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Proceedings of the Workshop on Radioactivity Associated with Coal Use

*Held in Santa Fe, New Mexico
September 15—17, 1981*

Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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Proceedings of the Workshop on Radioactivity Associated with Coal Use

Held in Santa Fe, New Mexico

September 15—17, 1981

Organized by

**Paul Wagner
N. Roy Greiner**

Sponsored by

**The Los Alamos National Laboratory
Under the Auspices of the
US Department of Energy
Assistant Secretary Environmental Protection
Safety and Emergency Preparedness
Office of Environmental Programs**

Coordinated by

**Los Alamos Technical Associates, Inc.
1650 Trinity Drive
Los Alamos, New Mexico**

Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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ABSTRACT

A workshop on radioactivity in coal use was held on September 15 through 17, 1981, under the auspices of the US Department of Energy, Office of Environmental Programs, and the Los Alamos National Laboratory. The purpose of the workshop was to identify research issues associated with radioactivity resulting from the use of coal for electric power generation.

The consensus of the 10 scientists participating in the workshop was that a moderate to strong need exists for research in solubility of fly ash in different fluids and for determination of radioactivity in construction materials. Several additional research issues were identified but were given a lower priority.

1.0 EXECUTIVE SUMMARY

The projected substantial increase in the use of coal to meet the nation's energy needs prompted concern about potential environmental and health effects. One of the concerns was the potential radioactivity emissions/releases and resultant adverse effects associated with increased coal use.

This workshop was organized to allow scientists studying radioactivity associated with coal use to present the findings of their research efforts and to allow for broad discussions of the relevant issues. Also, workshop participants were to develop a consensus document addressing the state of available data on radioactivity issues related to coal use and to identify those issues requiring the development of additional data.

Each of the participants was actively involved in research related to the potential effects of radioactivity emissions from increased coal use. During the workshop, technical papers were presented; in-depth discussions and dialogues were held; and priority issues/subissues were identified, discussed, and ranked in order of priority.

The order of priority was set through the use of a modified Delphi process aimed at achieving a consensus. The issues were ranked according to group determination of high need or low need for additional research data. The list of identified issues and the priority mean scores are presented in Table 1. These issues are thus rank-ordered according to the need for additional data.

Additionally, workshop participants felt that potential adverse environmental and health effects should be considered in ranking additional data collection needs. In this regard, it was the group consensus that (1) long- and short-term solubility of released fly ash in different fluids, (2) radionuclides in construction materials, and (3) stack releases were the most important.

TABLE I
 PRIORITIZATION ACCORDING TO RESEARCH NEEDS^a

	<u>Mean</u>
1. Long- and short-term solubility of released fly ash in different fluids	7.3
2. Radionuclides in construction materials	7.1
3. Stack releases	5.9
4. Activity and emission of waste at landfills and redistribution into populated areas	4.3
5. Long-term changes of leachability of coal and waste	4.1
6. Airborne radionuclide levels and external radiation levels inside power plants	3.4
7. Ground-level air concentrations	3.1
8. Radionuclides in waste products	2.9
9. Deposition from stack releases	2.9
10. Radionuclides in coal	1.6

^aScoring Key

- 10 - Critical importance
- 8 - Major importance
- 5 - Moderate importance
- 2 - Minor importance
- 0 - No importance.

2.0 INTRODUCTION

The increasing preference for coal over other fossil fuels for electric power generation has spurred investigations into the environmental and health effects of coal-fired power plants. This workshop was organized to bring together scientists studying radioactivity issues related to coal and to produce a consensus document based on discussion of these issues.

Formal papers were presented on (1) effects of coal combustion on the radiation environment, (2) low-level radiation in the coal used and ashes produced, (3) problems of radioactivity from coal use, (4) chemistry of radionuclides in coal preparation, (5) uranium daughters in natural atmospheric aerosols and coal-fired power plant emissions, and (6) the impact on water quality from radionuclides. Following the presentations, group discussions relative to the current state of knowledge produced a list of issues that in the opinion of the attendees needed continued and/or additional research. The order of priority for issues was determined by a modified Delphi process (Appendix). The criterion for ranking these issues is sufficiency or insufficiency of available data from experiments and monitoring. The issues having the greatest need for additional data were ranked highest. Through the ranking process, subissues were added under some of the radioactivity issues.

Note that, although an issue may require additional data and hence be ranked high, the potential for radiological impact from coal use on the environment and public health and safety must determine the final decision on future allocation of resources.

AGENDA

Monday, September 14

7:00-9:00 p.m. - Social gathering, "The Portal," Inn at Loretto

Tuesday, September 15

9:00-11:00 a.m. - Room A

<u>Speaker</u>	<u>Subject</u>
George E. Dials*	Introductory Remarks
Kevin M. Miller	Some Effects of Coal Combustion on the Radiation Environment
David T. Abbott and Clarence E. Styron	Radionuclides in Western Coal Efforts at Mound
Carol Hornibrook	Low-Level Radiation in Coals Utilized and Ashes Produced at New York State Electric Utilities
N. Roy Greiner	Radioactivity from Coal Use - Where Are the Problems?

11:00 a.m.-2:00 p.m. - Break for Lunch/Informal Discussions

2:00-4:00 p.m. - Room A

<u>Speaker</u>	<u>Subject</u>
John N. Ondov	Chemistry of Radionuclides in Coal Preparation
Lawrence E. Wangen	Uranium Daughters in Natural Atmospheric Aerosols and Coal-Fired Power Plant Emissions
C. Richard Cothorn	Possible Contributions of Coal Extraction and Utilization to Radioactivity Concentrations in Drinking Water
Paul Wagner	Impact on Water Quality from Radionuclides in Coal

*Los Alamos Technical Associates, Inc., coordinator.

AGENDA

Discussion - Establish Final List of Issues to be Addressed

7:30 p.m. - Dinner at the Steaksmith

Wednesday, September 16, 1981

8:30 a.m.-12 noon - Room A

Discussion and Preparation of Consensus Document

12 noon-1:00 p.m. - Sandwich and Salad Luncheon on the Patio

1:00-5:30 p.m. - Room A

Discussion and Preparation of Consensus Document

Thursday, September 17

9:00 a.m.-12 noon - Room A

Presentation of Consensus Document

1:00-3:00 p.m. - Room A

Final Discussion and Comments

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3.0 ISSUES AND DISCUSSION

3.1 Issue 1. Long- and Short-Term Solubility of Released Fly Ash In Different Fluids

Although leaching experiments of fly ash in water have revealed that in general radionuclides are not released in concentrations of environmental concern, all past work in this area has been performed in pure or acidified water solutions for times that are short in comparison with estimated residence times for the deposited fly ash. Very conservative estimates have been used in modeling data for inhalation and ingestion pathways. Data are needed for leaching over an extended time using pure water and body fluids to ensure that radioactive emissions over time will not create an environmental or health concern.

As an illustration of the current state of our knowledge in this area, models of vapor deposition and experimental evidence suggest that large portions of lead and uranium reside on the surface or nonmatrix portion of fly ash particles emitted from stacks. Further, the chemistry and physical properties of the stack-emitted aerosol are distinctly different from the bulk fly ash collected in control devices as the latter are dominated by the much larger particles. For example, electrostatic precipitator (ESP) hopper fly ash can contain up to 10% carbon, whereas stack fly ash typically contains only 0.1 or 0.2% carbon. The sizes of particles emitted from the stack are typically less than 10 or 20 μm , are predominantly submicrometer, and have high specific surfaces. Hopper fly ash contains particles ranging in size from the submicrometer region to a few hundred micrometers or larger. The solubility of stack-emitted fly ash has not been routinely measured as only very small quantities of ash are collected in most stack tests. Only a single group of collaborative researchers has collected sufficient quantities of stack fly ash and examined solubility properties in strong mineral acids. This material was collected at a single plant. Additional studies should be made of solubility of particles emitted from similar plants and those equipped with flue gas desulfurization (FGD), hot-side ESPs, etc.

3.2 Issue 2. Radionuclides in Construction Materials

With the expectation of greatly increased numbers of coal-fired generating plants, the problem of increased combustion waste has led to use of these by-products as a binder in building materials. Available data on radionuclide concentrations in ash raise questions concerning the levels of radionuclide exposure to workers and building inhabitants. Modern buildings are sealed against air leakage as a conservation measure, which increases the buildup and concentration of indoor air pollutants. This concentration could be sharply increased if materials containing ash had high radionuclide content.

