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Radioelemental characterization of fly ash from Chandrapur Super Thermal Power Station, Maharashtra, India

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Natural radioactivity due to the presence of ^{40}K, ^{238}U and ^{232}Th was measured in fly ash samples collected from economizer, aerator and electrostatic precipitator (EP) of the Chandrapur Super Thermal Power Station (CSTPS) using a NaI (Tl)-based gamma ray spectrometer. The study indicates an elevated concentration of these radionuclides, especially in the finer ash samples from EP, which may provide an exposure pathway through inhalation of airborne ashes and could probably cause severe environmental and human health problems. The present study gains significance as it provides the requisite basic data on the radionuclides concentration in fly ash from CSTPS for a detailed follow up of environmental monitoring and to formulate effective management strategies.

Keywords: Environmental monitoring, fly ash, radionuclides, thermal power plants.

COAL plays an increasingly important role to meet the ever-growing demand for energy, and coal-fired thermal power plants account for 54.3% (92,378.38 MW as on 31 December 2010) of the total electricity generation in India[1]. This shall remain the mainstay to meet the additional capacity requirements in the future[2]. But in this process of power generation, about 170 million tonnes (mt) of fly ash is produced annually in India alone, thus causing serious environmental problems[3]. Coal contains traces of naturally occurring radioactive elements like uranium, thorium and potassium, but their concentration depends on the composition and geological history of the coal[4]. Of the total 278,180 mt of coal deposits in India, approximately 276,810 mt comes from the Gondwana stratigraphic horizon and the rest from the Tertiary formations[2].

The Indian coal fields hold 77% of the sub-bituminous and bituminous coal reserves[5], with high ash content and low calorific value[6]. Therefore, thermal power plants in India are designed to use these huge volumes of bituminous or sub-bituminous varieties having 30–40% ash content, which are otherwise unsuitable for metallurgical industries[3]. Upon combustion of coal, the nonvolatile products form ash particles that follow the air stream and eventually condense on the fly ash particles. It creates disequilibrium in the decay chain of parent radionuclides having different physical and chemical characteristics[7]. During the combustion process, the volume of coal is reduced by over 85%, which increases the concentration of ^{40}K, ^{232}Th, ^{235}U and ^{238}U originally present in the coal[8]. Although significant quantities of ash are retained by precipitators, radionuclides such as uranium tend to concentrate on the tiny glass spheres that make up the bulk of fly ash. This uranium is released to the atmosphere with the escaping fly ash, at about 1.0% of the original amount[9]. The retained ash is enriched in uranium several times over the original uranium concentration in the coal[10,11].

Even though coal-fired power plants throughout the world are the major contributors of radioactive materials released to the environment[12], they have been neglected as a radiation source for a long time. Of late, due to advancement in the scientific knowledge of the biological effects of radiation on human beings[13,14], research activities are focused on increased concentration of naturally

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occurring radioactive materials due to anthropogenic activity known as 'TENORM' (Technologically Enhanced Naturally Occurring Radioactive Material) that has resulted in the increase of background gamma radiation level[15,16]. The mean natural radioactivity/specific activity reported in coal is 35 Bq kg^{-1} (range 17–60 Bq kg^{-1}) for ^{238}U, 30 Bq kg^{-1} (range 1–64 Bq kg^{-1}) for ^{232}Th and 400 Bq kg^{-1} (range 140–850 Bq kg^{-1}) for ^{40}K (refs 10 and 11). The radioisotopes in coal constitute a continuing source of radioactive nuclides released into the biosphere and per tonne coal combustion is estimated to contribute about 4.3 microcurie of radioactivity to the biosphere[17]. Although small in concentrations, these components are significant when the vast quantity of coal combustion is considered, and more so when collected over a long period of time[8]. Global accumulation of these long-lived radioactive species is projected to increase up to 828,632 tonnes of uranium, 2,039,709 tonnes of thorium and more than 30,000 tonnes of ^{40}K by 2040 (ref. 8).

