. Adults who lived near the coal-burning power plant with coal ash storage facilities were more likely to suffer from cough (AOR = 5.3, 95% CI = 2.60-11), shortness of breath (AOR = 2.59, 95% CI = 1.56-4.31), hoarseness (AOR = 4.02, 95% CI = 2.45-6.60), respiratory infections (AOR = 1.82, 95% CI = 1.14-2.89), and allergies (AOR = 1.62, 95% CI = 1.02-2.58).

Table 3. Logistic regression results for respiratory symptoms and health outcomes.

A gradient response of respiratory infections by time spent outside was assessed in both the exposed and non-exposed groups (Table 4). Among the non-exposed group, the trend test was not statistically significant. In the exposed group, participants who spent more time outside were more likely to report having a respiratory infection (p-value = 0.0004).

Table 4. Gradient response of respiratory infections by time spent outside.

4. Discussion

This study found a higher prevalence of cough, shortness of breath, hoarseness, respiratory infections, and allergies reported per person among participants living near a coal-burning power plant with coal ash storage facilities compared to a non-exposed group. Additionally, exposed participants reported more respiratory conditions overall. Furthermore, participants living near coal ash storage who spent more time outside were more likely to report having a respiratory infection.

Fugitive coal ash emissions from coal ash storage facilities can contribute to ambient air pollution, which can adversely affect the respiratory system [26]. Coal ash is an emerging environmental threat that can create increased air pollution in communities surrounding coal ash storage sites. The United States Environmental Protection Agency (EPA) has stated that without fugitive dust controls, there could be exceedances of the National Ambient Air Quality Standards for fine particulate matter in the air at residences near coal ash landfills [17].

Inhalable particular matter, which is characterized by the aerodynamic diameter reported in micrometers (μ m), are particles less than 10 μ m in size, also known as PM₁₀. In recent decades, a growing body of epidemiological research has been published on the association between ambient air PM and adverse health outcomes, including those pertaining to respiratory health [16,27,28,29,30]. Research also points to a dose-response relationship as the PM decreases in size; correlation strength increases as the aerodynamic diameter moves from PM₁₀ to PM_{2.5} [29,31,32], where PM_{2.5} is defined as particulate matter < 2.5 μ m. While PM₁₀ and PM_{2.5} both have the potential to cause damage to tissue, PM_{2.5} is capable of traveling deeper into the lungs by penetrating the alveolar gas-exchange region. Here, the particulate matter can enter the blood stream and travel throughout the body.

Limited research has been conducted assessing the respiratory health of populations exposed to coal ash from coal-burning power plants with on-site coal ash storage facilities. Cho, Cho, Shrivastave and Kapre (1994) [33] found that workers exposed to coal ash were more likely to report respiratory effects and Sears and Zierold (2017) [23] found an elevated, but not statistically significant odds ratio for respiratory conditions in children exposed to coal ash compared to children not exposed to coal ash.

The EPA reports that over six million people in the U.S. are exposed to coal ash, but the actual number may be much higher [34]. Regulations overseeing the disposal, storage, and transport of coal ash are minimal. In 2015, coal ash became considered a "special waste" and regulations were passed by the EPA, which set requirements for disposal and storage of coal ash. These regulations included requiring industry to report fugitive dust emissions. However, in 2018, the regulations were rolled back, allowing industry and the individual states to regulate their coal ash. Other countries where coal is a major source of energy have no standards regulating coal ash.

There are several limitations that need to be taken into consideration when discussing the results of this study. First, while the two source populations were comparable according to the census data, there were some differences between participants in the two study groups, specifically the gender ratios and years smoked. Just over half of the non-exposed population reported never smoking compared to 36.3% of the exposed population. Similarly, 25% of the non-exposed population reported smoking for more than 10 years compared with 36.1% of the exposed population. However, we do note that both groups did have similar current smoking rates: 31.9% among the non-exposed group and 34.9% among the exposed group. Differences between the two groups in the number of years smoked could also partially reflect differences in the average age of the two groups. Furthermore, nearly three-fourths of the non-exposed participants were female. It is likely that the comprehensive health clinic had more female visitors, over-representing women in this population. One study examining physician-child interaction reported that 80% of the time, the caretaker with the child was the mother [35], suggesting that women are more likely to accompany their child to a doctor's appointment than their male counterpart.

Sampling bias may exist. For this study, we wanted to obtain information from participants who lived adjacently to the power plants and coal ash storage facilities, therefore, we targeted recruitment in this population of four neighborhoods. Convenience sampling was used in the neighborhoods adjacent to the power plants. For the non-exposed population, to ensure that we did not get participants who were exposed to coal ash, we took a convenience sample from a local clinic in a region that was situated 60 miles away from the power plants and in a more rural setting.

Another limitation of this study was that a proxy was used for exposure. Since this study was a small pilot study and precursor to a much larger coal

ash study, living near the coal ash plant was used as the measure of exposure. The lack of exposure assessment should be taken into consideration when interpreting the results of this study.