3.2.1 Subissue 2a Exposure of Factory Workers Producing Blocks/Bricks

Dust resulting from the use of fly ash in concrete block- and brick-making plants may represent a radiologic as well as toxic health hazard to workers. Because there are no data available on the subject of radionuclide solubility of respirable fly ash particles, the data to be collected for Issue 1 as discussed in Subsection 3.1 are essential to the evaluation of worker health.

3.2.2 Subissue 2b. Gamma Radiation

The gamma radiation levels inside buildings are a potential health hazard and are determined to a large extent by the radionuclide content of the materials used in construction. The external exposure rate that results from the use of building materials containing waste products from coal combustion could in principle be measured directly or inferred from the analysis of radionuclide concentration of samples. Because a significant fraction of the exposure rate can be produced by the presence of lead-214 and bismuth-214, which are decay products of radon-222 in the uranium-238 series, the overall effect is coupled with the question of radon emanation.

3.2.3 Subissue 2c. Radon-222 Emission With Time

The demands of energy conservation call for tighter buildings to reduce the influx of outdoor air. Therefore, there is less fresh air to dilute any sources of radon-222 inside the building. Under these circumstances, it is

important to assess the effect of building materials that strongly emit radon-222.

Although the emanating power of raw fly ash or bottom ash is rather small (typically less than a few percent), the long-term behavior of these materials incorporated into building products is still unknown. Conceivably, the ash matrix could be degraded in the concrete curing process to cause increased release of radon gas from the final product. Effects of this sort could be measured experimentally with available apparatus and techniques. Because the concentration of radium-226 (the parent of radon-222) in coal ash is 5 to 10 times the likely concentration in the ingredients it will replace, it seems prudent to make measurements of radon-222 emission throughout the curing and aging process to verify the acceptability of the final product.

3.2.4 Subissue 2d. Activities in Ash Composite Materials as Compared to Conventional Construction Materials

Because the incremental radioactivity resulting from the use of ash in composite materials must be assessed to understand the nature and magnitude of potential environmental impact, data on radionuclide content of construction materials made of waste from coal-powered plants and the radionuclide content of conventional construction materials (cement block, wallboard, etc.) should be collected and compared.

3.3 Issue 3. Stack Releases

Stack releases are those particles that are suspended on fly ash and those gaseous emissions that disperse beyond the limits of the plant site. Data on concentrations of the various gas components and polonium-210 emissions have never been gathered.

3.3.1 Subissue 3a. Release Rate of Polonium

No reliable data on polonium-210 in emissions from coal-fired power plants exist. From the limited data available, it is uncertain whether or not we are actually measuring polonium-210 or lead-210 that decayed to polonium-210 after

combustion. Even these data suggest that 30 to 60% of polonium-210 may be emitted out the stack. However, given the uncertainty of these data, a worst case estimate of 100% polonium-210 emission results in a calculated annual average increase of $\cong 60\%$ in polonium-210 concentration at the location of maximum impact from a large coal-fired power plant. This gives a 6% increase from $\cong 100$ to 106 mrem/yr lung dose. If only 10% of the polonium-210 escapes, this dose is reduced to 0.6%. Thus, better data are needed on polonium-210 releases for more precise dose estimates.

3.3.2 Subissue 3b. Gaseous Emissions of Other Radionuclides

Estimates of ground-level concentrations of uranium-238, uranium-234, lead-210, and lead-212 have assumed that these radionuclides are totally associated with particles in coal combustion off-gases treated by emission control devices. Thermodynamic data suggest that this assumption is valid for plants using control devices located downstream from the intake air preheater (for example, cold-side ESPs). This is the case for the majority of utility coal boilers. However, in some western plants using low sulfur coal, ash resistivity limits the efficiency of fly ash collection at the lower preheater exhaust temperatures. These plants sometimes install hot-side ESPs (for example, the ESP is located upstream from the air preheater) to enhance charge carrier mobilization and fly ash collection. In hot-side ESPs, the gas temperature is typically 300°C greater than in cold-side ESPs. At these temperatures, the vapor pressures of uranium and lead compounds can be several orders of magnitude greater than their vapor pressures at flue gas temperatures (110 to 140°C). Depending on the amounts of lead and uranium present in coal, concentrations of lead and uranium in the vapor might be as large as 1 or 2% of the amount originally in the coal.

The vapor components of the radionuclides of lead and uranium in gases at 400°C have never been measured. Flue gas vapor phase levels of only 1 or 2% would easily produce emissions 10 times greater than the emission of lead and uranium associated exclusively with particles.

Estimates should not be based on stack tests of plants with cold-side ESPs. Few data have been reported on the composition of fly ash particles

emitted from plants with hot-side ESPs. Therefore, additional study may be warranted if accurate emission estimates of lead and uranium radionuclides for plants with hot-side ESPs are needed.

3.4 Issue 4. Activity and Emission of Waste at Landfills and Redistribution into Populated Areas

Mobilization, and resuspension, of radionuclides in waste and the use of released sites are areas needing further study to determine the long-term effects of radionuclides in the soil.

3.4.1 Subissue 4a. Mobilization of Radionuclides

The major concern associated with use of waste for landfills and similar type disposals is movement of the radionuclides out of the disposal site to regions where they may be accumulated. This may pose a potential radiation threat to human health and the environment.

At this time, no data exist for radionuclide movement by wind, water movement, plant or animal uptake, or allied paths. While this does not appear to present a severe impact at this time, the expected increase in coal use mandates an effort to fill the information void in this area.

3.4.2 Subissue 4b. Follow-Up Studies of Released Sites

States such as New York have strict regulations on the landfill disposal of fly ash, requiring that the ash be compacted, covered with 2 ft of soil (at the time of closure), and revegetated. Radon exhalation from aged landfills is an issue if aging increases the emanation powers of ash.

However, the regulations do not restrict the future use of these lands; the possibility exists for residential, recreational, or agricultural (for example, crop production or cattle grazing) use of the land. There is no information on the potential for radon accumulation in residences built on such areas, the potential for radionuclide exposure to transient populations, or the uptake of radionuclides by plants grown on soil above a landfill. This information would facilitate the successful reclamation of these lands in safe, productive ways.

3.4.3 Subissue 4c. Resuspension

The redistribution of ash from disposal sites by wind erosion may be significant and may contribute to the potential increase in downwind ground-level air concentrations of natural radionuclides along with direct stack emissions. Models for estimating resuspension exist but have not been applied to these sites nor tested against experimental data for these sites. Few such experimental data exist, and thus some additional field data may be required at such sites to determine the relative importance of this pathway.

3.4.4 Subissue 4d. Radiation Fields and Radon Release Rates as a Function of Time

The gamma radiation field may change as the site changes from active to inactive states, for example, covering with overburden, weathering, etc. The radon exhalation may also be modified by weathering, covering with soil, or drying out of waste ponds. Generally, the radon emissions from these sites are not currently an issue but could become one if the emanating power of the wastes increases with time. The gamma radiation fields might become an issue if the wastes were used for landfill or if the sites were reclaimed and used for housing sites.

3.5 Issue 5. Long-Term Changes of Leachability of Coal and Waste

There is a large quantity of data on the leachability, in both acid and alkaline mobilized aqueous systems, of trace elements and radionuclides from fossil fuels, mineral wastes associated with fuel extraction and processing, and combustion and utilization wastes. However, there are very little data on the changes in leachability of coals and coal combustion wastes as a function of time (that is, years).

The nature of minerals in the coals and chemical make-up of the various ashes could conceivably change upon weathering, (for example, freezing and thawing), oxidation, and hydrolysis over long periods of time, thus changing their leaching characteristics. Such morphological changes could lead to increased release of potentially hazardous radionuclides from these storage and disposal sites.

Because this is basically a question of chemical kinetics, traditional physical-chemical methods of representing time and temperature may be used to obtain preliminary information (over time periods of 1 to 2 yr) on changes to be expected in leaching behaviors over extended periods of time (centuries). This information is of special interest because increased use of coal will generate disposal problems of an unprecedented magnitude, and information on time-dependent release mechanisms should be made available to ensure the viability of adopted disposal options.

3.6 Issue 6. Airborne Radionuclide Levels and External Radiation Levels Inside Power Plants

Known available data of the rate of external radiation levels inside coal-generated power plants are limited. Data have probably been compiled by industry and regulatory agencies. A survey of these data and the related health effects should be taken.

To assess the radiologic impact on workers in a coal-fired power plant from inhalation of suspended coal dust in the work place, it is necessary to know the mass concentrations versus the size of airborne coal and ash dust.

It is also necessary to know the radionuclide content as a function of particle size. These latter data are available for ash but perhaps not for coal dust.

3.7 Issue 7. Ground-Level Air Concentrations

Ground-level concentrations of airborne radionuclides including polonium-210 need further study, particularly regarding site-specific atmospheric conditions and the increase of coal-fired power plants within a limited area.

