Review Article

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A review on fly ash from coal-fired power plants: chemical composition, regulations, and health evidence

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Abstract: Throughout the world, coal is responsible for generating approximately 38% of power. Coal ash, a waste product, generated from the combustion of coal, consists of fly ash, bottom ash, boiler slag, and flue gas desulfurization material. Fly ash, which is the main component of coal ash, is composed of spherical particulate matter with diameters that range from 0.1 μm to >100 μm. Fly ash is predominately composed of silica, aluminum, iron, calcium, and oxygen, but the particles may also contain heavy metals such as arsenic and lead at trace levels. Most nations throughout the world do not consider fly ash a hazardous waste and therefore regulations on its disposal and storage are lacking. Fly ash that is not beneficially reused in products such as concrete is stored in landfills and surface impoundments. Fugitive dust emissions and leaching of metals into groundwater from landfills and surface impoundments may put people at risk for exposure. There are limited epidemiological studies regarding the health effects of fly ash exposure. In this article, the authors provide an overview of fly ash, its chemical composition, the regulations from nations generating the greatest amount of fly ash, and epidemiological evidence regarding the health impacts associated with exposure to fly ash.

Keywords: air pollution; coal ash; coal-fired power plants; fly ash; particles.

Introduction

Coal is an abundant fossil fuel used for the generation of approximately 38% of the electricity used globally (1). From 2017 to 2018, the use of coal to produce power increased by 3% in the world, but was driven by increased use in China, India, and Southeast Asia. While Asia increased its electricity production from coal, power generated from coal in Europe and the United States decreased over the same year (–2% in Europe and –5% in the United States) (1).

The use of coal for energy production varies throughout the world. In 2016, China, India, and the United States ranked as the top three nations of the world for coal combustion (2). In the United States, coal-fired power plants account for approximately 27–30% of electricity generation (3, 4) as compared with China and India, where coal accounts for approximately 72% of the power generated in the nations (5–7).

When coal is burned for energy, coal combustion products (CCPs), often referred to as “coal ash”, are generated. CCPs include fly ash, bottom ash, boiler slag, and flue gas desulfurization material. Baig and Yousaf reported that for every four tons of coal that is burnt, one ton of coal ash is produced (8). Coal with high ash content will produce more coal ash (9). The composition of coal ash varies based on the geochemical properties of the coal being burned, the mining and preparation methods, the emission-control technology used, and the method of burning (10–17). There are four types of coal: anthracite, bituminous, subbituminous, and lignite. These coals produce coal ash that is composed predominately of silicon, aluminum, calcium, and iron and may contain heavy metals, such as arsenic and lead (18–34). The pH of coal ash, which is an important property for leaching of the trace metals, ranges from acidic to alkaline. However, coal ash from bituminous coal contain much less calcium than subbituminous coals, therefore generating ash that is slightly acidic to slightly alkaline on contact with water. Subbituminous coal ash tends to contain a higher concentration of calcium and generates alkaline solutions on contact with water (16, 35).

In 2016, throughout the world, approximately 1.2 billion tons of CCPs were produced (36). China, India, Europe, and the United States produce the most CCPs. In 2016, China produced over 565 million tons, India produced 197 million tons, and Europe produced 140 million tons (36). According

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to the U.S. Environmental Protection Agency (EPA), CCPs are one of the largest industrial waste streams in the United States (37). In the United States in 2016, more than 107 million tons of coal ash were produced (36, 38).

Coal ash is either beneficially reutilized in products such as concrete and wallboard, or stored in landfills, surface impoundments, or mines. Most power-generating facilities store the ash on site (39, 40). Of the total coal ash produced in the world, 64% of coal ash was beneficially used. Harris et al. reported that Japan had the highest utilization rate (99.3%), followed by Europe (94.3%), Korea (85%), and China (70%) (36).

The coal ash that is not used in products is disposed of in landfills or surface impoundments which may become a potential source of pollution (34, 41–43). In 2010, the EPA reported that "without fugitive dust controls, levels at nearby locations could exceed 35 $\mu g/m^3$ established as the level of the 24-h $PM_{2.5}$ National Ambient Air Quality Standard (US) for fine particulate" (41). Furthermore, the EPA noted that constituents of coal ash such as arsenic have leached at levels of concern from unlined and inadequately clay-lined landfills and surface impoundments (41).

Although some countries like the United States are phasing out the use of coal-fired power plants, other countries like India and Japan are building more coal-fired power plants to meet the supply of power needed for their country (44, 45). Fly ash, which is the CCP that is generated in the greatest quantities, is becoming an increasing environmental threat, as its management is becoming a greater concern among countries' governing bodies and populations living near coal ash storage facilities.

The purpose of this article is to provide an overview of fly ash, including its chemical composition, regulations that govern fly ash disposal, and the potential health effects from exposure to respirable fly ash and metals found in fly ash.

Methods

Peer-reviewed articles, presentations, and government websites from 1987 to 2019 were assessed using Google Scholar, PUBMED, and web resources. Search keywords included, but were not limited to, "coal ash", "fly ash", "coal ash regulations", "coal ash storage", "metals found in fly ash", "global fly ash", "groundwater and fly ash", "leaching behavior of fly ash", and "coal ash and health". For additional supportive information, search keywords included "air pollution and health" and "heavy metals and health". The reference sections of the selected articles were further examined for derivation of relevant articles. The information selected for this review is composed of web-based articles, government documents, review articles, papers presented at conferences, and full-text manuscripts, which were written in English. Overall, 231 references were utilized for this review.