This study assessed self-reported behavior and illness; therefore, recall bias is of concern. Healthcare personnel did not verify the respiratory conditions reported by the participants. However, previous research conducted in adult populations has found that self-report is well validated. Researchers have shown that there was high agreement between self-report and medical report for many symptoms and conditions [36,37,38,39]. Additionally, we expect recall bias to have had little effect on the results, as the questionnaire included personal health history recall and long-term habitual behaviors. Furthermore, because some of these behaviors were health-risk behaviors, such as smoking, the accuracy and truthfulness of the results could be of concern [40]. For example, participants may underreport rates of smoking because they believe it is socially undesirable. Both surveys of the exposed and non-exposed populations were anonymous, purposefully excluding personal identifying information in an effort to alleviate this risk

A final limitation of this study is the potential for selection bias. Participants who may have had more knowledge about coal ash or more respiratory symptoms may have been more likely to participate in the study. In order to attempt to reduce selection bias, we invited all the residents of the four neighborhoods to participate when we were recruiting participants. Additionally, we had tables set-up in central locations so that participants could answer the questionnaire in open spaces in the neighborhoods, where anyone could come and fill out the questionnaire.

One strength is that this study was community-based. Multiple community members helped design the questionnaire and recruit participants. Many researchers have shown that community-based research is improved by working with community members during data collection. This process allows for improved accuracy of results when community members are involved [41,42,43,44,45]. A final strength of the study was that the questionnaire was administered at multiple locations and times, allowing residents to have more opportunities to complete the questionnaire.

5. Conclusions

This study is one of the first non-occupational studies to assess respiratory health in adults exposed to coal ash. Although there are several limitations, the results suggest that living near coal ash storage sites could be associated with increased respiratory irritation and illness among the exposed population. Little is known about the health burden that coal ash has on the surrounding community. The results from this study indicate concern and charge future research to assess the impact of coal ash on ambient air pollution and the respiratory health effects. Future studies could help to further assess this relationship by measuring coal ash directly and adjusting for smoking status, years smoked and other potential confounders.

Author Contributions

Conceptualization, A.N.H. and K.M.Z.; Data curation, K.M.Z., A.N.H. and C.G.S.; Formal analysis, A.N.H. and K.M.Z.; Methodology, A.N.H., C.G.S. and K.M.Z.; Project administration, K.M.Z.; Writing —original draft, A.N.H. and K.M.Z.; Writing—review & editing, A.N.H., C.G.S. and K.M.Z.

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Conflicts of Interest

The authors declare no conflict of interest.

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Survey of the Potential Environmental and Health Impacts in the Immediate Aftermath of the Coal Ash Spill in Kingston, Tennessee

LAURA RUHL, † AVNER VENGOSH, *.†
GARY S. DWYER, † HEILEEN HSU-KIM, †
AMRIKA DEONARINE, † MIKE BERGIN, \$
AND JULIA KRAVCHENKO"

Division of Earth and Ocean Sciences, Nicholas School of the Environment, 205 Old Chemistry Building, Box 90227, Duke University, Durham, North Carolina 27708, Civil and Environmental Engineering, 121 Hudson Hall, Box 90287, Duke University, Durham, North Carolina 27708, School of Civil and Environmental Engineering and School of Earth and Atmospheric Sciences, 311 Ferst Drive, Georgia Institute of Technology, Atlanta, Georgia 30332, and Duke Comprehensive Cancer Center in Duke School of Medicine, 2424 Erwin Road, Hock Plaza, Suite 601, Durham, North Carolina 27705

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An investigation of the potential environmental and health impacts in the immediate aftermath of one of the largest coal ash spills in U.S. history at the Tennessee Valley Authority (TVA) Kingston coal-burning power plant has revealed three major findings. First, the surface release of coal ash with high levels of toxic elements (As = 75 mg/kg; Hg = 150 μ g/kg) and radioactivity (226Ra + 228Ra = 8 pCi/g) to the environment has the potential to generate resuspended ambient fine particles (<10 μ m) containing these toxics into the atmosphere that may pose a health risk to local communities. Second, leaching of contaminants from the coal ash caused contamination of surface waters in areas of restricted water exchange, but only trace levels were found in the downstream Emory and Clinch Rivers due to river dilution. Third, the accumulation of Hg- and As-rich coal ash in river sediments has the potential to have an impact on the ecological system in the downstream rivers by fish poisoning and methylmercury formation in anaerobic river sediments.

Introduction

On Monday, December 22, 2008, the containment structure surrounding the storage of coal ash at the Kingston coalburning power plant of the Tennessee Valley Authority (TVA) collapsed, which resulted in massive release of coal combustion products (CCP) ash to the environment near Harriman, Tennessee. The CCP material, consisting of fly ash and bottom ash, spilled into tributaries of the Emory River and directly

* Corresponding author phone: (919) 681-8050; fax (919) 684-5833; e-mail: vengosh@duke.edu.

[†] Division of Earth and Ocean Sciences, Nicholas School of the Environment, Duke University.

[‡] Civil and Environmental Engineering, Duke University.

