

Radioactivity From Coal-Fired Power Plants: A Review

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ABSTRACT

The radioactivity contents of coal and of the different types of ash formed during its combustion are reviewed. Also, the radiological impact of coal-fired plants and the plant features causing this impact are discussed. The depletion or enrichment of radionuclides in the different types of ash is interpreted in terms of the combustion temperature, the size of ash particles and the chemical forms of the radionuclides. The volatilization–condensation process as the postulated mechanism of radionuclide enrichment is also discussed.

Past studies of the release of radionuclides from coal-fired plants are compared and the environmental pathways of the radionuclides are highlighted. Individual and collective radiation doses calculated for various coal-fired plants are evaluated and the importance of the different pathways to man is outlined.

INTRODUCTION

Coal is one of the most important fuel sources for steam and energy production. Three facets of atmospheric pollution may be related to the combustion of coal in coal-fired plants (CFP): the release of relatively large amounts of gaseous and particulate pollutants, such as SO₂, NO_x, CO₂, hydrocarbons and flyash; the release of minor amounts of chemically toxic trace elements, such as arsenic, mercury and cadmium;

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and the release of minute amounts of radionuclides, such as uranium, thorium and their radioactive daughters, which may have a radiological impact on the environment.

This review deals with radioactivity from CFP and is limited to studies on flyash and flue-gases, to which some of the radionuclides present in coal are transferred following combustion. Radionuclides which are transferred to solid wastes are not considered.

Flyash, its chemical, physical, and toxic properties, and its emission, removal, and disposal, as well as its environmental effects, have been reviewed in several studies, e.g. Babcock & Wilcox (1975), Block & Danis (1976), Ray & Parker (1977), Fisher *et al.* (1978, 1979), Hansen *et al.* (1981), Quann *et al.* (1982) and Weissman *et al.* (1983).

Emphasis has also been placed on the study of trace elements in flyash and in coal, because of their toxicity and accumulation within the environment. These studies, which include uranium and thorium and their daughters as toxic materials, e.g. Berry & Wallace (1974), Lyon (1977), Horton *et al.* (1977), Hansen & Fisher (1980), McKveen (1981), IAEA (1982), estimate release rates and accumulations in different compartments of the environment. A comprehensive review of trace elements in coal and flyash, including the mechanisms of trace element enrichment, size dependence and volatility, was published by Smith (1980).

Studies of the radioactivity in coal and in flyash and evaluations of its release into the environment following the combustion of coal appeared in the sixties, e.g. Eisenbud & Petrow (1964), Martin *et al.* (1969). However, more in-depth evaluations of the radiological impact of the radioactivity released from CFP, including comparisons with the radiological impact of other energy production sources, were published more recently, e.g. Lee *et al.* (1977), McBride *et al.* (1977), Morris (1977), Coles *et al.* (1978), Cooper & Dakik (1978), Styron *et al.* (1979), US EPA (1979), Beck *et al.* (1980), Pensko & Geisler (1980), Camplin & Hallam (1980), Jacobi (1981), Jacobi *et al.* (1982), UNSCEAR (1982), Corbett (1983), De Santis & Longo (1984), Mishra *et al.* (1984) and Nakaoka *et al.* (1984, 1985).

The information contained in these and other studies is reviewed here. Particular emphasis is given to data which may influence evaluation of the radiological impact of CFP both on individuals living near such plants and on collective populations.

RADIONUCLIDES IN COAL

Small amounts of the uranium, thorium, and actinium series elements are contained in all coals used in CFP; combustion of the coal releases some of these radionuclides to the atmosphere. For example, it is estimated that about 1400 tons of uranium were released into the atmosphere in 1974 from coal-fired electric generating stations in the United States alone (Surprenant *et al.*, 1976). The different radionuclides of these series emit alpha, beta and gamma rays. In addition, coal contains minor amounts of ^{40}K , e.g. 1.4 to 2.9 pCi g⁻¹ coal (0.05 to 0.11 Bq g⁻¹ coal) (Beck & Miller, 1979). However, since this concentration is lower by at least a factor of 3 than the concentration of ^{40}K in soil, which is about 10 pCi g⁻¹ (0.37 Bq g⁻¹) (Beck & Miller, 1979), its environmental impact is insignificant and is not dealt with in this review.

There is a large variability, of up to several orders of magnitude, in the radioactivity, from the uranium, thorium and actinium series, in different coals. While the average concentration of these radionuclides in coals is of the order of a few parts per million (ppm), levels vary typically up to ~ 10 ppm and, exceptionally (mainly in low grade coal deposits), to as high as the order of 1000 ppm (Caldwell *et al.*, 1970; Los Alamos, 1976; Koester & Zieger, 1978; UNSCEAR, 1982; Jacobi *et al.*, 1982; Corbett, 1983; Valkovic, 1983; Nakaoka *et al.*, 1984; Papastefanou & Charalambous, 1984).

Table 1 shows the elemental and radioactivity concentrations of uranium and thorium, as well as the activities of ^{210}Pb , ^{210}Po and ^{226}Ra in coal, as found by different investigators. Most investigators (e.g. McBride *et al.*, 1977; Okamoto, 1980; Valkovic, 1983) assume that the entire uranium, thorium and actinium series are present in coal in secular equilibrium, except for radon and thoron, which escape because of their mobility and inertness and, in some cases (e.g. UNSCEAR, 1982; Corbett, 1983), except for ^{210}Pb and ^{210}Po . However, recent studies leave this question open. Thus, De Santis and Longo (1984) claim evidence against secular equilibrium and in the measurements performed by Papastefanou & Charalambous (1984) no radioactive equilibrium was found in Greek lignites.