Results

Fly ash

Fly ash constitutes the majority of CCPs representing about 40–90% of the total product (46–51). Pulverized fuel combustion has been utilized for over 100 years and most large plants utilize this method. Fly ash is generated when pulverized coal is blown into a combustion chamber. In the combustion chamber, the pulverized coal ignites, generates heat, and produces a molten mineral residue. As heat is extracted by the boiler tubes, flue gases are cooled and the residue hardens and forms an ash. Larger, heavier ash particles fall to the bottom of the combustion chamber. Lighter ash particles remain in the flue gases and are collected in air pollution control devices. These lighter ash particles are termed fly ash (35). Fly ash is a fine silt of spherical powdery particles with diameters that range from 0.1 μm to >100 μm. Particle sizes of most fly ash from bituminous coal are <75 μm (16). Fly ash from subbituminous coal tends to be coarser than fly ash from bituminous coal (47). Brown et al. reported that within the respirable range, the average sphere size ranged from 1.98 μm to 5.64 μm (11). The spherical particles that make up fly ash are grouped into plerospheres and cenospheres. Plerospheres are hollow spheres filled with smaller spheres, whereas empty spheres are termed cenospheres (35). The unique hollow sphere morphology allows for a range of specific gravities of fly ash. Specific gravities range from 1.6 to 3.1 and pH values can range from 1.2 to 12.5, with the majority of ashes being more alkaline (16, 46, 47, 51–60). Fly ash varies in color, based on the amount of unburned carbon and iron (46, 57). It can be orange to deep red, white to gray, or yellow or black (46, 47, 58, 61).

Once collected from the air pollution control devices, fly ash is beneficially used in products or as fill in roads or park construction. Countries vary widely on the amount of fly ash that they beneficially utilize. India, China, Canada, and the United Kingdom reutilize 50% or less of generated fly ash, while the United States utilizes approximately 65% (9, 43, 46, 58, 62, 63). Of the reutilized fly ash, the United States uses most of its fly ash (61%) in concrete, concrete products, grout, and in mining applications;

China uses most of its fly ash (67%) in cement, bricks, and tiles; India uses most of its fly ash (61%) in cement and reclamation; and the European Union uses most of its fly ash (62%) in reclamation, restoration, and as a concrete addition (57, 62).

Fly ash that is not utilized is predominately stored in landfills or surface impoundments. Due to the chemical composition of fly ash, the size distribution of fly ash, and the mobility of elements from fly ash, concerns have been raised that the fly ash storage methods, particularly older landfills and surface impoundments which are unlined, may harm the environment and impact human health.

Chemical composition of fly ash

Fly ash is composed mainly of silica, aluminum, iron, calcium, and oxygen and contains many other elements at trace levels (29, 46, 57, 61, 64, 65). Hatori et al. (29) reported that 80–95% of the sum of oxygen, silica, and aluminum make up the total mass of fly ash particles. Although oxygen, silica, and aluminum showed homogeneous distributions in the particles that Hatori and researchers studied, they also found that the trace elements were quite different in each particle (29). Some of the trace elements found in fly ash are heavy metals, such as arsenic, cadmium, chromium, and lead (11, 18, 29, 30, 46, 49, 64, 66, 67) and have become an environmental and health concern globally.

Multiple researchers have studied the composition of fly ash and have reported that the concentrations of metals are higher in fly ash than in the parent coal (68–70). Spencer and Drake assessed fly ash from Iowa, United States, and found that the trace elements of metals existed in concentrations 2 times greater than metals found in the original coal (68). Bhangare et al. found that in ashes from India, concentrations of lead, copper, cadmium, zinc, iron, manganese, chromium, nickel, magnesium, lithium, cobalt, mercury, and arsenic were higher in fly ash, compared to bottom ash or coal (69). Yao et al. reported that trace element levels in Chinese coal may be 4–10 times higher than those found in the original coal (51). Verma et al. (70) reported concentrations of five metals in coal and in the corresponding fly ash. The concentration of lead in coal was 4 ppm and was found to be 35 ppm in fly ash. The concentration of chromium was 8 ppm in the parent coal and 65 ppm in the fly ash (70).

Understanding the behavior of trace elements during combustion is important in understanding the chemical composition of fly ash. Temperature of the boiler impacts volatilization which effects the distribution of trace elements (16, 66). In understanding the behavior or trace elements, many researchers use the classifications developed by Rudd Meij which detail behavior according to their volatility and condensation. Based on the relative enrichment factor (RE) which is given in equation 1, Meij created three classifications of trace elements (See Table 1).

$$RE = \frac{\text{Concentration of element in ash} \times \text{Ash percentage in feed coal}}{\text{Concentration of element in coal} \times 100} \quad (1)$$

Table 1: Classification of trace elements.

	Description	Relative enrichment factor	Examples of elements	Distribution
Class I	Nonvolatile elements	RE ~1	Aluminum, calcium, iron, magnesium	More likely to be in bottom ash or equally distributed in fly ash and bottom ash
Class II	Volatile in boiler, condenses in electrostatic precipitator	RE < 0.7		
Class IIc			Chromium, manganese, barium, rubidium	
Class IIb			Beryllium, cobalt, copper, nickel	
Class IIc			Arsenic, cadmium, lead, zinc	
Class III	Very volatile, some do not condense in electrostatic precipitator	RE << 1	Chlorine, fluorine, mercury, selenium	Flue gas, fly ash, but not likely to be in bottom ash

Class I are nonvolatile elements, and Class II are elements that are volatile in the boiler, but condense on the fly ash particles in the electrostatic precipitator (ESP). Class II is divided into three groups with increasing volatility. Finally, Class III represents elements that are very volatile and may not condense on ash at all (27, 33, 66).

Elements like chlorine and fluorine are almost totally volatilized in flue gas and do not concentrate in bottom ash. However, as the flue gas cools down, some volatile elements may condense on the surface of the fly ash particles. For example, much higher quantities of arsenic, copper, and selenium are found in fly ash than are found in bottom ash or boiler slag (39).