§ Georgia Institute of Technology.

"Duke Comprehensive Cancer Center in Duke School of Medicine.

into the Emory River (Figure 1), which joins the Clinch River that flows to the Tennessee River, a major drinking water source for downstream users. The Kingston coal ash spill released over 4.1 million cubic meters of ash, which is one of the largest spills in U.S. history. Some previous coal ash spills in the United States include the 2000 Martin County spill in Kentucky, which released over 1.1 million cubic meters of coal slurry into abandoned mines and nearby creeks (1), and the 1972 Buffalo Creek incident in West Virginia, which released almost a half million cubic meters of coal mining residue slurry into a nearby town (2). Numerous studies have shown that coal ash contains high levels of toxic metals that can harm the environment (3–14) and some of these elements are soluble in water and easily leached in aquatic systems (13–16).

This paper aims to provide an initial assessment of the potential environmental impacts and health risks associated with the Kingston TVA coal ash spill. In particular, the paper examines the reactivity of trace metals known to be enriched in CCP ash (9, 13) in surface water and the potential ecological effects associated with the accumulation of CCP ash in river sediments. Furthermore, the paper emphasizes the relatively high content of radionuclides in CCP ash and the potential health impact of their resuspension in the atmosphere. While most studies have investigated the potential for radon emanation from cement and fly ash used as building materials (17-20), here we examine possible health risks associated with elevated radium activity in CCP ash. The study includes measurements of trace metals in solid ash, sediments from the river, and water samples that were collected in the vicinity of the coal ash spill. Given the limited data collection since the accident, this paper provides only an initial evaluation, and does not provide a comprehensive assessment of the overall environmental impacts of the TVA coal ash spill.

Analytical Methods and Materials

Coal ash, sediments from the rivers, and water samples from tributaries, the Emory and Clinch Rivers, and springs near the spill area in Kingston and Harriman, TN (Table S1; Figure 1) were collected in three fieldtrips on January 9-10, February 6-7, and March 27-28, 2009. The surface water samples were collected near the river shoreline from sites located upstream and downstream (at different distances) of the spill. Each location was determined by availability of public access and/or allowance by property owners. Water sampling strictly followed USGS protocol (21); trace metal and cation samples were filtered in the field (0.45 μm syringe filters) into new and acid-washed polyethylene bottles containing high-purity HNO3. Trace metals in water were measured by inductively coupled plasma mass spectrometry (ICP-MS); mercury in sediments and coal ash was measured by thermal decomposition, amalgamation, atomic absorption spectroscopy (Milestone DMA-80) (22); and radium isotopes were measured by γ -spectrometry (see supporting text S1).

Results and Discussion

Coal Ash and Sediments. A comparison of the chemical composition of the TVA coal ash and local soil in Kingston, Tennessee (Table 1) shows marginal enrichments of major elements of calcium (by a factor of 2), magnesium (1.3), and aluminum (1.5), and large enrichments of trace elements such as strontium (30), arsenic (21), barium (5), nickel (5), lithium (5), vanadium (4), copper (3), and chromium (2). The high arsenic concentration in the TVA coal ash (mean = 75 mg/kg) is consistent with previously reported As in ash

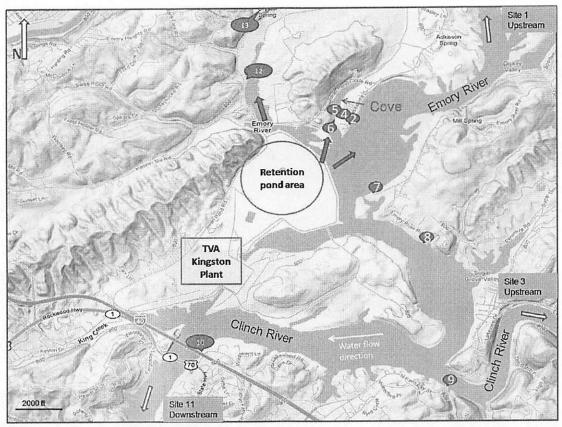


FIGURE 1. Map of the sampling sites of the TVA coal ash spill in Kingston, Tennessee. Site descriptions are reported in Supporting Information (Google maps provided the base map).

TABLE 1. Average Metals Concentrations (mg/kg) in TVA Coal Ash and Background Soil in Kingston, TN^a

material	n	AI	As	Ва	a	Ве	Cd	Ca	Cr	Co	Cu	Fe	Pb	Li	Mg	Mn	Mo	Ni	Se	Sr	٧	Zn
coal ash average (mg/kg) STD	12	14109 7264						3325 1142			46.2 16.6	13333 2807			1616 1531	102 54				201.0 39.3		
background soil average (mg/kg) STD	12	9367 3485	3.5	1000	11373		0.01 0.21	1418 933				15200 6729	16.5 8.4	0.000	1211 5580	1056 1007		4.4 4.7				29.7 11.7
ash/soil ratio		1.	5 21.4	4	1.8	9.4	3.0	2.3	2.1	1.9	2.9	0.9	1.1	4.6	1.3	0.1	13.4	5.2	0.2	29.8	3.7	1.4
^a Samples w					aı	naly	zed	by the	e Te	nnes	see	Departr	nent	of E	Enviror	ment	and	Con	serv	ation	and	the