It is seen from Table 1 that, while the average concentrations of uranium and thorium in coal are of the order of a few ppm, the possibility of exceptional concentrations of the order of tens of ppm or

TABLE 1
Concentrations of Radioelements and Radionuclides in Coal

<i>Radioelement or radionuclide</i>	<i>Concentration in coal</i>	<i>Type and origin of coal</i>	<i>Reference</i>
Uranium (ppm)	0.4 to 2.5	Appalachian coal	McBride <i>et al.</i> (1977)
	0.5 to 2.1	—	Ondov <i>et al.</i> (1979)
	1 (mean)	28 kinds of coal from Canada, China, Australia, Japan, S. Africa and USA	Nakaoka <i>et al.</i> (1984)
	1.4	NBS reference coal	Ondov <i>et al.</i> (1979)
	1.9	Bituminuous—Eastern or Western USA	USEPA (1979)
	1.7 to 3.3	Illinois and W. Kentucky coal	McBride <i>et al.</i> (1977)
	10	Western coals	Swanson (1972)
	25	South Texas lignite	Nucleonics Week (1979)
	0.8 to 39	—	Koester & Zieger (1978)
	0.2 to 43	Bituminuous coal	Cooper & Dakik (1978)
	10 to 140	Pennsylvania	Caldwell <i>et al.</i> (1970)
	20 to 1200	Spanish lignite	Valkovic (1983)
	up to 1800	N. Dakota lignite	Nucleonics Week (1979)
²³⁸ U (Bq g ⁻¹)	0.01 to 0.03	English coals	UNSCEAR (1982)
	0.02 to 0.04	West German coals	Jacobi <i>et al.</i> (1982)
	0.02 to 0.14	Polish coals	UNSCEAR (1982)
	0.02 to 0.16	US coals	USEPA (1979)
Thorium (ppm)	0.25	Sardinia lignite	Martinu (1980)
	0.3 to 3.6	Appalachian coal	McBride <i>et al.</i> (1977)
	2	—	Smith (1980)
	3 (mean)	28 kinds of coal from Canada, China, Australia, Japan, S. Africa and USA	Nakaoka <i>et al.</i> (1984)
	3.2	NBS reference coal	Ondov <i>et al.</i> (1979)
	5	Bituminuous Eastern or Western USA	USEPA (1979)
	40	South Texas lignite	Nucleonics Week (1979)
	2.2 to 47	Appalachian coal	Beck <i>et al.</i> (1980)
2.2 to 77	Bituminuous coal	Cooper & Dakik (1978)	
68 to 79	South Carolina and Georgia coals	Styron <i>et al.</i> (1979)	
²³² Th	0.002 to 0.02	English coals	UNSCEAR (1982)
	0.007 to 0.02	West German coals	Jacobi <i>et al.</i> (1982)

TABLE 1—*contd.*

Radioelement or radionuclide	Concentration in coal	Type and origin of coal	Reference
²³² Th	0.007 to 0.02	US coals	USEPA (1979)
	0.007 to 0.1	Polish coals	UNSCEAR (1982)
	0.06	US Western coal	Cooper & Dakik (1978)
²²⁶ Ra	0.01 to 0.02	West German coals	Jacobi <i>et al.</i> (1982)
	0.02 to 0.08	Indian coals	Mishra <i>et al.</i> (1984)
	0.1 to 2.6	Greek lignites	Papastefanou & Charalambous (1984)
²¹⁰ Pb	0.01 to 0.03	West German coals	Jacobi <i>et al.</i> (1982)
²¹⁰ Pb	0.01 to 0.03	West German coals	Jacobi <i>et al.</i> (1982)

even higher, in (low grade) coals of certain regions, should be considered when evaluating the (local) radiological impact of CFP. There is no information concerning the size of deposits of coal of high radioactivity content. However, since coals containing very high concentrations of uranium and thorium are rather exceptional, their use in CFP should be considered carefully and reduced as much as possible.

RADIONUCLIDES IN ASH

Previous investigators have not used consistent terminology for the ash sample types removed from various parts of CFP. In this study we use the following terminology for the different ash fractions; 'ash', in general, is defined as the mineral matter which becomes the dust loading flue gases in the furnace; 'bottom ash' is the ash component which forms a layer on the walls of the furnace; 'filter ash' is the ash which is separated from the flue gases in a dust collector system such as a fabric filter or an electrostatic precipitator (ESP); 'flyash' is the ash fraction which is released into the atmosphere from the stack and is of most interest from the standpoint of radiological impact.

Coles *et al.* (1979) analyzed the trace elements in four size fractions of flyash sampled in a large CFP. The concentrations of uranium and

TABLE 2
Uranium and Thorium in Fly Ash, by Size (Coles *et al.*, 1979)

Element	Particle mass median diameter (μm)			
	18.5	6.0	3.7	2.4
	Concentration (ppm)			
Uranium	8.8	16	22	29
Thorium	26	28	29	30

TABLE 3
Concentrations of Radioelements and Radionuclides in Different Fractions of Ash