If we have an understanding of stack releases, we can do a reasonable job of estimating ground-level concentrations by using dispersion modeling or other more empirical methods. If we know the ground-level concentrations, we can estimate deposition by using deposition velocity and precipitation scavenging efficiencies combined with appropriate meteorological data.

3.7.1 Subissue 7a. Polonium-210 if Released as Gas

As discussed in Subsections 3.3.1 and 3.3.2, polonium-210 may be emitted from coal-fired power plants in the gas phase. Although the polonium-210 activity is equal to that of its parent, the total polonium-210 inventory contains very few atoms in comparison to the numbers of atoms of more stable species. This small number of atoms may remain unattached to the particles in the plume, so that in environmental and lung deposition, polonium-210 would need to be treated as a gas. This could change the distribution of dose to the various organs.

3.7.2 Subissue 7b. Site-Specific Analyses

In previous ground-level air concentration analyses, consideration was given primarily to the issues and priorities important to promulgation of the national policy with regard to coal combustion for electric power. Therefore, results are directed toward evaluating average situations and applying to the general case. We recognize that a variation exists in factors affecting the release and downwind distribution of radionuclide emissions. For example, an atypical meteorological event could result in far less atmospheric dispersion than considered in previous studies. In general, questions regarding specific power plant sitings and assessments must be dealt with on a site-specific basis.

3.7.3 Subissue 7c. Regional Assessment of Radionuclides from Many Power Plant Operations

As the use of coal combustion for power generation increases nationally, there is a need for the development of a data base and long range transport models to predict changes in ground-level concentrations from many power plant operations. State air quality monitoring programs may need to incorporate radionuclide sampling in order to detect actual changes over time.

3.8 Issue 8. Radionuclides in Waste Products

Some waste products containing radionuclides may have a greater radon emanation than ash. Verification and understanding of this phenomenon would be desirable before waste is used in such items as building materials.

Possibly, a problem may exist with radon releases from building-type material made from FGD sludge. Although various investigations have considered coal ash, the area of FGD sludges has received much less consideration. Because FGD sludge is a major by-product of gas cleanup and a potential source of materials, namely, wall board, some additional work is needed.

3.9 Issue 9. Deposition from Stack Releases

As the plume from a power plant disperses, large and small particles are removed respectively by gravitational settling and by diffusion to surfaces on and near the ground. These processes reduce the ground-level air concentrations and control the amounts of radionuclides in soil and on plants.

3.9.1 Subissue 9a. Direct Deposition of Polonium-210

See Subsections 3.3.1, 3.3.2, and 3.7.1.

3.9.2 Subissue 9b. Effect of Intermodal Coagulation on Deposition and Settling of Radionuclides

By number, the principal mode of particle emissions from coal-fired power plants occurs at submicrometer diameters. The principal mass mode typically occurs at aerodynamic diameters between 1 and 15 μm . The populations of particles in the submicrometer and supermicrometer modes are typically on the order of 10^8 and 10^5 particles/ cm^3 of stack gas, respectively. Because the rate of particle coagulation of dissimilar size is greater than the rate for particles of the same size, intermodal coagulation is important. This effect leads to a net mass transfer of particles of the accumulation mode to the supermicron mode. In addition, the smaller particles contain greater concentrations of uranium and lead, so that intermodal coagulation can accelerate the mass transfer of these species to larger sizes. These larger particles settle much faster than the smaller particles, thus deposition rate of lead and uranium will be affected by intermodal coagulation.

3.9.3 Subissue 9c. Regional Assessment of Radionuclides from Many Power Plant Operations (Effects of Multiple Plants on Regional Scale)

This information has been addressed in Subsection 3.7.3.

3.10 Issue 10. Radionuclides in Coal

Sufficient information exists on the average radionuclide content of coal; however, continued studies of coal should be maintained to determine how coals with high uranium content are distributed in the coal-use cycle. Data from the US Geological Survey (USGS) show that 0.5% of coals presently used probably contain more than 10 times the uranium found in average coal (1.5 ppm uranium). If this type of coal were used for extended periods of time in one or two localities, a radiologic hazard to the public might possibly result. We have no evidence that this is indeed happening, and the likelihood of its happening is probably quite low because the likelihood depends on a combination of low-probability circumstances. (See Subissue 7b. on site-specific problems.)

4.0 STANDARDS ISSUES

The workshop was not specifically concerned with the identification of research and development needs or related data requirements for the development of standards. Nevertheless, several requirements/issues regarding standards development were identified in the course of the workshop. The most prominent ones are listed below.

1. Inhalation and Ingestion Transport Models - Development and/or Verification
2. Evaluation of Radionuclides from Coal-Fired Power Plant Waste Products and Pathways to Man
3. Emissions from Construction Materials and Pathways to Man (including the indoor air pollution issues)
4. Ingestion Transport Model from Coal and Power Plant Wastes Radionuclide Leachants
5. Migration Routes and Rates of Intrusions of Coal and Power Plant Wastes - Radionuclides into Ground Water and Drinking Water Supplies
6. Uptake of Coal-Derived Radionuclides in Plants, Animals, and Man

There is a gap between what is known and will be generated and the development of a standard. Perhaps the main need is to develop transport models that can estimate the amount and chemical form of a radioisotope that could potentially reach humans. The two paths of interest include both inhalation and ingestion. All of the various pathways from source to human must be examined and then relative contributions determined. These models need to be verified by field measurements.

Other information required for establishing standards includes the comparison of coal as a source of each radioisotope with other possible sources, the control technology methods or treatment needed to reduce the exposure to

humans, and determination of the solubility of radioisotopes in coal to develop the dosimetry to estimate exposure and risk.

5.0 CONCLUSIONS

Results of the issue ranking are presented in Table I. It is recognized that, in each of these issues and the previously discussed subissues, further studies or analyses could be performed; however, limited resources are available to effectively apply to all aspects of the research needs.

Thus, it is necessary to consider not only which outstanding issues require additional data, but also which issues are potentially most important with respect to assessing the impact of coal utilization on the population and on the environment. It was the consensus of the group that issues regarding long- and short-term solubility of released fly ash in different fluids, radionuclides in construction materials, and stack releases are also the most important in this second regard.

6.0 PRESENTATION SUMMARIES
(In Order of Presentation)

SOME EFFECTS OF COAL COMBUSTION ON THE RADIATION ENVIRONMENT

Kevin M. Miller and Harold L. Beck

US Department of Energy
Environmental Measurements Laboratory
New York, New York

Based on a review of the literature and our own sample analyses, we estimate mean concentrations in US coals of 1.7 $\mu\text{g/g}$ uranium, 4.5 $\mu\text{g/g}$ thorium, and 1,700 $\mu\text{g/g}$ potassium with only 5% of the samples exceeding these means by more than a factor of 3 (Refs. 1 and 2). Western coals are not, in general, more radioactive than eastern coals although small deposits of uraniferous coal do exist in some western states. In fact, typical western lignite coals are generally lower in radioactivity than eastern bituminous coals.

After combustion of the coal, the radionuclide concentration in the remaining ash would be expected to average a factor of 7.5 higher based on a mean ash content of 13.4% for US coals. Our own analysis of collected ash samples confirms this as well as the fact that some of the more volatile elements such as lead-210 are enriched on fly ash relative to bottom ash.

The calculated mean annual ground-level air concentration of radionuclides resulting from a 1000-MWe power plant burning an average coal varies from a fraction to a few times the mean ambient levels from resuspended soil depending upon the emission controls, stack height, and distance downwind. The deposition of fly ash on the surrounding land after 50 years would increase the natural background soil levels by a small fraction for a field plowed to a depth of 30 cm.

The dose to a maximum-exposed individual living near a power plant would result mainly from inhalation of the respirable fly ash particles. For a typical older plant releasing 10% of its fly ash from a 50-m stack, the dose equivalent to the lung would be less than 10 mR/yr. However, for a modern plant meeting EPA emission standards and releasing from a 150-m stack, the dose would be

less than 0.2 mR/yr. Although many investigators have suggested the ingestion pathway could result in doses of up to 20 mR/yr from ingestion of crops grown on surrounding land, we feel this is more unrealistic due to available evidence that indicates extremely low solubility of the radionuclides when bound in fly ash. This would result in minimal uptake by plants from the soil and, even in the case of direct foliar deposition and consumption without washing, minimal uptake by the body.

The release of radon-222 from ash piles and products does not appear to be a problem because measurements so far indicate a low emanating power (<2%). Also, the leaching of radionuclides into ground water from waste ponds does not appear to be a problem because of the low solubility.