Bhangare et al. (69) assessed 13 trace elements from five coal-fired power plants in India. The researchers found that chromium, manganese, lead, and iron had $RE > 1$ and that arsenic, mercury, zinc, and lithium had $RE < 0.7$. The remainder of the elements which included cadmium, nickel, cobalt, manganese, and copper had RE that ranged between 0.7 and 1.0, representing the semi-volatile nature of these elements (69).

Heavy metals often found in fly ash

Heavy metals such as arsenic, lead, chromium, and cadmium are often found in fly ash and may escape as fugitive dust or leachate from landfills and surface impoundments. Although concentrations of trace elements are low, usually in the parts per billion to parts per million range, concern about exposure is apparent in many countries throughout the world (41, 43, 46, 51, 64).

Arsenic (As)

Inorganic arsenic is one of the most toxic metals found in coal ash and it is a hazardous environmental pollutant. Arsenic concentrations in fly ash range from less than the detection limit to 1000 mg/kg, depending on the parent coal (31, 64, 69, 71–73). The toxicity of As is related to its form, valence state, solubility, the rate of absorption, and the rate of elimination. Trivalent arsenic (AsIII) is considered to be more toxic than pentavalent arsenic (AsV) (74–76). The United States Geological Survey (USGS) collected fly ash samples from three coal-fired power plants in the United States. They reported that 89% of arsenic was present as the more oxidized pentavalent arsenic (AsV), whereas only 11% was present as trivalent arsenic (AsIII) (77). Huggins et al. also found that AsV was present in much greater quantities, than AsIII (78).

Inorganic trivalent and pentavalent compounds are associated with multiple health effects. The International Agency for Cancer Research (IARC) classifies inorganic arsenic as a carcinogen (79). The evidence from drinking-water exposure allows IARC to state that carcinogenicity is related to exposure to AsIII and AsV. However, the evidence from inhaled arsenic mixtures only allows IARC to state that the carcinogenicity is related to inorganic arsenic compounds. Unlike drinking-water exposure, IARC states that the evidence for inhalation does not allow a separation of the carcinogenic risk associated with particular arsenic species that occur in these mixtures (79).

In addition to cancer, studies have shown that chronic exposure to arsenic is associated with heart disease (80–83), type 2 diabetes (81, 84–87), impairments in children's intellectual function (88–90), and respiratory conditions (91–94).

Lead (Pb)

Lead is a naturally occurring element that is found in the earth's crust; therefore, it can be found in the soil, dust, air, and water. Lead is found in parent coals throughout the world. Hence, Pb is also found in fly ash (95–97). Researchers have found Pb concentrations in fly ash that range from less than 1.4 mg/kg to 2120 mg/kg (31, 69, 72, 73, 98–100). Franus et al. reported that among the toxic elements in fly ash, Pb and As are found in the greatest amounts (101).

Exposure of children to Pb is of particular concern because their nervous system is still developing. Lead exposure can result in impaired learning, slow growth, behavioral problems, lower IQ, and hyperactivity in children (102–107). Chronic Pb exposure can also affect adults and result in health problems such as hypertension, hearing problems, poorer kidney function, cognitive impairment, and increased mortality for heart disease (108–112).

Although Pb is most noted for its effects on the central nervous system (CNS), IARC has classified inorganic Pb as a probable carcinogen to humans. IARC states that there is limited evidence in humans for the carcinogenicity of inorganic lead compounds and there is inadequate evidence in humans for the carcinogenicity of organic lead compounds (113).

Chromium (Cr)

Chromium is a naturally occurring element found in rocks, plants, and soil. It occurs in coal and is released into the air, soil, and water. Chromium exists in three forms: chromium(0), chromium(III), and chromium(VI). It is persistent in the environment, and is of concern because Cr(VI) is a known human carcinogen and certain compounds are highly soluble in water. Cr(III) is less toxic and less soluble than Cr(VI) (114, 115).

Chromium is more likely to occur primarily as Cr(III) in most bituminous coals (114, 116–118). There is limited presence of Cr(VI) (119). Researchers have found that Cr(VI) in coal is reduced in the flue gas by sulfur dioxide (SO_2), during the combustion process. It is estimated that the fraction of Cr(VI) in fly ash is less than 5% (116, 119). However, Huggins et al. reported that some samples of fly ash may contain up to 20% of Cr(VI) (119). Chromium concentrations range in fly ash from 7.82 mg/kg to 651 mg/kg (31, 69, 71–73, 99, 100).

Small amounts of Cr(III) are needed for biological processes in humans; however, exposure to Cr(VI) is of great concern. IARC classifies Cr(VI) compounds as known human carcinogens which can cause cancer of the lung, nose, and nasal sinus. There is less evidence for other cancers (120). Epidemiological studies have reported that Cr(VI) may also be associated with cancer of the stomach; however, the results are conflicting (120–123). In addition to cancer, there is some evidence that exposure to Cr(VI) is genotoxic. Lymphocytes of workers exposed to dusts of Cr(VI) compounds showed elevated occurrences of DNA strand breaks, sister chromatid exchange (SCE), and micronuclei (MN) (124–127).

Cadmium (Cd)

Cadmium is an element that is also found in the earth's crust. Although Cd is used in batteries, televisions, and paint pigments, the most significant source of cadmium exposure in humans is cigarette smoke exposure (128). Cadmium is found in coals and hence it is in fly ash. Concentrations of Cd in fly ash range from less than the detection limit to 17 mg/kg (31, 69, 71–73, 99, 100).

Exposure to cadmium may affect several organs in the body. IARC classifies Cd and its compounds as carcinogenic and reports that there is sufficient evidence that Cd is associated with lung, kidney, and prostate cancer (129). Cadmium exposure has been associated with osteoporosis, musculoskeletal pain, kidney failure, and hypertension (130–135).