Radioelement or radionuclide	Concentration	Type of ash	Reference
Uranium (ppm)	4.6-8.4	Bottom ash	Coles <i>et al.</i> (1978)
	5.6-11	Filter ash	
	16-36	Flyash	
	12	NBS reference flyash ^a	Ondov <i>et al.</i> (1975)
	1-14	Bottom ash	Ray & Parker (1977)
	15-100	Filter ash	
	7-20	Flyash	
	14.9-16.5	Bottom ash	Lyon (1977)
	20-30	Filter ash	
	17-95	Flyash	
²³⁸ U (Bq g ⁻¹)	0.05-0.1	Bottom ash	Coles <i>et al.</i> (1978)
	0.07-0.1	Filter ash	
	0.2-0.4	Flyash	Papastefanou & Charalambous (1980)
	0.3-0.7	Flyash	
Thorium (ppm)	3-20	Bottom ash	Ray & Parker (1977)
	10-23	Filter ash	
	7	Flyash	
	24.8	NBS reference flyash ^a	Ondov <i>et al.</i> (1975)
	15-20	Bottom ash	Lyon (1977)
	20-22	Filter ash	
	11-26	Flyash	
	14-22	Bottom ash	Coles <i>et al.</i> (1978)
	15-22	Filter ash	
	25-38	Flyash	

TABLE 3—contd.

Radioelement or radionuclide	Concentration	Type of ash	Reference
^{228}Th (Bq g ⁻¹)	0.05–0.08	Bottom ash	Coles <i>et al.</i> (1978)
	0.06–0.09	Filter ash	
	0.1	Flyash	UNSCEAR (1982)
	0.1	Flyash	
^{232}Th (Bq g ⁻¹)	0.04–0.1	Flyash	UNSCEAR (1982)
			UNSCEAR (1982)
^{224}Ra (Bq g ⁻¹)	0.02–0.05	Flyash	UNSCEAR (1982)
	0.07–0.9	Flyash	Mishra <i>et al.</i> (1982)
	0.2–0.5	Flyash	Papastefanou & Charalambous (1984)
^{210}Po (Bq g ⁻¹)	0.3–5.5	Flyash	UNSCEAR (1982)
^{210}Pb (Bq g ⁻¹)	0.02–0.03	Bottom ash	Coles <i>et al.</i> (1978)
	0.05–0.1	Filter ash	
	0.2–0.6	Flyash	UNSCEAR (1982)
	0.2–3	Flyash	

^aA composite sample of flyash from several CFP used as a reference standard by the US National Bureau of Standards.

thorium in the input coal used in this CFP were 2.1 and 6.2 ppm, respectively, and the ash content of the coal was 23%. Therefore, the concentrations of uranium and thorium in the mineral matter of coal were 9 and 27 ppm, respectively. Table 2 shows the concentration of these elements in the different size fractions of the stack flyash. Other investigations, however, indicate that there is a wide variation in the range of concentrations of these radioelements and related radionuclides in flyash, as can be seen in Table 3.

The large variation in the levels of radioactivity in the different fractions of ash is extremely relevant to the assessment of radiological impact and the possible ranges of radioactivity in ash should be considered fully in radiological impact studies. Several investigations have been performed, e.g. Furr *et al.* (1977), Smith (1980), in search of a correlation between the content of radionuclides in ash and different ash and coal parameters but no significant correlations have been found.

ENRICHMENT OF RADIONUCLIDES IN FLYASH

As indicated for radionuclides in Tables 2 and 3 and also in numerous studies performed on other trace elements released from CFP, e.g. Kaakinen *et al.* (1975); Klein *et al.* (1975); Coles *et al.* (1978, 1979), during the combustion of coal in a CFP there is an increase in the mass concentration of some trace elements in some fractions of ash. The mass concentration per unit weight of these elements is also higher in small size than in large size ash particles (see Table 2). We define here the ratio of concentration of an element in a certain material A, to that in

TABLE 4
Depletion or Enrichment of ^{210}Pb , Thorium and Uranium in CFP Ash

Radionuclide or radioelement	Ratio of radionuclide concentration in ash to that in coal ^a			
	Bottom ash	Mechanical collector ash	ESP ash	Flyash
^{210}Pb	0.2	0.6	2.1	3.5
Thorium	0.8	0.8	1.0	1.3
Uranium	0.7	NA	1.5	NM

^aMineral matter of input coal.

NA Not Applicable.

NM Not Measured.

another material B, as the enrichment or depletion factor of the element in A as compared with B.

Table 4 shows the depletion and enrichment factors of ^{210}Pb , uranium and thorium in different ash fractions formed in a CFP, as related to the mineral matter of the input coal and as calculated from measurements made by Kaakinen *et al.* (1975) and Klein *et al.* (1975). While bottom ash and mechanical collector ash are depleted in these elements, there is an enrichment in the ESP ash and flyash fractions, except for thorium in ESP ash.

Table 5 shows the variation of the enrichment factors of several radionuclides as a function of the size of the escaping flyash, as compiled by UNSCEAR (1982). These data indicate that there is almost no enrichment of ^{238}Th as a function of particle size of flyash. However,

there is an evident enrichment of ^{238}U , ^{226}Ra and ^{210}Pb in smaller and respirable size ($<10\mu\text{m}$) particles. The contents of ^{238}U and ^{226}Ra were also found by Papastefanou and Charalambous (1984) to be greater by a factor of 4 in respirable flyash particles than in $100\mu\text{m}$ size particles. Studies in the Federal Republic of Germany (Chatterjee *et al.*, 1980; Jacobi, 1981) show that the highest enrichment factor (11) is encountered for ^{210}Po in flyash.