Fly ash is the fine powdery residue of the coal combustion process, and by virtue of its lightness it has the tendency to fly for longer distances along with the gases through the stacks to the atmosphere. The elevated concentration of ^{238}U, ^{232}Th and ^{40}K in these fly ash particles is likely to pose serious ecological problems if locally accumulated for a considerable period of time[8,12]. This would also affect human health, as it provides an exposure pathway through inhalation of airborne ashes containing such long-living radionuclides[13]. Therefore, the present study endeavours to characterize the radioelemental concentration of fly ash generated from the Chandrapur Super Thermal Power Station (CSTPS) and to provide the requisite basic data for a detailed follow up of environmental monitoring[16].

The CSTPS is located near Chandrapur (Figure 1) and functions with four units of 210 MW and three units of 500 MW each. It is the biggest pit head thermal power station of the Maharashtra State Power Generation Company Limited.

Physical and chemical variations in the composition of the fly ash samples are mainly dependent on the thermal gradient along the stack height at different points[18]. Hence, fly ash samples of about 10 kg each were collected from the economizer (EC), aerator (AE) and stack vent electrostatic hopper of the power plant. They were initially dried at a temperature of 105°C. About 100 g ash samples of EC, AE and the electrostatic precipitator (EP) was taken and the magnetic materials were separated from the non-magnetic fractions using wet magnetic separation methods. The quantitative fractions of magnetic and non magnetic portions of EC, AE and EP fly ash samples were 7.96 and 92.04 g, 9.82 and 90.18 g, and 5.77 and 94.23 g respectively. These ash samples were further separated into 300, 150, 106 and 45 μm size using a standard set of sieves and expressed in weight percentages (Figure 2).

Laboratory measurements of ^{238}U, ^{232}Th and ^{40}K in fly ash samples were undertaken at the National Geophysical Research Institute, Hyderabad using a low-level gamma-ray spectrometer consisting of a 5″ diameter and a 6″ high BICRON NaI (Tl) crystal. It is hermetically sealed and coupled to a photo-multiplier tube, dynode chain and preamplifier which together constitute the detector assembly.

The concentration of ^{40}K was measured directly by its own gamma-ray peak at 1.46 MeV, whereas ^{238}U and ^{232}Th were estimated with the help of their gamma-ray emitting daughter products, i.e. ^{214}Bi (1.76 MeV) and ^{208}Tl (2.62 MeV). Fly ash samples were placed in plastic containers (1 mm in wall thickness) with a diameter of about 45 mm, and a height of 45 mm, as close as possible to the detector window. The background count rates counts/ks were obtained by counting in the above-mentioned windows by keeping a dummy sample[19,20].

Coal contains primordial radionuclides ^{40}K, ^{232}Th, ^{235}U and ^{238}U, and the members of the decay series of the last