Areas that need further investigation include the leaching of radionuclides during coal mining and cleaning operations and any possible long-term weatherization effects on the ash solubility and emanation power. Also the extent of ash use in building materials and its effect on gamma radiation levels should be investigated.

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RADIONUCLIDES IN WESTERN COAL EFFORTS AT MOUND

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Concern over radiation hazards of nuclear power and the increasing shift towards the preferential use of coal over other fossil fuels for electrical generation have spurred investigations into the environmental and health effects of coal-fired power plants. Mound Facility personnel have been assessing the environmental and health impacts of radionuclides in western coal.¹⁻³ The first step was to categorize the western coals presently used. Nineteen coal seams at 18 mines representing 65% of regional production were sampled. Contrary to expectation, these coals were generally low in radionuclide content, as exemplified by an average uranium-238 content of 0.3 ± 0.1 pCi/g. This is significantly less than the national average of 0.6 pCi/g.

Mound also participated in an interlaboratory comparison to evaluate the applicability of various radioassay techniques to coal. The results showed that the radon emanation technique is adequate to estimate radium content with the proviso that care is taken in the selection of standards. It was also evident the lead-210 analytical procedures need further development.

The possibility that radium is not in equilibrium with uranium-238 and thorium-228 in coal was of concern with regard to radioanalytical methodology. Mound performed a study on western coals and found that radium is in secular equilibrium with its parent nuclides in coal.

The next step in our investigation was an in-depth field study of a typical power plant burning western coal. It was found that most radionuclides remain with the ash fraction. With possible exceptions of lead-210, polonium-210, and radon-222, existing pollution control devices prevent the escape of significant

*Mound Facility is operated by Monsanto Research Corporation for the US Department of Energy under Contract No. DE-AC04-76-DP00053.

quantities of radioactive materials out the stack. Stack sampling and environmental sampling confirmed this conclusion. Therefore, it was decided that further efforts should concentrate on pathways from the ash ponds. Radon emanation studies showed that fly ash has an emanation power of less than 0.01, which is very small when compared to typical soil values of about 0.2. Leaching experiments also resulted in very low values; typically less than 1% of the radionuclide content of ash could be leached.

Ground-water and airborne gas modeling was performed to estimate radionuclide concentrations. Ground-water monitoring indicated that the leachates from ash ponds at the power plant were diluting concentrations of radionuclides in the background ground water. Radon gas emanation from the ash ponds was estimated at 0.09 pCi/m²/s, which is less than typical soil values of about 0.5 pCi/m²/s. Dispersion and dosimetry modeling indicated that the radiation equivalent dose from inhalation to the population surrounding the plant would be less than 1% of natural background.

Windblown ash resuspension, deposition, plant uptake, and human ingestion modeling showed that a dose equivalent maximum of 40 mrem/yr to bone tissue was possible. The maximum ingestion dose to other organs was less than 4 mrem/yr. A total body dose of 26.7 mrem/yr was predicted. Inhalation doses were less than 2 mrem/yr. Very conservative assumptions were made in these estimates, particularly with respect to plant uptake, an area that needs further investigation. Ongoing work has centered on flue gas desulfurization (FGD) at five power plants and fluidized bed combustion (FBC) at one demonstration boiler and their effects on potential radionuclide hazards associated with coal combustion.

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LOW-LEVEL RADIATION IN COALS UTILIZED AND ASHES
PRODUCED AT NEW YORK STATE ELECTRIC UTILITIES

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New York State is actively pursuing programs that would decrease the State's dependence on foreign oil. The increased use of coal at electrical generating and industrial facilities is being promoted by regulation, legislation, and research. At the present time approximately 800 000 tons of fly ash and 400 000 tons of bottom ash would be produced if eight power plants currently using oil were converted to coal. This would be a sizable increase in the amount of ash to be disposed of. If these facilities were required to use scrubbers, an almost equivalent amount of flue gas desulfurization sludge would be produced and require disposal.

Since 1979, the New York State Energy Research and Development Authority (ERDA) has been developing a coal-combustion waste program. This program aims at decreasing the quantities of ash produced while improving the potential for utilization and disposal of these wastes in ways that would most benefit the State while incurring minimal environmental impact. A major project within this program has been the Coal Waste Artificial Reef Project. This five-year study, which is presently completing its third year, has developed an engineering scheme for commercial production of blocks the size of typical concrete blocks without holes. A quarter-mile reef, 10 ft high and 20 ft wide, made of these coal waste blocks, is located 1 mile off Fire Island in the Atlantic Ocean.

The study of low-level radioactive contamination from coal consumption was undertaken by ERDA in this area of coal waste management as a first step towards gathering a data base for New York State.

The objectives of this study were to

- (1) collect representative samples of coals burned and ashes produced at New York State power plants and two blocks that are representative of those in the reef off Fire Island;
- (2) analyze those samples (dry and the fly ash leachates) for uranium-238, uranium-235, uranium-234, radium-226, radium-228, lead-210, thorium-230, thorium-232, and polonium-210; (Radon emanation rates were measured on dry samples only.)
- (3) determine whether or not these ash samples comply with federal and state standards; and
- (4) create a data base that would be the foundation upon which future programs could be built.

In order to achieve these goals, the study was arranged as follows.

Sampling Program

There are currently 10 coal-fired power plants in New York State. Eight of these were sampled for coal, fly ash, and bottom ash. Our sample collection strategy was to obtain a sample from the coal being burned and ash samples resulting from the combustion of that coal. Each sample was to represent the "average" characteristics of the coal or ash. Fly and bottom ash sample collection from the combustion of coal sampled proceeded with mixed success. Success depended on a knowledge of plant operations by our contact person. For example, a contact person would agree to spend the morning with the sampling crew to obtain the necessary samples from two stations. However, to get a sample of coal representing the ash collected, the contact person must know that a coal sample should have been collected during the night shift.

A second problem, thought to be more significant, was the impact of the 1981 coal strike on coal quality during our sampling program. However, the data comparing the sulfur, ash, and British thermal unit content of 1981 coals with those burned the previous year indicate that the coals sampled fall within the range of previously used coals.

Table I shows where each coal, fly ash, and bottom ash sample was collected at each of the eight cooperating facilities.

Two coal waste blocks representative of the specific types now being studied in the Atlantic Ocean off Long Island were brought in for analysis. One block was a mixture of 4:1 fly ash to scrubber sludge with 3% cement added. The other was 1:1 fly ash to scrubber sludge with 3% cement added.

Analytical Program

The radionuclides selected for this study were uranium-238, uranium-235, uranium-234, thorium-232, thorium-230, radium-226, radium-228, lead-210, polonium-210, and radon-222. The leachate of six fly ash samples was analyzed for all of the above except radon-222.

Sample Analysis

Determination of Uranium and Thorium

Uranium-232 and thorium-224 tracers were added to the sample to determine chemical recovery. Samples of coal, coal ash, or waste blocks were decomposed by ashing and digested with HNO_3 and HF and then HCl. Aqueous leachate samples were evaporated to dryness and the residue digested with HCl.

The sample dissolved in an HCl solution passes through an anion exchange column, which absorbs the uranium, while the thorium passes through. The uranium was eluted from the column and electrodeposited onto a stainless steel disk for measurement by alpha spectrometry.

The solution containing the thorium was evaporated to dryness and the residue dissolved in dilute HCl. Thorium is coprecipitated with lanthanum fluoride, dissolved in HNO_3 , and passed through an anion exchange column to purify the thorium. The thorium was electrodeposited onto a stainless steel disk for measurement by alpha spectrometry.

TABLE I
SAMPLING LOCATIONS

Power Plant	Coal	Bottom Ash	Fly Ash
1	Conveyor to bunker	Prior to sluice pit base of furnace	Precipitator hoppers
2	Base of crushed coal hopper	Truck loading from hydrobin	Base of ash hoppers
3	Conveyor to bunker	Sluicing from furnace	Truck loading from ash silo
4	Before pulverizer	Truck loading from hydrobin	Silo
5	Before pulverizers	Boiler discharge pipe ash pond	Boiler precipitator hoppers
6	Before pulverizers	Discharge pipe to ash pond	Active ash pond discharge pipe
7	Grate before boiler	Discharge pipe to ash pond	Top of ash silo
8	Grate before boiler	Discharge pipe to ash pond	Top of ash silo

The plated disks were counted with an alpha spectrometry system using a silicon surface barrier detector. The sensitivity of the method was approximately 0.01 pCi per sample for the uranium and thorium isotopes measured. The counting efficiency was approximately 0.25 counts per minute/ disintegrations per minute (cpm/dpm) and the chemical recovery 70%.