From the aforementioned and additional data from Natusch *et al.* (1974) and Horton *et al.* (1977), the following conclusions can be drawn. There is a large variation in radionuclide enrichment factors in different

TABLE 5

Variation of the Enrichment Factors of Radionuclides as a Function of the Size of the Escaping Flyash Particles (UNSCEAR, 1982)

Particle Size ^a (μm)	Enrichment factors ^b			
	^{238}U	^{226}Ra	^{228}Th	^{210}Pb
2	2.8	2	1.2	4.8
10	1.6	1.3	1.1	2.1
17	1.3	1.1	1.1	1.4

^aParticle sizes of $<10\mu\text{m}$ are considered to be within the respirable range.

^bRelated to the mineral matter of coal.

fractions of ash. This variation may reflect the different compositions and origins of the coals and ashes analyzed and the use of different firing systems which are characterized by specific distributions of ash among the various fractions (see Table 6, based on data from EPRI, 1979). However, in all these studies, there is either depletion or enrichment of the radionuclides in ash as follows; (a), the bottom ash and mechanical collector ash are depleted, while the ESP ash, scrubber ash and flyash are enriched in some radionuclides, when compared with the mineral matter of the input coal; (b), the enrichment factors of the radionuclides in flyash are higher than in any filter ash; (c), the enrichment factors also increase with decreasing particle size; and (d), enrichment factors appear to be element dependent. In flyash, ESP ash and scrubber ash, the lowest enrichment factor is found for thorium and the highest for ^{210}Po . The same ranking is valid for the depletion factor in bottom ash and mechanical collector ash.

TABLE 6
Characteristics of CFP Furnaces

Parameter	Furnace type		
	Pulverized coal	Underfed or travelling grate stoker	Cyclone
Combustion temperature (°C)	1 200–1 600	—	>1 650
Ash content in flue gases (%)	80	10–20	20–30
Particles finer than 10 μm ^a in flue gas ash	65	5	90

^aParticle sizes of < 10 μm are considered to be within the respirable range.

THE MECHANISM OF ENRICHMENT OF RADIONUCLIDES IN ASH

Since radionuclides are most highly enriched in the respirable size particles of flyash which is released into the atmosphere, it is desirable to clarify the enrichment mechanism. A comprehensive study of the enrichment of trace elements during combustion of coal has been performed by Smith (1980). Based also on previous investigations, he suggests that enrichment of the trace elements may be caused by their volatilization at the coal combustion temperature, followed by a condensation process occurring at lower temperatures, mostly on small particles of relatively high specific surface area. Elements that are associated with organic compounds are postulated to be more efficiently transferred to the gaseous phase during combustion and, thus, to have a higher probability of being enriched in flyash. Organometallic compounds of some trace elements, having a high vapour pressure, may not condense until after the filtration stage or may even be released into the atmosphere in a gaseous (vapour) state.

Solvent leaching and surface analysis techniques seemed to indicate that some trace elements appear in ash as a surface adsorbed layer (Cleaver *et al.*, 1976; Linton *et al.*, 1977), a phenomenon which strengthens the hypothesis that some chemical species condense on the particle surface during the volatilization–condensation process. Hansen

and Fisher (1980), using a differential solubility technique for flyash in HCl and HF, found that uranium is mostly associated with the surface material of the flyash, while thorium appears to be associated with the aluminosilicate matrix.

The possibility of volatilization of uranium and radium compounds during the combustion of coal is mentioned by several investigators. Coles *et al.* (1978) indicate that, in an oxidizing combustion environment leading to temperatures of 1500 to 1600°C, volatile compounds of UO_3 could be formed, while radium, in the presence of steam, may form a volatile species such as $Ra(OH)_2$, similar to that found for barium.

Klein *et al.* (1975) and Lyon (1977) indicate the possibility that elements initially volatilized or dispersed in the flue gas stream are subsequently oxidized to form less volatile species which condense or are adsorbed on the ash. The volatilization–condensation process is suggested by Kaakinen *et al.* (1975) as the prevailing mechanism of enrichment of trace elements in coal combustion ash. For different elements, they found a linear relationship between the enrichment factor and the specific surface area of ash. Hence, they postulated that smaller particles, having a higher specific surface area, will be richer in trace elements, because of the availability of a larger surface area for the condensation of trace elements. However, Smith (1980) concludes that the mechanism of enrichment of trace elements in ash is still obscure and requires further investigation of both the gaseous and particulate phases of the combustion process.

VOLATILIZED SPECIES OF RADIONUCLIDES IN ASH

From the scattering of the data on particle size distributions in ash and the enrichment factors of the radionuclides there are no unequivocal conclusions. However, based on the most up-to-date data, it is suggested that the difference in particle size distribution between bottom ash, filter ash and flyash on the one hand, and the difference in enrichment factors between the particle sizes on the other hand, are not sufficient to explain the observed sequential enrichment of radioactive elements within the filter ash and flyash.

In studies by Gladney *et al.* (1976), Natusch *et al.* (1974) and Klein *et al.* (1975), the enrichment of lead in the bulk of flyash compared with that of the bulk of filter ash was found to be higher than the enrichment

of any particle size composing the flyash. Natusch *et al.* (1974) found that filter ash of particle size below about $5\ \mu\text{m}$ has a lead content of 980 ppm, while a similar particle size of flyash has a lead content of about 6500 ppm. It is, therefore, suggested that within the volatilization–condensation process of trace elements, including radionuclides, some trace elements may still be in the volatilized form at the filtration stage. Condensation of a fraction of these trace element compounds may occur, at the post-filtration stage, within the flyash or on other media. A similar phenomenon was found to occur for organic matter contained in the ash (G. L. Fisher, 1980, pers. comm.). Fisher also found that flyash is mutagenic, while filter ash is not, indicating that the mutagenic material escaped filtration and condensed after filtration of the ash.

