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SOME RADIOLOGICAL ASPECTS OF COAL COMBUSTION

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Summary

Data on the radionuclide content of coal, bottom ash and fly ash are reviewed. Estimates are made of the quantities of various radionuclides released to the environment from coal combustion and the probable resultant radiation doses to the population. Factors that influence these dose estimates, such as particle size, solubility and radon emanating power are discussed.

Introduction

The combustion of coal results in the release of small quantities of radionuclides into the atmosphere. Unlike nuclear power, these releases result entirely from a redistribution of already existing radionuclides rather than the creation of new radionuclides. Whether or not such releases constitute a potential health hazard and whether the releases of radioactivity are comparable to or greater than those from a nuclear fuel cycle has become a matter of some public and scientific concern. This question is particularly relevant as a result of recent government policy decisions to increase the utilization of coal relative to other fuels.

We have made an extensive review of data on the radioactivity of coal and coal ashes and the releases of radionuclides from typical power plants, and have also analyzed the results of a number of published assessments of potential health hazards.(1) In this paper we summarize some of our findings and discuss some of our conclusions as well as some caveats with respect to other published assessments, and present some additional data.

Reference 1 contains a comprehensive list of literature citations, and we refer the reader to that paper for more complete documentation on some of the results summarized here.

Radioactivity of U. S. Coals

Measurements of uranium, thorium, and potassium in coal have been reported in the literature for almost 1000 different samples obtained directly from the mines that provide most of the coal used in the U. S. today as well as from most of the proven reserves. Mean concentrations were found to be 1.7 μg/g for natural uranium, 4.5 μg/g for thorium and 1700 μg/g for potassium, equivalent to the mean radioactivity levels for U-238, Th-232, and K-40 shown in Table 1. Although a few coals were found to have fairly high concentrations of radionuclides, over 95 percent of the samples had activities less than a factor of three times the means. Contrary to some published reports, western U.S. coals are not, in general, more radioactive than eastern U. S. coals, nor are lignites, although small pockets of uraniferous coal do exist in some western states.

TABLE 1

RADIOACTIVITY OF U. S. COALS IN pCi/g

Nuclide:		^{238}U	^{232}Th	^{40}K
Samples collected from mines (see Reference 1)	mean:	0.60	0.50	1.4
	range:	< 0.1-15	< 0.1-5.3	0.02-20
	# samples:	(910)	(910)	(983)
Samples collected from power plants and analyzed by EML	mean:	0.58	0.24	1.9
	range:	0.13-1.8	< 0.1-0.47	< 1 -5.3
	# samples:	(14)	(14)	(14)
Other reported analyses of samples from power plants*	mean:	0.34	0.56	2.8
	range:	< 0.1-0.8	0.17-2.3	0.73-5.6
	# samples:	(15)	(10)	(10)
Soil[3,6] (for comparison)	mean:	0.70	0.70	10
	range:	0.3-1.4	0.2 -1.3	3-20

*Values for mine samples are for coal as mined, for power plants for air-dried samples.

Since a large fraction of the coal burned in U.S. power plants is "cleaned" prior to use to reduce ash and sulfur content, we have compared the activities of the mine samples with those of samples obtained directly from power plants. These results are also given in Table 1. Data for samples analyzed by us are reported separately from other data in the literature, since some of the latter were ambiguous with respect to numbers of different samples, origins of the samples and accuracy of analysis. Our data, most of which have not been previously reported, were obtained by performing high resolution gamma-ray spectrometry on samples obtained from 14 different power plants, three of which burn lignite.

Uranium-238 ahd thorium-232 have generally been found to be in secular equilibrium with their decay products in coal. Our own analyses generally confirm this. Thus, the data in Table 1 for U-238 and Th-232 also represent the activities of their respective decay products. In Reference 1, we referred to a preliminary report from Mound Laboratory which suggested enhanced Pb-210 and Po-210 levels in some coal samples. In an updated report, however, this group reports secular equilibrium does appear to exist for their samples.(2)

Considering the relatively small number of samples from power plants as opposed to mines, the agreement in means and ranges of activities is excellent and tends to support our previous conclusion that the mean activities inferred from the mine samples are representative of coal burned in actual power plants.

