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Identification and Quantification of Radionuclides in Coal Ash

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FHWA/IN/JTRP-98/01

Final Report

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July 1998

FINAL REPORT

Identification and Quantification of Radionuclides in Coal Ash

FHWA/IN/JTRP-98/1

by

.

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Joint Transportation Research Project

Project No: C-36-68G
File No: 4-7-7

Prepared as Part of an Investigation Conducted by the

Joint Transportation Research Project
Engineering Experiment Station
Purdue University

funded by the

Indiana Department of Transportation
and the
Federal Highway Administration

School of Civil Engineering
Purdue University
West Lafayette, Indiana 47907-1284

9 July 1998

TECHNICAL REPORT STANDARD TITLE PAGE

1. Report No. FHWA/IN/JTRP 98/1	**2. Government Accession No.**	**3. Recipient's Catalog No.**
4. Title and Subtitle		**5. Report Date** 9 July, 1998 **6. Performing Organization Code**
7. Author(s) J.E. Alleman, F.M. Clikeman, and T. Skronski		**8. Performing Organization Report No.** FHWA/IN/JTRP 98/1
9. Performing Organization Name and Address Joint Transportation Research Project Civil Engineering Building Purdue University West Lafayette, Indiana 47907-1284		**10. Work Unit No.** **11. Contract or Grant No.** SPR-2175
12. Sponsoring Agency Name and Address Indiana Department of Transportation State Office Building 100 North Senate Avenue Indianapolis, IN 46204		**13. Type of Report and Period Covered** Final Report **14. Sponsoring Agency Code**

15. Supplementary Notes

Prepared in cooperation with the Indiana Department of Highways and Federal Highway Administration.

16. Abstract

One of the important environmental issues raised recently in regard to coal ash reuse for highway construction purposes (e.g., embankment development) is that of worker, and public, exposure to radiation which might possibly be emitted by these types of residuals. Radiation emission is, in fact, a natural phenomenon for most materials, both natural and man-made, but in the case of coal ash residuals the process of combustion produces an inevitable concentration of radionuclides from the original virgin coal. INDOT's corresponding environmental concern consequently focuses on the following basic question: *does this magnification of radionuclides found within these coal ash residuals cause sufficiently high levels of radiation to impose harmful effects due to exposure?*

This research project subsequently addressed the associated issue of radiation emission by coal ash residuals generated within the State of Indiana, covering both fly ash and bottom ash materials. Samples were obtained at sixteen (16) different coal-fired power generating facilities within Indiana and subjected to a quantitative analysis of their associated gamma-ray emission levels. After identifying the responsible radionuclides, a conservative approximation was then developed for the worst-case potential occupational exposure with construction employees working on this type of high-volume, coal ash embankment. In turn, these potential emission levels were compared to those of other traditional construction materials and other common sources.

The observed results indicated that these coal ash residuals did contain levels of gamma-ray emitters that were, in fact, higher than those of traditional construction materials (i.e., clay, sand, brick, and limestone). However, these levels of gamma-ray emission were not excessively high, and considerably below the limits respectively promulgated for public exposure and occupational exposure by the Environmental Protection Agency (100 mrem/yr for a single man-made source) and the Nuclear Regulatory Commission (5 rem/yr).

17. Key Words coal ash; radiation emission; environmental quality; waste reuse; highway construction	**18. Distribution Statement** No restrictions. This document is available to the public through the National Technical Information Service, Virginia, 22161

19. Security Classif. (of this report) Unclassified	**20. Security Classif. (of this page)** Unclassified	**21. No. of Pages** 77	**22. Price**

Form DOT F 1700.7 (8-69)

INDOT Research

TECHNICAL Summary

Technology Transfer and Project Implementation Information

TRB Subject Code: 36 Waste Materials *July 1998*
Publication No.: FHWA/IN/JTRP-98/1, SPR-2175 *Final Report*

Identification and Quantification of Radionuclides in Coal Ash

Introduction

Coal ash residuals, including both fly ash and bottom ash, are routinely generated in large quantities within the State of Indiana, and significant efforts are underway to find constructive options for their reuse. However, as with any residual material, there are associated concerns regarding the environmental quality of these materials which must be resolved prior to pursuing any reuse strategies. Chemical-specific issues (e.g., relative to leachate elements such as arsenic, selenium, etc.) have commonly been cited in this respect, but additional concerns have also been expressed regarding radiation emission levels and their potential impact on humans. This study, therefore, was developed to quantify the emission levels which could arise with these materials and to develop a projection of the radiation dosage levels which might be experienced by workers involved with the constructive reuse (i.e., building embankments) of these materials.

Findings

Representative coal ash residuals were obtained from 16 power plants in the state of Indiana and tested for their gamma-ray emission levels. In addition, a number of traditional building materials (e.g., sands, clays, etc.) were also tested in order to develop a relative comparison of their emission levels. The observed results indicated that both bottom ash and fly ash contained levels of gamma-ray emitters that were higher than those of traditional construction materials (i.e. clay, sand, brick, and limestone). However, these levels of gamma-ray emitters were considerably below any level which would warrant undue concern. Excluding any exposure whatsoever to coal ash residuals, the average adult human will have a cumulative, natural 'background' radiation dose of about 320 mrem/yr. By comparison, construction workers involved with fly ash or bottom ash over a one-year period (i.e., a worst-case scenario, involving a full 2,000 hours of annual exposure) would have projected radiation exposure levels less than one-fifth of their 'natural' exposure. By analogy, their radiation exposure to these coal ash residuals would be approximately equal to the dose equivalent received by living in a brick home for one year. Although there are no specific criteria for permissible levels of radiation for these coal ash materials, the results found

with this study strongly suggest that they bear no adverse or unacceptable risk to human health. The projected levels of radiation which might be realized with their constructive reuse, either on an occupational or public basis, are below the limits for safe exposure established by the US EPA and the Nuclear Regulatory Commission.

Implementation

Pursuant to these findings, the following suggestions are being provided as a means of implementing and extending this research effort:

- ***Continued Full-Scale Implementation Projects:***

  During the recent few years, INDOT has conducted three 'proof-of-concept' full-scale embankment projects using coal ash materials. The findings of this report strongly support further INDOT efforts to extend the scale of this reuse activity, working in consort with similarly motivated coal-fired power plants to develop mutually favorable arrangements for material preparation, stockpiling, and conveyance.

- ***Development of Pro-Active INDOT Reuse Perspective for Indiana Residents:***

  The fact that 'environmental groups' within the state of Indiana have openly raised concerns about coal ash radiation levels is a circumstance which underlies the importance and direction of public sentiment relative to their perception of environmentally sensitive technical matters. Backed by real data which negates the associated level of concern stemming from coal ash radiation levels, a constructive dialogue with these types of groups would appear to be beneficial to the long-term success of INDOT's reuse policies for coal ash materials.

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ACKNOWLEDGMENTS

During the course of this project, the following individuals and organizations provided considerable, and enthusiastic, support to the development and completion of the research effort detailed within this report:

- **Barry Partridge**, INDOT, Chief of Research Division,
- **David Ward**, INDOT Section Manager,
  [*INDOT Applied and Environmental Research, Electrical Engineering and Technical Support*]
  …for their technical assistance, professional insight, and financial support

- **Participating Indiana Power Generators,**
  - AEP
  - Hoosier Energy
  - IPL
  - NIPSCO
  - PSI
  - Purdue University
  …without their cooperation and understanding, the gathering of samples would have been much more difficult, and this project would not have been as comprehensive

- **Brandon Ogilvie,**
  [*M.S. Candidate, Environmental and Hydraulic Engineering Area, Purdue University]*
  …for writing a computer code that saved several hours of sorting data

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Acronyms, Symbols, and Abbreviations

Organizational Acronyms

HEC	Hoosier Environmental Council
INDOT	Indiana Department of Transportation
EPA	Environmental Protection Agency
NRC	Nuclear Regulatory Commission

Mathematical Symbols and Technical Abbreviations

ESP	electrostatic precipitator
E	energy of a nuclear particle, i.e. gamma rays (keV)
A	calculated specific activity of a material--rate at which a radioisotope decays (Bq/g)
A_{meas}	measured activity of a material (Bq/g)
M	sample mass (g)
ϕ_{ash}	gamma ray flux at a point on an air/coal ash interface (gamma rays/s*cm^2)
b	thickness of self-attenuating volume-distributed source (cm)
ρ	density of material (g/cm^3)
ε	gamma-ray emission probability (unitless)
μ	linear attenuation coefficient (cm^{-1})
μ_a	linear absorption coefficient (cm^{-1})
E_2	second order exponential integral function
ϕ_{air}	gamma ray flux in air at a point above a source (gamma rays/s*cm^2)
z	distance from a radiation source (cm)
D	dose rate (mrad/yr)
H	dose equivalent rate (mrem/yr)
QF	quality factor

Unit Abbreviations

rad	radiation absorbed dose
mrad	10^{-3} rads
rem	radiation equivalent mammal
mrem	10^{-3} rem
Bq	Bequerel
keV	kilo electron volt
pCi	picoCurie, equivalent to 0.037 Bq

Elemental Abbreviations

Ac	Actinium		
Ba	Barium	Pb	Lead
Bi	Bismuth	Po	Polonium
Ca	Calcium	Ra	Radium
Cs	Cesium	Rn	Radon
Fr	Francium	Th	Thorium
K	Potassium	Tl	Thallium
Pa	Palladium	U	Uranium

IMPLEMENTATION REPORT

Coal combustion by-products (i.e., coal ash) have been considered as a alternative material for use in the construction of large embankments for highways. One of the concerns raised with using these coal ash residuals, though, is the potential for exposure to the public by radiation resulting from naturally occurring radionuclides that are concentrated in ash during the burning of coal. However, the findings presented within this report indicate that this concern is unwarranted since the levels of exposure observed with residuals taken at sixteen different facilities have been found to be consistently low. While the dose equivalent of coal ash (i.e., due to the emission of gamma radiation) is higher than that of several other traditional construction materials, it is comparable to, and often lower than, a number of other radiation exposures that are present in everyday life.