Determination of Polonium-210

Coal, coal ash, and waste block samples were ashed in a low-temperature asher, so that volatile polonium was not lost. Polonium-208 tracer was added to the ash to determine chemical recovery, and a nitric acid-hydrofluoric acid dissolution was performed.

Polonium-208 tracer was added to samples of aqueous leachates.

The polonium was extracted from the sample with 0.5% trilauryl amine (TLA) in xylene. Bismuth, iron, and copper are removed from the TLA with sulfuric acid. After backextracting the polonium into perchloric acid, it was coprecipitated with lanthanum hydroxide. The precipitate was dissolved in dilute HCl from which the polonium was spontaneously deposited onto a silver disk. The plated disks were counted with an alpha spectrometry system using a silicon surface barrier detector.

The sensitivity of the method was approximately 0.008 pCi per sample with a counting efficiency of 0.25 cpm/dpm and a chemical recovery of 75%.

Determination of Radium-226 and Radium-228

Chemical recovery was determined by addition of barium-133 tracer. Barium carrier was also added as a precipitating agent. Aqueous leachate samples were acid digested. Coal, coal ash, and waste block samples were fused with Na_2CO_3 , and the water insoluble residue was dissolved with HCl.

RaSO_4 was coprecipitated with BaSO_4 . The $\text{Ba}(\text{Ra})\text{SO}_4$ precipitate was dissolved with ethylene diamine tetracetic acid (EDTA), the chemical recovery determined by gamma counting the barium-133, and the solution transferred to a sealed radon bubbler. After radon-222 has grown back into the solution, the gas was transferred into a ZnS-coated scintillation cell. The activity of radon-222 in equilibrium with its daughters was measured at two-day intervals to follow the radon-222 decay. Radon-222 activity was determined by using a least-squares method to fit the counting data to the theoretical decay curve for radon-222. The radium-226 concentration was calculated from the radon-222 activity.

$\text{Ba}(\text{Ra})\text{SO}_4$ was reprecipitated from the EDTA solution used to measure the radium-226. The precipitate was filtered, sealed, and the chemical recovery redetermined by measuring the barium-133 activity. After an ingrowth period of approximately 17 hr, the actinium-228 was separated from solution and measured by beta/gamma-coincidence counting. This is the optimum time for

measuring actinium-228 activity when radium-226 is present, because it allows for the ingrowth of actinium-228 while minimizing interference from the radium-226 daughters.

The sensitivity of the method for radium-226 was approximately 0.04 pCi per sample with a counting efficiency of 2.4 cpm/dpm, a background count rate of 0.3 cpm, and a chemical recovery of 95%. The sensitivity of the method was defined as that value at which the error at the 95% confidence limit equals the measured value.

The sensitivity of the radium-228 measurement depends on the radium-226 concentration in the sample. If the radium-226 is not detectable (<0.04 pCi), then the sensitivity of the radium-228 measurement was approximately 0.08 pCi per sample using a counting efficiency of 0.05 cpm/dpm, a background counting rate of 0.008 cpm, and a chemical recovery of 80%. A 100-fold increase in the radium-226 concentration produces a 10-fold increase in the sensitivity for radium-228 measurements.

Determination of Radon-222 Emanation

Samples of coal, coal ash, or waste block (ground to 10 mesh) were transferred to a radon bubbler. The sample and bubbler were purged and sealed for at least 17 days to allow radon-222 to emanate from the sample. The gaseous radon-222 was transferred from the bubbler to a ZnS-coated scintillation cell. The activity of the radon-222 in equilibrium with its daughters was measured at two-day intervals in order to follow the radon-222 decay. The radon-222 activity was determined by using a least-squares method to fit the counting data to the theoretical decay curve for radon-222.

The sensitivity of the method was approximately 0.05 pCi per sample or 0.005 pCi/g if a 10-g sample was measured. The sensitivity of the method was defined as that value at which the error at the 95% confidence limit equals the measured value.

Determination of Lead-210

Lead carrier was added to the sample to determine chemical recovery. If the sample contained a significant amount of lead, the concentration was determined by x-ray fluorescence.

Samples of coal, coal ash, or waste blocks were decomposed by ashing and digested with HNO_3 and HF acids and then HCl. Aqueous leachate samples are evaporated to dryness and the residue digested with HCl.

The residue was dissolved in dilute HCl, and PbS is precipitated. The PbS was dissolved in HCl, and most of the iron was removed by extraction into isopropyl ether. The aqueous phase was evaporated to dryness, the residue dissolved in HBr, and the sample passed through an anion exchange column. After allowing lead-212 to decay, the sample was eluted from the column. PbCrO_4 was precipitated, and the activity of the bismuth-210 daughter was measured by β -counting at two-day intervals in order to follow its ingrowth. The bismuth-210 activity was determined by using a least-squares method to fit the counting data to the theoretical ingrowth of bismuth-210. The lead-210 concentration is calculated from the bismuth-210 activity.

The sensitivity of the method was approximately 1 pCi per sample with a counting efficiency of 0.4 cpm/dpm, a background of 1 cpm, and a chemical recovery of 80%.

Discussion

The data available in this report were compared with data contained in the Los Alamos Scientific Laboratory Progress Report LA-8363-PR entitled "Radioactive Emissions from Coal Production and Utilization," October 1, 1979 through September 30, 1980. At this point, there appears to be good correlation.

The only exceptions have occurred in the lead-210 and polonium-210 concentrations for fly ash samples 7F and 8F. In Table II, for both species, there appears to be a sizable difference in concentration relative to the other

TABLE II
FLY ASH
(PC/G)

Fly Ash	^{238}U	^{235}U	^{234}U	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po	^{232}Rn	^{232}Th	^{230}Th
1F				3.7 ± 0.4	1.9 ± 0.7	3.22 ± 0.14	3.1 ± 0.04	0.028 ± 0.011		
2F				3.6 ± 0.4	1.9 ± 0.9	3.76 ± 0.17	5.3 ± 0.4	0.030 ± 0.011		
3F				3.3 ± 0.3	1.6 ± 0.6	3.1 ± 0.3	3.7 ± 0.4	0.060 ± 0.013	0.7 ± 0.4	0.8 ± 0.4
4F				2.7 ± 0.3	0.70	3.06 ± 0.15	2.6 ± 0.3	0.011		
5F	2.7 ± 0.9	0.12 ± 0.06	2.8 ± 0.9	3.0 ± 0.3		2.79 ± 0.13	3.0 ± 0.1	0.119 ± 0.015		
6F						2.37 ± 0.09	2.4 ± 0.3	0.634 ± 0.007		
7F						38.4 ± 0.9	32 ± 6	0.004 ± 0.002		
8F						55 ± 2	64 ± 10	64 ± 0.06		

samples. The raw coal data from the utilities indicate that the sulfur content is lower than for most coal at these two facilities whereas the ash content is a bit higher (Table III). Additional information on these coals and a lab analysis of the coal samples 7C and 8C will be obtained from the utilities.

These fly ash samples were taken from facilities that use stoker boilers. Because a stoker boiler burns unpulverized chunks of coal, the composition of coal waste from stoker boilers is usually 20% fly ash and 80% bottom ash. Because less fly ash is produced by a stoker boiler than a pulverized coal boiler, the concentration of radionuclide on available ash may increase proportionately. Until further data are available for analysis, the noticeably higher polonium-210 and lead-210 in the fly ash seem reasonable.

Federal and State Regulations

The Federal regulations considered in this study are the Resource Conservation and Recovery Act and Title 10, Code of Federal Regulations.

TABLE III
COALS BURNED IN NEW YORK STATE
JUNE 1981

<u>Samples</u>	<u>Percent Sulfur</u>	<u>Percent Ash</u>	<u>Btu/lb</u>	<u>Coal Region</u>
1	2.9	12.79	12,387	W. PA
2	2.67	13.1	11,990	PA/WV
3	2.49	11.0	12,350	PA/WV
4				
5	2.26	17.46	11,166	W. PA
6	2.71	17.25	11,533	W. PA
7	0.87	20.23	11,203	W. PA
8	1.11	21.07	11,178	W. PA

Parts 50 and 20. Only Federal sources are listed here because the New York State Solid Waste Disposal Regulations, Part 360, are the same as the Federal regulations.

Conclusions

From the available data, radionuclide concentrations in coals and ashes analyzed from New York State power plants appear to be within the range for eastern coals as found in the published literature.