For perspective, the activities of typical soils are also given in Table 1. (3)

Radioactivity of Coal Ash

When coal is burned in power plants, non-combustible trace elements are concentrated in the ash. Based on a mean ash content in U. S. coals of 13.4 percent, we would expect the specific activity of this ash to average 7.5 times that of the coal, i.e., 4.5 pCi/g for U-238 and each of its decay products, 3.7 pCi/g for Th-232 and each of its decay products, and 10 pCi/g for K-40. Studies have shown, however, that some of the more volatile trace elements are preferentially recondensed on smaller particles. This results in an enrichment of fly ash relative to bottom ash, and particularly on the smaller flyash particles, which are less efficiently removed by the emission control equipment. Radionuclides which have been demonstrated to exhibit this enrichment are Pb-210 and Po-210, in particular, and to a lesser extent U-238, U-234 and Ra-226.

In Table 2 we summarize available data on radionuclides in ash samples obtained from U. S. power plants. Except for one set of data by Coles et al., (4) these are all samples of contained rather than escaping ash. The Coles et al. data are for 4 different size fractions of the same stack sample and illustrate the progressive enrichment on smaller sized particles for certain radionuclides. Again we have listed our own data separately from those of the other investigators. Both sets of data clearly indicate the enrichment of Pb-210 in fly ash relative to bottom ash. The enrichment of other uranium and radium nuclides is too small to be seen in collected ash, particularly for this small number of samples. Our own data include 6 lignite ashes and 5 bituminous coal ashes obtained from 11 different power plants. No significant differences in activity were observed between lignite and non-lignite ashes.

Considering the relatively small number of samples, the average activities shown in Table 2 are in excellent agreement with those expected based on the coal data. We thus conclude that the mean enriched ash activities derived from the coal mine samples can be used as a basis for estimating typical radionuclide emissions from coal-fired power plants.

Radioactivity Emissions from Power Plants

The amount of fly ash, and thus the amount of radioactivity released to the atmosphere, depends on the efficiency of the plant's emission control equipment. Although the total amount of ash generated by the plant depends on the type and amount of coal burned, a typical modern 1000 MWe plant operating 80 percent of the time will consume about 2.3×10^9 kg of coal per year and produce about 3×10^8 kg of ash per year. If this plant just meets the EPA particulate emission standard of 0.1 lb per 10^6 BTU it will release 3.1×10^6 kg/y of fly ash. Based on the Coles et al. (4) data, as well as other measurements reviewed in Reference 1, we have estimated fly ash escaping from such a plant will be enriched in Pb-210 and Po-210 by about a factor of five over the concentration expected based on the ash content of the fuel, in U-238 and U-234 by a factor of two, and in Ra-226 by a factor of 1.5. These values will of course vary

TABLE 2

RADIOACTIVITY OF COAL ASHES COLLECTED FROM U. S. POWER PLANTS in pCi/g

	Nuclide:	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{40}K
Samples analyzed by EML:						
Flyash	mean:	4.2	3.6	9.5	2.1	10
	range:	2.3-6.6	2.3-6.1	2-19	1.2-2.8	2-20
	# samples:	(11)	(11)	(6)	(11)	(11)
Bottom ash	mean:	4.2	4.0	2.2	1.9	8.0
	range:	1.8-8.6	1.6-7.6	0.5-5	1.1-2.8	0.6-20
	# samples:	(10)	(10)	(5)	(10)	(10)
Scrubber sludge	mean	2.9	2.6	3.4	0.5	5.6
	# samples	(1)	(1)	(1)	(1)	(1)
Other analyses:*						
Flyash	mean:	2.4	2.8	4.5	3.4	14
	range:	1.4-6.5	0.8-5.0	1.4-11	0.4-7.5	1.2-29
	# samples	(27)	(17)	(5)	(39)	(33)
Bottom ash	mean	2.3	1.7	2.0	1.9	6.5
	range:	1.5-2.8	0.6-2.5	0.6-5.6	1.5-2.2	6.2-6.8
	# samples:	(3)	(3)	(4)	(2)	(2)
Stack sample[4]	range:	5.4-12	3.3-5.9	4.3-17	2.8-3.3	7.0-7.4

*See Reference 1 for details.

