Radioactivity aspects of Indian coals

M. P. Rajan, M. A. R. Iyengar and T. V. Ramachandran*

Environmental Survey Laboratory, Health Physics Division, Bhabha Atomic Research Centre, Kalpakkam 603 102, India
*Environmental Assessment Division, Bhabha Atomic Research Centre, Bombay 400 085, India

This paper presents the results of a study of the radioactivity aspects of Indian coals. The measurements were carried out by gamma spectrometry and chemical methods for estimation of U-nat, Ra-226, Ra-228, K-40, Po-210 and Pb-210 in the samples. A brief description of sampling and measurement procedures are also given. Radiation dose estimates of the population from two typical thermal power stations at Neyveli and Ennore in Tamil Nadu have been attempted using the model developed by UNSCEAR and ORNL reports as guidelines, with suitable correction for local population density, plant load factor, fly-ash release, coal consumption per day, etc.

THE global energy needs have enormously increased with rapid strides in technology and to a large extent these have been met from fossil fuels. India has to depend largely on coal reserves for her energy needs to meet the present demand and possibly in future also. The annual requirement of coal is expected to go beyond 425 million tonnes by 2000 AD compared to current annual production of about 250 million tonnes. The total energy-generating capacity in the country is expected to be about 106 GW(e) by 2000 AD. Thermal power generation, as on 1994, is about 66% of the total generating capacity, i.e. about 65000 MW(e).

Combustion of coal in thermal power stations (TPSs) gives rise to gaseous and particulate pollution in the environment, such as SO_2, NO_x, particulate matter, etc., which have long-term effects on vegetation, fish, animal life, human beings and even on buildings, structures, etc. This has also resulted in the release of large quantities of CO_2 adding to its continuous build-up of its level in the environment. It has been estimated that a 1000 MW(e) coal-fired power station consumes about 3 million tonnes of coal per year, producing 7 million tonnes of CO_2, 0.12 million tonnes of SO_2, 0.02 million tonnes of NO_x and 0.75 million tonnes of ash[1]. The present coal consumption in TPS in the country results in adding an estimated 12.2 million tonnes of fly-ash into the environment every year, of which nearly a third goes into the air and the rest is dumped on land or discharged in waterbodies.

In India, most of the coal available is of bituminous type. The quality of Indian coal is poor with 5–50% ash, 1–30% moisture, 79–82% carbon, 3.5–6.0% hydrogen, 0.8–2.4% nitrogen, 1.3–10% oxygen, 13–50% volatile matter, 0.2–2.0% sulphur and 0.01–0.4% phosphorus. Besides, coal also contains radioactivity due to primordial uranium and thorium series nuclides and K-40; during its combustion these natural radionuclides are readily released which disperse over a large area in the vicinity of TPSs[2,3]. Unlike most of the nuclear and hydroelectric power stations, coal-fired power stations in India are generally located in areas which are thickly populated and hence the environmental impact experienced by the neighbourhood population may be of significance. Due to the high ash content of coal and relatively poor fly-ash control measures, particulate pollution as well as natural radioactivity deposition due to TPSs in India are higher than in some of the western countries. However, recently built TPSs in India have installed modern pollution control technologies which mitigate the pollution levels in the environment to a considerable degree.

In the light of the possible radiological significance in the environment, studies have been carried out by several investigators in different parts of the world to assess the nature and quantity of radioactivity releases from thermal power stations using coal as fuel. Some of these studies have shown that the radiation hazard in the environment of the coal-fired power stations is comparable and sometimes even higher than that of nuclear power stations[4–6]. There is thus considerable interest in investigating the radioactivity aspects of coal and fly-ash in Indian TPSs and their significance in terms of environmental impact and public radiation exposures. This paper presents the results of a study carried out to assess the radiological aspects of certain selected TPS units in the country using both coal and lignite.