Pursuant to these findings, therefore, the following suggestions are being provided as a means of implementing and extending this research effort:

- **Continued Full-Scale Implementation Projects**

  During the recent few years, INDOT has conducted three 'proof-of-concept' full-scale embankment projects using coal ash materials. The findings of this report strongly support further INDOT efforts to extend the scale of this reuse activity, working in consort with similarly motivated coal-fired power plants to develop mutually favorable arrangements for material preparation, stockpiling, and conveyance.

- **Development of Pro-Active INDOT Reuse Perspective for Indiana Residents**

  The fact that environmental groups within the State of Indiana have openly raised concerns about coal ash radiation levels is a circumstance which underlies the importance and direction of public sentiment relative to their perception of environmentally sensitive technical matters. Backed by real data which negates the associated level of concern stemming from coal ash radiation levels, a constructive dialogue with these types of groups would appear to be beneficial to the long-term success of INDOT's reuse policies for coal ash materials.

CHAPTER 1

Introduction

Given the fact that one of the largest problems facing the coal-fired electric power industry is that of the massive buildup of coal combustion by-products (i.e. coal ash), there is a significant on-going search to find acceptable uses for these residuals. While several beneficial reuse applications have been considered (e.g., concrete and asphalt additives, roofing material preparation, etc.), the high-volume use of these coal ash residuals with embankments for highways and bridge overpasses appears to represent one of the most promising management options.

Indeed, the Indiana Department of Transportation (INDOT) currently lists coal ash as a qualified material for road construction and has arrangements with several power companies to obtain coal ash for this purpose. Within recent years, INDOT has conducted three such full-scale, *'proof-of-concept'* road construction projects, including:

- US Highway-50 near Vincinnes, IN,
- US Highway-12 in Gary, IN, and the
- 56th Street overpass across Interstate Highway-465 in Indianapolis, IN.

Using coal ash in this type of application, however, may present a number of concerns for public safety and environmental protection. Although chemical-specific issues (e.g., relative to leachate elements such as arsenic, selenium, etc.) are typically cited in this respect, additional concerns have also been expressed in the technical literature regarding radiation emission levels and their potential impact on human health (e.g., Myrick, *et al*., 1983; Tadmor, 1986; and Roeck, *et al*., 1987).

On the one hand, radiation is usually perceived as a problem because of its perceived potential for harming humans, and the corresponding anxiety over unknown and misunderstood facts. However, this perception is normally unwarranted, especially at the low levels of radiation experienced in the natural environment.

In the case of coal ash residuals, though, radiation levels found within virgin coals will inevitably be concentrated during the course of firing. While there may well be some loss of radionuclides through the stack during firing, the expected magnification level for these radionuclides (increasing from virgin coal to

fired ash levels) could well estimated to be a factor a ten, and possibly higher (i.e., based on the fact that raw coal typically has an inert, non-volatile ash content of approximately 10%).

INDOT's corresponding environmental concern consequently focuses on the following basic question: *does the magnification of radionuclides found within these coal ash residuals reach levels sufficiently high to impose harmful risks of exposure?*

This research project subsequently addressed the associated issue of radiation emission by coal ash residuals within the State of Indiana, covering both fly ash and bottom ash. Samples were obtained at sixteen (16) different coal-fired power generating facilities within Indiana and subjected to a quantitative analysis of their gamma-ray emission levels.

After identifying the responsible radionuclides, a conservative approximation was then developed for the worst-case potential occupational exposure to construction employees working on this type of high-volume, coal ash embankment. In turn, these potential emission levels were compared to those of other traditional construction materials and other common sources.

CHAPTER 2

Objectives

This project was developed to experimentally quantify the level of radiation hazard (or lack thereof) associated with the projected use of coal ash residuals for highway-related embankment construction. Overall, the project had seven (7) sequential objectives, as follows:

- **First**, to develop and secure technical contacts at each of Indiana's principal coal-fired power plant facilities who, in turn, would be able to provide authorization and access for subsequent sampling visits,
- **Second,** to secure a representative set of real-world samples from a range of coal-fired power plants within the State of Indiana, including both residual (i.e., fly ash and bottom ash) and virgin coal specimens,
- **Third,** to obtain another group of natural materials (e.g., clays, sands, limestone, granite, etc.) to serve as testing benchmarks for comparative purposes against which the coal ash residuals could be compared,
- **Fourth,** to analytically quantify the gamma-ray emission spectrums for each of the aforementioned coal, coal ash, and natural materials,
- **Fifth,** to develop a corresponding, conservative analysis of the projected radiation emission levels which could be experienced by occupational workers using these coal ash residuals during the construction of embankments,
- **Sixth,** to compare these latter coal ash emission levels against radiation dose equivalents commonly associated with other natural and commonplace sources routinely linked to our workplace and home environments, and
- **Seventh,** to compile and disseminate these findings within this technical report.

CHAPTER 3

Basic Concepts in Radiochemistry and Nuclear Physics

3.1 Technical Definitions Associated with Radiation Testing and Analysis

The technical vernacular used with the science and engineering of radiation testing and emission analysis tends to be rather unfamiliar to most individuals. As a result, a special *'Key Definitions Listing'* has been provided in Appendix F in order to provide a quick guide to many of the terms used throughout this report (i.e., supplementing the *'Acronyms, Symbols, and Abbreviations'* list given earlier on pg. vii).

3.2 Radionuclides in Natural Materials

Coal and coal ash, as well as any other solid material that is naturally derived from the earth's crust, contain trace elements that undergo radioactive decay. Most radionuclides that are naturally common in raw materials are derived from a few long-lived isotopes. Three of these isotopes, Uranium-238 (^{238}U), Uranium-235 (^{235}U), and Thorium-232 (^{232}Th), are the parents of a series of alpha and beta decays called decay chains. These decay chains are shown in Figure 1. After the emission of alpha and beta particles, gamma rays are frequently emitted to release extra decay energy. Table 1 shows the half-lives and significant gamma-rays of each isotope. Each of these chains results in a stable isotope of lead.

The ^{235}U and ^{232}Th decay chains exhibit a phenomenon called branching. Branching occurs when a radionuclide has two options of decay. The percentages shown next to the decay modes, alpha or beta, of Francium-223 and Actinium-227 are the branching ratios for those modes.