Mercury (Hg)

Mercury exists in different forms as organic, elemental, and inorganic. All these forms are toxic. Humans can be exposed to Hg via food, air pollution, industrial, occupational, and agricultural activities (136, 137). Mercury has a low excretion rate; little of it is excreted while the rest is absorbed by different organs in the body (e.g. kidney and liver) to produce neurotoxic and deteriorating effects (136, 138). The effects of Hg on neurobehavioral development in early years have also been reported (138–141). Mercury is found in fly ash with concentrations ranging from less than the detection limit to 2.13 mg/kg (69, 72, 73).

Regulations associated with the storage of fly ash

Regulations regarding fly ash disposal vary by country and reflect whether countries consider fly ash as a hazardous waste (16, 36) or another form of waste. In most countries, fly ash is not considered a hazardous waste, thus there are limited rules and regulations that govern its storage and disposal.

Disposal regulations in the United States

Prior to 2015, there were very limited regulations on disposal and storage of fly ash in the United States, because it was never considered a "hazardous waste". In 1978, the Resource Conservation and Recovery Act (RCRA) was enacted by the United States' Congress, which mandated that the EPA identify and regulate hazardous wastes. When EPA defined "hazardous wastes" under Subtitle C of RCRA, coal ash and five other large waste streams were termed "special wastes" until more research could be conducted on the human health and environmental impact of these wastes. In 1980, the United States' Congress enacted the Solid Waste Disposal Act Amendments including the Bevill Amendment. The Bevill Amendment exempted "special wastes", specifically fossil fuel combustion waste, from regulation until additional assessment of risk was conducted. After studying the wastes, the EPA determined that coal combustion waste did not belong under Subtitle C of RCRA and was therefore not declared "hazardous". As the waste was not hazardous, its disposal falls under Subtitle D of RCRA, which means that it was not regulated by the federal government; instead, each state in the United States was to regulate the waste.

Years after the final decision on the classification of coal ash, there were occurrences of large coal ash spills from surface impoundments in Kingston, Tennessee and Eden, New York which led to damage of property and environmental pollution. Collapse of the dike used to contain coal ash by the Tennessee Valley Authority plant at Kingston Tennessee resulted in spillage of over 4 million cubic meters of coal ash into nearby rivers and surrounding areas. Elevations of various neurotoxic and teratogenic metals and compounds, especially methyl mercury, were reported in the area (142–144).

Based on studies that reported the possible harmful effects that could result from improper disposal and storage of coal ash, in 2015 the EPA released a Coal Combustion Residuals (CCR) Rule that set requirements for the disposal of coal ash from coal-fired power plants (37). The CCR Rule was established under the RCRA, Subtitle D. However, although regulations were set forth, coal ash was still not considered a hazardous waste. The CCR Rule addressed regulations of ponds and landfills, plant's location, safety practices, ground water protection, transfer of particles into air as dust, and rules for ash impoundment sites (37). In addition, the rule required facilities to document and make public any information or changes they make regarding the ruling (37). The rule provided a comprehensive and more rigorous design, monitoring, operating, corrective action, closure, and post-closure requirements for CCR landfills and surface impoundments. The CCR Rule was applicable to old, new, and existing facilities and did not apply to plants and facilities that were already closed or no longer being used to generate energy. The CCR Rule was in existence until July 2018, when the current government of the United States started to roll back the regulations to allow more flexibility to the industry.

Disposal regulations in India

Approximately 70–75% of electricity generated in India comes from coal-fired power plants (9, 62, 64, 145). The quality of coal is poor, having a low calorific value and a high ash content (9, 64, 145). The use of high ash coal produces fly ash in large quantities, and disposal and storage have become a problem, throughout India, although fly ash is not currently considered to be a hazardous waste.

The government of India has attempted to regulate fly ash based on utilization. In 1999, the Ministry of Environment and Forests (MoEF) issued a regulation mandating the need for the reutilization of fly ash. According to the regulation, it was mandatory for existing coal-fired power plants and new coal-fired power plants to utilize 100% of the generated fly ash in a specified time period. New coal-fired power plants were required to use 100% of fly ash within 9 years of start-up. Old plants were required to achieve 100% utilization within 15 years from the MoEF regulation (9). This MoEF regulation also mandated that fly ash be provided free of charge to potential users until 2009. Utilization rates throughout India increased, but could not keep up with the amount of ash that was generated. One-hundred percent re-utilization has never been achieved. In response to not being able to meet the 100% utilization rate, the Ministry of Environment, Forests and Climate Change (MoEFCC) mandated that in addition to being required to utilize 100% of fly ash, the power plants were required to pay for the transportation of fly ash to sites within a radius of 300 km of road construction projects and programs of the government involving construction of buildings, roads, dams, and embankments. Furthermore, the MoEFCC granted permission for fly ash to be used in agriculture. Plants are required to give free fly ash within a 300-km radius (62).

Disposal regulations in China

The rapid economic and population growth in China comes with a high energy demand and enormous production of CCPs (60, 146). In China, the increased production of fly ash alongside with its environmental impact has necessitated an increase in the utilization of fly ash, as a regulatory measure (147, 148). China does not consider fly ash a hazardous waste; instead, the country classifies it as a solid industrial waste (16, 60). In China, two agencies are responsible for coal ash management: the National Development and Reform Commission (NDRC) and the Ministry of Environmental Protection (MEP). NDRC is responsible for overseeing fly ash reutilization, and MEP is responsible for ensuring that fly ash does not pollute the environment (16). Subsequently, a series of standards, regulations, and requirements were introduced including the Management Measure for Comprehensive Utilization of Fly Ash, which entails special measures regarding the utilization of fly ash (60, 148).