Materials and methods

Coal samples for the study were obtained from West Bengal, Chanda (Maharashtra), Singareni (Andhra Pradesh) and Talcher (Orissa) coal fields. Lignite samples were collected from various locations of the Neyveli Lignite Corporation in South Arcot District of Tamil Nadu. The fly-ash and slag (bottom ash) samples were obtained from Singareni, Ennore and Neyveli power stations. Coal, fly-ash and slag samples thus obtained were thoroughly mixed, dried and ground to a fine powder and nearly 200 g each of the sample was filled and topped up in an air-tight plastic container of 6.5 × 7.5 cm size and stored for nearly a month before

counting to ensure that nat. U and Th attain equilibrium with their respective daughters[2].

Standard sources of uranium and thorium have also been prepared in the same geometry by mixing known quantities of uranium ore and thorium nitrate salt with an inactive matrix having density close to those of the samples. For preparing K-40 standard, potassium chloride of Analar grade was used directly. Source strengths were selected in a way that would yield a low statistical error in channel counts for nearly one-hour counting. Sodium oxalate was used as a dummy matrix because it has low radioactivity content, is nonhygroscopic and has density close to the sample matrix. The overall errors due to sampling and source preparation were estimated to be ± 15% (ref. 2). The thorium nitrate and the uranium ore used for the source preparation were more than 15 years old and hence it was ensured that radium and thorium-228 had attained radioactive equilibrium with their daughters.

The selection of the 12.5 × 10.0 cm NaI (Tl) detector of Harshaw make for the present work was made keeping in view the low specific activity of the samples to be counted and the maximum energy of interest[7]. The detector offers the best compromise if the energy of interest is up to 3 MeV. Similarly, the selection of the multichannel analyser for the analysis is also based on the criteria outlined by Schonfeld[8]. The choice of the internal dimensions of the lead shield and of lining it with cadmium and copper were also made to reduce the low-energy scattered background.

Some of these samples were also analysed by radiochemical methods to determine concentrations of radionuclides like U-nat, Ra-226, Ra-228, Po-210 and Pb-210. Details regarding the radioanalytical procedures used in the study are described elsewhere[9,10].

Results

From the gamma spectrometric analysis of the samples, it was observed that the coal and its fly-ash samples contained radionuclides from both uranium and thorium series, while it was interesting to note that the lignite and fly-ash samples from Neyveli showed predominantly radionuclides from uranium series.

Tables 1 and 2 give the results of the radiochemical analysis of the samples investigated. From Table 1 it can be seen that the coal samples from different regions showed more or less similar activity content, whereas the lignite samples showed a considerable variation in the activity levels. From Table 2, it can be seen that the lignite fly-ash contains higher amount of U-nat, Ra-226, Po-210 and Pb-210 compared to the respective concentrations of these nuclides in coal fly-ash.

Among the samples investigated, the samples from Neyveli region showed evidence of higher concentration of uranium. The carbonaceous clay, which is present as a layer above the lignite seam, contains small amounts of U_3O_8 and is the main constituent which may be responsible for the observed higher concentration of uranium in the lignite samples[2,3]. Table 3 gives the

Table 1. Radioactivity content in coal and lignite samples

Source	U-nat	Ra-226	Ra-228	Po-210	Pb-210
	(Bq/kg)				
Coal					
Singareni	5.7–21.5	6.7–15.5	8.9–17.0	2 6–21 8	4 9–24.5
Bengal	22.3	14.2	17.4	17.5	16.3
Chanda	20 6	15.7	18 5	18 1	14 4
Talcher	40 3	38 9	40.2	35.8	36.1
Singrauli	53 8	54 4	45 5	50.1	48 6
Ennore	40.4–56 8	8 1–31.8	6 6–28.9	18.9–43.7	17.4–45.0
Lignite					
Neyveli	0.7-3.2	BDL-5.4	BDL-1 3	BDL-9 8	1.1–7 6
Neyveli (power station)	74.5–99.5	37.3–109.9	BDL-7 5	67.1–87.7	30.7–85.8