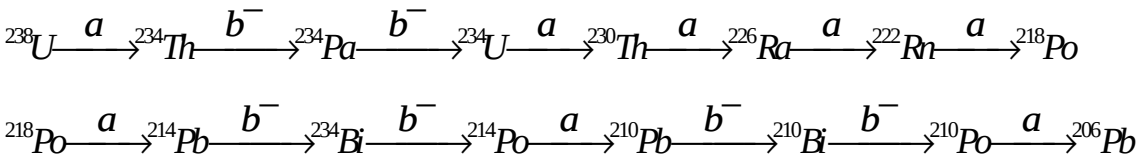

Figure 1a: Uranium-238 Decay Chain

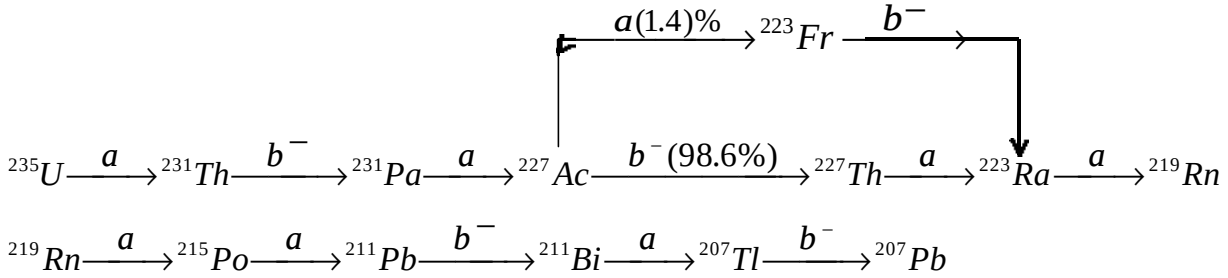

Figure 1b: Uranium-235 Decay Chain

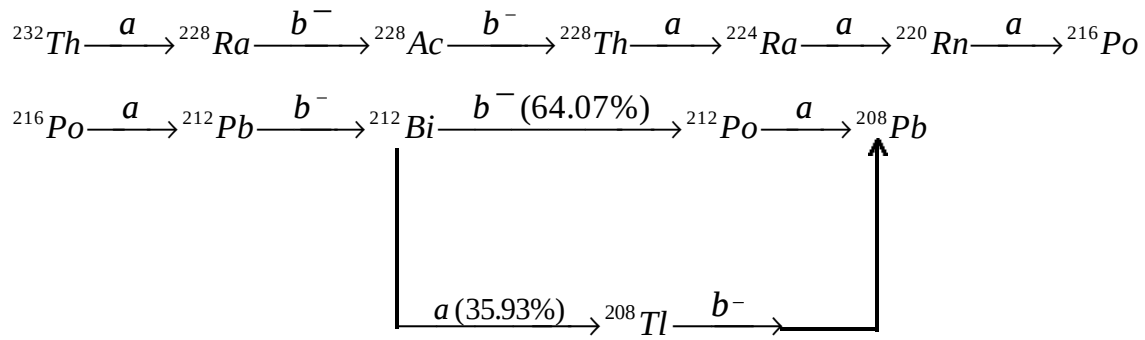

Figure 1c: Thorium-232 Decay Chain

Table 1a: Half-lives and Significant Gamma-rays of ^{238}U Decay Chain

Isotope	Half-life	Energies of significant gamma rays (keV)
U-238	4.47E9 yr	none
Th-234	24.1 d	none
Pa-234	6.7 hr	none
U-234	2.46E5 yr	none
Th-230	7.54E4 yr	none
Ra-226	1600 yr	186
Rn-222	3.82 d	none
Po-218	3.05 min	none
Pb-214	26.8 min	295, 351
Bi-214	19.9 min	609, 768, 1120, 1155, 1238, 1378, 1408, 1509, 1764, 1784, 2119, 2448
Po-214	164 μsec	none
Pb-210	22.3 yr	none
Bi-210	5.01 d	none
Po-210	138 d	none
Pb-206	stable	none

Table 1b: Half-lives and Significant Gamma-rays of the ^{235}U Decay Chain

Isotope	Half life	Energies of significant gamma rays (keV)
U-235	7.038E8 yr	144, 163, 186, 202, 205
Th-231	25.52 hr	none
Pa-231	3.276E4 yr	none
Ac-227	21.77 yr	none
Fr-223	22 min	none
Th-227	18.718 d	none
Ra-223	11.434 d	none
Rn-219	3.96 sec	none
Po-215	1.778 msec	none
Pb-211	36.1 min	none
Bi-211	2.13 min	none
Tl-207	4.77 min	none
Pb-207	stable	none

Table 1c: Half-lives and Significant Gamma-rays of the ^{232}Th Decay Chain

Isotope	Half life	Energies of significant gamma rays (keV)
Th-232	1.405E10 yr	none
Ra-228	5.75 yr	none
Ac-228	6.13 hr	338, 463, 795, 911, 965, 969
Th-228	1.9132 yr	none
Ra-224	3.62 d	none
Rn-220	55.61 sec	none
Po-216	0.146 sec	none
Pb-212	10.643 hr	238, 300
Bi-212	60.55 min	727, 1079, 1620
Po-212	0.298 μsec	none
Tl-208	3.053 min	277, 583, 861, 2615
Pb-208	stable	none

One isotope that is naturally occurring but not a daughter or parent of a decay chain is Potassium-40 (^{40}K), whose decay is shown in Figure 2a. The beta decay of ^{40}K results in Calcium-40.

Other radioactive isotopes that are present in many natural materials that are located near the surface of the earth (in top few meters) result from fallout of nuclear tests. One major isotope of this type is Cesium-137 (^{137}Cs). The beta decay of ^{137}Cs results in Barium-137, shown in Figure 2b. Half-lives and gamma rays for ^{137}Cs and ^{40}K are in Table 2.

$$^{40}K \xrightarrow{b^-} {}^{40}Ca$$

Figure 2a: Potassium-40 Decay

$$^{137}Cs \xrightarrow{b^-} {}^{137}Ba$$

Figure 2b: Cesium-137 Decay

Table 2: Half-life and Significant Gamma Rays of ^{40}K and ^{137}Cs

Isotope	Half-life	Energies of significant gamma rays (keV)
K-40	1.28E9 yr	1461
Cs-137	30.17 yr	662

3.3 Gamma Radiation

Along with the alpha and beta particles that are emitted, there are many gamma rays released from each decay. In most situations outside of the body, it is gamma radiation that presents the greatest potential for harm to human health. This is because photons are much more difficult to attenuate than alpha or beta particles. For example, alpha particles can be effectively shielded by a sheet of paper or a layer of dead skin cells on the human body, beta particles by several millimeters of concrete or a layer of skin, and gamma rays by several centimeters of lead. Of course, shielding is only necessary when large amounts of radiation is being produced.

Although radioisotopes emit a total sum of hundreds of gamma rays, many of these gamma rays are undetectable because they rarely occur or have a low energy. In fact, some of the radionuclides above actually emit twenty or more gamma rays during a decay, but many of them are undetectable and present no significant risk to humans. Some gamma rays are undetectable because their energies are too low. Other gamma rays are rarely emitted but can be detected in concentrated samples of the source radionuclide. Ten radionuclides that emit significant numbers of high-energy gamma rays are singled-out for this study.

Despite the amount of shielding required to stop each type of radiation and lack of significant gamma rays, it should be noted that beta and alpha radiation can be problematic. If particles are inhaled that have radionuclides attached to them, alpha and beta radiation could be quite hazardous, because no shielding exists inside the body to protect internal organs. For this reason some of the radionuclides in the decays shown above, that do not have significant gamma rays, could still cause some damage when inhaled.

The detectable gamma rays can be represented on an energy spectrum. An energy spectrum is used to quantify the energy and frequency of gamma-ray emission. This information can be used to identify which radionuclides are present in an unknown sample. Since each radionuclide emits a specific energy and number of gamma rays that can be counted, the presence of a radionuclide can be determined by collecting data in the form of an energy spectrum and analyzing the spectrum for specific patterns of gamma rays.

CHAPTER 4

Materials and Methods

4.1 Coal and Coal Ash Sample Collection

Five power companies, encompassing fifteen power stations, along with the Purdue University power plant, were visited to collect samples of coal, bottom ash, various types of fly ash, and commingled ash, a mixture of bottom and fly ash. Table 3 lists the types of samples acquired from each plant (**NOTE**: *a map depicting the location of the sixteen plants sampled during this project is given in Appendix G*).

Table 3: Evaluated Power Plants and Sample Types

Plant Name	Output (MWe)	Sample Type
IPL		
Stout	900	BA, ESP, Coal
Petersburg	1300	BA, ESP, Coal
Perry	100	BA, ESP, IA, Silo, Coal
Pritchard	400	BA, ESP, Coal
NIPSCO		
Schaeffer	1100	BA, ESP, Coal
Michigan City	800	BA, ESP, Coal
Bailly	700	BA, ESP, Coal
Mitchell	600	BA, ESP, Coal
Hoosier Energy		
Merom	1300	BA, ESP, APHA, EA, Coal
Ratts	300	BA, ESP, EA, Coal
PSI		
Cayuga	1100	BA, ESP, Coal
Gibson Station	3400	BA, ESP, Coal
Wabash River	1000	BA, ESP, AP, Coal
AEP		
Breed	N/A	AP
Rockport	1300	BA, ESP, Coal
Purdue University	50	BA, BH, MC, LS, HA, Coal

Key: BA=bottom ash, BH=baghouse fly ash, LS=limestone, AP=ash pile, APHA=air preheat fly ash, ESP=electrostatic precipitator fly ash, HA=hopper ash, MC=mechanical collector, EA=economizer fly ash, IA=intermediate ash

In most cases the coal and bottom and fly ashes comprise a "complete set" of freshly-produced samples. At some power plants other samples of bottom and fly ash were collected along with the "complete sets." Coal was sampled from bunkers or pulverizers just before being burned. Bottom and fly ash samples, that correspond to the coal were taken from ash hoppers. Representative commingled ash specimens were taken from ash piles, ponds, or silos.

4.2 Comparison of Ash to Natural Materials

The activities and exposures of ash samples were compared to those of several traditional building materials and other common materials. Granite, limestone, clay, brick, marble, and medium-grained sand were among the building materials included in the comparison, and diatomaceous earth, dried bananas, and potassium chloride were among the other materials tested.

4.3 Sample Analysis

Before analysis each sample was oven dried, to determine the dry density of material and to remove the effects of water during radiation detection, and ground into fine particles, in the case of bottom-ash clinkers, if required.

Each sample was tested for photon emissions in a one-liter Marinelli beaker using a Germanium-crystal detector system, as schematically depicted in Figure 3.