Tennessee Department of Health.

residue of both hard coal (anthracites, bituminous, and white minous A and B: As = 50 mg/kg) and brown goal wg/kg in sediments from the downstream Clinch River at

subbituminous A and B; As = 50 mg/kg) and brown coal (lignites and subbituminous C; As = 49 mg/kg) (7). In addition, the mercury level of the TVA coal ash (an average of 151.3 \pm 15.9 μ g/kg; Table 2) is higher than background soil in Tennessee (45 µg/kg) (23). These concentrations are consistent with the range of values reported in fly ash $(100-1500 \,\mu\mathrm{g/kg})$ (24). Likewise, the ²²⁶Ra (a mean of 4.4 \pm 1.0 pCi/g) and 228 Ra $(3.1 \pm 0.4 \text{ pCi/g})$ activities of the coal ash are higher than those in local soil in Kingston (1.1 \pm 0.2 and 1.4 ± 0.4 pCi/g, respectively; Table 3). The Ra activity of the TVA coal ash is similar to the levels reported previously for fly and bottom ash from a Kentucky utility (Table 3) with a consistent activity ²²⁸Ra/²²⁶Ra ratio of ~0.7 (25). The potential impact of Ra on the environment and human health is an important consideration in remediation of the spill and is discussed below.

The Hg concentration increases from $16-54 \mu g/kg$ in upstream sediments, collected at the shoreline of the Emory and Clinch Rivers, to a level of $53 \mu g/kg$ directly across

μg/kg in sediments from the downstream Clinch River at Sites 9 and 10 (Tables 2 and S1). The Hg concentrations of the upstream sediments are consistent with previously reported Hg data for the overall Tennessee River (Table 2) (23). A historical massive release of Hg from the Oak Ridge Y-12 plant into East Fork Poplar Creek has resulted in accumulation of Hg in the sediments from the downstream Clinch River (26, 27), which could have provided a Hg legacy source for the Clinch River sediments. Our direct sampling of two sites in the upstream Clinch River (relative to the coal ash spill; Table 2) and downstream of the Y-12 source in Oak Ridge resulted, however, in low Hg contents of the river sediments (16 \pm 5 upstream and 54 \pm 11 μ g/kg downstream of Poplar Creek), which are similar to background values we report for the Emory River (Table 2). In contrast, sediments from the downstream Clinch River (sites 9 and 10) have higher Hg content (>100 μ g/kg), which suggests a significant contribution of Hg from the coal ash to the river sediments. We therefore conclude

TABLE 2. Hg Results (µg/kg) in Coal Ash and River Sediments Associated with the Spill Area in Kingston, TN°

			total Hg, μ g/kg average \pm SD ($n=3$)					
sample ID	site no. in Figure 1	description	Feb 7	March 28				
		upstream						
LR2	site 2	upstream Emory River	43 ± 0.5	10 (n = 1)				
LR11	upstream of site 3	upstream Clinch River (just downstream of Oak Ridge)	NA	16 ± 5				
LR10	site 3	upstream Clinch River	NA	54 ± 11				
LR1	site 7	close to the spill on Emory River	29.7 ± 3	22 ± 0.2				
		coal ash						
RC8S1	site 6	spilled ash pile	139 ± 5	NA				
RC8S2	site 6	spilled ash pile	145 ± 12	NA				
		downstream						
RC3	site 8	across from spill on Emory River	53 ± 3	NA				
LR9	site 9	convergence of Clinch and Emory Rivers	130 ± 5	104 ± 12				
LR8	site 10	downstream Clinch River (I-40)	115 ± 9	92 ± 32				
LR6	site 11	downstream Clinch River	NA	81 ± 38				
LR7	downstream of site 11	downstream Clinch River (before convergence with TN river)	NA	51 ± 10				
		Background soil, Tennessee						
		Lower Clinch River (n = 9)	45 ± 12					
		Upper Tennessee hydrological unit $(n = 73)$	47 ± 27					
		Roane County	56 ± 23					
^a Backgrou	und data of Hg in Tenn	essee soil from ref 23.						