He et al. (60) reported that the Law of the People's Republic of China on the Prevention and Control of Environmental Pollution by Solid Waste further divides solid wastes into class I and class II. Fly ash is marked as a class II solid waste, and therefore the fly ash that is not reutilized can be placed in storage facilities. For ash that is not reutilized, the Standard for Pollution Control on the Storage and Disposal Site for General Industrial Solid Wastes applies (16, 60). This standard provides directions for where the waste should be stored and measures to prevent fugitive dust and leakage from the storage sites (60). However, according to investigations by Greenpeace, the majority of coal ash storage sites did not meet minimal standards. For example, one of the fly ash storage sites was found to be within 500 m of residential homes, which violates one of the regulations in the Standards for

Pollution Control on the Storage and Disposal of Sites for General Solid Waste (149).

Although there are other regulations that can be applied to fly ash storage facilities, most are voluntary. He et al. claim that the standards are ineffective in constraining coal-fired power plants (60).

Exposure pathways to humans

Inhalation

As fly ash contains particles in the respirable range, unused fly ash stored in landfills and surface impoundments can lead to air pollution and resulting health conditions associated with exposure to particulate matter (41, 58, 60). Poorly managed storage facilities that hold dry ash are susceptible to winds that create fugitive dust that can impact communities living near the storage facilities. The smaller particles, those with diameters less than 10 μm when inhaled, can penetrate deep into the lungs leading to inflammation of cells and irritation of the mucous membrane of the respiratory tract leading to respiratory and cardiovascular conditions associated with exposure to particulate matter (150–153).

In addition to respiratory and cardiovascular conditions, chronic exposure to particulate matter has been found to cause chronic inflammation and elevated levels of cytokines in the body and brain increasing the risk for diseases related to the CNS (154, 155). Researchers have shown that metals can enter the brain through several pathways, including the nasal olfactory pathway and by crossing red blood cell membranes.

The research by Calderon-Garciduenas et al. (155) showed that particulate matter <2.5 μm ($PM_{2.5}$) is capable of passing through the nasal olfactory pathway into the circulatory system and brain. Researchers hypothesize that the large ratio of surface to volume allows the particles to penetrate cell membranes and pass through the lung tissue and the blood-brain barrier. Particulate matter that crosses the blood-brain barrier introduces the components in the particles to the brain and bloodstream. Researchers call this the Trojan Horse Effect (154, 155). This effect may potentially allow concentrations of metals from fly ash access to the brain, producing CNS disease.

Ingestion

In addition to the threat of air pollution from fugitive dust, long-term storage of fly ash in surface impoundments may impact the environment and human health. Water infiltration, leaky storage sites, and unlined sites may result in metals and other elements leaching from fly ash into the environment. The leaching of elements from fly ash has the potential to impact groundwater and surface water, which is a major concern for humans, especially populations that rely on well water as their drinking source.

Elements that leach from fly ash particles tend to be positioned on the surface of the particle, where there is a gradient of element concentrations (156, 157). Although the surface layer is only microns in thickness, it contains a significant amount of leachable elements (156). The surface layer of fly ash particles is more likely to contain trace elements such as As, boron (B), Hg, Cr, selenium (Se), Cd, copper (Cu), molybdenum (Mo), antimony (Sb), vanadium (V), and zinc (Zn). Elements that tend to distribute throughout the core of the particle, such as manganese (Mn) and Pb, are not directly exposed to leaching (158). The releases of elements distributed throughout the core are controlled by diffusion and are dependent on the dissolution rates of the surface layers (157).

The leaching behavior of trace metals from fly ash has been well studied. The ability of elements to leach from fly ash depends on properties including the pH of the fly ash, pH of the water, amount of lime present in the fly ash, mineralogy of other compounds, particle size, time, reduction-oxidation conditions, liquid-to-solid ratio (L/S), and temperature (34, 72, 98, 100, 145, 159–164). However, it is reported that pH is the greatest factor for mobility of trace metals in water, and that the greatest leaching of metals occurs under an acidic environment (34, 160, 163, 165–168). As different coals contain different trace elements and have different pHs, leaching of metals from fly ash can vary widely (72).

Leaching of As, which is oxyanionic and positioned on the surface of the fly ash particle, is a major threat from disposal sites because it is mobile throughout a range of pH values (116). Van der Hoek et al. reported that releases from acidic fly ash increase with pH, whereas in alkaline fly ash this trend is reversed (169). In India, where ash content in coal is high, Kapoor and Christian reported that fluorine (F), As, magnesium (Mg), and Zn showed solubility with water and were leached in higher concentrations at acidic pH and higher temperature (170). Researchers have reported that AsIII is more mobile in the environment, because it is more weakly bound to mineral surfaces, compared with AsV (72, 171–173).

The mobility of chromium is highly dependent on its oxidation state and pH (114). Hexavalent chromium is much more soluble than trivalent chromium. As previously stated, most research has assessed CrIII and reported that lower amounts are leached with near-neutral pH, but shows a leachability plateau from pH 8 to 12. Dubikova et al. also reported slightly higher mobility for alkaline ash and increasing releases with increasing pH (174).

In experiments reported by the USGS, two fly ash samples were placed in small volumes of oxic and anoxic freshwater (pH = 7) and measured over time. Arsenic dissolved into the freshwater under both oxic and anoxic conditions and increased over time. At the end of the experiment, concentrations were above the EPA Maximum Contaminant Level (10 ppb) for total As. In the same experiment, it was observed that Cr dissolved from one fly ash sample in oxic freshwater only. Chromium did not dissolve from the second sample of fly ash. Researchers reported that Cr was probably present in insoluble species. During the study time, Cr concentrations were below the EPA Maximum Contaminant Level (100 ppb) (77).

Although Cd is located on the surface of fly ash particles, it is not very mobile in neutral or alkaline conditions, and only somewhat soluble in acidic conditions. Researchers have shown that despite the fact that Cd is located on the surface of particles, concentrations leached in water rarely surpass 0.01 mg/kg in alkaline-natured fly ash (61, 175, 176), while values for acidic fly ash are approximately 0.1 mg/kg at pH = 4 (177, 178).

