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PRELIMINARY ASSESSMENT OF THE IMPACT OF RADIONUCLIDES IN WESTERN COAL ON HEALTH AND ENVIRONMENT*

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4) assess the possible need for additional control technology or standards. This report will summarize results of initial studies at the power plant.

MATERIALS AND METHODS

Samples of coal, ash, stack effluents, airborne particulates, soil, and vegetation associated with a power plant using Western coal were prepared and analyzed by alpha pulse height spectroscopy. The data were then used in an effort to calculate the radionuclide balance for the power plant, to model radiation dose to the population adjacent to the power plant, and to estimate radiation doses for power plant workers. Details of sampling and analytical procedures are presented elsewhere [8].

INTRODUCTION

The goal of the U.S. government to reduce dependency on imported fossil fuels, as well as other factors, is moving the country toward an economy based on coal as the primary fossil fuel [1]. Use of coal as a fuel is expected to triple between 1975 and the year 2000, from approximately 10 quads (1 quad = 10¹⁵ Btu) to 30 quads [2]. It seems almost certain that the U.S. will turn increasingly to the vast coal deposits of the Western states, since Western coal reserves (198 billion tons) represent 72% of the identified U.S. coal resources and this coal has a low sulfur concentration. Furthermore, 50% of the coal minable at current prices is in the West [3].

RESULTS AND DISCUSSION

The George Neal Steam Electric Generating Station (Figure 1), located on the east bank of the Missouri River approximately 17.7 km south of Sioux City, Iowa, began generating electricity in 1964. At present there are three coal-fired units at the station with generating capabilities of 150, 330, and 540 MWe, respectively. A fourth coal-fired unit is under construction, and it will have a generating capacity of 640 MWe. Unit #4 should become operational in 1978 [9]. The Neal Station used coal from the Hanna #5 mine in Wyoming, and coal at the power plant contained concentrations of radionuclides (uranium-234 averaged 0.51 pCi/g; uranium-238, 0.48 pCi/g; and polonium-210, 1.10 pCi/g) higher than the mean for 19 Western mines (uranium-234 averaged 0.31 pCi/g; uranium-238, 0.29 pCi/g; and polonium-210, 0.82 pCi/g) sampled during other aspects of this program. (For uranium-234, 1 pCi/g = 0.16 ppb; for uranium-238, 1 pCi/g = 3.0 ppm; for polonium-210, 1 pCi/g = 0.22 x 10⁻⁶ ppb.)

Governmental agencies, environmentalists, and scientists have become increasingly concerned over potential problems associated with large additions of trace elements in the environment from burning fossil fuels [4,5]. The process of coal combustion releases trace elements to the atmosphere as vapors and particles. These particles have relatively greater concentrations of certain trace elements than the feed coal [6] or the collected fly ash. In addition to the trace elements, coal contains uranium and its decay products. Early reports [2,3,7] suggested that uranium is found chiefly in low-rank and impure coal, including lignite, sub-bituminous coal, and carbonaceous shale of the northern Great Plains and Rocky Mountains regions, and this caused the U.S. Environmental Protection Agency (EPA) and Department of Energy (DoE, formerly the Energy Research and Development Administration and the Federal Energy Administration) to jointly initiate a project with Monsanto Research Corporation at Mound Facility (formerly Mound Laboratory) to assess the radiological impact associated with utilization of Western coal.

Bottom ash and fly ash contained higher concentrations of all radionuclides than did the feed coal. Samples of bottom ash contained uranium-234 and uranium-238 ranging from 2.33 to 2.74 pCi/g, polonium-210 ranging from 4.98 to 5.11 pCi/g, lead-210 ranging from 2.31 to 5.71 pCi/g, and thorium-230 ranging from 2.65 to 3.95 pCi/g. Fly ash contained uranium-234 and uranium-238 ranging from 4.04 to 4.68 pCi/g, polonium-210 ranging from 5.25 to 5.37 pCi/g, lead-210 ranging from 4.68 to 5.76 pCi/g, and thorium-230 ranging from 3.66 to 4.24 pCi/g. With an ash content of 11.91%, one would expect an increase in concentration of nonvolatile elements in ash over feed coal by a factor of 8.4. The actual ratios of radionuclide concentrations in bottom ash to feed coal are 6.7 for uranium-234, 6.3 for uranium-238, 6.3 for polonium-210, 4.5 for lead-210, and 6.1 for thorium-230. Similar ratios for the fly ash are 10.9 for uranium-234, 10.6 for uranium-238, 6.6 for polonium-210, 6.4 for lead-210, and 7.3 for thorium-230. Ratios less than 8.4 for bottom ash suggest that some of the radionuclides are preferentially entrained in stack effluents.