TABLE 3. Radioactivity Data (pCi/g Unit) and Activity Ratios of Coal Ash and Background Data from the Spill Area in Kingston, TNP

sample/site	material	²²⁶ Ra	²²⁸ Ra	210-Pb	total Ra	²²⁸ Ra/ ²²⁶ Ra
the cove RC8S1 RC5S RC8S2	coal ash coal ash coal ash	4.9 2.6 4.9	3.2 2.1 3.1	3.57 3.54 5.01	8.1 4.6 7.9	0.65 0.79 0.63
TDEC data average for coal ash $(n = 12)$ average background soil $(n = 15)$	coal ash soil	4.4 ± 1.0 1.1 ± 0.2	3.1 ± 0.4 1.4 ± 0.5		7.5 ± 1.4 2.6 ± 0.7	0.70 ± 0.07 1.21 ± 0.28
Kentucky coal ash fly ash (average, $n = 17$) bottom ash (average, $n = 6$)	fly ash bottom ash	4.4 ± 0.6 4.0 ± 1.2	3.4 ± 0.6 2.9 ± 0.9	6.5 ± 1.9 2.2 ± 2.9	7.8 ± 0.9 7.3 ± 1.3	0.77 ± 0.15 0.71 ± 0.21

^a TDEC data are from the Tennessee Department of Environment and Conservation and the Tennessee Department of Health, Kentucky coal ash data are from ref 25.

that ash transport and deposition in the Clinch River has increased the Hg content in the river sediments.

Water Contamination. Results show that the tributary that was dammed by the coal ash spill and turned into a standing pond ("the Cove" in the area of Swan Pond Circle Road: Figure 1) has relatively high levels of leachable coal ash contaminants (LCAC), including arsenic, calcium, magnesium, aluminum, strontium, manganese, lithium, and boron (Table 4; Figure 2). Some of these elements are highly enriched in coal ash (6, 8) (Table 1), and are known to be highly soluble in aquatic systems (8). Among the LCACs, arsenic stands out with concentrations of up to 86 μ g/L in the Cove area. Groundwater data from the other tributary (Figure 1) show negligible LCAC levels, thus indicating that the shallow groundwater is not contaminated. In this hydrological setting, noncontaminated groundwater discharges into the dammed tributary and causes leaching of LCAC from the coal ash. Under restricted water exchange, the formation of standing water in the Cove resulted in contaminated surface water. In contrast, surface waters from the Emory River and Emory-Clinch River downstream from the breached dam show only slight LCAC levels, and all river inorganic dissolved constituents concentrations are below the EPA Maximum Contaminant Levels (MCL) and EPA Criterion Continuous Concentration (CCC) for aquatic life (Table 4) (28, 29).

The upstream Clinch River has a distinct chemical composition (higher Na^+ , Ca^{2+} , Mg^{2+} , Sr^{2+} , and SO_4^{2-}) relative to the upstream Emory (Table 4), and thus the major inorganic constituents show mixing relationships between these two water sources (Line M1 in Figure 2) downstream from the confluence of the Emory and Clinch rivers (Figure 1). The concentrations of arsenic, strontium, and boron in the downstream river samples deviate toward higher values, however, relative to these river mixing relationships (Figure 2). These geochemical shifts reflect a small but traceable indication of leaching of contaminants from coal ash that was spilled into the river and further mixing with the uncontaminated river water (Line M2 in Figure 2). The data show that the river flow is effective in reducing the LCAC's contents by an order of magnitude relative to directly contaminated water measured in the Cove.

The water samples were collected near the shoreline of the river, which may be an underestimate of the concentration of dissolved elements throughout the river vertical profile. The spatial distribution of contaminants (dissolved and suspended particulate fractions) in a river water column depends on a number of factors including particulate size,

