HAZARDOUS ELEMENTS IN AQUEOUS LEACHATES OF COAL FLY ASH AROUND THERMAL POWER PLANTS DISPOSAL AREA

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The fossils fuels are main resources of energy worldwide. Huge production of coal fly ash creates water and air pollution. Coal fly ash generally contains trace toxic metals like Ni, Hg, As, Cr, Cd, Ba, Mn, and Pb etc., so coal fly ash is considered as an environmental hazard worldwide. These toxic elements leached from coal fly ash into soil and water which contaminate soil, ground water and other water sources. In present paper is the study of water samples collected from different locations near fly ash disposal area. We found that the water samples contain hazardous trace metals in concentration higher than the permissible limits of drinking water standard. Coal fly ash and other constituents which are generated from coal can create harmful effect on environment, society, health and life. A better method is required to disposal of coal fly ash generated from thermal power plant in India to stop contamination of environment as it is a matter of great concern to human health.

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INTRODUCTION

The human life depends a lot on energy and its resources. The fossils fuels are the main resources to generate energy world-wide apart from nuclear and other renewable energy resources. Coal and oil are two fossils fuels are present world-wide and coal is used extensively to generate energy worldwide.[1]

Mainly coal-based thermal power plants meet energy requirements of developing countries and play major role to meet global energy demand. Its low cost and easy availability made it the fuel that launched industrial revolution in world. The geological resources of coal reserve of India have so far been estimated at about 326.05 billion tonnes in 2018. Total annual production reached from 675.40 million tons (Mt) in 2017-18 to 730.35 Mt in India during 2018-19. The import of coal reached up to about 235.24 Mt in 2018-19 and it was 166.86 Mt in 2013-14.[2] Coal contributed about 60 % (194,402.88 MW) of total installed capacity (329,204.53 MW) power generation in India as on 30 April 2017. Total 624.88 million tons coal was consumed by 167 coal-based power plants in 2017-18. Coal-based power generation produces massive amounts of fly ash worldwide. About 196.44 Mt coal fly ash generated in 2017-18 as the coal comprises of 31.44 % of fly ash and 67.13 % of utilization. Management of fly ash in coal based thermal power stations in country is a challenging task in view of large quantity of fly ash generated and target of achieving 100% utilization.[3,4]

India together China and US accounted for more than two thirds of global increase in energy demand. India imports fossil fuels equivalent to 373.3 Mt oil equivalent (Mtoe) which is equal to 46.13 % of total primary energy consumption in 2018. It is anticipated that amount of coal consumed, in India, per year will be increase from 3840 Mtoe in 2015 to 4032 Mtoe in 2035.[5] India is largely dependent on fossil fuel imports to meet its energy demands by 2030, India's dependence on energy imports is expected to exceed 53 % of the country's total energy consumption.[6]

Carbon, sulphur, oxygen, hydrogen and nitrogen are the main components of coal. In addition, coal has some traces of heavy metals. Coal fly ash is one of the major concerns for the scientific community due its huge amount of production worldwide and its toxicity.[7] With increasing poisoning incidents related with hazardous trace elements in recent years, the adverse effects of these elements on human health and eco-environment have been highlighted.[8] The U. S. Clean Air Act Amendments,[9] European Union and the Canadian Environmental Protection Agency[10] have listed some hazardous trace elements such as Hg, As, Pb, etc., as the main environmental concerns. Coal combustion for power generation is considered as one of the major emission sources of hazardous trace elements.[11] The phase transformations that mineral matter in coal undergoes during high temperature combustion may render the toxic trace elements in the original coal matrix susceptible to leaching, likely to be release into environment after encountering water on application/storage/disposal in landfills or lagoon.[12] However, the borderless nature of the environment could result in the transfer of pollutants into groundwater and river systems, either in dissolved or particulate form. This may pose serious threats for aquatic organisms, while metal inputs in groundwater resources may entail a significant health hazard. The worldwide standard leaching tests can be found in various studies.[13-15].The hazardous trace elements can be leached out from the fly ash and may contaminate soil, groundwater, and surface water.[16,17]