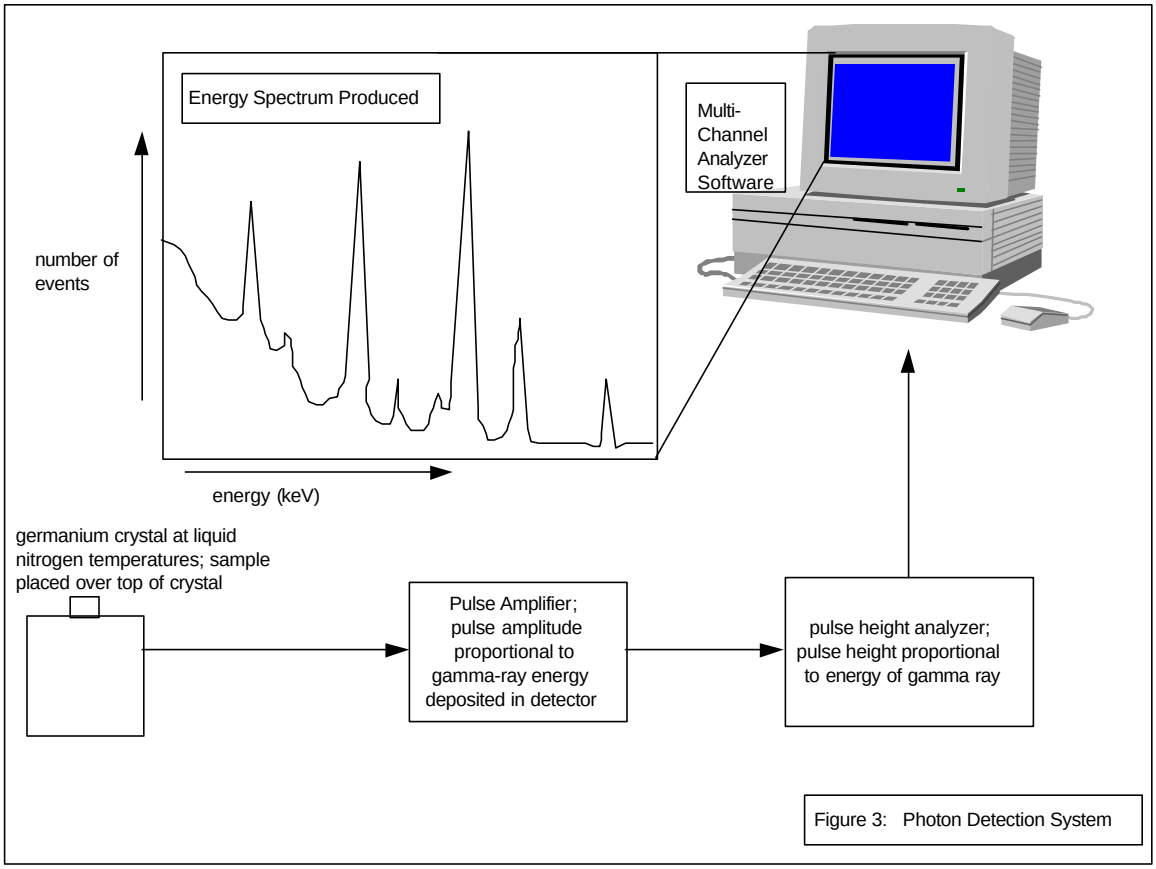

Figure 3: Photon Detection System

The Germanium crystal within this device was sensitive to incident photons at temperatures below 100^{o}K. Through a series of conversions these incident photons were represented by electronic impulses which were recorded on an energy spectrum.

Each test produced a gamma-ray spectrum that was used to identify and quantify the radionuclides present in each sample. Sample spectrums for the fly ash, bottom ash, and coal of plant number 4 are shown in Figures 4, 5, and 6.

The spectral data obtained from testing was analyzed using a gamma ray analysis program, GANAAS, to determine which radioactive species were present in the samples and to calculate the isotopic activities of each species, based on the energy and intensity of the gamma rays listed in Tables 1 and 2.

The pattern of gamma-ray energies in the energy spectrum of each sample was compared to a standard library of gamma rays to determine which radionuclides were present. In turn, the activities provided by GANAAS were used to calculate exposure levels from gamma radiation.

It should be noted that some gamma-ray energies are emitted by two or more isotopes, as is the case with ^{235}U and ^{226}Ra (E=186 keV). In these cases special attention is required to determine the contributions of each isotope to the spectral peak of interest. By using the activities of ^{210}Bi and ^{210}Pb and the emission probabilities of the 186 keV gamma ray for ^{235}U and ^{226}Ra, the contribution to the 186 keV peak of each of the two isotopes can be determined.

4.4 Modeling Methods to Characterize Coal Ash Embankment Radionuclide Emission and Exposure

4.4.1 Basis of Physical Model

Figure 7 and 8 show the physical condition of an embankment of coal ash and the scenario for exposure, respectively. The embankment can be from several centimeters to several meters thick. It is assumed to be comprised of incremental volumes of silicon (coal ash composition can be best approximated by silicon for gamma-ray attenuation purposes) that isotropically emit gamma rays. The gamma rays, that can “escape” an embankment, are those that travel in the positive z-direction. Each volume also has the ability to attenuate gamma rays, removing them or, at least, reducing their energy.

It is also assumed that no soil, asphalt, or concrete is covering the surface of the coal ash. This assumption provides a worst-case scenario, since anything covering the coal ash would have the ability to attenuate gamma rays.