It is now apparent that Western coal being mined today does not contain significantly greater concentrations of radionuclides than Eastern coal and that Western coal does not have a unique geochemical affinity for radionuclides. The first phase of the project was designed to 1) delineate the scope of the potential environmental and human health problem associated with radioactivity in Western coal; 2) establish a data base on the radionuclides found in Western coal; 3) study the release, fate, and accumulation of radionuclides from a power plant burning Western coal; and

Effluents at the input to the electrostatic precipitator contained uranium-234 averaging 9.99 pCi/m³, uranium-238 averaging 9.15 pCi/m³, polonium-210 averaging 9.95 pCi/m³, lead-210 averaging 13.2 pCi/m³, and radon-222 averaging 560 pCi/m³. Effluents at the output of the electrostatic precipitator contained uranium-234 averaging 7.48 pCi/m³, uranium-238 averaging 7.56 pCi/m³, polonium-210 averaging

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9.17 pCi/m³, lead-210 averaging 14.6 pCi/m³, and radon-222 averaging 290 pCi/m³. These limited data indicate quantities of radon-222, uranium-234 and uranium-238 are reduced by the electrostatic precipitator, while polonium-210 and lead-210 are not affected significantly. Data on efficiency of electrostatic precipitators in reducing particulate emissions suggest the need for additional stack sampling.

The radionuclide concentration of airborne particulates collected at the five air-sampling stations around the power plant averaged at each station from 28.6 to 48.8 aCi/m³ of uranium-234, 36.8 to 54.2 aCi/m³ of uranium-238, 983 to 1300 aCi/m³ of polonium-210, and 5410 to 24200 aCi/m³ of lead-210. No trend could be detected from the levels of uranium-234 and uranium-238 found at the five air-sampling locations. It appears that two weeks of sampling does not yield a true representation of airborne particulates for determining trends in atmospheric dispersion. Values for polonium-210 concentrations in airborne particulates at air-sampling station #1 are equivalent to values reported by NCRP Report No. 45, Natural Background Radiation in the U.S. [10], and differences in polonium-210 concentrations of airborne particulates at stations #1-5 were not statistically significant.

Radionuclide concentrations in soil samples were not significantly above background levels reported by NCRP Report No. 45, Natural Background Radiation in the U.S. [10]. Concentrations of uranium-234 and uranium-238 ranged from 0.5 pCi/g at monitoring site #4 to 1.05 pCi/g at a cornfield 96 km south of the power plant; polonium-210 ranged from 1.09 pCi/g for the lower strata of soil at monitoring site #2 to 2.93 pCi/g for the upper strata of soil at the same site. Concentrations of lead-210 ranged from 0.70 pCi/g at monitoring site #3 to 3.15 pCi/g in the upper strata of soil at monitoring site #2. Concentrations of thorium-230 ranged from 0.58 pCi/g at site #3 to 1.94 pCi/g in the lower strata of soil at monitoring site #4.

There was no apparent pattern in distribution of uranium-234, uranium-238, and thorium-230 relative to the power plant. If the stack plumes were dispersing radionuclides to the environment, one or more sampling sites should have evinced consistently high concentrations of all radionuclides present in stack effluents in significant quantities.

Further evidence on the possible enrichment of soil in radionuclides from the power plant may be derived by comparing concentrations of radionuclides in upper (0 to 5 cm) vs lower (5 to 30 cm) soil strata. Presumably, the upper stratum of undisturbed soil would have a higher concentration of radionuclides than the lower stratum if deposition were the predominating mechanism. Tilled soils would not show a significant difference in concentrations of radionuclides in upper vs lower soil strata. Ratios of measured concentrations of radionuclides in upper vs lower soil strata (Table 1) indicate no significant difference (ratio \approx 1) for uranium-234, uranium-238, and thorium-230.

Ratios for lead-210 and polonium-210 are significantly different [$P < 0.05$ (probability of no significant difference is $< 5\%$)] from unity, as would be expected from characteristics of the radon-

222 decay scheme [11]. The radon-222 diffuses upward through soil to the atmosphere and decays to lead-210 which deposits on soil. Subsequently, lead-210 decays to polonium-210 (via bismuth-210), the lead-210:polonium-210 ratio depending on the length of time lead-210 and its matrix coexist and whether the polonium-210 is selectively removed by geochemical mechanisms. Ratios of measured concentrations of lead-210 and polonium-210 in soil are not significantly different from unity except at site #3. It is suggested that agricultural activities at this site could have sufficiently modified the natural state of this site to account for the radionuclide imbalance. The remaining sites have apparently remained undisturbed long enough for establishment of a lead-210/polonium-210 equilibrium.