The leached concentrations of elements expressed in terms of absolute value may certainly differ, but the leaching behavior appears relatively common patterns. It can cause water and soil pollution, disrupt ecological cycles and pose environmental hazards.[17] Coal fly ash has many heavy metals including As, Cu, Ni, Zn, Pb, Cd, Cr, Hg, that are usually implicated in causing adverse effect on living systems. The effects have been attributed to various oxidative stress reactions.[18,19]

The coal fly ash contains Si, Al, and Fe as major elements. Ca in coal fly ash is found as the dominant cation followed by Mg, Na, and K.[20] It also has great adverse effects on environment as it releases substantial amounts of trace metals like Mo, Mn, Zn and Cu along with significant amount of other toxic elements (i.e. As, Cr, Co, Pb, Ni, Se) which are major source of soil and water pollution.[21,22] Arsenic contamination has turned into a major global concern with constantly growing pollution of soil, water, and crops. The fuel gas cools down after combustion and volatile elements such as As, B, Hg, Cl, Cr, Se and most prominently S, condense on the surface of the fly ash particles, forming compounds with a high leachability. Furthermore, coal fly ash contains a high amount of soluble salts which support contaminants to move from soils into groundwater through leaching and results in water pollution.[20] Thus, disposal of huge amount of coal fly ash leads to the release of toxic elements which in turn contaminate soil and groundwater resulting in a potential environmental hazard and great effect on the environment throughout the globe. The extensive amount of coal fly ash imposes serious environmental threat[23]. The human health may be negatively influenced by leachate and its noxious compounds in the vicinity of a coal fly ash landfill site[23-25] and some studies have shown that leachate produced by landfills can be toxic to groundwater, and even to humans in the proximity to the landfill.[26] It is imperative to develop new techniques for reuse, recycling and recovery of various elements from coal fly ash.[27,28] Huge amounts of coal fly ash are produced globally but only 1/4th of the total production of coal fly ash is utilized.[29] Toxins present in coal fly ash contaminate air quality when it exposed to the environment. Oversized ash pond unable to control huge amount of ash and breach, causing numerous ecological problems and severe below the ground and above the ground distress to environment and local communities.[30]

STUDY AREA

The study area is near Sardar Gobind Ballabh Pant Sagar also known as Rihand Dam near village Pipri in Sonbhadra district of the state of Uttar Pradesh, India. Its reservoir area is located on the border of Madhya Pradesh and Uttar Pradesh sates. Rihand Dam is based on the Rihand River, a tributary of the Son River. The catchment area of this dam extends over Uttar Pradesh, Madhya Pradesh and Chhattisgarh states whereas it supplies irrigation water in Bihar state located downstream of the river. The water stored in Rihand Dam is released into River Son periodically for irrigation purpose throughout the year. The Dam lies between latitude 24^0 00′ 00″ and 24˚ 12′43 ″N and longitudes 82^0 38′00″ and 83^0 00′00″ N. It has power house of 300 MW (6 units of 50 MW each) installed capacity. The water collected in the Govind Ballabh Pant Sagar reservoir is diverted to the Son canal which irrigates about 2.5 lakh hectares of the agricultural land in Champaran, Darbhanga and Muzaffarpur districts of Bihar (India).

The southern region of Sonebhadra district, Eastern Uttar Pradesh is referred as Energy Capital of India because the region has large reserves of coal and numbers of power plants of UPRVUNL and NTPC are installed around the Govind Ballabh Pant Sagar. The region produces more than 10,000 MW of electricity. This area is used for coal mines, coal-fired power plants, coal fly ash and coal slurry disposal areas. Many super thermal power stations are located in the catchment area of the dam. These are Singrauli, Vindyachal, Rihand, Anpara and Sasan super thermal power stations and Renukoot thermal station. The highly contaminated water from the ash dumping area of these coal-fired power stations ultimately collects in this reservoir enhancing contamination of its water.