Lead is dispersed throughout the core of fly ash particles and as a result, Pb is highly insoluble and virtually immobile (<1% and often <0.1%) in both acidic- and alkaline-natured fly ash samples. Researchers have shown that the immobility and insolubility occurs irrespective of the pH (175, 176, 179, 180). As with other cations, acidic conditions slightly enhance Pb leaching (154). Since Pb is a cation, acidic conditions slightly improve Pb leaching (158, 161); however, the concentrations remain at very low levels. Multiple researchers have shown that among 30 samples of fly ash with pH 11–13, none of them leaches Pb levels greater than 0.6 mg/kg (34, 174–176). Researchers from the United States reported that <1 mg/kg of Pb was released from fly ash using a range of extractants and tests (179, 181).

In addition to pH, the L/S ratio is an important factor when assessing leaching behavior. A lower L/S ratio is indicative of a material with a restricted flow, and a higher L/S ratio is associated with a material having a greater flow. Researchers evaluating the impact of L/S ratios on mobility report that as the L/S ratio increases, concentrations of elements reduces. Da Silva et al. (72) showed that when the L/S ratio was increased from 0.5 to 10, concentrations of As, Cd, Cr, Hg, Pb, and Se decreased. The researchers stated that the elements reached their maximum leaching at an L/S of 0.5 (72). Dandautiya et al. (182) assessed L/S ratios from 5 to 50 and reported that Zn, Iron (Fe), Cr, Pb, Se, strontium (Sr), V, titanium (Ti), aluminum (Al), calcium (Ca), and Mg showed maximum leachability at an L/S ratio of 5. The maximum concentration of As occurred at an L/S of 10 (182). Zandi and Russel (165) showed that leaching of most elements is not based on the L/S ratio when the L/S ratios are higher than 50, which is in agreement with the work of Jones (158).

Contamination of surface and ground water with metals has been reported in several studies carried out at locations near fly ash disposal facilities (41, 70, 144, 162, 170, 183–186). In these cases, concentrations of elements have been reported to be higher than country or the World Health Organization (WHO) environmental standards. While many studies report elevated levels of metals, Dandautiya et al. assessed groundwater contaminants around a coal-fired plant in India and found that the concentrations of Zn, As, Cu, Nickel (Ni), Pb, Ca, Mg, Cd, Cr, and Mn were below the WHO standards (182).

Fly ash exposure and human health

Limited epidemiological studies exist that assess the relationship between exposure to fly ash and human health. Some research has focused on the health impacts among coal-fired power plant workers exposed to “dust”, without directly studying fly ash exposure. In these studies, researchers measured dust levels in different locations, including where the coal was handled and where coal ash was removed. In some cases, they created job exposure levels to account for exposure. Few researchers have directly assessed exposure to fly ash on health among occupationally exposed workers. Those who did have reported increased oxidative stress, DNA damage, genotoxic damage, and decreased respiratory function.

Research addressing the direct exposure to fly ash on communities does not exist. Most research investigates the impact of coal-fired power plants and health in communities by utilizing distance from the coal-fired power plant as a proxy for fly ash exposure. This section highlights some of the limited epidemiological research conducted among coal-fired power plant workers, workers directly exposed to fly ash or other CCPs, and community findings.

Health outcomes among coal-fired power plant workers

Few researchers have assessed cancer morbidity and mortality (187–189), markers of oxidative stress (190), and immunological profiles (191) in employees of coal-fired power plants.

Forastiere (188) determined that there was a slight excess in total cancer mortality among 406 workers employed in two power plants in Italy. This excess was mainly due to the increased respiratory cancer that was not statistically significant. However, when assessing lung cancer, researchers determined that the observed number of lung cancer cases was greater than the expected number of cases. Although the standard mortality ratio (SMR) was increased (SMR = 178), it was not statistically significant as the confidence intervals (CIs) contained the null (90% CIs = 88–321). However, the SMR was significantly elevated for lung cancer in workers <60 years old. Furthermore, the SMRs were increased when the length of exposure and latency period from first employment exceeded 10 years (188). These results were similar to findings from two other epidemiological studies of workers and lung cancer (192, 193). However, the sample size in all three studies was small and the SMRs were not statistically significant (188).

Kaur et al. (190) studied 200 males who were divided into four groups: coal handling workers, turbine unit workers, boiler unit workers, and city electricians. They found that malondialdehyde (MDA) levels were increased in workers in the coal handling unit, turbine unit, and boiler unit compared to the electricians (190). MDA is a byproduct of lipid peroxidation during oxidation stress. Oxidative stress has been implicated in many diseases, including neurodegenerative diseases such as Alzheimer's disease (194–198), atherosclerosis and cardiovascular disease (199–203), diabetic neuropathy (204, 205), and chronic obstructive pulmonary disease (COPD) (206, 207).

Bencko et al. (189) conducted a retrospective study of mortality from cancer among workers exposed to arsenic in coal-fired power plants. The researchers assessed cancer mortality patterns among male workers in one coal-fired power plant where high arsenic content coal was combusted (exposed group) vs. male workers from three coal-fired power plants where low arsenic content coal was combusted (unexposed group). The rate of mortality in the exposed group was higher than the mortality in the unexposed group (38% exposed vs. 23% unexposed), but the difference was not significant. However, compared to the unexposed group, exposed workers died of cancer at shorter exposure intervals and younger age (189). In the same population, Bencko et al. examined the immunological profiles of workers and reported that the exposed population had significantly higher levels of carrier proteins, ceruloplasmin, transferrin, and orosomucoid, compared with the unexposed population (191).