These data will be useful to the ERDA Coal Waste Reef project in the following ways. (1) By knowing the concentrations of radionuclides in the blocks, we can compare this information with the concentrations of radionuclides in the sediment. (2) Based upon the known chemistry of these species, we can calculate the release rates of radionuclides from the surfaces of the blocks. Therefore, we can deduce the amount of radionuclides being released from the blocks. (3) If concentrations look particularly high, we could conduct organism accumulation studies and compare those radionuclides found in the organisms with those found in the blocks.

New York State is interested in pursuing radiological research using this data as a premise upon which to identify, justify, and design future research efforts.

RADIOACTIVITY FROM COAL USE - WHERE ARE THE PROBLEMS?

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The Los Alamos National Laboratory has been studying radioactivity from coal use for about three years.^{1,2} Our experimental program has involved samples from 21 power plants, 5 coal cleaning plants, and 4 western coal fields. We can summarize the results as follows.

- The coals are about as radioactive as soils.
 - The ashes are about 10 times as radioactive as soils.
 - Lead-210 and polonium-210 tend to concentrate in fly ash.
 - All radioactivity tends to concentrate on the finer fly ash.
 - The undiluted leachates are about as radioactive as rainwater.³
- The coals and ashes give off less radon than soils.

Our coal samples have the same average uranium content (1.8 ppm) as samples from 800 US mines analyzed in a recent USGS program.⁴

Exposure to concentrations of radioactivity 10 to 100 times that observed in our samples might become a matter of serious health or environmental concern. Factors that could contribute to such exposure might include

- extensive localized use of coals with 20 ppm or greater uranium content (approximately 0.5% of the present supply),
- exposure to large concentrations of respirable fly ash particles (1- μ m range),
- enhanced leachability of coal or wastes,
- massive entry of concentrated leachate into water supplies,
- enhanced radon emanation, and
- long-term exposure to ash products as in building materials.

We believe that further definition and study of these factors are necessary to understand the problem sufficiently to assess the impact of radioactivity from coal use on health and the environment.

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CHEMISTRY OF RADIONUCLIDES IN COAL PREPARATION

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Radionuclides in materials forming coal were sealed in the earth for long periods in comparison to the half-lives of their shorter-lived daughters. Thus, with the exception of the naturally occurring radionuclides that decay directly to stable daughters, the radionuclides of the three principal decay chains can be considered to be in secular equilibrium in coal. In secular equilibrium, the activities of each daughter nuclide are equal to the parent. The activities of all of the primary parent radionuclides found in nature are readily and accurately calculated from their known abundance ratios and activity constants if their corresponding elemental concentrations are known. Although some 25 primary parent radionuclides occur in nature, only uranium-235, uranium-238, thorium-232, and potassium-40 need be considered. The remaining 21 nuclides have extremely low specific activities and decay to stable daughters so that their activities are negligible in comparison with the 4 nuclides above.

During coal combustion, secular equilibrium is disrupted by physical/chemical redistribution of the radionuclides in accordance with their individual chemical properties. For example, radon gas, which is originally mechanically trapped within coal particles, is released to the gas phase where its decay must be treated as a newly purified parent nuclide. Chemical measurements indicate that most of the lead is vaporized during combustion. Some lead apparently remains with the mineral residue. In cooler regions of the boiler, the volatilized lead condenses onto the surfaces of the suspended fly ash particles. Condensing vapors preferentially become associated with flying particles leading to a chemical enrichment with respect to size.

Emission control devices typically pass submicrometer particles more readily than particles of greater size; thus, the emitted aerosol is chemically enriched with respect to coal. Individual radionuclides that were originally present in coal will be enriched in fly ash in accordance with their volatility

and their origin in the decay chain. Therefore, calculation of radionuclide emissions from coal combustion must consider chemical enrichment and decay kinetics that occur within the confines of the plant.

In our presentation, Dr. Ondov will discuss the redistribution and enrichment of potassium-40 in the various radionuclides of the thorium and uranium decay chains that occur during coal combustion including the available experimental data. The unknown enrichments of important radionuclides will be estimated. An algorithm for calculating radionuclide emission based on the decay systematics and chemical enrichment data in estimates will be presented. Inputs for the algorithm are

- concentrations of uranium, thorium, and potassium;
- the coal feed rate;
- electrical output;
- the bottom ash in control device efficiencies; and
- the relative penetration (enrichment factors of the pertinent chemical elements).

Transit time in a typical coal-fired power plant is on the order of 1 min. However, maximum ground-level concentrations of emitted materials occur later as dictated by atmospheric conditions. Then the algorithm activities of emitted radionuclides are computed for decay times of 10 hr.

Finally, emission calculations for four control scenarios are presented. These are a power plant with an electrostatic precipitator (ESP) of efficiency nearly that of the national average, a plant with a high efficiency ESP, a plant with a high efficiency venturi wet scrubber system, and a plant with a hot-side ESP.

URANIUM DAUGHTERS IN NATURAL ATMOSPHERIC AEROSOLS AND COAL-FIRED POWER PLANT EMISSIONS

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Over the past decade, research on aerosol dynamics in the atmosphere has led to a better understanding of the sources and distributions of uranium daughters in the atmosphere. Generally, there are two sources of these radionuclides in the atmosphere: (1) windblown dust and (2) atmospheric decay of radon-222 subsequent to its emanation to the atmosphere from the earth's surface. The relative contribution of these two sources to concentrations of lead-210 and polonium-210 is determined by their lifetimes and those of their radiogenic parents and by the lifetimes of aerosols in the atmosphere. Atmospheric radon-222 over the continents originates nearly completely from emanation from the earth's surface with a negligible contribution from decay of radium-226 in windblown dust. Current evidence suggests that average lifetimes of atmospheric aerosols of 0.1 to 1.0- μm diameter range between 2 and 10 days. This is enough time for a substantial decay of radon-222 ($t_{1/2} = 3.8$ d) and subsequent accumulations of lead-210 on atmospheric particulates prior to their removal by various atmospheric particulate scavenging mechanisms. Thus, like radon-222, most lead-210 in the atmosphere originates from radon-222 emanations from the earth's surface with only a relatively small contribution from windblown dusts. In contrast, there is not enough time for much polonium-210 ($t_{1/2} = 138.4$ d) formation from decay of lead-210 ($t_{1/2} = 22$ yr) prior to removal of aerosols by atmospheric scavenging processes. Thus, most atmospheric polonium-210 originates from windblown dust, and as a consequence, atmospheric activity of polonium-210 is much less than that of radon-222 or lead-210.

Concentrations of uranium and its daughters in coals are about the same as in typical subsurface soils, that is, soils removed from a large contribution of atmospheric fallout. Generally, we observe equilibrium between uranium daughters in coals. Thus, the concentrations of lead-210 and polonium-210 in

windblown coal would be comparable to or less than those of windblown dust. This indicates that, in terms of natural concentrations of these radionuclides, coal-fired power plant emissions cannot greatly impact concentrations of lead-210 (or radon-222) but could conceivably have a measurable impact on polonium-210 concentrations.

Based on a "worst case" calculation (that is, all polonium-210 in coal escapes out the stack into the atmosphere), we estimate an upper limit increase of 60% to the natural annual average polonium-210 concentration at the location of maximum impact near a hypothetical coal-fired power plant. This amounts to an increase in the annual average lung dose from all natural sources of 6%. The impact on lung dose from the other radionuclides resulting from coal-fired power plant emissions would appear to be negligible.

The necessity for further evaluation of polonium-210 emissions from coal-fired power plants should be determined.

POSSIBLE CONTRIBUTIONS OF COAL EXTRACTION AND UTILIZATION TO RADIOACTIVITY CONCENTRATIONS IN DRINKING WATER

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Radioactivity occurs naturally in one of three series of isotopes involving beta and alpha emitting radionuclides. These series and isotopes are present in coals in varying amounts, and thus, coal extraction and utilization can lead to the release of radioactivity. It is suggested by a recent study¹ that the key radioisotopes in these processes are uranium-238, radium-226, bismuth-214, lead-210, polonium-210, thorium-232, and thallium-208. Of these, radium-226, lead-210, and polonium-210 were detected in leachates from coal, bottom ash, and fly ash. The concentrations ranged from less than detected in the blank to a few picocuries per liter.

The potential impact of such concentrations can be seen Table I.² Although the Environmental Protection Agency (EPA) has no defined level of acceptable risk, the risk of existing regulations is a guide. The existing maximum contaminant level (MCL) for combined radium-226 and radium-228 is 5 pCi/l, which leads to an excess lifetime cancer risk range of 10^{-4} to 10^{-5} . Table I shows concentrations that lead to risks of 10^{-5} per lifetime. These estimates were calculated using the ICRP 30 model³ and a lifetime approach.^{4,5} A potential health hazard may exist as a consequence of coal extraction and utilization.