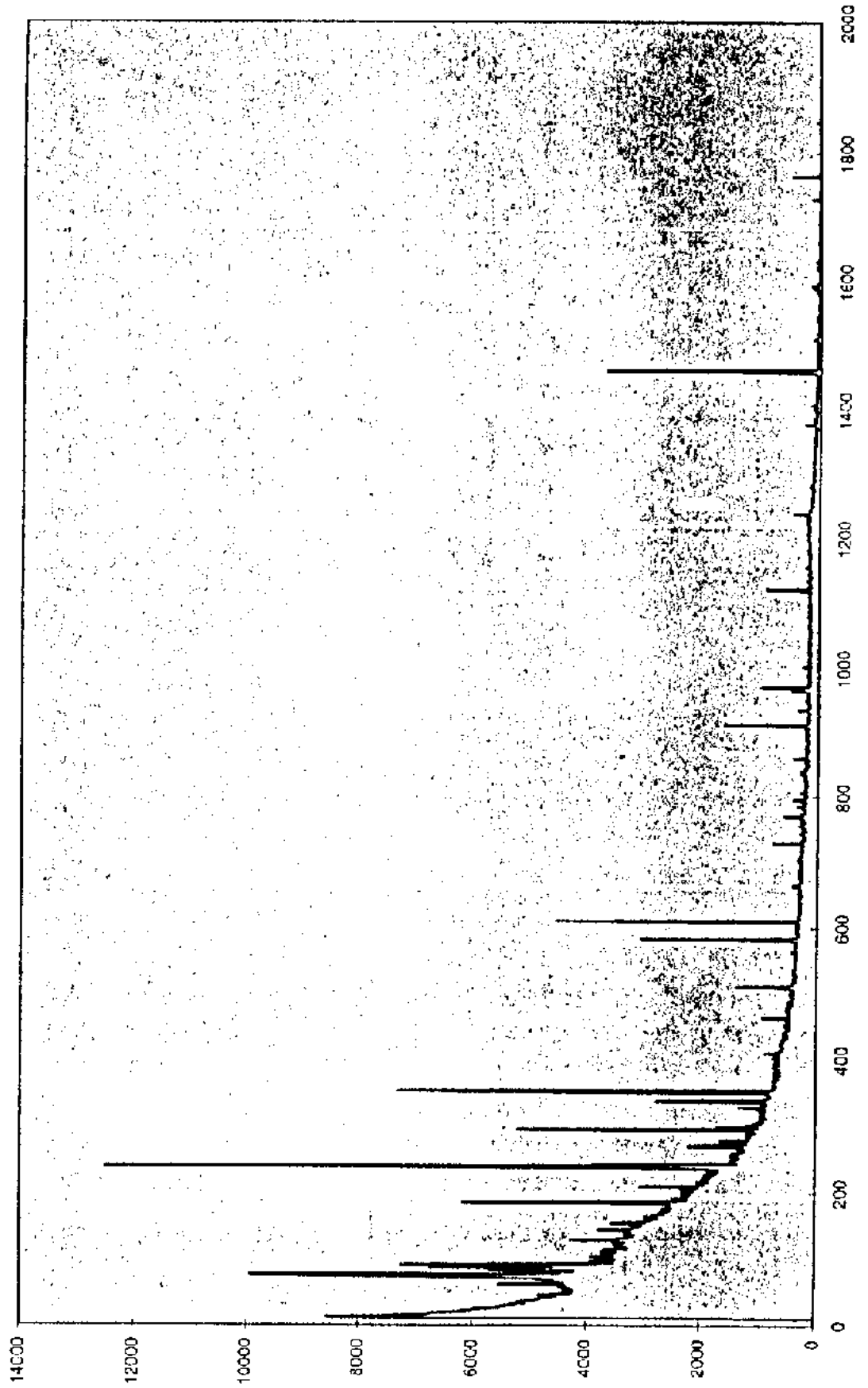

Figure 4: Gamma-ray Energy Spectrum of Fly Ash 04f

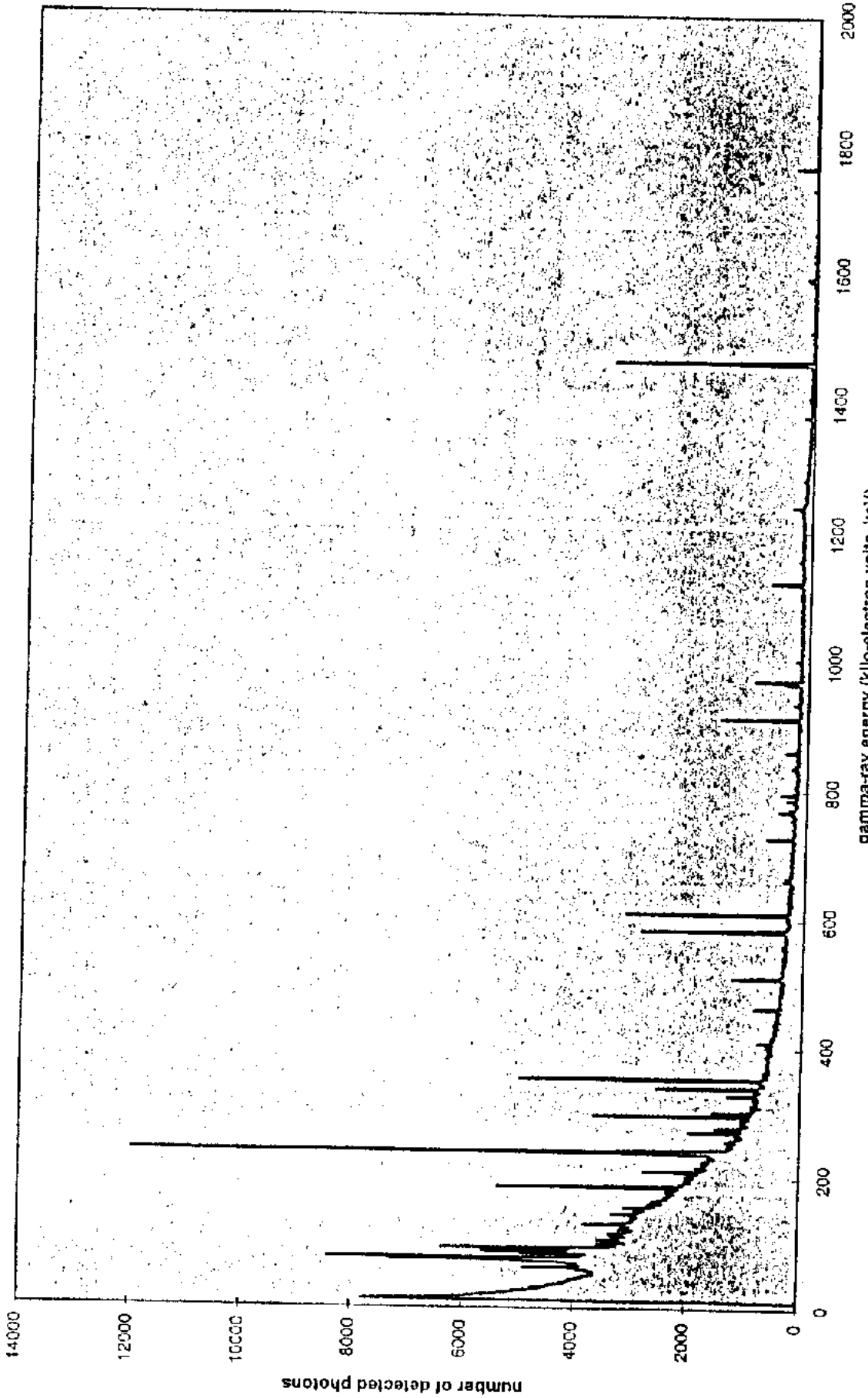

Figure 5: Gamma-ray Energy Spectrum of Bottom Ash 04b

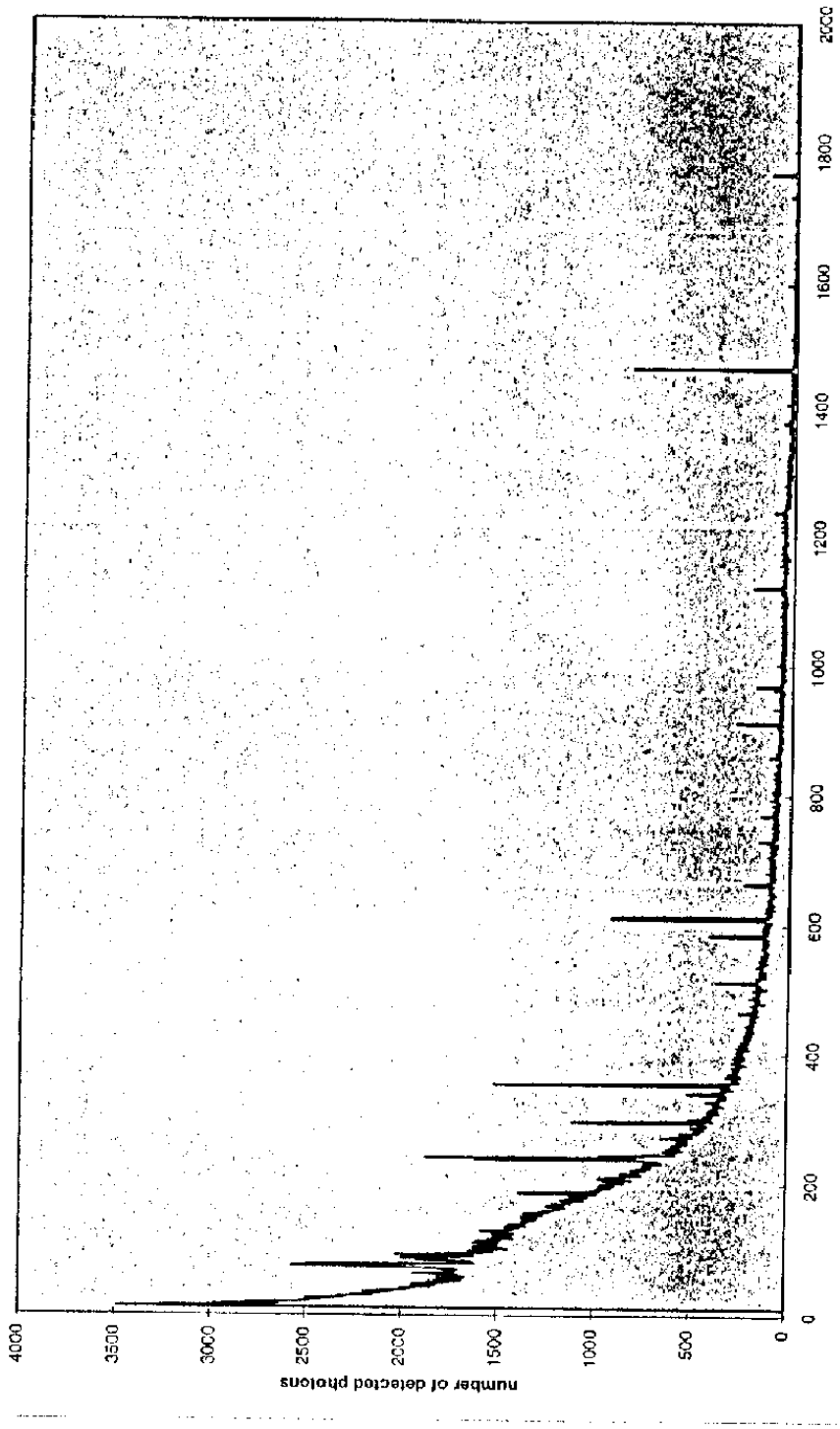

Figure 6. Gamma-ray Energy Spectrum of Coal 04

The air surrounding the embankment also attenuates some gamma rays, but attenuation in the air is much smaller than attenuation in the ash.