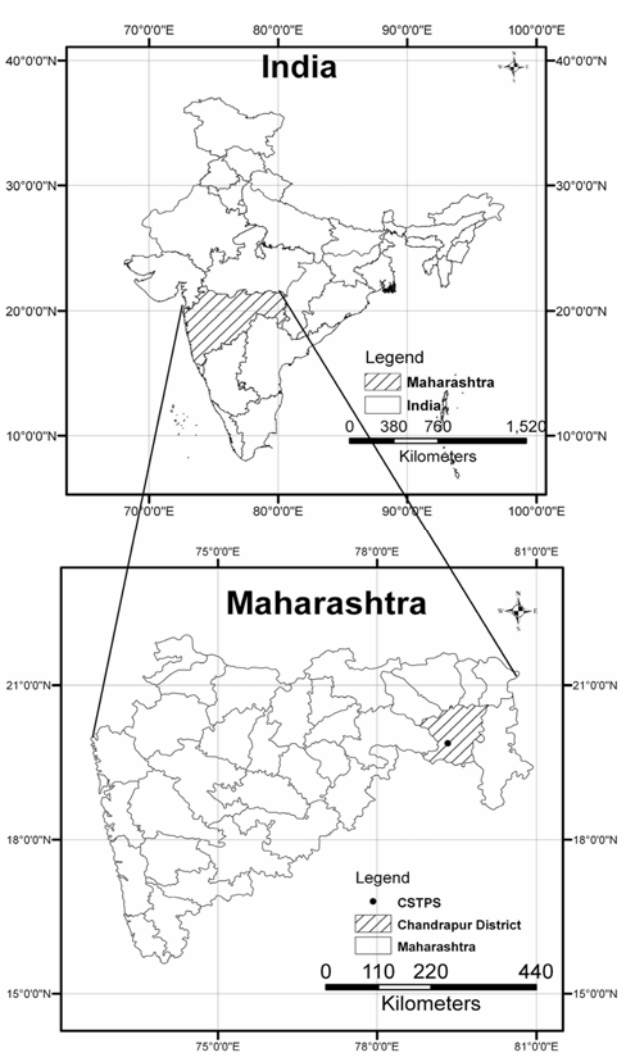

Figure 1. Location map of Chandrapur Super Thermal Power Station.

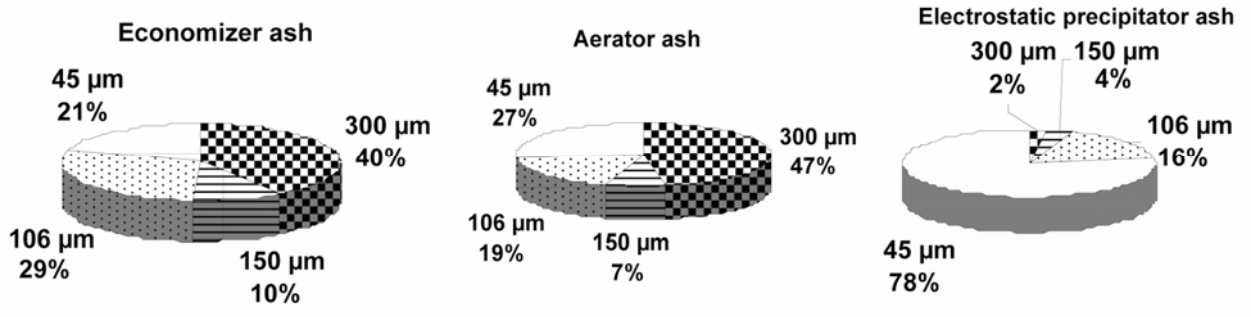

Figure 2. Weight percentage of different size fractions in fly ash samples.

Table 1. Radioactivity concentration in fly ash samples of Chandrapur Super Thermal Power Station

	^{40}K (Bq kg^{-1})			^{238}U (Bq kg^{-1})			^{232}Th (Bq kg^{-1})		
Size (µm)	EC	AE	EP	EC	AE	EP	EC	AE	EP
Fly ash (magnetic)									
300.00	52.92	131.04	320.04	97.40	24.35	84.54	38.42	134.73	544.33
150.00	52.92	65.52	282.24	17.67	27.32	83.92	33.73	50.94	205.82
106.00	63.00	60.48	315.00	43.01	24.35	120.39	52.84	56.36	227.69
45.00	68.04	42.84	640.08	28.06	27.32	207.03	44.04	43.67	176.44
Fly ash (non-magnetic)									
300.00	153.72	95.76	163.80	12.85	34.24	95.05	24.32	56.44	228.01
150.00	201.60	209.16	161.28	24.47	33.00	96.04	45.05	67.95	274.53
106.00	186.48	221.76	173.88	85.04	55.25	89.73	146.65	140.43	567.34
45.00	171.36	312.48	171.36	79.97	88.00	97.64	132.11	171.78	693.99

EC, Economizer; AE, Aerator; EP, Electrostatic precipitator.

three nuclides[21]. The main sources of radiation released from coal combustion include not only uranium and thorium, but also their daughter products, such as radium, radon, polonium, bismuth and lead, as well as ^{40}K (refs 19, 20). Most of the radionuclides get concentrated into ash as solid and gaseous combustion products and are finally discharged into the environment.