As can be seen in Figure 1, radon and uranium are specifically excluded from the current regulations. Thus, of the list of radionuclides mentioned earlier, only radium-226 and radium-228 are currently regulated. A Suggested Action Guidance (SAG) is being developed for uranium.* This guidance is not a legally enforceable standard. It is our current assessment based on health

*Contact the authors for more information.

TABLE 1
 CONCENTRATIONS OF SOME RADIONUCLIDES
 IN DRINKING WATER THAT LEAD TO A
 RISK LEVEL OF 10^{-5} PER LIFETIME
 (From Reference 2)

Isotope	Concentration ^a (pCi/l)
Uranium-238	4
Radium-226	2
Radium-228	4
Bismuth-214	10,000
Lead-210	5
Polonium-210	2
Thorium-232	3
Thallium-208	50,000

^aThe numbers have been rounded off to one significant figure.

and occurrence. We have no immediate plans for developing standards for the other radionuclides listed above. However, we do plan to consider the health implications of these isotopes as soon as we have developed an adequate information base.

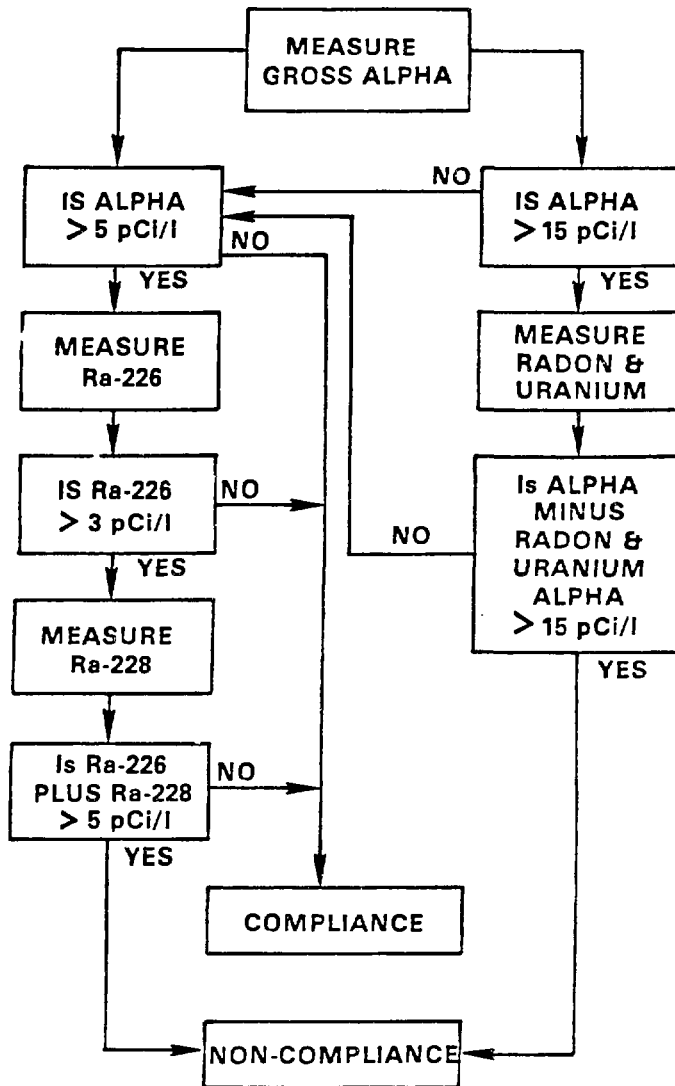


Figure 1. Flow chart for gross alpha particle activity monitoring. Note that it is not a requirement that radon and uranium be measured if the gross alpha particle activity is greater than 15 pCi/l. (From Reference 5).

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IMPACT ON WATER QUALITY FROM RADIONUCLIDES IN COAL

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For the past three years, Los Alamos National Laboratory has been pursuing a research program on radioactive emissions from coal extraction, storage, and use. One of the major components of this program has been a quantitative assessment of the concentrations of uranium and thorium and their radiogenic daughters in aqueous effluents associated with coal mineral wastes, coals, bottom ashes, fly ashes, and scrubber sludges. One important activity necessary to the accomplishment of the program objectives has been a series of "Round Robin" analyses on trace concentrations of uranium-238, radium-226, polonium-210, and thorium-232 in coals, bottom ash, and fly ash by Los Alamos National Laboratory, Mound Facility, and the Environmental Protection Agency Laboratory at Montgomery, Alabama. The results from these interlaboratory analyses established the validity of the data and analytical methods used.^{1,2}

Results of our determinations of the concentrations of the listed radionuclides in laboratory-prepared leachates from coal cleaning plant solids, coals, bottom ash, and fly ash,³ as well as the field-monitored effluents from these sources² show no radionuclides to be in excess of the maximum permissible soluble concentrations as per the Code of Federal Regulations 10, Part 20, Appendix B (Rev. 1-1-80). Thus, our current view must be that these leachates present a minimum radiation threat to the environment. In expanding our information base to all available data sources, we have only reinforced this conclusion.

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APPENDIX

DETAILED METHODOLOGY AND ANALYSIS OF RESULTS

This section describes the approach used in identifying and ranking the radioactivity issues and identifying their subissues. It also describes the results and statistical analysis of the issue ranking exercise.

A.1 Issue Identification

Each participant was asked to bring a list of radioactivity-from-coal issues to the workshop. These lists were used to prepare a tentative issues list. Although it was initially accepted by the participants, this tentative list was later rejected in favor of an alternative list. With minor modifications, this alternative list was adopted by the participants as the final list of radioactivity-from-coal issues. The final list of issues is shown in Table A-1. The definition of each issue on this list has been presented in Section 3.0 of this document.

A.2 Issue Ranking

Once the final issues were identified and agreed upon, the participants were asked to assign scores to the issues according to their relative importance. Importance was defined as data needs; issues for which radiological impact data are lacking were given higher scores than those issues for which data are sufficient.

A modified Delphi technique was used to achieve a consensus on the importance score assigned to each issue. Score sheets, like the one in Figure A-1, were distributed to the participants. The participants were asked to individually assign each issue a value between 0 and 10, where 0 indicated that current data are sufficient and 10 indicated that there are critical data needs associated with the issue. After they were completed, the score sheets were collected, and the mean and range of scores for each issue were calculated and put on a Vu-Graph. The Vu-Graph was projected to the group and the participants were asked to discuss the mean and range of scores that were assigned to each issue. Of particular interest in the discussion were those

TABLE A-I
FINAL LIST OF ISSUES

1. Long- and Short-Term Solubility of Released Fly Ash in Different Fluids
 2. Radionuclides in Construction Materials
 3. Stack Releases
 4. Activity and Emission of Waste at Landfills and Redistribution into Populated Areas
 5. Long-Term Changes of Leachability of Coal and Waste
 6. Airborne Radionuclide Levels and External Radiation Levels Inside Power Plants
 7. Ground-Level Air Concentrations
 8. Radionuclides in Waste Products
 9. Deposition from Stack Releases
 10. Radionuclides in Coal
-

issues with a wide range of scores, because a wide range indicated that there was considerable disagreement among the participants as to the data needs associated with that issue.

At the conclusion of this discussion, a second score sheet was distributed to each participant. The participants were asked to rescore each issue based on the preceding discussion, the mean and range indicated on the Vu-Graph, and the score that the participants individually gave previously to each issue. The purpose of having the participants rescore the issue was to come closer to a consensus on issue scores; by having the participants discuss the basis of their scores for each issue and by comparing their initial scores with the mean and range of scores for the participants as a group, it was hoped that the range of scores on this second iteration would be narrower than on the first.

Name _____
 Iteration No. _____

ENVIRONMENTAL ISSUES

Instructions

Please score each issue according to its environmental importance, relative to the other issues. Each issue should be given a score between 0 and 10 based on the scoring key. If you have no knowledge regarding an issue, please do not score it but, instead, check the box next to that issue.