TABLE 3

ESTIMATED ANNUAL RADIONUCLIDE EMISSIONS FROM COAL-FIRED POWER PLANTS*

	Modern Plant		1972 Plant	
Nuclide	Flyash Activity (pCi/g)	Annual Emissions (mCi /y)	Flyash Activity (pCi/y)	Annual Emissions (mCi /y)
^{238}U	9.0	28	4.5	126
^{234}U	9.0	28	4.5	126
^{230}Th	4.5	15	4.5	126
^{226}Ra	6.8	21	4.5	126
^{232}Th	3.7	11	3.7	104
^{228}Th	3.7	11	3.7	104
^{228}Ra	3.7	11	3.7	104
^{210}Pb	22.5	70	9.0	252
^{210}Po	22.5	70	9.0	252
^{40}K	10	31	10	280
^{222}Rn	-	1500	-	1500
Flyash (kg/y)	-	3.1×10^6	-	2.8×10^7

*Modern plant is a plant meeting EPA particulate emissions standards of 0.1 lb per 10^6 BTU, 1972 plant is one releasing at a rate equal to mean for all U. S. plants operating in 1972.

from plant to plant, and are perhaps slightly greater for plants using scrubbers instead of electrostatic precipitators, since the former plants release smaller-size particles for the same total mass release.(5) No enrichment is expected for other radionuclides. The final assumed mean activities of escaping fly ash and the resultant total annual release from a typical modern plant for each long-lived radionuclide are given in Table 3.

For comparison, we considered a typical older power plant, i.e., one that releases at a rate equal to the average for all power plants operating in the U. S. in 1972. Such a plant, which would release $2.8x10^7$ kg of fly ash per year per 1000 MWe, might be considered a present-day worst case situation. Since, as the mass of released ash increases, the fraction made up by small particles decreases, the relative enrichment of volatile elements on escaping fly ash is much smaller. We have assumed an enrichment of a factor of two for Pb-210 and Po-210, consistent with the measurements made on collected fly ash, and no significant enrichment for other nuclides. The fly ash activities and annual releases for this plant are also given in Table 3.

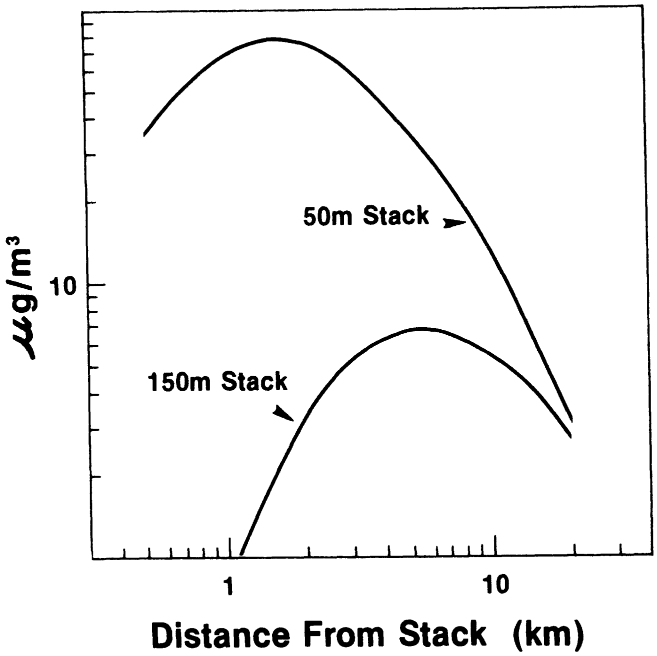
The estimated annual emissions of Rn-222 given in the Table assume that essentially all the available Rn-222 in the coal is released during pulverizing and combustion.