Health outcomes of workers exposed to fly ash and CCP

Research assessing direct exposure to fly ash or other CCPs is limited. Recent occupational studies have found that exposure to coal ash may cause genotoxic effects and disrupt biological processes. A case-control study conducted by Celik et al. (208) investigated the genotoxic damage among workers in a coal-fired power plant compared with healthy controls. Investigators found that the mean frequencies of chromosomal aberrations (CA), SCEs, MN, and polyploidy were all significantly elevated when compared to the controls. Compared with controls, cases had increased frequency of CA ($p < 0.01$) and increased percentage of cells with aberrations ($p < 0.01$) in peripheral lymphocytes. Researchers reported that the most common type of aberration in cases and controls was the chromatid break, followed by a chromosome break. In addition to increased CA in the cases, there were also significant increases in SCE ($p < 0.01$), MN ($p < 0.05$), and the frequency of polyploidy ($p < 0.01$), compared with controls (208). Celik et al. (208) further investigated the genotoxic damage with respect to exposure time and age of the workers. They found a positive correlation between years of exposure and CA ($p < 0.05$) and MN ($p < 0.05$), but not with SCE. Age was not correlated with genotoxic damage.

In a more recent study, Zeneli et al. (209) examined levels of metals and oxidative stress in 70 male workers exposed to fly ash (cases) compared with 27 men (controls) residing in a rural area of Kosovo. To measure levels of oxidative stress, researchers measured glutathione peroxidase (GPx), superoxide dismutase (SOD), and ascorbic acid in blood. Metals were also measured in blood. Compared with controls, cases had significantly higher levels of As ($p < 0.02$), mercury (Hg) ($p < 0.01$), Zn ($p < 0.007$), and Se levels ($p < 0.0002$), but lower levels of Cu ($p < 0.002$). The concentration of Cd in the cases was higher than that in controls, but it was not statistically significant ($p > 0.25$). Researchers reported that the concentration of Hg in the power plant workers was 4.3 times greater compared to the control group. Arsenic was 1.7 times higher in the cases compared to the controls. Zeneli et al. (209) also found that workers had significantly lower levels of SOD ($p < 0.002$) and GPx ($p < 0.001$), and levels of ascorbic acid was significantly lower ($p < 0.0001$). From their research, the authors stated that the antioxidant potential of the cases significantly differed from the controls as manifested by significant changes in the activity of SOD and GPx and ascorbic acid (209).

Since fly ash consists of respirable particles, it would seem that researchers would have thoroughly investigated the impact of fly ash on respiratory function; however, this is not the case (65, 210–212). Schilling et al. (210) assessed lung function among 268 worked who were exposed to fly ash for more than 10 years. They created three exposure groups (low, medium, and high), based on job and years of exposure. Researchers found that those workers who were in the high-exposure group had a greater prevalence of chronic cough, chronic phlegm, wheeze, chest tightness, and difficulty in breathing. Researchers highlighted that these results needed to be interpreted with caution, because of the large number of smokers and ex-smokers in the sample. To further investigate lung function and fly ash, researchers measured concentrations of fly ash exposure and performed lung function testing. Concentrations of fly ash ranged from 0.08 to 21.8 mg/m^3 in the low-exposure group, from 0.12 to 73.31 mg/m^3 in the middle group, and from 0.07 to 98.62 mg/m^3 in the high-exposure group (210). Researchers reported that as fly ash exposure increased, an increasing effect on several lung function tests occurred. Specifically, they reported that modest effects were shown on forced vital capacity (FVC), vital capacity (VC), forced expiratory volume in one second (FEV1), peak flow (PF), and gas transfer (DCO) among the workers in the high-exposure group (210).

In two case reports, men who were exposed to large amounts of fly ash developed lung disease. Cho et al. reported that a man unloading his truck and exposed to a large amount of fly ash developed an acute case of silicosis within 2 weeks of exposure (211). Interestingly, no other research has investigated silicosis in workers exposed to fly ash. In a second case report, Davison et al. (212) reported that a 27-year-old worker exposed to fly ash developed asthma due to his exposure at work. The man's peak expiratory flow (PEF) was taken every 2 h for 28 days, which got worse during work, but improved when he was not working. During an inhalation test, the man responded to exposure to fly ash, but not to the placebo (212).

Community health in relation to coal-fired power plants

The effect of direct exposure to fly ash or other CCP has not been studied in communities. Researchers have predominately assessed the health impacts from coal-fired power plant emissions based on proximity to plants. Other researchers assessed health conditions after a power plant closed. In addition to particulate matter, many researchers have assessed multiple pollutants that are emitted from coal-fired power plants, such as sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and mercury (Hg). Although there is no research on direct exposure to fly ash, there is extensive epidemiological evidence reporting that emissions from coal-fired power plants deleteriously impact the health of neighboring communities. Researchers have reported significant associations between power plant emissions and mortality, poor respiratory health, cancer (lung, larynx, and bladder), skin conditions, higher urinary markers for polycyclic aromatic hydrocarbons (PAHs) and metals, poor birth outcomes, and neurobehavioral problems (213–228).

In a recent study, Lin et al. (219) assessed the impact of coal-fired power plants and the burden of lung cancer. To assess the burden globally, researchers evaluated annual lung cancer incidence rates from 83 countries with coal-fired power plants by utilizing the per capita coal capacities for each country. Researchers reported that over 860,000 male and over 540,000 female lung cancer cases were attributed to power plants that used coal as the primary energy source (219). Collarile et al. (217) investigated the risk of lung and bladder cancer in people living near a coal-oil-fired power plant. Based on tertiles of exposure to benzene, NO_2, SO_2, and PM_{10}, incidence rate ratios (IRR) by sex, age, and histology were computed among 1076 incident cases of lung and 650 cases of bladder cancers. For lung cancer, no excess risk was found in men, but an excess risk was found in women aged ≥75 years in the highest tertile of exposure to benzene, NO_2, SO_2, and PM_{10}. No excess risk for bladder cancer was found in men, and only women aged ≥75 years in the highest tertile had an excess risk for bladder cancer. A major limitation of this study was that smoking data were not collected on the participants, which may have confounded the relationship between exposure and disease (217).