EXPERIMENTAL

Coal fly ash sample collection

The detail information studied three coal-fired power plants is shown in Table 1. The coal fly ash samples were collected from electrostatic precipitator (ESP) of the three power plants, transported and mixed uniformly. The fly ash was collected four times at every two hours from each unit of all power plants to get representative sample. Total 104 number of samples collected from all 26 units of different capacity of all three plants. The fly ash collected from electrostatic precipitators has temperature about 60-70 °C, fly ash was allowed to cool down at ambient temperature before mixing. All samples were mixed thoroughly to get representative sample. A composite sample has prepared by mixing the separate samples in equal proportion. About 250 g of composite sample of coal fly ash was used for elemental analysis.

Table 1. List of locations of sample collection.

Power	Location	Capacity	Total
No. 1	NTPC, Shaktinagar, U.P.	2000 MW	5x200 2x500
No. 2	NTPC, Rihandnagar,U. P.	3000 MW	6x500
No. 3	NTPC, Vindhyanagar, M. P.	4760 MW	6x210 7x500

Coal fly ash elemental analysis

Coal fly ash is hygroscopic in nature. Approximately 250 g of composite sample was put in muffle furnace (M/s Usha Instrument Pvt Ltd.) at108 ± 2 ˚C for one hour to one and half hour, as per IS 1350 part 2, to dry fly ash and to determine the moisture contain on air dried basis. Sample was sent to ATMY Analytical Labs Pvt. Ltd. (NABL/ISO 17025 Accredited Laboratory), I-38, DLF Industrial Area phase-1, Faridabad (Haryana) India for fly ash elemental analysis and rest of the composite sample was stored for future elemental analysis.

Fly ash collected from ESP is solid in nature, it was required to convert in solution form to run sample in ICP-OES. The trace metal concentrations in coal fly ash sample were determined by ICP-OES after acid digestion. To digest coal fly ash, 0.10 g of coal fly ash sample was digested in microwave with aqua regia at 220 ˚C for 30 min and made up with ultrapure distilled water. Analysis of coal fly ash done by using instrument ICP-OES model Optima™ 3300 RL ICP-OES make PerkinElmer® equipped with WinLab32™ for ICP Version 4.0 software for simultaneous measurement of all wavelengths. Plasma, auxillary and Nebulizer gas flows (argon) were 16, 1 and 0.95 L min^{-1} L, respectively. RF power was 1100 W.

Standard solution prepared by Perkin Elmer and VHG NIST® traceable quality control standards for ICP-OES (N9302946, 987841-2) standard used as the stock standards for preparing different working standards. Millipore water acidified with 5 % nitric acid was used as blank. All standard was prepared in ultrapure distilled water acidified with nitric acid with a range of 0.05 ppm to 25 ppm, the standard solution was prepared from 1000 ppm stock solution.

Unburned carbon content

Unburned carbon content in coal fly ash sample was determined by weight loss as per ASTM D 7348. The sample were placed in muffle furnace make at 815 ± 10 ˚C for 1 h after being dried at 108 ± 2 ˚C.

Water sample collection from different locations near fly ash pond

Sonebhadra of Uttar Pradesh known as power capital, contributed more than 10000 MW power generation, huge coal is consumed in this area and coal fly ash is disposed in Ash ponds near Rihand dam. Huge amount of coal extracted by Northern Coal Fields Ltd in this area also contribute to water pollution. Water sample collected from different locations near fly ash pond has been shown in Table 2.

RESULTS AND DISCUSSION

Coal fly ash characterization

The concentration of elements presents in the samples collected from various locations around fly ash disposal area is shown in Table 2. The elements are present mainly in the form of oxides/silicates. The three major constituents of fly ash are silicon, aluminium and iron, which are presents in the form of their oxides. Figure 1 shows silicon is the main constituents of coal fly ash, 28.69 % silicon is present in coal fly ash, second major components of coal fly ash is aluminium which contribute 14.48 %. Various studies have been conducted to develop technologies to clean extraction of aluminium from coal fly ash which has high alumina content. High-alumina coal fly ash would be beneficial to achieve better overall coal fly ash utilization, environmental and economic benefits. Alumina extraction from sulphuric acid leachate of high-alumina coal fly ash by electrolysis is nonhazardous.[27] The elements, present in coal fly ash meet the specific requirements of ASTM C-618 class F coal fly ash (ASTM, C-618, 2015) and Indian Standard (IS:3812, 1981) to use as pozzolana in cement, cement mortar and concrete; total percentage sum of SiO_2, Al_2O_3 and Fe_2O_3 content in coal fly ash should not be more than 70 % and CaO content should be less than 10 %. Similar results have been shown in other studies.[31] The three major constituent elements make up 96.8 % of coal fly ash. Minor constituents are calcium, magnesium, phosphorous in the form of oxides, calcium oxide (CaO), magnesium oxide (MgO), phosphorous pentoxide (P_2O_5) and sulphur trioxide (SO_3) are present as oxides of silicates and aluminosilicates.[32]