Releases for actual plants may, in a given year, deviate from the estimates of Table 3 as a result of temporarily burning coal or releasing ash containing activities significantly greater or less than our assumptions. However, since most plants, particularly those in populated areas remote from coal mines, will burn coal from a variety of mines over their lifetime, we would expect the results in Table 3 to fairly reflect the long term average releases to be expected from both older and newer U. S. power plants.

Perturbations on Ambient Natural Radiation Levels

The actual number of curies released of each nuclide is not in itself particularly significant. The important question is how much do these releases increase ambient natural background radiation levels in air and soil and thus potentially increase the radiation exposure of the surrounding population. One important factor which influences the extent of these perturbations is the height at which the effluents are released. We therefore calculated expected average annual ground-level activities resulting from the releases given in Table 3, for both a 50 meter stack and a 150 meter stack (see Reference 1 for details). The results obtained for the distance of maximum concentration are given in Table 4. Even for the older "dirtier" 1972 plant, releasing from a short stack, the calculated maximum ground-level activities are at most only a few times ambient natural background levels while for a modern plant releasing from tall stacks the calculated maximum activities are small fractions of ambient background levels. These maximum levels for the 5o meter stack occur at about 2 km from the plant but drop off quite rapidly as shown in Figure 1. Thus only a small area is actually exposed to these levels. Conversely, the maximum for the tall stacks occurs at about 6 km and drops off more slowly. Background levels were estimated by assuming a mean ambient air dust loading (resuspended soil) (6) of 100 $\mu g/m^3$ and the average soil activities given in Table 1.

Figure 1. Mean annual ground level airborne fly ash concentration versus distance from power plant for release rate of 1 kg/s.



For short periods of time, of course, actual air activities can be considerably higher than these annual averages, depending on meteorological conditions. The annual average values may also vary from year to year due to variations in coal burned, annual average meteorology, and other factors. Clearly even large such variations will not result in significant perturbations of ambient radiation levels for modern plants, releasing from tall stacks.

The increases in concentration for Rn-222 have not been given in Table 4 since in all cases they are insignificant relative to ambient levels. The amount of Rn-222 being released is equivalent to that which is released from only about 0.1 km^2 of soil surface.(1)

Assuming a deposition velocity of 1.0 cm/s each, for both dry deposition and wet deposition, we can estimate the average long term deposition onto the ground from the air concentrations given in Table 4. (See Reference 1 for a discussion of the appropriateness of these deposition velocity estimates.) The total depositions after 50 years of operation are given in Table 5. Again, these represent values at the distance of peak air concentrations and the average deposition in the environs of the plants would be about a factor of 70 less for releases from 50 meter stacks and a factor of 10 less for 150 meter stacks if averaged out to a distance of 50 km.

As an illustration of the net effect of these depositions on ambient soil activities in fields where

TABLE 4

ESTIMATED MAXIMUM MEAN ANNUAL GROUND LEVEL AIR ACTIVITY IN aCi/m^3 RESULTING FROM COAL-FIRED POWER PLANT EMISSIONS

Nuclide	Modern Plant 50 meter stack	Modern Plant 150 meter stack	1972 Plant 50 meter stack	1972 Plant 150 meter stack	Natural Background
^{238}U, ^{234}U	70	6	310	27	70
^{230}Th	35	3	310	27	70
^{226}Ra	52	4.5	310	27	70
^{232}Th, ^{228}Th, ^{228}Ra	29	2.5	250	22	70
^{210}Pb	180	15	620	54	14000
^{210}Po	180	15	620	54	3300
^{40}K	78	6.7	690	60	1000
Ash ($\mu g/m^3$)	7.8	0.67	69.4	6.0	100

crops might be grown, we have assumed that the activities given in Table 5 are mixed uniformly down to a depth of 30 cm due to periodic plowing and have compared the resultant increase in soil specific activity with the mean ambient soil activities given in Table 1. The results are shown in Table 6. Even for the 1972 plant releasing from a short stack, the increase over background is only about 3 percent. For modern plants, releasing from tall stacks, it is clear that the increase in soil specific activity would be negligible even for a much smaller penetration or mixing of the fly ash into the soil.