Another mechanism which might explain the higher enrichment factors of the bulk flyash compared with those of the filter ash may be the existence of certain very conductive or high-resistivity chemical species of the trace elements within the flue gases at the inlet of the ESP filter. These might escape filtration (Katz, 1980) and appear again within a wide range of particle sizes composing the flyash.

These postulates deserve further investigation. If the assumption is valid that condensation of some trace elements, including some of the radionuclides, occurs at the post-filtration stage, then no improvement in the efficiency of the filtration system, except scrubbing, would significantly diminish the amount of these volatile radionuclides in flyash. However, if the other assumption is valid, i.e. that some chemical species of the trace elements are not filtered by the ESP because of their high resistivity or high conductivity, then the use of another filtration system, instead of ESP, e.g. fabric filters, might significantly reduce the amounts of some radionuclides in the flyash which escape into the atmosphere.

Based on a material balance performed on the coal and the different ash fractions of a CFP and on laboratory experiments, Papastefanou & Charalambous (1984) suggest that part of the uranium and radium contained in the coal is released from CFP in gaseous (vapour) phase.

A preliminary study was performed by the author of this review (Tadmor, 1986) to investigate the assumption that some condensation (sorption) of volatilized radioelements may occur on flyash after the filtration stage. Material balances were calculated for ^{210}Pb , uranium and thorium which were measured by Kaakinen *et al.* (1975), Klein *et al.* (1975) and Horton *et al.* (1977) at the various stages of coal combustion and ash formation and separation, at different CFP. The results indicate

that a significant part of the radioelements contained in flyash (e.g. 71% of the ^{210}Pb in flyash) may have condensed after filtration. Furthermore, this study indicates that only partial condensation of the volatilized radioelements occurs on the ash within the CFP. A ^{210}Pb inventory which is 2.5 times larger than the amount contained within the flyash was calculated to be in a phase distinct from that of the flyash at the stack outlet. The ^{210}Pb calculated to be in this separate form represents about 33% of the total content of the radioelement in the input coal.

Trace elements and organic matter in a form distinct from flyash have been found to be released as a vapour from CFP stacks (Carpenter, 1979). Mercury, selenium and boron have been mentioned as being released from CFP in the vapour phase (Billings *et al.*, 1973; Gladney *et al.*, 1978; Smith, 1980). Up to 76% of the uranium contained in coal and 36% of the thorium are organically bound in coal (Smith, 1980). The partial occurrence of uranium and thorium, as well as other trace elements, as metal-organic complexes and chelates is also emphasized by Gluskoter *et al.* (1977). This might account for their volatility during and following the combustion of coal.

Some radionuclide compounds of relatively low boiling points may be formed during coal combustion in a CFP, following reactions with components of the flue gases. Chlorides, which have low boiling points, may be formed. Table 7 shows the boiling temperatures of some

TABLE 7
Boiling Temperatures of Some Elements of the Uranium and Thorium Series and of Their Compounds

<i>Element or compound</i>	<i>Boiling point (°C)</i>
$\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	118
PoCl_4	390
TlNO_3	430
BiCl_3	447
UCl_4	792
ThCl_4	Subl. 820
PoCl_2	950
AcCl_3	Subl. 960
Po	962
Ra	<1 140
Tl	1 457
Bi	1 560

elements of the uranium and thorium series and of their compounds (Weast, 1980). All these boiling points are below the combustion temperature of coal in boilers ($\sim 1600^{\circ}\text{C}$) and might explain the formation and persistence of volatile compounds of radionuclides in CFP. Organometallic compounds of the radionuclides of the uranium and thorium series also have low boiling temperatures, e.g. trimethyl bismuth (110°C) and trimethyl thallium (147°C). Consequently it should be taken into account that a (gaseous) phase of the radioelements distinct from ash may exist within and at the outlet of a CFP, and that this component may not be filtered by dust collectors such as fabric filters or electrostatic precipitators.

RELEASE OF RADIONUCLIDES TO THE ATMOSPHERE

Even with efficient collection systems, a certain amount of flyash is still released into the atmosphere from CFP. In addition, radon is released from the CFP stack and from ash piles stored over long periods of time. Lee *et al.* (1977) indicated that up to 10 Ci d^{-1} ($3.7 \times 10^{11}\text{ Bq d}^{-1}$) of ^{222}Rn may be released from an ash storage pile resulting from 30 years of operation of a 1000 MWe CFP.

Table 8 shows estimates from the literature of release rates of radionuclides contained in flyash from different CFP normalized to 1000 MWe. There are significant differences; unfortunately, however, only some of the pertinent data and assumptions which might explain these differences are presented in the publications. These data indicate that modern CFP, i.e. those equipped with sophisticated ash-filtration devices, release about an order of magnitude less radioactivity to the atmosphere than do older CFP. After averaging the radioactivity released at different CFP, it was concluded (US EPA, 1979; IAEA, 1982; UNSCEAR, 1982) that, for a content in coal of 1–2 ppm uranium and 3–5 ppm thorium, a release rate of 7.5×10^5 to $1.5 \times 10^6\text{ Bq}$ of ^{238}U per MWe year, with a corresponding release of the other radionuclides, is a plausible assumption for CFP.