Heavy trace elements

The ICP-OES elemental analysis has been shown, 13 heavy trace metals also present in composite coal fly ash sample. All thirteen toxic trace heavy metals were found in detectable concentration in coal fly ash sample. Seven hazardous trace elements have been studied in coal and corresponding fly ash from four power plants in China and it is reported the elements concentration in coal fly ash is more than that in coal. The concentration of Hg, As, Cr, Cd, Ba, Mn and Pb in the four samples of coal fly ash are in the range of 0.17–1.26, 5.15–25.74, 43.25–64.61, 0.56–0.56, 777.05–970.70, 163.83-831.47 and 28.94–119.57 mg kg^{-1}, respectively.[33]

Water sample analysis

ICP-OES study of water samples collected from various locations around coal fly ash disposal area has been shown in Table 2, all elements which are included in table are present in all samples. Elements are present in all 08 samples collected from different locations around coal fly ash disposal area has been shown in Figure 2, highest concentrations of mercury and titanium are present in No. 4 sample (Raja Paraswar Hand Pump 120 feet deep) which is approximate 500m distance from fly ash coal disposal area. Mercury concentration is much more than the permissible limit of drinking water, many studies has been shown mercury is very toxic in nature and hazardous for human. In sample No. 6 (NTPC Vidyut Vihar SBI) barium concentration is highest. Antimony concentration is highest in sample No. 8 (Rihand Dam Upstream). Many studies have shown that, near coal fly ash disposal area, leaching of heavy toxic metals is main reason for water contamination. The TCLP leaching experiments has been done from coal fly ash and coal fly ash bricks which is found leaching trend of heavy metals as Al > Fe > Mg > Zn > Cr > Cu > B > Pb > Ni > Co > As > Cd. The SPLP test also shows the trends of heavy metal leaching similar to that of TCLP.[31]

Elements in samples below permissible limit as per IS 10500:2012

Water sample analysis has been shown, calcium, magnesium and zinc concentrations are under permissible limits in all water samples collected from different locations, concentration of barium is under permissible limit except Rihand Dam water and NTPC Vidyut Vihar SBI hand pump, chromium is under permissible limit in Rihand upstream, AWRS and Raja Paraswar 50 feet deep

Table 2. Concentrations of elements in fly ash and water samples.