sample (D	location	site	Ca	Mg	Na	CI	SO ₄	Li	В	Al	Mn	Co	Ni	Ca	Zn	As	Se	Rb	Sr	Cr	U
								ups	tream r	iver											
TN3	upstream Emory River	1	5.8	1.3	1.6	2.7	9.9	0.5	5.9	50	23	0.1	1.1	0.1	5.7	0.1	0.4	1.4	26.4	0.2	-
TN3U	upstream Emory River	1	5.5	1.2					7.2	131	15	0.1	-	0.4	8.0	-	0.3	1.4	23.8	-	-
RC9	upstream Emory River	1	7.4	1.6	2.6	3.6	12.7	0.4	5.8	19	72	0.4	1.1	0.2	4.8	0.1	0.1	1.0	31.3	0.2	0.1
RC10	upstream Clinch River	3	36.6	11.2	9.5	6.8	30.5	3.0	13.4	5	5	0.2	1.0	0.5	9.2	0.4	0.4	1.0	134.5	0.4	0.4
								1	he Cov	е											
TN1	The Cove	4	38.9	7.0	2.8	16.8	98.1	13.2	431.9	-	847	2.1	4.0	1.5	16.5	69.6	2.4	15.6	578.4	1.9	0.1
TN1U	The Cove	4	42.5	8.0					425.9	344	974	3.1	-	5.0	42.4	95.2	0.4	17.1	632.6	-	-
RC5	The Cove	4	93.2	12.7	3.2	12.6	260.3	19.6	470.8	22	3014	7.0	9.0	1.6	47.2	85.6	3.8	23.8	1244.5	1.9	1.1
TN9	The Cove	5	13.0	3.1	1.7	5.0	20.4	3.1	84.9	43	296	0.3	4.3	0.8	12.2	9.3	0.5	5.0	108.3	6.6	0.0
TN9U	The Cove	5	14.9	4.5					112.9	197	332	1.2	-	3.5	24.9	12.6	0.0	6.2	120.1	-	0.0
RC8	The Cove	6	35.2	6.1	3.0	5.5	76.7	7.4	229.6	40	556	1.9	1.7	2.8	36.6	20.7	1.8	6.3	455.9	0.5	0.5
								dow	nstream	river											
TN2	riverside of Lakeshore Dr.	2	6.4	1.4	1.8	3.1	9.8	0.5	8.7	58	34	0.1	1.3	1.1	11.3	0.6	0.2	2.0	31.0	0.2	-
TN2U	riverside of Lakeshore Dr.	2	6.5	4.8					23.4	154	28	0.4	_	2.1	14.4	0.9	0.1	2.0	28.8	_	-
RC6	riverside of Lakeshore Dr.	2	9.1	2.1	2.4	3.7	13.1	0.7	38.1	40	60	0.3	0.9	0.5	5.1	3.1	0.6	1.3	59.3	0.3	0.2
TN5	Emory River	7	7.3	2.0	1.6	2.7	9.4	0.4	7.9	51	15	2.6	1.0	0.4	1.5	0.3	0.4	1.2	27.6	0.4	-
TN5U	Emory River	7	7.3	2.1					9.7	466	16	0.4	_	0.5	4.3	0.3	0.1	1.9	26.5	_	_
RC2	Emory River	7	7.3	1.9	2.5	3.5	11.0	0.4	6.6	23	42	0.3	1.1	34.0	8.0	0.1	0.1	1.0	28.1	0.3	0.1
RC3	Emory River	8	16.5	4.8	4.8			1.2	10.1	34	22	1.2	1.0	0.8	5.1	0.3	0.2	1.1	61.6	0.4	0.2
TN6	convergence of Emory and Clinch	9	37.3	11.9	8.6	5.9	31.9	2.5	19.3	56	5	0.0	1.1	0.6	1.8	0.6	0.6	1.0	137.5	0.4	0.4
TN6U	convergence of Emory and Clinch	9	37.9	12.1					14.2	56	17	0.2	_	0.9	3.2	0.4	0.3	1.2	144.2	_	0.1
RC1	convergence of Emory and Clinch	9	37.3	11.5	8.6	6.9	29.7	3.0	14.1	19	5	0.2	1.2	1.1	6.5	0.4	0.2	1.0	130.4	0.4	0.4
TN7	Clinch River at I 40 Bridge	10	23.4	7.3	5.4	4.6	21.7	1.6	14.8	34	13	0.0	1.2	0.6	2.1	0.7	0.5	1.1	89.0	0.3	0.2
TN7U	Clinch River at I 40 Bridge	10	24.7	7.5					13.7	149	20	0.1	_	0.9	2.1	1.2	0.2	1.5	94.1	_	_
RC4	Clinch River at I 40 Bridge	10	35.1	10.3	8.7	7.5	27.0	2.8	15.4	20	18	0.2	1.2	0.9	5.5	0.7	0.4	1.0	121.3	0.5	0.4
TN12	city of Kingston Gravel pit park	11	24.9	7.8	5.5	4.7	22.6	1.8	20.2	32	17	0.0	1.0	1.1	3.2	0.8	0.5	1.2	92.4	0.4	0.3
TN12U	city of Kingston Gravel pit park	11	25.6	7.9					15.3	176	20	0.2		1.0	6.2	1.3	0.5	1.5	97.8	_	0.1
RC11	city of Kingston Gravel pit park	11	29.8	8.9	7.7	6.2	25.7	2.9	14.7	12	17	0.2	1.0	0.9	6.5	0.5	0.5	1.0	110.6	0.4	0.4
								ar	oundwa	ter											
TN4	spring water flow to the Cove	12	34.1	8.2	2.6	5.8	18.1	0.1	12.3	15	109	2.1	0.9	0.1	3.9	0.2	0.6	8.0	61.3	0.4	0.2
TN4U	spring water flow to the Cove	12	34.5	8.5					14.6	38	108	0.7	-	0.1	4.7	0.1	0.9	0.8	59.0	-	_
TN8	spring water flow to the Cove	12	14.7	7.6	0.6	1.7	3.1	0.3	7.1	5	0.1	0.1	0.3	0.2	5.3	0.1	0.2	0.7	15.5	0.3	-
TN8U	spring water flow to the Cove	13	15.4	8.0			-	_	7.4	18	0.3	0.0	_	0.1	5.3	_	0.4	0.7	15.6	_	_
RC7	spring water flow to the Cove	13	19.9	10.5	0.6	1.3	1.4	0.2	2.6	4	0.4	0.1	0.3	0.1	4.8	0.1	0.0	0.7	15.9	0.4	0.3
								EPA	regulat	tions											
EPA MCL	Maximum Contaminant Level for													1300		10	50			100	30
L. A WICL	drinking water													1300		10	50			100	50
EPA CCC	Criterion Continuous Concentration									87			52	9	120	150	5			(III):74,(VI):11	4

[&]quot;"" refers to unfiltered samples. EPA MCL is the Maximum Contaminant Level for drinking water and EPA CCC is the Criterion Continuous Concentration, which is an estimate of the highest concentration of a material in surface fresh water to which an aquatic community can be exposed indefinitely without resulting in an unacceptable effect (28, 29). For Site location see Figure 1 and Supporting Information.