TABLE 5

ESTIMATED MAXIMUM DEPOSITION IN nCi/m^2 FROM 50 YEARS OF POWER PLANT OPERATION

Nuclide	Modern Plant 50 meter stack	Modern Plant 150 meter stack	1972 Plant 50 meter stack	1972 Plant 150 meter stack
^{238}U, ^{234}U	2.25	0.19	10	0.85
^{230}Th	1.35	0.10	10	0.85
^{226}Ra	1.7	0.14	10	0.85
^{232}Th, ^{228}Th, ^{228}Ra	0.9	0.08	8	0.70
^{210}Pb, ^{210}Po	5.5	0.50	20	1.7
^{40}K	2.5	0.22	22	1.9

TABLE 6

INCREASE IN SOIL ACTIVITY IN fCi/g FOR PLOWED FIELDS FROM 50 YEARS CUMULATIVE DEPOSITION

Nuclide	Modern Plant 150 Meter Stack	1972 Plant 50 Meter Stack	Natural Soil Background
^{238}U, ^{234}U	0.42	22	700
^{230}Th	0.21	22	700
^{226}Ra	0.30	22	700
^{232}Th, ^{228}Th, ^{228}Ra	0.18	18	700
^{210}Pb, ^{210}Po	1.1	44	~1,000
^{40}K	0.49	49	10,000

Dose to Individuals

The major potential pathways which might result in increased radiation doses to humans from coal fired power plant emissions are inhalation of fly ash, ingestion of food grown in contaminated soil, or direct radiation exposure from the increased deposited radioactivity. We concluded in Reference 1 that the latter two pathways could be completely neglected and that the only potentially significant pathway is direct inhalation, and the only organ potentially at risk is the lung.

Ingestion and direct radiation doses are negligible since, as we have shown, the probable increases in soil specific activity are very small, even over the small area of maximum deposition. Even if direct deposition on foliage is considered, the available evidence indicates fly ash is very insoluble (1,7) and thus its activity is not likely to be incorporated into the vegetation, or into body tissues and organs if ingested. Also it is unlikely any individual would obtain more than a small fraction of his diet from crops grown in the small area of maximum contamination, and even less likely he would do so for the extended period necessary to build up an increased body burden of naturally occurring radionuclides.

The dose-equivalent to the lung from inhalation of fly ash particles can be estimated from the airborne activity levels given in Table 4. These estimates, which are described in more detail in Reference 1, are summarized in Table 7. They assume, conservatively, that all the airborne fly ash is respirable and insoluble, and are for the maximum exposed individual, i.e., one who is exposed continuously at the levels given in Table 4 for a sufficient period of time to build up equilibrium levels in the lung. Even for the older 1972 plant with a 50 meter stack, the maximum dose equivalent is less than 10 percent of that from natural background. The dose equivalent for the older 1972 plant is additionally conservative in that, as larger quantities of ash are released, the relative fraction in the respirable range decreases. Even for plants with the most efficient emission control systems, a considerable fraction of the released fly ash is not respirable. (5, 7-9)

TABLE 7

ESTIMATES OF DOSE EQUIVALENT TO LUNG OF MAXIMUM EXPOSED INDIVIDUALS FROM INHALATION OF COAL PLANT EMISSIONS

Type Plant	Dose Equivalent (mrem/y) 50 Meter Stack	150 Meter Stack
Modern Plant	1.6	0.14
1972 Plant	8.7	0.75
(Natural Background)[6]	~ 100	

The dose-equivalents to other organs from inhaled radionuclides are considerably smaller than those to the lung, since the insolubility of most of the fly ash results in very little transfer of radionuclides from the lung to other organs or to soft tissue. We thus conclude that the effluents from even "dirty" coal-fired power plants releasing from short stacks will result in negligible increases in radiation exposure to the general population or to any individual.