S.No.	Elements	(1), %	(2), ppm	(3) ppm	(4) ppm	(5) ppm	(6) ppm	(7) ppm	(8) ppm	(9) ppm	(10) ppm
1	Silicon	28.69	75.5	60.2	47.8	56.3	75.6	52.3	68.2	41.2	-
2	Aluminium	14.84	7.5	2.5	1.9	6.3	4.6	1.3	6.9	1.5	0.03/ 0.2
3	Magnesium	0.391	91	42	32	81.2	49.6	23	54	31	30/100
4	Phosphorus	0.028	0.4	0.1	0.1	0.2	0.1	0.1	0.3	0.2	-
5	Sulphur	0.053	0.6	0.1	0.1	0.7	0.4	0.2	0.4	0.1	-
6	Calcium	0.634	120.6	47.5	64	112	93	96	88.5	34.6	75/200
7	Titanium	0.802	0.8	0.1	0.06	0.9	0.2	0.2	0.2	0.1	-
8	Manganese	0.052	0.7	0.5	0.4	0.7	0.5	0.4	0.8	0.4	0.1/0.3
9	lron	4.48	16.2	17.2	11.5	16.5	18.3	13.6	17	14.1	0.3/0.3
10	Nickel	0.007	0.3	0.3	0.2	0.4	0.1	0.2	0.1	0.1	0.02/0.02
11	Copper	0.005	3.1	0.8	0.6	2.5	1.8	1.1	0.8	0.5	0.05/1.5
12	Chromium	0.007	0.2	0.07	0.05	0.2	0.09	0.08	0.04	0.03	0.05/0.05
13	Vanadium	0.032	0.6	0.1	0.1	0.6	0.3	0.1	0.1	0.1	-
14	Zinc	0.021	7.1	1.8	0.4	5.6	7.2	1.2	0.12	0.09	5/15
15	Lead	0.007	0.04	0.03	0.04	0.06	0.03	0.05	0.06	0.01	0.01/0.01
16	Mercury	0.002	0.006	0.04	0.06	1.7	0.3	0.1	0.05	0.01	0.001/0.001
17	Barium	0.001	0.2	0.8	0.4	0.1	0.1	5	0.4	0.5	0.7/0.7
18	Arsenic	0 002	1.3	0.9	1.5	1.5	0.9	1.4	1.4	1	0.01/0.05
19	Selenium	0.002	0.02	0.01	0.01	0.03	0.02	0.01	0.01	0.01	0.01/0.01
20	Cadmium	0.004	0.01	0.01	0.02	0.01	0.02	0.01	0.03	0.01	0.003/0.003
21	Antimony	0.002	0.004	0.001	0.001	0.002	0.002	0.003	0.01	0.001	-

(1) Fly Ash (2) Ash Dyke Khadia Hand Pump (within 10-meter range) (3) Rihand dam (Near Ash Pond) (4) Raja Paraswar Hand Pump 50 feet deep (within 500-meter range) (5) Raja Paraswar Hand Pump 120 feet deep (within 500-meter range) (6) Tarapur Hand Pump 120 feet deep (within 1000-meter range) (7) NTPC Vidyut Vihar SBI (within 5000-meter range) (8) AWRS Water (9) Rihand Dam Upstream (10) Acceptable/ permissible limit by IS 10500

hand pump samples, copper is under permissible limit except in Ash dyke, Raja paraswar and Tarapur hand pump sample, lead is under permissible limit only in Rihand upstream sample.

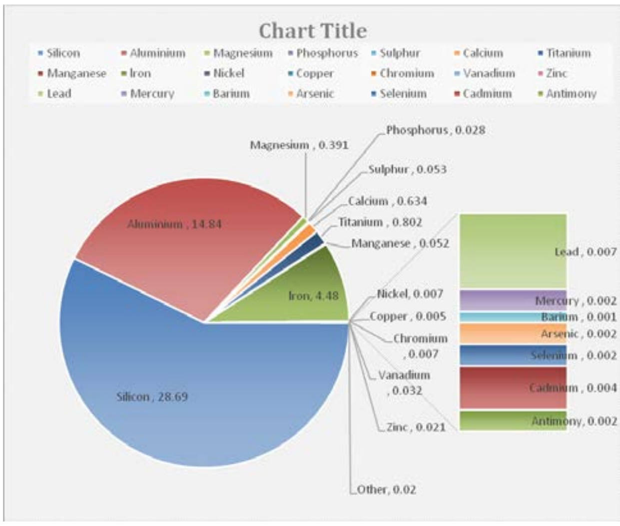

Figure 1. Concentration of elements present in coal fly ash.

Elements above permissible limit as per IS 10500:2012

The concentrations of Fe, Hg, As and Cd are above the permissible limit in all water sample. These all are toxic trace metals in nature, only Pb is under limit in upstream sample of Rihand Dam.

Physico-chemical characteristics

Indian coal has high sulphur contents, which causes acid rain after it enters in environment. Sulphur content is also high in coal fly ash, which brings the pH down as when coal fly ash is mixed with water, oxides of sulphur adsorbed on highly reactive and extremely fine ash particles produce H_2SO_3 or H_2SO_4 in solution. Acid formation results in low pH of fly ash slurry. Several factors may influence pH behaviour of leachate, as quantity of CaO and MgO increases, pH of leachate may increase with time.