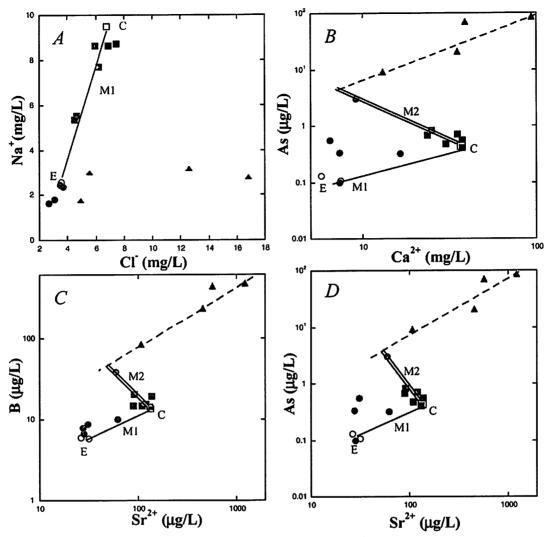


FIGURE 2. Variations of Na⁺ and Cl⁻ (A), Ca²⁺ and As (B), Sr²⁺ and B (D), and Sr²⁺ and As (D) in water samples from the Cove (triangles), Emory River upstream (open circles) and downstream (closed circles), and Emory—Clinch upstream (open squares) and downstream (closed squares). Mixing of Emory and Clinch Rivers (Line M1) is identified by the major ion composition (A). Elements that are enriched in coal ash, as reflected by the high concentrations in the Cove area (dashed line), show higher concentrations in the downstream Emory—Clinch river samples relative to the expected Emory—Clinch river mixing composition (Line M1). Line M2 reflects possible mixing of contaminants derived from coal ash leaching near the spill area (dashed line) and the uncontaminated Clinch River composition (Line M1).

turbulent flow conditions, seasonal flow changes, and channel morphology (30). The dissolved phase, which was measured in this study, is typically homogeneously distributed in a river, but can vary with the proximity to a point source of pollution (30). Assuming that metal mobilization in the river derives from both suspended ash and bottom sediments, the distribution of dissolved constituents in the water column would depend on numerous factors such as differential river velocity, rate of mobilization, and water depth (31). Further investigation is therefore required to determine the vertical distribution of metals in the river water, and whether sampling of the upper river section represents the most diluted segment of the river flow.

Potential Environmental Impacts. While the downstream river water shows only trace levels of LCACs (at the surface), the downstream river sediments show high Hg concentrations similar to the coal ash levels ($115-130~\mu g/kg$; Table 2). The ecological effects of Hg in the coal ash and sediments depend on the chemical lability of Hg in the solids and the potential for mercury methylation in the impacted area. While previous studies have demonstrated that Hg in CCP ash is not readily soluble through acid-leaching protocols (32), Hg has a high

affinity for natural organic matter (33, 34), which can promote desorption if the Hg is associated with weaker binding sites on metal-oxide minerals in the ash material (35). Furthermore, the transformation of Hg to methylmercury by anaerobic bacteria in river sediments is a concern because of bioaccumulation of methylmercury in food webs. Previous studies have shown that sulfate addition can promote methylation in freshwater ecosystems by stimulating sulfatereducing bacteria (36), the primary organisms responsible for producing methylmercury in the environment (37). In coal-ash-containing waters, a 10- to 20-fold increase in SO₄²⁻ concentrations was observed in the Cove area relative to unaffected upstream sites (Table 4). Therefore, the methylation potential of mercury from this material could be high because the coal ash also provides an essential nutrient (SO₄²⁻) that encourages microbial methylation. In addition, accumulation of arsenic-rich fly ash in bottom sediment in an aquatic system could cause fish poisoning via both food chains and decrease of benthic fauna that is a vital food source (7, 38).

Potential Health Impacts. Of particular concern to human health is the wind-blown resuspension of fly ash into the

atmosphere. It is well-known that wind-blown dust can travel long distances, as exemplified by Asian dust storms that result in transport to locations as far away as the U.S (39). It is possible that coal ash exposed to the atmosphere can be resuspended and transported to populated areas where human exposure may occur. Fly ash-airborne particles with diameters less than 10 μ m (PM10) are regarded as respirable and may affect the human lung and bronchus (40-42). The process of particulate resuspension will depend on a variety of factors, including the fly ash particulate size and related chemical and physical properties, wind speed and atmospheric turbulence, and likely the relative humidity and surface moisture (43, 44). The particles that are of most importance for human health are in the fine particulate (PM_{2.5}) mode, which readily deposit deep in the lung (45). Past work has shown that CCP ash has particulate sizes ranging from less than 1 μ m to 100s of micrometers in size (46, 47). In addition, there is a compositional relationship as a function of fly ash particle size (46). Several studies have also measured ambient fine particulate matter associated with elevated concentrations of toxic metals in the vicinity of coal-fired power plants (42, 48-50). In some cases, fly ash-airborne particles were also found in remote areas (up to 30 km from power stations) (42, 51). Overall, past work indicates that coal ash contains inhalable particulate matter, and that fly ash emitted from the burning of coal is readily transported in the atmosphere.