The ratios of concentrations of lead-210 in surface vs subsurface soil strata and similar ratios for polonium-210 appear to decline with increased distance from the power plant. However, a regression analysis of these ratios on distance from the power plant indicated that there is no statistically significant decline in concentrations of lead-210 or polonium-210 in upper soil strata with increasing distance from the plant; and it may be concluded that the power plant is not the source of these radionuclides.

Radionuclide concentrations in samples of vegetation ranged from 0.0001 to 0.045 pCi/g of uranium-234, from 0.0007 to 0.020 pCi/g of uranium-238, from 0.025 to 0.64 pCi/g of polonium-210, and from 0.023 to 5.70 pCi/g of lead-210. Concentrations of uranium-234, uranium-238, and polonium-210 in vegetation are considerably lower than in the soil, suggesting that these isotopes are either geochemically unavailable to the vegetation or that the plants discriminate against them. On the other hand, lead-210 was present in or on vegetation at concentrations frequently exceeding concentrations in soil. The elevated concentration of lead-210 on vegetation is most likely to have occurred as a result of peculiarities of the radon-222/lead-210/bismuth-210/polonium-210 decay scheme, previously described for radionuclides in soil. Upon the decay of atmospheric radon-222 to lead-210, the lead-210 tends to return to the surface of the earth where broadleaved plants which present convenient surfaces for deposition are enriched in lead-210 [11]. Furthermore, the lead-210:polonium-210 ratio will vary with the length of time lead-210 has been deposited, equilibration requiring about one year. The ratios of concentrations of lead-210 and polonium-210 measured in grass and corn are significantly greater ($P < 0.05$) than unity, as would be expected since their leaf surfaces were not available for deposition long enough for ingrowth of polonium-210. To further test this point (deposition from air vs uptake from soil), shucks were removed and the corn cob and kernels were analyzed for lead-210 and polonium-210. Concentrations of both isotopes in cob and kernels were lower than levels in leaves and stalk by two orders of magnitude, and the isotopes were nearly in secular equilibrium. Concentrations are also lower than those in soil. The elevated concentrations of lead-210 observed for broadleaved grass (*Bromus* sp.) and corn plants may thus be explained by deposition rather than uptake from soil.

A central, or point, source for the lead-210 or its parent radionuclides is not suggested by the distribution of elevated concentrations in vegetation. The highest concentrations of lead-210 and its daughter polonium-210 were found at the park 96 km from the power plant. The second highest concentration of lead-210 occurred 3.6 km from the plant at site #5; the second highest concentration of polonium-210 occurred at site #2, closest to the power plant. The absence of a clear trend of declining concentrations of lead-210 and polonium-210 in vegetation with increasing distance from the power plant strongly suggests that the Neal Station is not the sole source of these radionuclides. More extensive sampling will be required to determine whether these radionuclides are emanating from other than the locally derived, naturally occurring parents.

Atmospheric diffusion calculations were made of concentrations of radionuclides in air, of ground deposition, and of inhalation doses around the Neal Station. The isotopes uranium-234, uranium-238, polonium-210, lead-210, and radon-222 were investigated. The emission of these isotopes from the power plant stack did not produce significant doses or ground depositions. This is most likely the result of the utilization of electrostatic precipitators on the stacks, since the precipitators removed over 70% of the radionuclides entering the stack. Detailed input and output for the atmospheric diffusion model are presented elsewhere [8].

The calculated air concentrations for uranium-234 and uranium-238 are higher than the measured values, but are within an order of magnitude. This is considered satisfactory because of the short duration of sampling involved and because of the approximations used in calculations. Background concentrations of uranium in air have been reported [10] ranging from 120 aCi/m³ at Argonne, Illinois, to 400 aCi/m³ in New York state. The air concentrations of uranium around the Neal Station are, therefore, not significantly higher than background and the calculated release from the stack is on the order of magnitude of background.

The calculated and measured values of polonium-210 and lead-210 differ markedly. Reported [10] background levels of polonium-210 are on the order of 1000 aCi/m³, and those for lead-210 may vary between 8000 and 26000 aCi/m³. The measured values of polonium-210 are obtainable from a summation of the background and the calculated power plant emissions. The measured lead-210 values are on the order of magnitude of background, while the calculated emissions from the power plant are about 1% of the background.