Scoring Key

- | | | | |
|---|------------------|----|---------------------|
| 0 | No Importance | 5 | Moderate Importance |
| 1 | | 6 | |
| 2 | Minor Importance | 7 | |
| 3 | | 8 | Major Importance |
| 4 | | 9 | |
| | | 10 | Critical Importance |

Issue	Score		Comments
1. Long- and Short-Term Solubility of Released Fly Ash in Different Fluids	0 5 10 	<input type="checkbox"/> No Knowledge	_____
2. Radionuclides in Construction Materials	0 5 10 	<input type="checkbox"/> No Knowledge	_____
3. Stack Releases	0 5 10 	<input type="checkbox"/> No Knowledge	_____
4. Activity and Emission of Waste at Landfills and Redistribution into Populated Areas	0 5 10 	<input type="checkbox"/> No Knowledge	_____
5. Long-Term Changes of Leachability of Coal and Waste	0 5 10 	<input type="checkbox"/> No Knowledge	_____
6. Airborne Radionuclide Levels and External Radiation Levels Inside Power Plants	0 5 10 	<input type="checkbox"/> No Knowledge	_____
7. Ground-Level Air Concentrations	0 5 10 	<input type="checkbox"/> No Knowledge	_____
8. Radionuclides in Waste Products	0 5 10 	<input type="checkbox"/> No Knowledge	_____
9. Deposition from Stack Releases	0 5 10 	<input type="checkbox"/> No Knowledge	_____
10. Radionuclides in Coal	0 5 10 	<input type="checkbox"/> No Knowledge	_____
11. _____	0 5 10 	<input type="checkbox"/> No Knowledge	_____

Figure A-1. Score sheet for ranking issues.

The completed second iteration score sheets for the participants were collected, and the mean and range for each issue were calculated and placed on another Vu-Graph. These revised means and ranges of scores were discussed to determine what differences of opinion still existed between participants. A third score sheet was handed out to each participant for recording their final score for each issue. This third iteration was used to further narrow the range of scores. It was the last iteration used because it is commonly believed that any greater consensus cannot be achieved by going beyond a third or fourth iteration when using the Delphi technique.

Table A-II shows the 10 radiological issues ranked by the mean scores assigned by the participants in the third iteration. The mean, or average, scores range from 7.3 for the Long- and Short-Term Solubility of Released Fly Ash in Different Fluids issue to 1.6 for the Radionuclides in Coal issue. The mean scores fall into four groups. The first group is clustered around a value of 7 and includes the two highest scored issues. The second group, which consists of only the third highest scored issue, is centered around 6. A third group occurs around 3.5 to 4 and includes the next six issues. The final group is clustered around 1.5 and consists of only the lowest scored issue.

Next to the mean scores in Table A-II, the ranges of scores assigned during the third iteration are shown. The range gives some indication of the degree of consensus achieved on the score assigned each issue. As Table A-II shows, the range is narrow around some issues like Ground-Level Air Concentrations (2 to 5) and very wide around others like Stack Releases (4 to 10).

A more useful measure than the range of consensus is the standard deviation. The standard deviation is often used in statistical analyses to measure the degree of variability or dispersion about the mean score. In the case of the participant responses from the third iteration, the greater the spread of individual participant scores about the mean score for an issue, the larger the standard deviation for that issue.

The standard deviation is a more useful measure of agreement or consensus than the range, because the range is more susceptible to outlying or extreme

TABLE A-II

ISSUES AND STATISTICAL DATA

	<u>Mean</u>	<u>Range</u>	<u>Standard Deviation</u>	<u>Median</u>
1. Long- and short-term solubility of released fly ash in different fluids	7.3	5 to 10	1.7	8
2. Radionuclides in construction materials	7.1	4 to 9	1.7	7
3. Stack releases	5.9	4 to 10	1.8	5
4. Activity and emission of waste at landfills and redistribution into populated areas	4.3	2 to 7	1.4	4
5. Long-term changes of leachability of coal and waste	4.1	2 to 8	1.9	4
6. Airborne radionuclide levels and external radiation levels inside power plants	3.4	1 to 5	1.1	4
7. Ground-level air concentrations	3.1	2 to 5	1.5	2
8. Radionuclides in waste products	2.9	1 to 7	1.8	3
9. Deposition from stack releases	2.9	2 to 5	1.2	2
10. Radionuclides in coal	1.6	0 to 3	0.9	2

scores. For example, seven participants may each assign a score of 6 to an issue, whereas an eighth assigns it a score of 1 and the ninth a score of 10. The range of this issue is 1 to 10 and therefore makes it appear that little agreement was reached. Although the standard deviation does not ignore extreme scores, it does take into account the high level of agreement among the seven participants.

The standard deviation for each issue is shown in the fourth column of Table A-II. Most of the issues have small standard deviations, and therefore there appears to be a fairly strong consensus among the participants regarding the score for each issue.

The final column of Table A-II shows the median scores for each issue. Just as the standard deviation is less susceptible to extreme scores than the range, the median is less influenced by extremes than the mean. The median is often used as a measure of the average group score instead of the mean, when extreme values are evident. Extreme values are not a significant problem in the participant scores, because the rank order of the issues by median is not very different from the rank order by mean.

Before accepting the rank ordering of the issues in Table A-II as being truly indicative of the relative data needs for the issues, it is important to determine if there is enough difference between the mean scores of each pair of issues to conclusively state that one issue scores higher than the other. For example, it is logical to question if the highest ranked issue, Long- and Short-Term Solubility of Released Fly Ash in Different Fluids, actually has more important data needs than the second highest-scored issue, Radionuclides in Construction Materials, when the scores of these two issues differ by only two-tenths of a point.

One way to determine which issues have greater data needs than others is to conduct statistical tests using Repeated Measures Analysis of Variance paired comparisons of issues. Time constraints prohibit conducting such tests for inclusion in this proceedings. Figure A-2, which shows the mean and standard deviation for each issue, gives a rough indication of how significant the differences are between the issue scores. This figure suggests that the highest and second highest scored issues have scores that are so similar that there probably is no statistical difference between them. The third highest scored issue has a considerably lower mean score than the two highest scored issues, and it is possible that it has a statistically significantly lower score. It also probably has a statistically significantly higher score than most of the lower scored issues. All of the issues with means clustered around 3 and 4

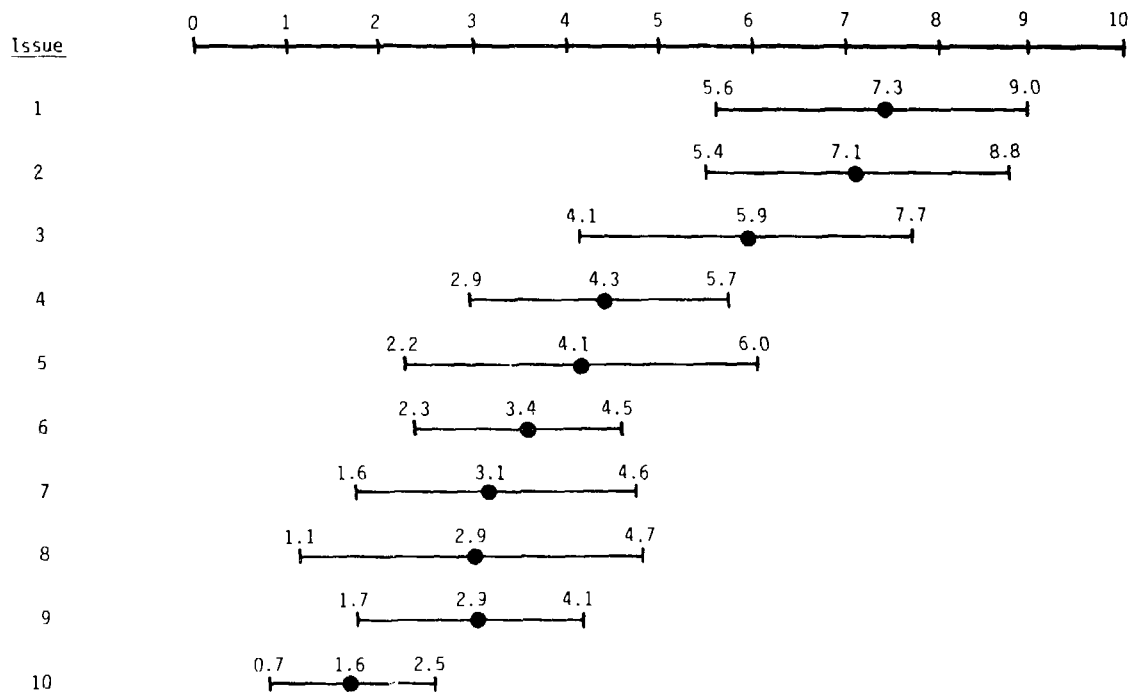


Figure A-2. Mean and standard deviation for each issue.

probably do not have sufficiently different scores to distinguish one or more as being statistically significantly higher or lower than the others. Finally, the lowest scored issue may have a statistically significantly smaller score than all of the other issues.

A.3 Subissue Identification

After radioactivity issues were ranked, the participants were asked to identify and justify subissues under each issue. This information was compiled to create tentative lists of subissues under each issue. Each tentative list was then discussed by the participants to determine additions, deletions, and modifications to the subissues. The final lists of subissues agreed upon by the participants are presented in the text of this document.