The high concentrations of trace metals (Tables 1 and 2) and radioactivity (Table 3) reported in this study for the bulk TVA coal ash are expected to magnify, as fine fractions of fly ash (which may be resuspended and deposited in the human respiratory system) are typically 4-10 times enriched in metals relative to the bulk ash and the coarse size fraction (7, 46). The toxic metal content in coal ash, the sizes of fly ash particulates, and the ionizing radiation (IR) exposure (both incorporated and external) may act synergistically or, less frequent, antagonistically, affecting human health directly (predominantly through inhalation of contaminated air) and indirectly through the food chains (consuming contaminated agricultural products) (14). Coal ash was recognized as a Group I human carcinogen (based on occupational exposure studies) associated with increased risks of skin, lung, and bladder cancers (52). Arsenic and radium exposures in humans are associated with increased risks of skin, lung, liver, leukemia, breast, bladder, and bone cancers (53) for exposure predominantly due to chronic ingestion or chronic inhalation, with the dose-response curve dependent on location, sources, and population susceptibility and/or tolerance.

Health impacts of CCP ash have been predominantly studied on animal models and human cell lines, with few short-term epidemiological follow-ups. CCP ash particulates affect lung epithelial and red blood cells in animal studies and human in vitro models, causing inflammation, changing the sensitivity of epithelia, altering immunological mechanisms and lymphocyte blastogenesis, and increasing the risk of cardiopulmonary disease (e.g., pulmonary vasculitis/ hypertention) (54-57). Individuals with pre-existing chronic obstructive pulmonary disease, lung infection, or asthma are more susceptible to the coal ash affliction (58). Several epidemiological studies have proved the significant health hazards (such as enhanced risk for adverse cardiovascular events) of fine-particulate air pollution for individuals with type II diabetes mellitus and people with genetic and/or disease-related susceptibility to vascular dysfunction, who are a large part of the population (59).

Radium-226 and ²²⁸Ra, which are the main sources of low-dose IR exposure in coal ash, can remain in the human lung for several months after their inhalation, gradually entering the blood circulation and depositing in bones and

teeth with this portion remaining for the lifetime of the individual. When inhaled, the radionuclides can affect the respiratory system even without the presence of the other coal ash components. Thus, the airborne particles containing radioactive elements inhaled by cleanup workers of the nuclear accident at the Chernobyl nuclear power plant caused bronchial mucosa lesions, in some cases preneoplastic, with an increased susceptibility to the invasion of microorganisms in bronchial mucosa (60, 61). Consequently, the combined radioactivity of coal ash at the TVA spill, together with other enriched trace metals such as Ni, Pb, and As, may increase the overall health impact in exposed populations, depending on duration of exposure, and particularly for susceptible groups of the population. It is important to underscore the fact that at this time it is not possible to estimate the health impacts of CCP ash resuspended particulates due to a lack of information on the rate at which they are entrained into the atmosphere, as well as their chemical, physical, and synergistic properties linked to morbidity and mortality. Clearly future studies are needed linking ambient element and radionuclide concentrations with ground level CCP ash characteristics, ambient meteorological characteristics, and human population exposure.

This study has provided an initial assessment of the environmental impacts and potential health effects associated with the TVA coal ash spill in Kingston, Tennessee. The study shows that the high metals contents of coal ash and their high solubility resulted in contamination (e.g., As) of surface water associated with the coal ash spill in areas of restricted water exchange. In the downstream Emory and Clinch Rivers the leaching of trace metals is significantly diluted by the river flows. While the levels of contaminants in the downstream Emory and Clinch Rivers are below the MCL levels, high concentrations of Hg found in the river sediments pose a serious long-term threat for the ecological system of these rivers. This study also highlights the high probability of atmospheric resuspension of fine fly ash particulates, which are enriched in toxic metals and radioactivity, and could have a severe health impact on local communities and workers. Based on these initial results, this study provides a framework for future and long-term monitoring of the TVA coal ash spill and remediation efforts. Future studies should focus on evaluating the ecological ramifications, such as methylmercury formation in the sediments in the downstream Emory and Clinch Rivers, and the composition of particulate matter in the air in the vicinity of the spill area. Finally, future prognoses of the health impacts of residents exposed to coal ash requires long-term follow-ups of various population groups, including children and adolescents, pregnant women, persons exposed in utero, and individuals with pre-existing broncho-pulmonary diseases and diabetes mellitus. All these factors must be included in remediation efforts for the TVA Kingston coal ash spill.

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Supporting Information Available

Supplementary description of the analytical techniques and sample location. This material is available free of charge via the Internet at http://pubs.acs.org.

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