In evaluating the contribution of radionuclide release from CFP to the environment, consideration should also be given to the negative 'Suess effect'. Because of the age of the fossil fuel and of the comparatively short half-life of ^{14}C (5730 y), coal is free from ^{14}C and, therefore, the release of large amounts of CO_2 from CFP dilutes the 'natural' ^{14}C within the atmosphere. Thus, a 'negative radioactivity emission credit'

TABLE 8
Release Rates (Bq y⁻¹) of Radionuclides Contained in Flyash, from CFP, Normalized to 1 000 MWe

Reference	Release rates of radionuclides (Bq y ⁻¹)							
	²³⁸ U	²³² Ra	²¹⁰ Pb	²¹⁰ Po	²¹⁰ Rn	¹³⁷ Cs	¹³⁴ Cs	⁹⁰ Sr
Eisenbud & Petrow (1964)		6.4 × 10 ⁸				4.0 × 10 ⁸		
McBride <i>et al.</i> (1977)	3.0 × 10 ⁸	3.0 × 10 ⁸	3.0 × 10 ⁸	3.0 × 10 ⁸	3.0 × 10 ¹⁰	1.8 × 10 ⁸	1.5 × 10 ¹⁰	1.2 × 10 ⁷
Beek & Miller (1979) ^a	10 ⁹	7.8 × 10 ⁸	2.6 × 10 ⁹	2.6 × 10 ⁹	5.5 × 10 ¹⁰	4.1 × 10 ⁸		
US EPA (1979)—old model ^b	7.8 × 10 ⁹	7.8 × 10 ⁹	1.6 × 10 ¹⁰	1.6 × 10 ¹⁰	5.3 × 10 ¹⁰	6.7 × 10 ⁹	4.2 × 10 ¹⁰	3.7 × 10 ⁸
US EPA (1979)—new model ^c	10 ⁹	7.3 × 10 ⁸	2.6 × 10 ⁹	2.6 × 10 ⁹	6.3 × 10 ¹⁰	6.7 × 10 ⁸	5.3 × 10 ¹⁰	3.0 × 10 ⁹
Okamoto (1980)			7.4 × 10 ¹⁰	7.4 × 10 ¹⁰				
Camplin & Hallam (1980)	10 ⁹	10 ⁹	10 ⁹	10 ⁹	10 ⁹	10 ⁹	10 ⁹	3.7 × 10 ⁷
Teknekron (1981)—old model ^b	2.3 × 10 ⁹		4.8 × 10 ⁹	4.8 × 10 ⁹	3.2 × 10 ¹⁰	1.7 × 10 ⁹	2.4 × 10 ¹⁰	
Teknekron (1981)—new model ^c	7.0 × 10 ⁸	5.2 × 10 ⁸	1.7 × 10 ⁹	1.7 × 10 ⁹	3.6 × 10 ¹⁰	2.5 × 10 ⁸	2.7 × 10 ¹⁰	
IAEA (1982) and UNSCEAR (1982)—average values	1.5 × 10 ⁹	1.5 × 10 ⁹	5.0 × 10 ⁹	5.0 × 10 ⁹	1.5 × 10 ⁹	1.5 × 10 ⁹		
UNSCEAR (1982) exceptional values	7.0 × 10 ⁹	1.8 × 10 ¹⁰	2.6 × 10 ¹⁰		2.6 × 10 ¹⁰			
Corbett (1983)—modern CFP ^a	1.8 × 10 ¹⁰							
Mishra <i>et al.</i> (1984)	8.0 × 10 ⁸	8.0 × 10 ⁸	8.0 × 10 ⁹	8.0 × 10 ⁹	4.0 × 10 ⁸	4.0 × 10 ⁸		
		4.5 × 10 ⁹						
		2.6 × 10 ¹⁰						
De Santis & Longo (1984)	2.2 × 10 ⁸	2.2 × 10 ⁸	3.7 × 10 ⁹	3.7 × 10 ⁹	2.2 × 10 ⁸	2.2 × 10 ⁸	2.2 × 10 ⁸	2.2 × 10 ⁷
Nakaoka <i>et al.</i> (1984)	7.6 × 10 ⁷	7.6 × 10 ⁷	3.7 × 10 ⁹	3.7 × 10 ⁹	8.1 × 10 ⁷	8.1 × 10 ⁷	8.1 × 10 ⁷	8.1 × 10 ⁷
	9.0 × 10 ⁷							

^aModern CFP, i.e. equipped with sophisticated devices, from which only about 1% of the ash is released into the atmosphere.

^bCFP which releases about 10% of the ash into the atmosphere.

should be allocated to the emission of CO₂ from CFP (Suess, 1955). The negative emission credit is estimated to be 4×10^{11} Bq y⁻¹ for a CFP of 1000 MWe (Camplin & Hallam, 1980; Okamoto, 1980; De Santis & Longo, 1980).

ENVIRONMENTAL RADIOACTIVITY FOLLOWING THE RELEASE OF RADIONUCLIDES FROM CFP INTO THE ATMOSPHERE

Flyash containing radionuclides of the uranium, thorium and actinium series or the radionuclides released in a separate phase from flyash diffuse within the atmosphere and at a certain distance from the release point reach ground level, thereby increasing the natural concentration of these radionuclides. Thus, the radionuclides in a separate phase from flyash, or the radionuclides accompanying the flyash particles in the respirable size range ($<10\mu\text{m}$), present an inhalation hazard to man. It should be noted that 80–90% of the flyash particles escaping from the outlet of an ESP at a CFP are of a size smaller than $10\mu\text{m}$ (Gabrielson *et al.*, 1978). In addition, during their atmospheric transport, flyash and the radionuclides deposit on the ground either by dry deposition or by precipitation scavenging. The ground surface then becomes enriched in these radionuclides and causes a potential external radiation hazard to man. Deposition may also occur directly on vegetation, thus producing a potential ingestion hazard to man. From the ground surface, the radioactivity may leach into ground waters or be transferred into vegetation growing on the contaminated soil and into other food products, thereby presenting a further ingestion hazard to man. It is therefore important to assess the environmental radioactivity enhancement stemming from the release of radionuclides from CFP.