Respiratory outcomes have been well studied. In one study on respiratory complaints and spirometric parameters, Karavus et al. (213) compared residents who lived 5 km around a coal-fired power plant to residents who lived at least 30 km away from the power plant. Residents who lived closer to the power plant were significantly more likely to report chest tightness and coughing attacks lasting for more than 1 year, compared to the residents who lived farther away (chest tightness, 46.2% vs. 29.2%, $p = 0.001$; cough, 28% vs. 20.8%, $p = 0.024$). However, productive coughing was not found to differ among the two groups (13.3% vs. 8.4%, $p = 0.89$). Among non-smokers, Karavus et al. found that spirometric results differed between the people residing near the power plant and those who lived farther away. People residing closer to the power plant had lower FEV1, FVC, and mean forced expiratory flow during the middle half of the FVC (FEF25–75%) ($p < 0.001$ for all tests) (213).

Hagemeyer et al. (214) assessed respiratory health in adults living within four neighborhoods adjacent to a coal-fired power plant with coal ash storage facilities and compared the results to unexposed comparison population. In this cross-sectional study, the researchers found that exposed adults were statistically more likely to report cough [adjusted odds ratio (AOR) = 5.30, 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), and respiratory infections (AOR = 1.82, 95% CI = 1.14–2.89). Furthermore, the researchers found that the exposed population had a significantly lower mean overall respiratory health score, compared with the non-exposed population (2.82 vs. 3.87, $p < 0.0001$) (214).

Researchers investigating the impact of coal-fired power plant emissions on children's respiratory health have been consistent in showing its health impact. Children are considered a vulnerable population because they exhibit behaviors that will increase their risk of exposure.

Children tend to breathe through their mouths and have larger lung surface area per body weight, compared to adults. Children's lungs do not develop completely until late teen years, and developing organs like the lungs are more susceptible to toxic effects and environmental pollutants. In addition, children absorb more pollutants than adults and are more likely to retain them over time because they tend to spend more time outside. Children tend to be mouth breathers as well (not just nasal breathers) which increases their ventilation rate, thereby increasing the amount of pollutants taken up by the lungs. Undeveloped immune system could also pose health risks in children (229, 230).

Rodriguez-Villamizar et al. (215) conducted a spatial cross-sectional study to assess the association between emergency department (ED) visits for asthma and residential proximity to a coal-fired power plant and a petrochemical facility. The researchers found that children who lived closer to the power plant were 10 times more likely to be admitted to the ED for asthma, than children who lived farther away. After adjusting for age, socioeconomic status, and gender, an inverse association between asthma visits and distance from the power plant was determined. Interestingly, there was no association when examining relationships with the nearby petrochemical facility (215).

Sears and Zierold (224) utilized a cross-sectional epidemiological study to investigate health in children living adjacent to a coal-fired power plant with coal ash storage facilities and in children living 60 miles away. Although the researchers found associations with other health conditions [attention-deficit hyperactivity disorder (ADHD) and gastrointestinal problems], they found an elevated but not significant AOR for the prevalence of asthma (AOR = 2.52, 95% CI = 0.8–7.6) (224).

Studies on neurodevelopment in children exposed to power plant emissions are few, but indicate a relationship. Tang et al. (221) assessed the effects of prenatal exposure to coal-burning pollutants on children's development in China and found that exposure to pollutants from power plants harmed the development of children living in the study region. The researchers concluded that the level of PAH-DNA adducts in cord blood of newborns was associated with reductions in developmental quotients in both motor and language areas. Additionally, the researchers found that in utero exposure to lead from the plant deleteriously affected social development of the children (221).

A study by Liang et al. determined that coal combusted fly ash is a dominant source of Pb exposure for children living in Shanghai (231). The researchers estimated that Pb pollution in air is caused mainly by coal combustion (50%), metallurgic dust (35%), and vehicle exhaust (15%). As previously noted, Pb is associated with many neurobehavioral heath disorders in children. Furthermore, the researchers stated that based on the Pb isotopes of the children's blood, they believed that Pb from fly ash is much easily absorbed into the blood than Pb from vehicle exhaust or metallurgic dust (231).

In a study conducted in the United States, Sears and Zierold (224) assessed the relationship between proximity to a coal-fired power plant with coal ash storage facilities and ADHD, learning difficulties, and emotional and behavioral problems. The researchers found that children living adjacent to a coal-fired power plant had an increased prevalence of ADHD compared with unexposed children living 60 miles away (AOR = 3.6, 95% CI = 1.2–10.7). The logistic regression model used to determine the odds ratio was adjusted for secondhand smoke exposure, age, and gender (224).

To date, there is no research on community populations directly exposed to fly ash; however, there is extensive epidemiological evidence reporting that emissions from coal-fired power plants deleteriously impact the health of neighboring communities. Research is needed to advance our understanding of the health impact of fly ash, particularly as storage continues to increase in some countries while other countries battle to maintain ash that is stored in unlined landfills and surface impoundments.

Conclusion

Fly ash, which results from the combustion of coal, is of environmental concern. Although fly ash can be beneficially reutilized in products such as concrete, a large percentage of it is stored in landfills and surface impoundments. Fugitive dust emissions and leaching of elements into groundwater are pathways for which human populations may be exposed to the respirable fraction of fugitive dust and the trace elements in fly ash. Unfortunately, limited research on the health impacts of fly ash exists. Longer-term monitoring and direct exposure assessments of communities impacted by fly ash are needed. Although researchers have proven that many of the trace elements in fly ash, such as lead and arsenic, are harmful to human health, questions such as the concentrations to which people are exposed, duration of exposure, and the reaction of these elements synergistically need to be answered to understand the health impact of exposure to fly ash. Epidemiological and environmental research on the impact of fly ash and other CCPs is needed to further policy and regulations on storage, disposal, transportation, and clean-up.

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