Table 2. Radioactivity content in fly-ash samples

Source	U-nat	Ra-226	Ra-228	Po-210	Pb-210
	(Bq/kg)				
Singareni	18 8–27 9	15 5–66 6	13.7–44 4	24 1–27 4	25 7–30 2
Ennore	73.1–121 4	16 0–94 1	15 8–79 9	5 9–55 9	7 0–69 6
Neyveli	207.5–635.3	177.5–596 4	24 4–59 4	98 9 -276.4	155 8–271 4

observed levels of radionuclides in lignite from different layers as seen at the mine. The increase in the activity content of lignite fly-ash can be clearly traced to the mixing of the relatively higher activity of carbonaceous clay with lignite during the mining process and pulverization, prior to combustion in the power station.

Dose assessment

In view of the range of activities and respective levels and the wide dispersion potential of radioactivity from coal-fired TPSs, it is of interest to calculate the doses received, if any, by the public inhabiting the neighbouring environment. In our earlier publications[11,12], the radiation dose commitments to the population living within 80 km radius of thermal power stations were calculated following the 1980 UNSCEAR report. This was calculated on the basis of an assumed plant load factor of 50%, average coal consumption of 15 tons per day per MW(e) and 30% ash content of coal. A good deal of work has been done in the past few years by several investigators on assessing the doses and dose commitments from energy production in coal-fired and nuclear power plants. A review of the methods used for such calculations by De Santis[5] has been taken into account and the method described by McBride *et al.*[4] has been adopted. The method described in this report gives a clear procedure for dose assessment from a model 1000 MW(e) coal-fired and nuclear power plants.

In the present calculations the actual average plant load factor during the period of sampling, the coal consumption, and the ash content of the coal used in a particular power plant, have been used[13,14] (Table 4). These calculations were also modified in the light of ICRP recommendations of 1977, i.e. change of quality factor for alpha particles from 10 to 20 and the concept of collective effective dose equivalent commitments. The Ennore TPS near Madras is equipped with electrostatic precipitators, for which an average fly-ash control of 97% was assumed. The Neyveli TPS was equipped with mechanical or cyclone collector at the time of sample collection and a fly-ash control of 82% was assumed for this plant. These factors have been applied in deriving the dose estimates.

McBride *et al.*[4] assumed that all the daughters of the uranium and thorium series were in equilibrium and had calculated the radiation doses in the population living within 88.5 km of the plant under normal climatic conditions using the AIRDOSE computer code for the atmospheric dispersion calculations. The annual precipitation assumed in these calculations is about 900 mm. The TPSs under the present study fall in the range of 600–1000 mm annual rainfall. Hence, the data of McBride *et al.*[4] were used for calculation of dose with appropriate corrections for population density[15,16], installed capacity, coal consumption and ash content, fly-ash control factor, average load factor for the sampling period and lastly the activity release ratio of the actual activity released in Indian TPSs to that of the model TPS.

Using the above correction factors and assumption

Table 3. Radioactivity content in lignite samples of Neyveli

Sample no.	Type of sample	Bq/gm Ra-226	Bq/gm Ra-228	Bq/gm K-40	Sample no	Type of sample	Bq/gm Ra-226	Bq/gm Ra-228	Bq/gm K-40
ASC1	Carbonaceous clay	1 20	0 07	0 37	AML1	Lignite	0.06	0 01	0.06
ASC2	Ball clay	0.02	0 02	0.02	AML2	Lignite	0.05	0.02	0.07
ASL1	Lignite	0 05	0.07	0 17	AML3	Lignite	0.05	0.03	0 08
ASL2	Lignite	0 03	0.03	0.12	ANC1	Carbonaceous clay	0.33	0.08	0.09
ASL3	Lignite	0.03	0.03	0.21	ANC2	Ball clay	0 02	0 04	0.08
AMS1	Sandstone	0 05	0.09	0.16	ANL1	Lignite	0.05	0.07	0.17
AMC1	Carbonaceous clay	0.26	0.09	0.08	ANL2	Lignite	0 04	0.03	0 12
AMC2	Ball clay	0.02	0.04	0.08	ANL3	Lignite	0.03	0.03	0 21
						Fly-ash	0.26	0.07	0.14