Because of the wide variation in the data found by different investigators for the release of radionuclides from CFP, large discrepancies in the environmental radioactivity data around CFP would also be expected. Indeed, the data presented by different investigators differ by orders of magnitude (Martin *et al.*, 1970; Beck & Miller, 1979; Papastefanou & Charalambous, 1980; UNSCEAR 1982; Nakaoka *et al.* 1984, 1985). Thus, for example, UNSCEAR (1982) cites measurements performed downwind of a CFP in Alabama, USA, in which the following atmospheric (ground-level) concentrations were found: 5×10^{-3} Bq m⁻³ of ²²⁶Ra, 10^{-5} Bq m⁻³ of ²³²Th and 10^{-4} Bq m⁻³ of ²³⁸U.

On the other hand, Nakaoka *et al.* (1984, 1985) calculated the following concentrations of radionuclides in air at the maximum concentration points around a 1000 MWe CFP: 4.8×10^{-9} Bq m⁻³ for ²³⁸U, ²³²Th, ²²⁶Ra and ²²⁴Ra; 1.9×10^{-7} Bq m⁻³ for ²¹⁰Pb, ²¹²Pb and ²¹⁰Po; 1.3×10^{-6} Bq m⁻³ for ²¹⁴Pb and ²¹⁴Bi. These data should be compared with the normal background concentration of about 10^{-6} Bq m⁻³ for each one of the aforementioned radionuclides (UNSCEAR, 1982). Some, but not all, of the differences between the environmental concentrations of radioactivity around the various CFP might be explained by the differences in filtration systems and stack heights. Thus, Nakaoka *et al.* (1984) indicate that their low radioelement concentration data are probably the result of the high removal rates of flyash in Japanese CFP.

Enrichments of uranium and thorium in the soils around CFP have been observed by Ray and Parker (1977) and Horton *et al.* (1977) and have also been reported in industrialized areas of Poland and New Mexico (Styron *et al.*, 1979). The radionuclide concentrations in soils around the Allen CFP divided by the world average concentrations were found (Ray & Parker, 1977) to be 4 for uranium and 2.2 for thorium. Therefore an estimate of the radiation doses caused by the radionuclides released from a CFP is appropriate.

RADIATION DOSES AND PATHWAYS TO MAN

Radiation doses have been estimated by several investigators and the major exposure pathways evaluated for radionuclides released from CFP. Since, as described earlier, there are discrepancies between investigations over the input parameters for evaluation of radiological impact, these discrepancies are similarly found in the evaluated radiation doses and exposure pathways.

Radiation doses

Table 9 shows the maximum whole-body radiation doses caused to an individual at a distance of about 500 m from a CFP, as calculated by different investigators. The original values were normalized for a CFP of 1000 MWe and expressed as effective radiation dose equivalent, using the weighting factors recommended by ICRP (1977). Different values for some of the parameters (content of radioelements in coal and stack height of CFP) which might partly explain differences between the

radiation doses are also indicated. Maximum collective population doses, wherever available, are also given. The radiation doses shown are the maximum calculated doses in each study. For example, the US EPA (1979) radiation doses for an individual and for the population at large have been selected from different case studies, in which each is at maximum value, i.e. a rural site for the individual radiation doses and an urban site for the population doses.

Table 9 shows that even taking into account the differences in the uranium and thorium contents of coal as assumed by the various investigators, the results of their evaluations of the individual and population radiation doses are still quite different. These radiation doses were computed for a range of uranium and thorium concentrations in coal of up to 25 and 40 ppm, respectively.

As aforementioned the radiation doses resulting from the release of radionuclides from CFP should be compared to the reduction of radiological risks resulting from the negative Suess effect. Such a comparison of dose estimates should be made on the basis of the same population assumptions and over the same time-frame of exposure. Table 10 shows such a self-consistent comparison based on several different studies. It is seen that, when the comparison is made on the same population and time-frame basis, the reduction of radiation doses due to the Suess effect amounts to a small percentage (about 3 to 16%) of the doses caused by release of radioactivity from CFP.

Pathways to man

There is a general consensus among different investigators that air immersion is insignificant and that ground surface irradiation makes only a minor contribution to the radiation dose caused by CFP. However, there are some differences of opinion as to the predominance of the inhalation or ingestion pathways in the generation of dose to an individual or a nearby population. Table 11 shows some data on exposure pathways as presented by different investigators.

Whether ingestion or inhalation is the more important irradiation mechanism depends on the radionuclides considered, the site characteristics, the food and food acquirement habits, other parameters related to the radionuclides and to the environment and the methods and parameters used in the evaluations.