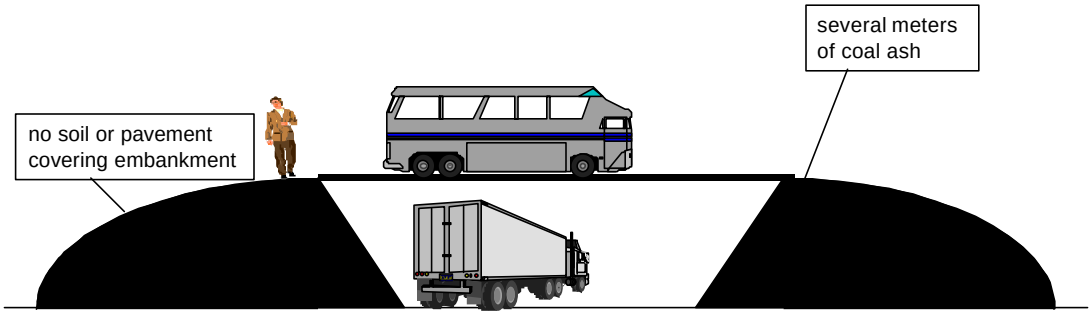

Figure 7: Physical Representation of a Coal Ash Embankment (overpass)

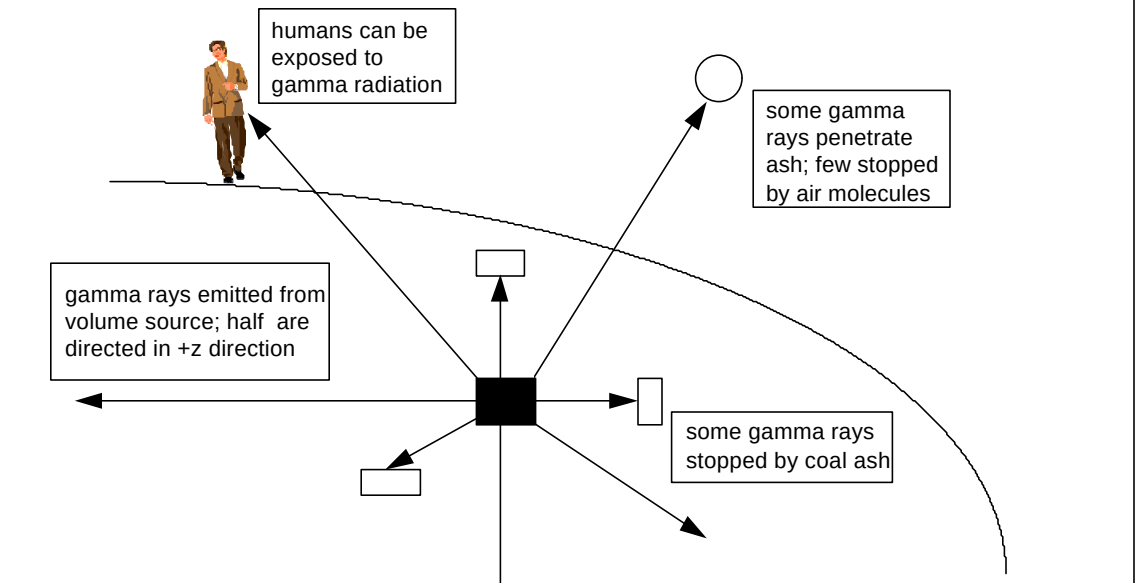

Figure 8: Fate of Gamma Rays in Coal Ash

4.4.2 Basis of Mathematical Model

The numerical output of GANAAS is the isotopic activity given in the metric unit of Bequerels (Bq), or decays per second, of a one-liter sample of material. The activity of an isotope is the rate at which

a radionuclide decays or the rate at which alpha or beta particles are emitted from a source. Activity is not the rate of exposure or the dose from gamma rays. The following is the method used to relate the activity to the dose equivalent experienced by the public from an embankment of coal ash.

4.4.2.1 Specific Activity

For purposes of this report the term "activity" is used to refer to the activity of those radionuclides that emit gamma rays. The inclusion of the other isotopes, i.e. all alpha and beta emitters, in the decay chains described would dramatically increase the actual activity of each sample. For analysis of gamma-ray emission and exposure, the activity of those radionuclides that emit gamma-rays is used.

The specific activity is the isotopic activity of a material per unit mass of material. It is given by:

$$A_i = \frac{A_{meas,i}}{M}, \qquad (1)$$

where A_i (Bq/g) is the specific activity of isotope, i, $A_{meas,i}$ (Bq) is the measured activity of isotope, i, and M is the mass of the one-liter test sample. The sum of the contributions of n gamma-emitting isotopes is the composite specific activity of gamma-emitting isotopes, A, of the sample:

$$A = \sum_{i=1}^{n} A_i. \qquad (2)$$

4.4.2.2 Gamma-ray Flux at the Surface of a Volume-distributed, Self-attenuating Source

An embankment of coal ash can be modeled as a volume-distributed, self-attenuating source of gamma rays. The volume is comprised of point sources that each contribute a small part of the gamma-ray emissions from the entire embankment. The integration of all of these point sources results in the total gamma-ray flux of the embankment.

The gamma-ray flux at the ash/air interface is the rate of gamma-ray emission per unit surface area. The surface flux of a gamma ray with energy, j, $\phi_{ash,j}$ (gamma rays/cm^2*sec), as a function of ash depth, b (cm), and gamma-ray energy, E (keV) is:

$$f_{ash,j}(b,E_j) = \frac{A_i * r * e_j}{2 * m(E_j)}\left[1 - E_2(m(E_j) * b)\right], \quad \text{(Glasstone/Sesonske, 1981)} \qquad (3)$$

where A_i is the specific isotopic activity, ρ (g/cm^3) is the uncompacted density of the test sample, E_j is the energy of gamma ray, j, μ_j (cm^{-1}) is the linear attenuation coefficient that corresponds to E_j, ε_j (gamma rays/decay) is the probability of emission of gamma ray, j, and E_2 is the second-order exponential integral function.

E_2 is the result of the mathematical integration of the contribution of point sources to the gamma-ray flux of a volume source. $E_n(x)$ is given by:

$$E_n(x) = x^{n-1}\int_x^\infty \frac{e^{-p}}{p^n}dp. \qquad \text{(Glasstone/Sesonske, 1981)} \qquad (4)$$

E_2 can be approximated by:

$$E_2(x) = \frac{e^{-x}(x+3)}{(x)^2 + 5x + 4}. \qquad \text{(Glasstone/Sesonske, 1981)} \qquad (5)$$

As the argument of E_2 becomes large, E_2 quickly approaches zero, and equation (3) simplifies to:

$$f_{ash,j}(b,E_j) = \frac{A_i * r * e_j}{2 * m(E_j)}. \qquad \text{(Glasstone/Sesonske, 1981)} \qquad (6)$$

As mentioned, μ is the linear attenuation coefficient. The value of $e^{\mu x}$ is the probability that a gamma ray will pass through an absorber with thickness x without interaction. Dividing μ by the absorber density, ρ, results in the mass attenuation coefficient (cm^2/g). Both coefficients are a function of gamma-ray energy. Table 4 contains the mass attenuation coefficients of silicon at relevant energies.

Table 4 also contains the gamma-ray emission probabilities for relevant gamma rays.

Table 4: Nuclear Data for Relevant Isotopes and Energies

Isotope	Energy (keV)	Probability of gamma-ray emission	Mass absorption coefficients for tissue (cm^2/g)	Mass attenuation coefficients for silicon (cm^2/g)	Mass attenuation coefficients for air (cm^2/g)
Ac-228	338	0.1126	0.0314	0.1026	0.1019
Ac-228	463	0.0450	0.0319	0.0900	0.0899
Ac-228	795	0.0434	0.0311	0.0708	0.0708
Ac-228	911	0.2660	0.0305	0.0667	0.0667
Ac-228	965	0.0505	0.0302	0.0647	0.0648
Ac-228	969	0.1633	0.0302	0.0646	0.0647
Bi-212	727	0.0664	0.0314	0.0741	0.0742
Bi-212	1079	0.0061	0.0296	0.0614	0.0614
Bi-212	1620	0.0149	0.0271	0.0500	0.0500
Bi-214	609	0.4460	0.0319	0.0798	0.0800
Bi-214	768	0.0476	0.0312	0.0721	0.0722
Bi-214	1120	0.1470	0.0294	0.0602	0.0603
Bi-214	1155	0.0170	0.0293	0.0593	0.0593
Bi-214	1238	0.0578	0.0289	0.0570	0.0570
Bi-214	1378	0.0411	0.0282	0.0541	0.0541
Bi-214	1408	0.0249	0.0280	0.0535	0.0535
Bi-214	1509	0.0222	0.0276	0.0516	0.0516
Bi-214	1764	0.1510	0.0265	0.0480	0.0479
Bi-214	1784	0.0300	0.0265	0.0477	0.0476
Bi-214	2119	0.0117	0.0252	0.0437	0.0435
Bi-214	2448	0.0155	0.0240	0.0411	0.0406
Cs-137	662	0.8520	0.0317	0.0772	0.0774
K-40	1461	0.1067	0.0278	0.0525	0.0525
Pb-212	239	0.4350	0.0300	0.1181	0.1164

Pb-212	300	0.0325	0.0312	0.1070	0.1060
Pb-214	295	0.1820	0.0311	0.1079	0.1069
Pb-214	351	0.3510	0.0315	0.1011	0.1005
Ra-226	186	0.0351	0.0290	0.1289	0.1260
Tl-208	277	0.0245	0.0308	0.1111	0.1099
Tl-208	583	0.3058	0.0319	0.0813	0.0815
Tl-208	861	0.0448	0.0308	0.0684	0.0685
Tl-208	2615	0.3588	0.0234	0.0398	0.0391
U-235	144	0.1096	0.0281	0.1431	0.1361
U-235	163	0.0508	0.0285	0.1353	0.1311
U-235	186	0.5720	0.0290	0.1290	0.1261
U-235	202	0.0100	0.0293	0.1246	0.1226
U-235	205	0.0501	0.0294	0.1240	0.1221

4.4.2.3 Gamma-ray Flux at a Distance above the Ash

The gamma-ray flux is slightly attenuated by air. It can be considered to be the flux at a point above an infinite plane source, i.e. the ash embankment. The flux, $\phi_{air,j}$ (gamma rays/cm^2*s), for gamma ray, j, at a point that is a distance, z (cm), above the ash/air interface is:

$$f_{air,j}(z, E_j) = f_{ash,j}(z, E_j) * e^{-z\mu(E_j)}, \qquad (7)$$

where μ (cm^{-1}) is the linear attenuation coefficient for air. Mass attenuation coefficients for air are analogous to those of ash and are listed in Table 4.