The calculated radon-222 air concentration values produced by the power plant do not appear to be significant. Background levels are reported [10] to vary between 50 and 1040 pCi/m³ with an average of about 150 pCi/m³ (150000 aCi/m³), so that the power plant emission levels are on the order of 10% of background.

The calculated and measured ground depositions are shown in Table 2. The conversions from pCi/m² to pCi/g were obtained by assuming that the plowlayer areal density of soil is 224 kg/m² [12]. The depositions shown are dry depositions and do

not include the effects of washout produced by rainfall. The deposition velocity was assumed to be 0.01 m/sec [12]. The calculated depositions are for one year of power plant operation. The maximum calculated accumulated depositions over a 20-yr period are also shown in Table 2.

The calculated depositions from the plume of the power plant are significantly below the measured values, indicating once again that the plume has very little effect on the ground activity. Measurements of activity taken 96 km south of the power plant have higher values than corresponding measurements taken within the vicinity of the power plant. The measured values for uranium agree fairly well with the natural background level of uranium in soil [10] of 0.6 pCi/g.

The maximum calculated inhalation dose for each isotope evaluated in the power plant plume is about 0.2 mrem/yr produced by lead-210 in bone. Maximum doses occurred 4.5 km southeast of the power plant, with slightly lower doses 1.5 km north of the plant. The maximum calculated ingestion dose for lead-210 from the power plant plume (including direct deposition and ingrowth from radon-222) on vegetation was 0.24 mrem/yr to the kidney. Both the inhalation and ingestion pathways are of no dosimetric significance in comparison with natural background radiation.

Potential inhalation doses to power plant workers were evaluated by collecting samples of airborne particulates inside the Neal Station and extrapolating doses from quantities of airborne particulates. In 30.5 hr a total of 1.220 g of fly ash was deposited on a Microsorb filter from 4974 m³ air, indicating 2.452 x 10⁻⁴ g/m³. Using previously reported values for radionuclide concentrations in fly ash, it is possible to estimate concentrations of radionuclides in air. The highest doses to workers based on exposures of 8 hr/day are 0.12 mrem/yr to bone from uranium-234, 0.12 mrem/yr to kidney from lead-210, and 0.10 mrem/yr to bone from uranium-238. These doses are not considered significant in relation to natural background levels and in relation to current standards of exposure for the general public.

CONCLUSIONS

The greatest fraction of uranium-234 (87%), uranium-238 (87%), polonium-210 (87%), and lead-210 (82%) in feed coal remained with the ash after combustion. Electrostatic precipitators on the stack of Unit #2 of the Neal Station removed over 70% of the radionuclides entering the stack in association with fly ash, and thus the precipitators appear to be of value in controlling radionuclide emissions. Since the precipitators were operating at below normal efficiency during the sampling period, an even greater reduction in radionuclide emissions should be routinely achieved. Other particulate emission control devices, e.g., bag houses, should also be very effective in removing radionuclides that enter the stack in association with fly ash.

Atmospheric diffusion modeling of stack effluents for uranium-234, uranium-238, lead-210, polonium-210, and radon-222 identified no radionuclides of dosimetric significance for inhalation or ingestion pathways. Occupational exposures of

workers inside the Neal Station are also of no dosimetric significance. Furthermore, specific trends in distribution of radionuclides measured in the environment (airborne particulates, vegetation, and soil) relative to the power plant were not apparent.

Use of Western coal being mined today in a modern power plant with effective emission controls (ESP) has led to no significant environmental contamination by radionuclides.

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Table 1 - RATIOS OF RADIONUCLIDE CONCENTRATIONS
IN 0 TO 5-cm AND 5 TO 30-cm SOIL STRATA

| Sample Location ^a | U-234 | U-238 | Po-210 | Pb-210 | Th-230 |
|--------------------------------------|-------|-------|--------|--------|--------|
| Site #2 | 1.11 | 0.94 | 2.63 | 2.41 | b |
| Site #4 | 0.92 | 1.15 | 1.88 | b | 0.49 |
| Park, 40 km south of Neal Station | 1.02 | 0.92 | 1.72 | 1.44 | 1.09 |
| Park, 96 km south of Neal Station | 1.08 | 1.08 | 1.49 | b | 0.87 |

^aRatios not appropriate for Sites 1, 3, and 5, since these sites are tilled fields.

^bNo data.