Radioactivity of Waste Materials

Only a small fraction of the ash produced in coal plants is released through the stack. The remainder is usually sluiced to holding ponds. About 20 percent is currently sold for use as land fill or in manufacturing building materials. The use of scrubbers to remove sulfur from flue gases results in an additional waste product in the form of sludge. As shown in Table 2, these wastes have radioactivities several times that of the average soil. Because of the insolubility of the ash, leaching of radionuclides into ground water does not appear to present a serious problem, although there have been few studies of long-term weathering effects. The use of ash in building materials can result in potential increases in direct external radiation exposure to occupants of the resultant buildings depending on the activity of the materials with which the ash is mixed. There have been only a few scattered measurements of the activity of building materials made with ash and slag and no systematic studies have been made of the potential increase in population exposure.

One serious concern has been the potential release of Rn-222 from ash waste piles and ash products. We reported previously that radon did not appear to emanate significantly from fly ash and bottom ash. Since then, we have analyzed a number of additional samples for radon emanation. These results are summarized in Table 8. As shown, the emanation from ash averages less than 2 percent. This compares to about 15 percent for the average soil (3) and 20 percent for uranium mill tailings. (10) Bituminous coal appears to emanate at a rate comparable to soil, however, the three lignite coal samples we have evaluated emanated at much higher rates. Unless long term weathering somehow modified the physical composition of the impermeable glassy fly ash particles, we would conclude that emanation of radon from ash does not constitute a significant potential perturbation to ambient Rn-222 levels.

TABLE 8

^{222}Rn EMANATION* FROM COAL AND ASH SAMPLES

Type of Sample	No. of Samples	Emanation (%) Mean	Range
bituminous coal	6	20	11-37
lignite coal	3	53	44-63
flyash	11	< 2	< 1-3
bottom ash, slag	10	< 2	< 1-5
scrubber sludge	1	4	-

*Ratio of escape to production rates.

Other sources of wastes from the coal fuel cycle, in particular coal mining refuse and coal cleaning residues, have been reported to result in contamination of streams and rivers and may constitute a more significant potential problem area. (1)

Discussion

The preceding discussion illustrates that if one uses realistic, yet conservative, estimates of activities in coal and fly ash, power plant release rates, atmospheric diffusion and subsequent deposition, one concludes that the combustion of fossil fuels, even in the less controlled "older" plants that release waste products from short stacks, will not result in significant perturbations on natural environmental radiation levels or significant increases in doses to any individual. One can also see, however, that by compounding worst cases or unrealistic assumptions, one arrives at fairly large estimates of dose. Unfortunately,numerous recent publications containing such estimates has resulted in unnecessary public and press concern regarding this topic. This is regrettable since it tends to divert attention from the other potential health hazards of coal combustion, some of which are likely to be far more consequential.

Another controversial, although not particularly relevant, question has been the relative radiological impact of coal versus nuclear power. We examined this question in Reference 1 and concluded that, when the entire fuel cycles are considered, the radiological impact of coal is far smaller than that of nuclear. Most noteworthy, however, is that routine releases from both cycles produce negligible increases in radiation exposure compared to background or other sources of exposure. The public has been confused with widely differing published comparisons. Some of these simplistically compared the total activities (in curies) released, a very misleading exercise, since, obviously, different radionuclides with different half-lives, radiations, radiation energies, ecological transport properties, dose pathways to man, and biological effectiveness are involved. Others compared the effects of emissions from power plants only, ignoring the fact that most of the routine radiological impact of nuclear power derives from other parts of the fuel cycle.

Although some further research on the long term fate of certain waste products might be beneficial, we feel that it is clear that the radioactivity released by the combustion of coal in modern plants meeting EPA's particulate emission standards is not a matter of concern.

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