4.4.2.4 Dose Rate to the Human Body

The dose rate, $\dot{D}$, is the rate at which radiation deposits energy into an absorbing material, which, in the case of this study, is human tissue. It is a function of photon flux and energy and is given by:

$$\dot{D}_j(E_j) = 5.76*10^{-5}\,\frac{f_{air,j}(z,E_j)*E_j*m_a(E_j)}{r}. \text{ (Glasstone/Sesonske 1981)} \qquad (8)$$

μ_a is the linear energy absorption coefficient (cm^{-1}). It is analogous to the linear attenuation coefficient except it represents the rate of energy deposition, not the rate of particle interaction. Table 3 lists the mass energy adsorption coefficients, μ_a/ρ, for soft human tissue. E_j is the gamma-ray energy in keV. Dose rate, $\dot{D}_j(E_j)$, is expressed in units of mrads, or 10^{-3} rads (radiation absorbed dose), per hour. The dose rates of each gamma-ray energy, E_j, of a single source may be summed to obtain the total dose rate of the source.

4.4.2.5 Dose Equivalent

The dose equivalent rate, $\dot{D}$, of radiation is a measure of the amount of energy the body absorbs per gram of tissue, not the biological effect of the absorbed radiation. Since the biological effect depends on the type of radiation and other factors, the dose equivalent is used to quantify the biological effect. The unit of dose equivalent is the rem (radiation equivalent in mammal). The dose equivalent rate, $\dot{H}$, is:

$$\dot{H}(E) = \dot{D}(E)*QF. \text{ (Glasstone/Sesonske 1981)} \qquad (9)$$

QF is the quality factor. It represents the potential for a specific type of radiation to deposit energy over a short distance in tissue. For gamma rays of interest to this study, QF ≈ 1, and for alpha particles, QF ≈ 10. The dose equivalent rates for all energies of a single source may be summed to obtain the total dose equivalent rate of the source.

4.5 Significant Qualifications and Assumptions Associated with the Applied Model

4.5.1 Model Assumptions

There were three important assumptions used during the course of this modeling effort, as follows:

1. For calculating dose levels, a 2000 hour work year was assumed for occupational exposures (i.e., project workers with the highest possible exposure levels). In contrast, public exposure stemming from infrequent contact with these types of embankments (measured in time increments of seconds or, at most, minutes) would be much less than the occupational exposure levels.

2. For calculation of doses the embankment of coal ash was represented as a self-attenuating volumetric source with a finite thickness without any clay, topsoil, or asphalt covering. This represents a conservative assessment, however, given the fact that this type of attenuating cover will almost certainly be employed (e.g., as an erosion control measure).

3. The vital (i.e., critical organ location) zone of the human body was assumed to be one meter above ground level when in the standing position.

4.5.2 Model Qualifications

The applied modeling approach was based on ‘single sample’ results obtained at random from ash piles, steam generators, and coal piles. It should be noted, however, that the observed activities might well change from location to location in a single coal mine, depending on geologic history and deposit formation. Since the location inside a coal seam and, in many cases, the specific coal mine is unknown, the values presented in this report would be somewhat representative of each specific coalmine and its by-product ash. If a sampling scheme were to be developed and exact sampling information (i.e. location in the mine), a “radiation map” could be constructed showing larger activities and lower activities within each seam of coal.

It is also possible that burning processes and steam generator types change the activities of the ash. For example, if the exact same coal were burned in a stoker boiler and a tangential boiler, the activities of the resulting ashes could be different.

In many cases, the measured activities appear to be large in comparison to other materials, but in reality the activities are very minuscule in comparison to doses experienced from many other sources of radiation. (see, for example, the KCl salt substitute activities, as well as the ‘background radiation’ information)

Finally, these exposures and doses were calculated using uncompacted densities determined in the laboratory. Under actual conditions the ash would be compacted to about 1.5 times the uncompacted density. Exposures generally decrease as densities increase.

CHAPTER 5

Results and Discussion

5. 1 Activities of Coal, Bottom Ash, Fly Ash, and Commingled Ash

Figures 9, 10, and 11 show the isotopic activities of the ten radionuclides that emit significant numbers of gamma rays for samples of coal, bottom ash, and ESP fly ash which belong to "complete sets." Figure 12 shows the isotopic activities of commingled ash samples. The labeling scheme used to establish sample ID's in these figures was as follows:

1) the first number of the sample ID identifies the respective power plant being tested,

2) the following letter (f, b, or c) establishes the form of the material, respectively including 'fly ash,' 'bottom ash,' or 'coal,' and

3) a concluding number was then added, if necessary, when multiple samples of the same type were obtained and tested at each location. For example, sample ID '10c2' corresponds to the second of two 'coal' samples tested from power plant #10.

By comparing the vertical scale of the four plots, it can be seen that the activities of coal were much less than those of any type of ash. This was expected because the burning of coal removed all of the carbon and volatiles, leaving the impurities, including radionuclides, in the ash. The combustion of coal concentrated the activity by an order of magnitude or more, since the ash content of coal was about 10%. In most samples there were four gamma-ray emitting isotopes, ^{214}Pb, ^{214}Bi, ^{226}Ra, and ^{40}K, that regularly appeared as significant contributions to the activity of the sample. ^{214}Pb and ^{214}Bi appeared because they emitted many significant gamma rays. They will always have similar activities because they were part of the same decay chain, ^{238}U, and have relatively equal half-lives. The appearance of ^{226}Ra coincided with ^{214}Pb and ^{214}Bi since it was also part of the ^{238}U decay chain. ^{40}K was present in the greatest quantities because it was much more abundant in nature than almost any other radionuclide.

When looking at these plots, it is tempting to draw conclusions that a certain coal, that may or may not have a high activity when compared to other coals, produces an ash with a high activity. Caution should be taken because these sets of samples do not represent a perfect conversion from coal to ash.