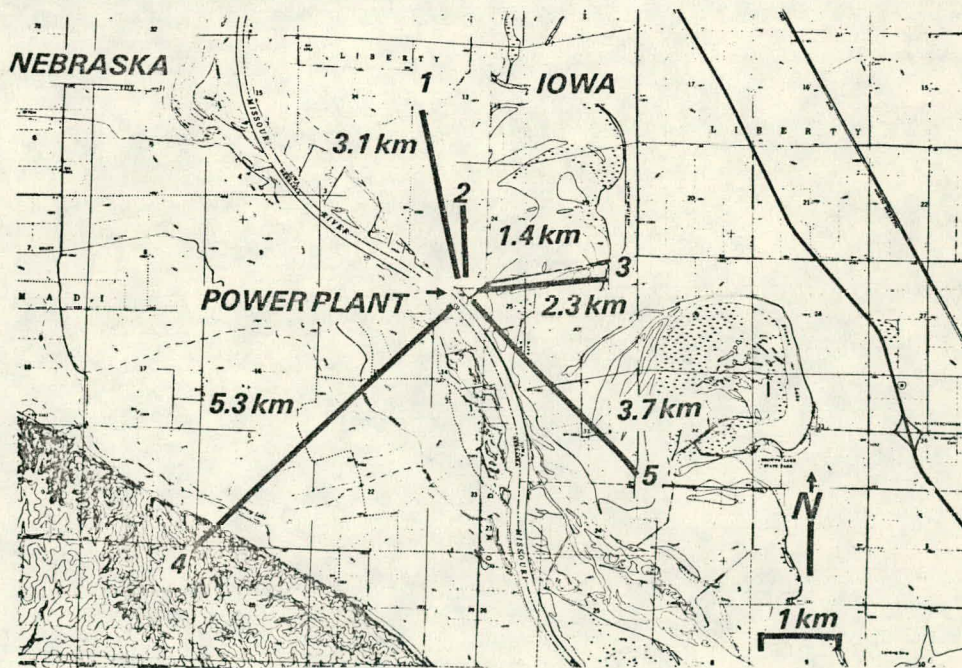


Table 2 - CALCULATED SOIL DEPOSITIONS FROM THE POWER PLANT PLUME
AND MEASURED RADIOACTIVITY OF THE SOIL

| Sampling Station | Location from Power Plant | U-234 | U-238 | Po-210 | Pb-210 | Rn-222 |
|--|---------------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|
| A. Calculated Soil Deposition (pCi/m ² /yr) | | | | | | |
| Site #4 | 5.3 km, SW | 18.7 | 18.7 | 10.5 | 35.7 | 19.3 |
| Site #3 | 2.2 km, E | 26.0 | 26.0 | 14.6 | 49.5 | 26.7 |
| Site #5 | 3.6 km, SE | 64.6 | 64.7 | 36.4 | 123.1 | 66.5 |
| Maximum calculated value | 4.5 km, SE | 68.4 | 68.5 | 38.5 | 130.3 | 70.3 |
| B. Calculated Soil Deposition (pCi/g/yr) ^a | | | | | | |
| Site #4 | | 8.37×10^{-5} | 8.38×10^{-5} | 4.71×10^{-5} | 1.59×10^{-4} | 8.62×10^{-5} |
| Site #3 | | 1.16×10^{-4} | 1.16×10^{-4} | 6.54×10^{-5} | 2.21×10^{-4} | 1.19×10^{-4} |
| Site #5 | | 2.88×10^{-4} | 2.89×10^{-4} | 1.63×10^{-4} | 5.50×10^{-4} | 2.97×10^{-4} |
| Maximum calculated value | | 3.05×10^{-4} | 3.06×10^{-4} | 1.72×10^{-4} | 5.82×10^{-4} | 3.14×10^{-4} |
| C. Calculated Maximum Deposition (pCi/g) over 20-yr Operation ^b | | | | | | |
| | | 6.10×10^{-3} | 6.12×10^{-3} | 2.05×10^{-4} | 1.05×10^{-3} | 3.14×10^{-4} |
| D. Measured Values (pCi/g) | | | | | | |
| Site #4 | | 0.50 | 0.52 | 2.63 | | |
| Site #3 | | 0.67 | 0.86 | 1.85 | 0.70 | |
| Site #5 | | 0.70 | 0.75 | 1.73 | | |
| Park | 96 km, S | 0.86 | 0.81 | 1.95 | 2.04 | |

^aValues obtained by dividing pCi/m²/yr by the areal density of soil, 2.24×10^5 g/m².

^bValues obtained by multiplying maximum values by the factor $(1 - e^{-20\lambda})(1 - e^{-\lambda})$, where λ is the physical decay constant of the isotope in inverse years.