Radioelemental concentrations of ^{40}K, ^{238}U and ^{232}Th (Table 1) indicate that there is a significant increase in the radioactivity of ^{40}K from EC to EP in the different size fractions of the magnetic portion. In the magnetic fraction of the economizer (EC_m) and aerator (AE_m) ash, ^{40}K increased with decreasing particle size. The non-magnetic fraction (EC_{nm}) did not follow any specific trend in economizer, whereas it showed increasing trend with decreasing ash size fraction in the aerator (AE_{nm}). Higher activity concentrations were recorded in the EP ash, i.e. 282.24–640.08 Bq kg^{-1} and 161.28–173.88 Bq kg^{-1} in the magnetic and non-magnetic fractions respectively. Antipathetic relationship was observed in specific activity and particle size in both EP_m and EP_{nm} ash samples. Elevated specific activity concentration of ^{238}U (97.4 Bq kg^{-1}) was found in 300 µm fraction of EC_m, whereas AE_m samples showed near-uniform specific activity in all size fractions. Radioactivity due to ^{238}U concentration in AE_{nm}, EP_m and EP_{nm} showed increasing trend with decreasing particle size. The EP_m and EP_{nm} fractions also contained elevated concentration of ^{232}Th.

In general, increasing specific activity of ^{40}K, ^{238}U and ^{232}Th was observed with decreasing particle size classes, except for a few anomalous values in aerator ash samples. Higher radioactivity concentration of ^{40}K, ^{238}U and ^{232}Th, especially in the 45 µm EP ash, attributed to its volatility and higher surface to mass ratio[22]. The radionuclides that accumulate on the smaller fly ash particles are assumed to be volatile at the temperature of combustion (1300°–1600°C). As the fuel gas cools, the volatiles are adsorbed, particularly on the surface of smaller fly ash particles. Therefore, these fly ash particles are mostly enriched in many such elements in finer $EP_{m–nm}$ ash than in $EC_{m–nm}$ ash samples[23].

Abundance of ^{232}Th in the fly ash is approximately 2.5 times that of ^{238}U, and this could be due to its occurrence in the inorganic phase of coal and due to its low solubility[24]. Uranium is comparatively more soluble than thorium because of its oxidation states, i.e. +4 and +6. The ratio of U and Th in ash is more or less in accordance with the general abundance of these radioelements in coal (U = 1.7 ppm and Th = 3.2 ppm; Th/U ratio = 2). However, actual U and Th contents may vary in areas specific to

coal. There is a three to four-fold increase in the radioactivity concentration of ^{232}Th in the EP_{m-nm} ash fractions compared to ^{238}U in fly ash.

The enrichment of radionuclides in fly ash is due to the volume reduction in coal during combustion. Poor fly ash control devices and the age of the thermal power plants are the main causes for the higher concentration of radionuclides emission[9]. Hence, effective advanced filters should be used in EP to reduce the fly ash emission[9]. The elevated concentrations of ^{238}U and ^{232}Th in the finer ash fraction may well be the exposure pathway in the form of inhalation that will go into the respiratory system of human beings and will be hazardous to health[12,13]. This may also be deposited onto the soil as well as settle over the water bodies, which in turn infiltrate into the underlying soil[25–27]. Accumulation of ^{238}U, ^{232}Th and ^{40}K in the fly ash deposits in the vicinity of a thermal power plant over a considerable period of time may pose significant ecological burden through atmospheric pollution. Direct addition of radionuclides to surface waters and their subsequent leaching into the soil system will ultimately reach humans through the drinking water–crop pathways. Therefore, caution should be exercised in dumping the fly ash and essential preemptive steps should be taken prior to the alarming increase in concentration of radionuclides in fly ash, which will have irrevocable and disastrous effects on the fragile ecology and environment. In addition, the potential threat of these radionuclides in the long run should not be ignored and regular environmental monitoring is required to put a check on the increasing level of pollution in and around CSTPS.

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