CONCLUSION

The present study is an assessment of leaching from coal fly ash disposal area and its effect on nearby water sources. Leaching is likely to deteriorate soil and ground water quality in the vicinity of the thermal power plant.

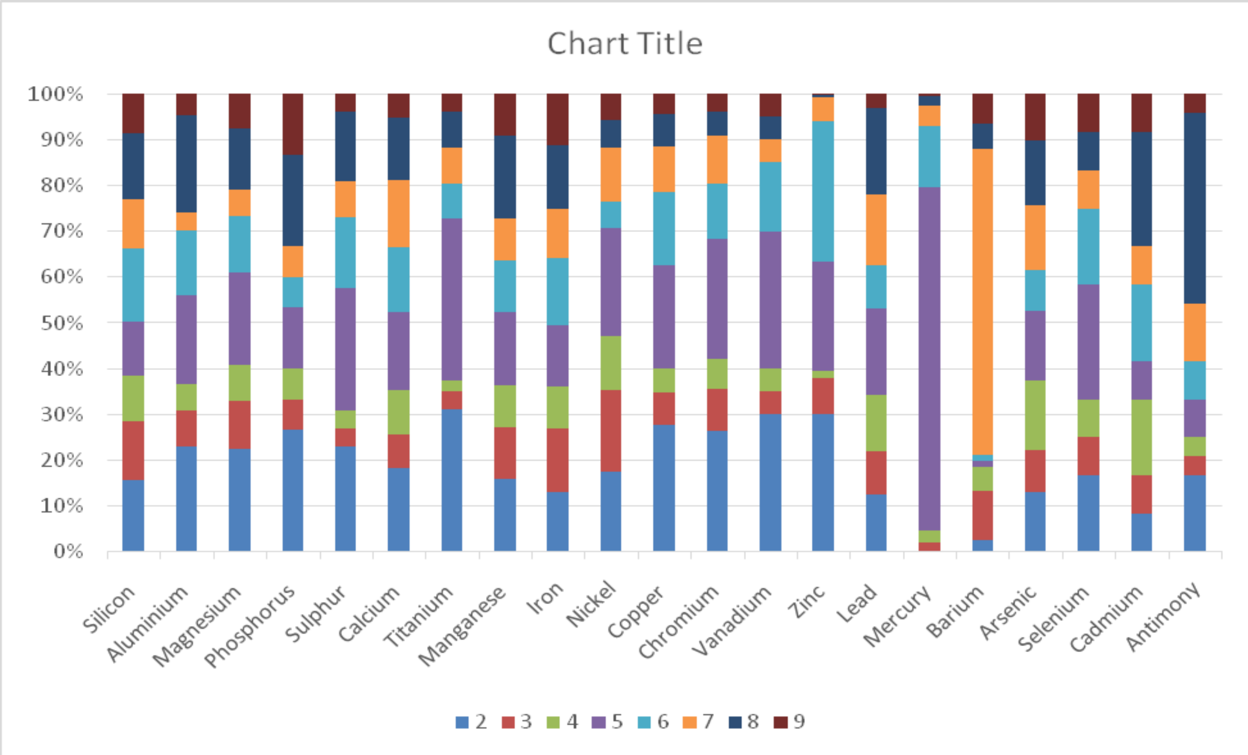

Figure 2. Concentration of toxic elements in water samples.

This research work provides significant evidence to the scientific community for taking adaptive measures to dispose of coal fly ash in better manner and application of appropriate processes for its uses or disposal to stop contamination of ground water and soil. The major conclusions are as follows:

Coal fly ash contain various heavy metal which are toxic in nature as As, Cr, Hg, Pb, Cd and Ni, etc. The most of the samples contains concentration higher than the permissible of drinking water, hazardous trace elements Ni, Pb, Hg, Cd, Mn and As are detected in samples above the prescribed limit. Heavy toxic metals leach into water from coal fly ash disposal area and contaminate ground water and soil also which is dangerous to human health. A better and full proof plan required to use and disposal of such huge amount of fly ash to stop contamination of environment. It is a matter of great concern to human health.

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