Table 4. Estimated dose commitments from Neyveli and Ennore thermal power stations

Name of power plant	Population density (km^{-2})	Inst. capacity (MW(e))	Ash content (%)	Avg. PLF*	Activity released (MBq/yr) Ra-226	Th-228	K-40	Collective effective dose equivalent commitment (man-Sv. yr^{-1}) W.B.	Bone (1)	Lung (2)	Thyroid (3)	Total (1+2+3)
Ennore	330	330	35	28 0	518	637	1409	0.07	0.44	0 14	0.004	0 584
Neyveli	370	600	4	59.3	3684	524	262	0.36	2.49	0.60	0.022	3.112

*PLF – plant load factor.

for release considered by McBride *et al.*[4], it was found that, of the total dose received, uranium daughters contribute 75, 77, 61 and 76% of the total dose to the whole body, bone, lungs and thyroid, respectively. The corresponding figures for Ra-228, Th-228 of thorium series were 22, 18, 33 and 22% of the total dose. The contribution to the doses and dose commitments from the airborne releases of U and Th daughter products due to inhalation and direct gamma irradiation from surface deposition are 6.5, 17, 38 and 6.4% to the whole body, bone, lungs and the thyroid, respectively, the remainder arising through ingestion pathways. The doses and dose commitments from the airborne releases from thermal power stations were calculated only for the whole body, bone, lungs and thyroid as these are the doses usually considered in the context of nuclear power plants. They are also used elsewhere[12] for comparison between these two energy production technologies.

Dose estimates from K-40 have not been included because the levels of K-40 in man are homeostatically controlled. The lung is the critical organ for Rn-222 but the Rn-222 contribution to the total lung dose is only about 1 part in a million[4]. Hence, doses due to radon and its daughters are neglected in the present dose assessment and the dose assessment is done only for Th-230, Ra-226, Po-210 and Pb-210 from the U series and Th-232, Ra-228, Th-228 from the Th series.

The resulting equations based on the above considerations and assumptions for assessment of doses and dose commitments (in man-Sv. Y^{-1}) are as follows:

$$\text{Wholebody and thyroid} = 9.5 \times 10^{-8} P\ (mx + ny) \cdot d,$$
$$\text{Bone} = 2.7 \times 10^{-6} P\ (mx + ny) \cdot d,$$
$$\text{Lungs} = 7.7 \times 10^{-7} P\ (mx + ny) \cdot d,$$

where x and y refer to the radionuclides concentrations of Ra-226 and Th-228 in Bq/kg of fly-ash, d is the population density per km^2 in an 88.5 km radius environment and P is the total installed capacity in MW(e). The constants m and n are defined as follows:

m = (annual ash released from the stack in kg for 1000 MW(e)) × (dose conversion factor from U series) × (296 MBq of Ra-226)$^{-1}$,

n = (annual ash released from the stack in kg for 1000 MW(e)) × (dose conversion factor from Th series) × (185 MBq of Th-228)$^{-1}$

The values of dose conversion factors for the uranium and thorium series for different body organs have been reported earlier[17]. Plant load factors are incorporated in the computation of m and n.

The conversion of the collective dose equivalent commitments into collective effective dose equivalent commitments allows a better evaluation of the hazard of radionuclides exposures for risk estimates. Hence, all the doses estimated in the foregoing to different body organs have been converted to collective effective dose equivalent commitments and the calculated results of the dose assessments are given in detail in Table 4 for the TPSs studied.