TABLE 9
Maximum Individual and Collective Radiation Doses Resulting from a 1000 MW_e CFP

<i>Whole-body maximum individual doses (mSv y⁻¹) at about 500 m from CFP, according to:</i>						
<i>McBride et al. (1977)</i>	<i>Cooper & Dakik (1978)</i>	<i>US EPA (1979)</i>	<i>Campbell & Hallam (1980)</i>	<i>Teknickron (1981)</i>		
<i>S. Texas coal Worst case</i>		<i>rural site</i>			<i>old^a (mean)</i>	<i>new^b (mean)</i>
0.04	0.1	0.4	0.1	0.08	0.08	0.01
1.5	1	214 ^c	110	1.3	0.2	
Collective doses (effective – person Sv y ⁻¹) up to about 80 km (for the population of Great Britain, 500 y commitment from 30 y operation)						
Content (ppm) of radioelements in coal						
U	5	25	1.9		1.2 (mean)	
Th	7	40	5		3.1 (mean)	
50–300	200	97	185	500 (effective)	150	
Physical stack height (m) of CFP						

^aCFP which releases about 10% of the ash into the atmosphere.

^bCFP equipped with sophisticated devices, from which only about 1% of the ash is released into the atmosphere.

^cUrban site.

TABLE 10
 Comparison of Collective Radiation Doses Resulting from a 1 000 MWe CFP, with the Reduction of Doses due to the Stress Effect

Reference	Collective radiation dose estimates			
	Population for which dose estimates were made	Time-frame for which dose estimates were made	The release of radio-activity from CFP (person-Sv) (GW) ⁻¹	
			The reduction due to the Stress effect from CFP (person-Sv) (GW) ⁻¹	
Corbett (1983)	Local ^a	1 y	2	0.15
	Global ^b	1 y		2
Campbell and Hallam (1980)	Great Britain	500 y	170	11
Okamoto (1980)	Japan	1 y	29-170	4.6

^aPopulation in the 'vicinity' (a few tens of km) of the CFP.

^bWorld population.

CONCLUSIONS

The data reviewed in the present study indicate that minute amounts of radionuclides of the uranium, thorium and actinum series are released from CFP into the atmosphere and irradiate the population in the vicinity of the CFP, mainly through the inhalation and ingestion irradiation modes.

A maximum individual radiation dose of 0.01 to 0.05 mSv y^{-1} and a collective dose of 2 to 4 person. Sv y^{-1} are considered as representative for the radiation doses resulting from a modern 1000 MWe CFP equipped with sophisticated devices to retain 99% or more of the ash within the CFP. The individual radiation dose amounts to 1–5% of the natural background radiation, while the stochastic radiological risk inherent in the collective radiation dose amounts to 1 lethal cancer in 25 to 50 years. This radiological risk is relatively small. However, the diversity of data should be taken into consideration. This diversity derives from differences in coal supply, CFP features, environmental conditions, as well as from uncertainties in the models and parameters used in the evaluations. Amongst these, the coal radioactivity content most strongly influences the radiological risk. The aforementioned risks were evaluated for a CFP using a coal supply in which it is assumed that the contents of uranium and thorium are of the order of unit ppm. However, although exceptionally, the contents of uranium and thorium in various coal supplies may differ by orders of magnitude, increasing the (local) radiological risks almost linearly to a level of concern. Therefore, it is suggested that even infrequent use of coals of high uranium and thorium contents cannot be disregarded. This implies that the radioactivity content of the coal supply to a CFP should be monitored and the use of coal of high uranium and thorium contents should be avoided or reduced to such a level that—on an average annual basis—the use of low radioactivity coal would limit the annual radiological risk caused by a CFP.

Other main parameters which should be considered to minimize the possible radiological risks are: the height of the CFP stack (increase of stack height reduces the individual dose) and site characteristics, such as population density (low population densities reduce the collective dose).

Although these strategies are mentioned as separate alternatives, combinations and trade-offs may be envisaged to minimize the radiological risks caused by CFP.

TABLE II
 Pathway Parameters Related to the Release of Radionuclides from CFP

<i>Reference</i>	<i>Predominant pathway</i>	<i>Most important contributors among the radionuclides</i>	<i>Most important food chain</i>	<i>Evaluation made for</i>	<i>Remarks</i>
Martin <i>et al.</i> (1969)	Inhalation				Ingestion was not considered in evaluation. Secular equilibrium in ash was assumed.
McBride <i>et al.</i> (1977)	Ingestion	^{226}Ra ^{228}Ra	Beef, milk, vegetables	Individual population	100% solubility of radionuclides in environmental compartments was assumed. Higher release heights increase the contribution of ingestion. Secular equilibrium in ash was assumed. All food was assumed to be produced by reference area.
Lee <i>et al.</i> (1977)	Inhalation	^{222}Rn from ash piles (1.5% release)		Population	Secular equilibrium in ash was assumed.

Styron <i>et al.</i> (1979)	Ingestion	^{210}Pb ^{210}Po	Vegetables	Individual	No rain scavenging was considered. Only ^{238}U , ^{234}U , ^{210}Pb and ^{210}Po were considered.
Beek <i>et al.</i> (1980)	Inhalation			Individual	Secular equilibrium in ash was assumed. Most radioactivity was assumed to be insoluble in human body fluids.
Camplin & Hallam (1980)	Ingestion	^{210}Pb ^{210}Po ^{231}Pa		Individual	Food was assumed to be grown at point of maximum concentration.
	Inhalation	^{228}Th ^{230}Th ^{232}Th		Population	Dose commitment for the population was evaluated for 500 y, from 30 y operation of the plant.
Okamoto (1980)	Ingestion	^{210}Po ^{210}Pb	Fish, leafy vegetables	Individual and population	Secular equilibrium in ash was assumed.
Nakaoka <i>et al.</i> (1985)	Ingestion		Vegetables	Individual and population	The vegetable pathway contributes about 75% of the total dose.

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