Discussion

Among the samples analysed, the lignite and its fly-ash samples from Neyveli were found to contain more radioactivity due to uranium series radionuclides, compared to coal and its fly-ash samples from other parts of India. This gives rise to higher collective doses at Neyveli as is clear from Table 4.

The doses reported in the paper are due to the inhalation of radionuclides contained in the fly-ash released through the stack. However, for ingestion route, an investigation of environmental samples such as water, air, vegetation, fish, etc., from Ennore and Neyveli TPSs, revealed that the activities due to Ra-226, Ra-228, U-nat, etc., in these samples were in the same range as that of the samples collected from a control area free from any type of industrial pollution. Therefore, it can be concluded that the collective dose is coming essentially from the inhalation route and the dose from the ingestion route is negligible.

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Composition and enzyme activities in *Aspergillus flavipes* grown on crude petroleum oil and glucose

J. Savitha* and C. V. Subramanian**

Centre for Advanced Study in Botany, University of Madras, Guindy Campus, Madras 600 025, India
*Present address: Department of Microbiology, Bangalore University, Bangalore 560 056, India
**'Anjaneya', 62, Dr Ramaswamy Salai, West K. K. Nagar, Madras 600 078, India

Aspects of the growth, cellular composition and enzymology of *Aspergillus flavipes* grown on a medium with crude oil or glucose as the sole source of carbon are reported. When grown on crude oil, the total lipid content of *A. flavipes* was considerably greater than on glucose medium. A large number of fatty acids, particularly saturated fatty acids, were observed when grown on crude oil and more lipid was excreted into the growth medium. The total mycelial carbohydrate and glycogen contents were considerably less in crude oil medium, whereas the mycelial protein content was similar in both media. The activity of some enzymes like NAD^+-alcohol dehydrogenase, $NADP^+$-aldehyde dehydrogenase, iso-citrate lyase and catalase was greater when crude oil was the carbon source. Conversely, the activity of succinate dehydrogenase was greater after growth on glucose medium.

THE subject of degradation and utilization of hydrocarbons by fungi has received considerable attention[1–3] but much more remains to be done[4]. A strain of *Aspergillus flavipes* isolated from the effluents of the Madras Oil Refineries that can degrade hydrocarbons in crude petroleum oil was reported earlier[5]. Work on utilization of hydrocarbons has mainly been carried out on a few selected bacteria, and some fungi, notably species of *Candida* and the kerosene fungus *Cladosporium resinae*[6–8]. Much of this work has used specific hydrocarbons such as alkanes of known chain length. In the present study, however, we have examined the potential of *A. flavipes* isolated from the effluents of the Madras Oil Refineries to degrade crude petroleum oil, which often forms around 2% (v/v) or more of the effluent from the refinery. The emphasis in the study has been on the utilization of crude oil *per se* rather than particular components of the oil.

We have examined the amount of total protein, total carbohydrate, glycogen, total lipids, total sterol, fatty acids (saturated and unsaturated) in *A. flavipes* grown with either crude oil or glucose as the sole source of carbon. As a preliminary to understanding the physiology of utilization of crude oil, some data are given on the enzymology of crude oil and glucose utilization.

Materials and methods

Organism. A single spore isolate of *A. flavipes* isolated from the effluents of the Madras Oil Refineries Ltd., Manali, Madras, was used throughout the study. It was maintained on potato dextrose agar (PDA).

Culture techniques. Standard microbiological techniques were used in the preparation of media, sterilization etc.[9]. The fungus was grown in a mineral salts solution of Bushnell and Haas[10] supplemented with 0.1% (w/v) yeast extract, adjusted to pH 6.0. Glucose (10 g l^{-1}) was added to mineral salts solution before the autoclaving. Crude oil was filter-sterilized (pore size, 0.41 μm) and added to sterile mineral salts solution to give 10 g l^{-1}. Flasks (250 ml) containing 100 ml medium were inocu-