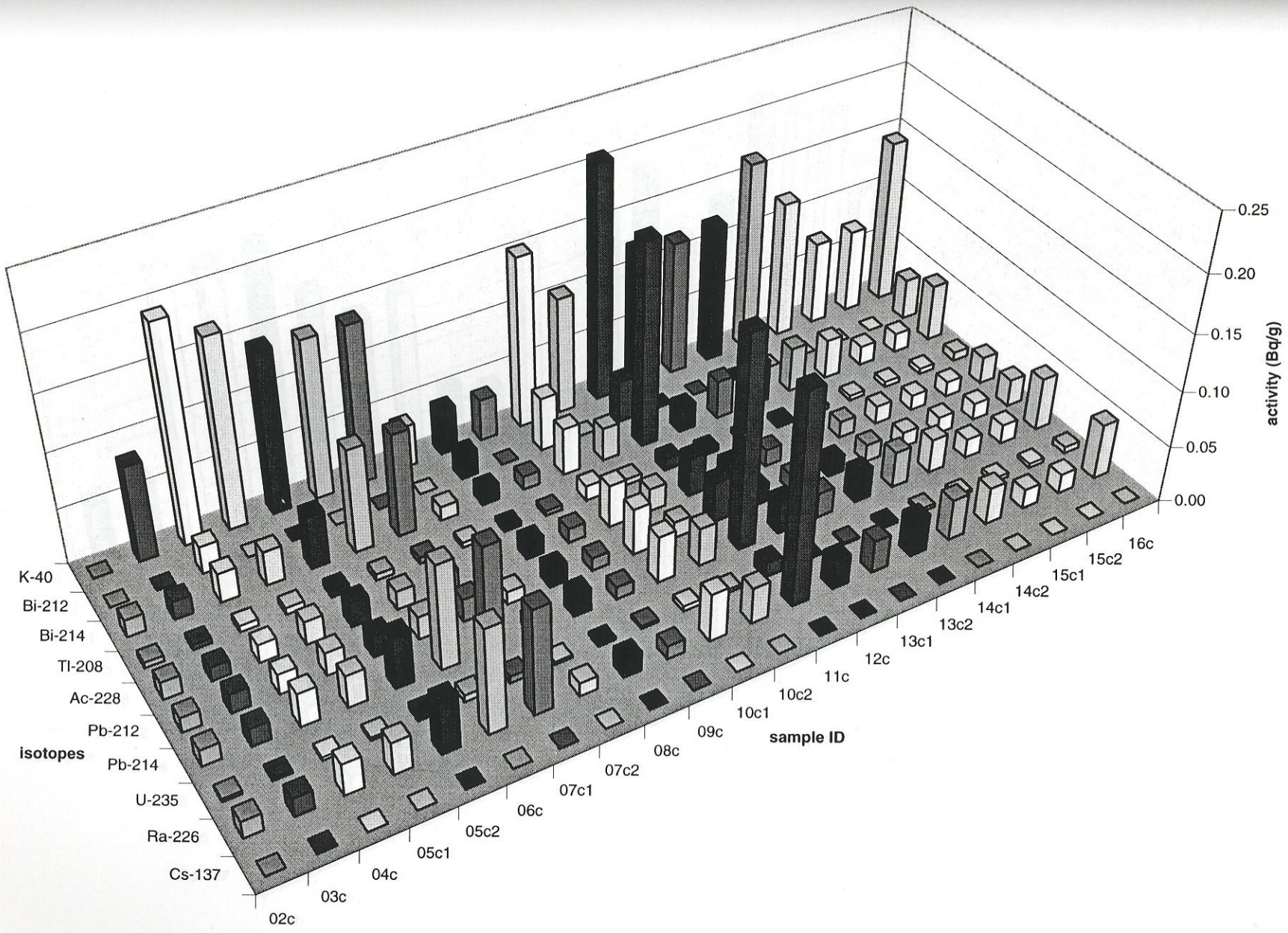

Figure 9: Isotopic Activities of Gamma-Ray Emitters in Coal

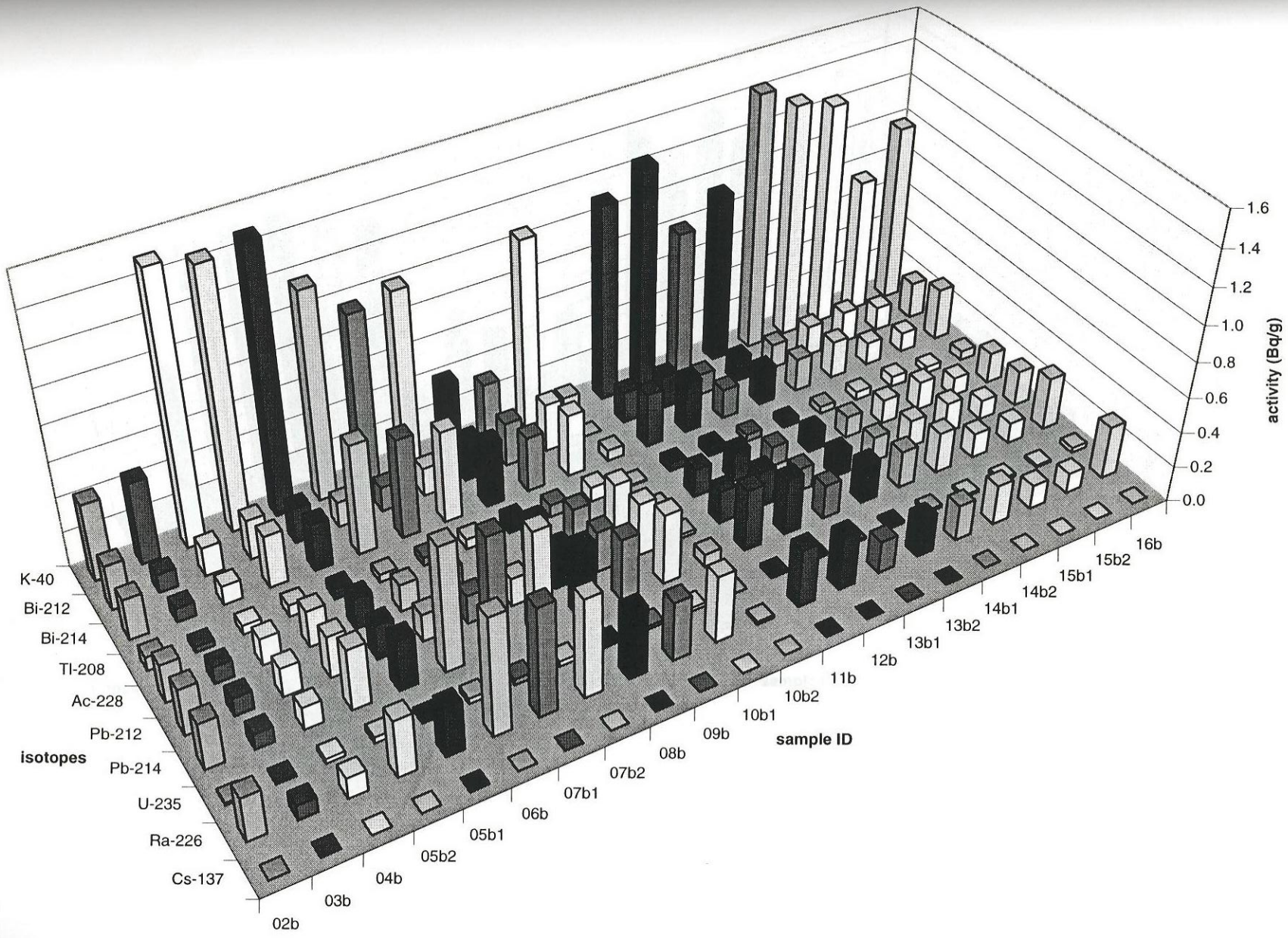

Figure 10: Isotopic Activities of Gamma-Ray Emitters in Bottom Ash

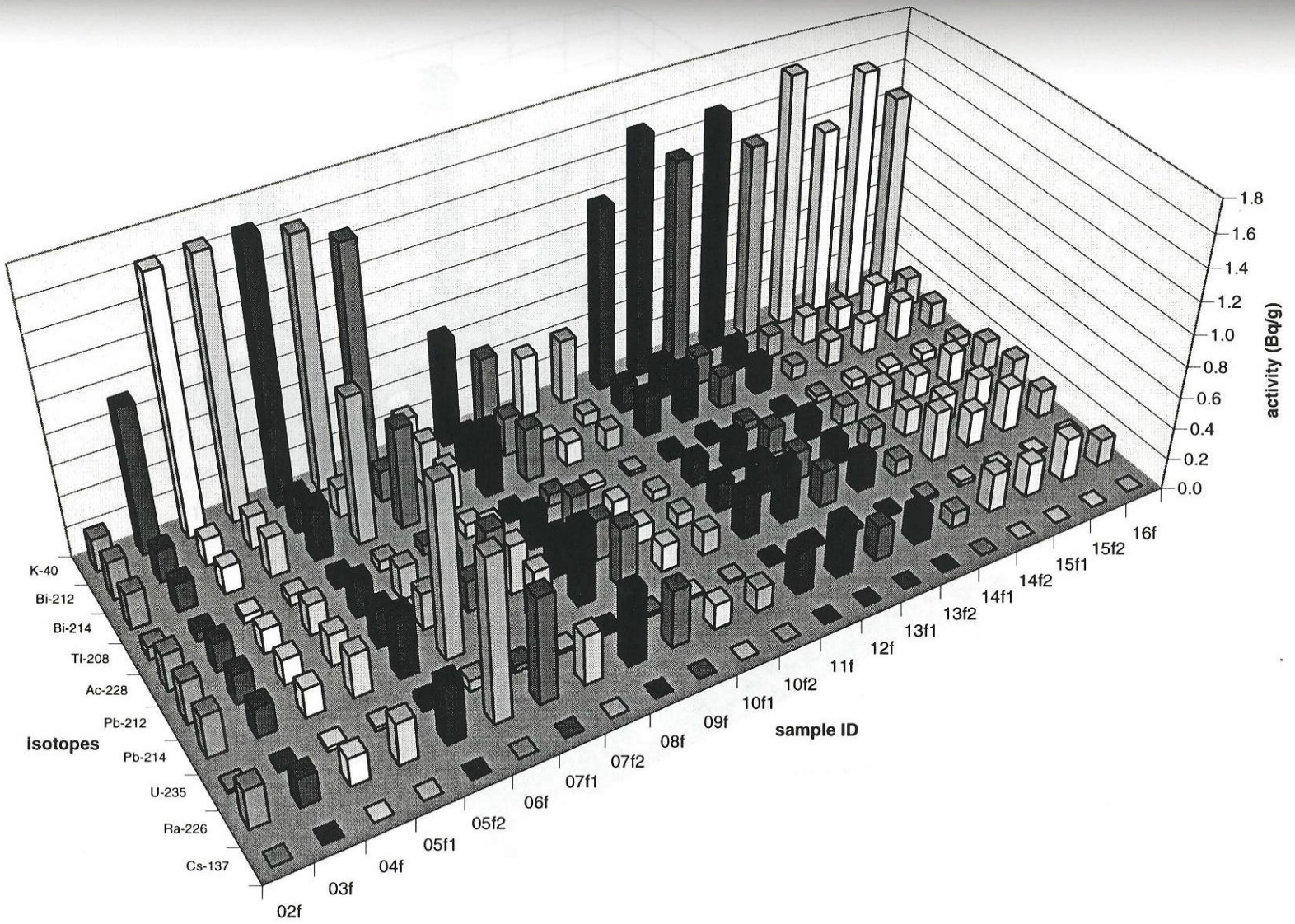

Figure 11: Isotopic Activities of Gamma-Ray Emitters in Fly Ash

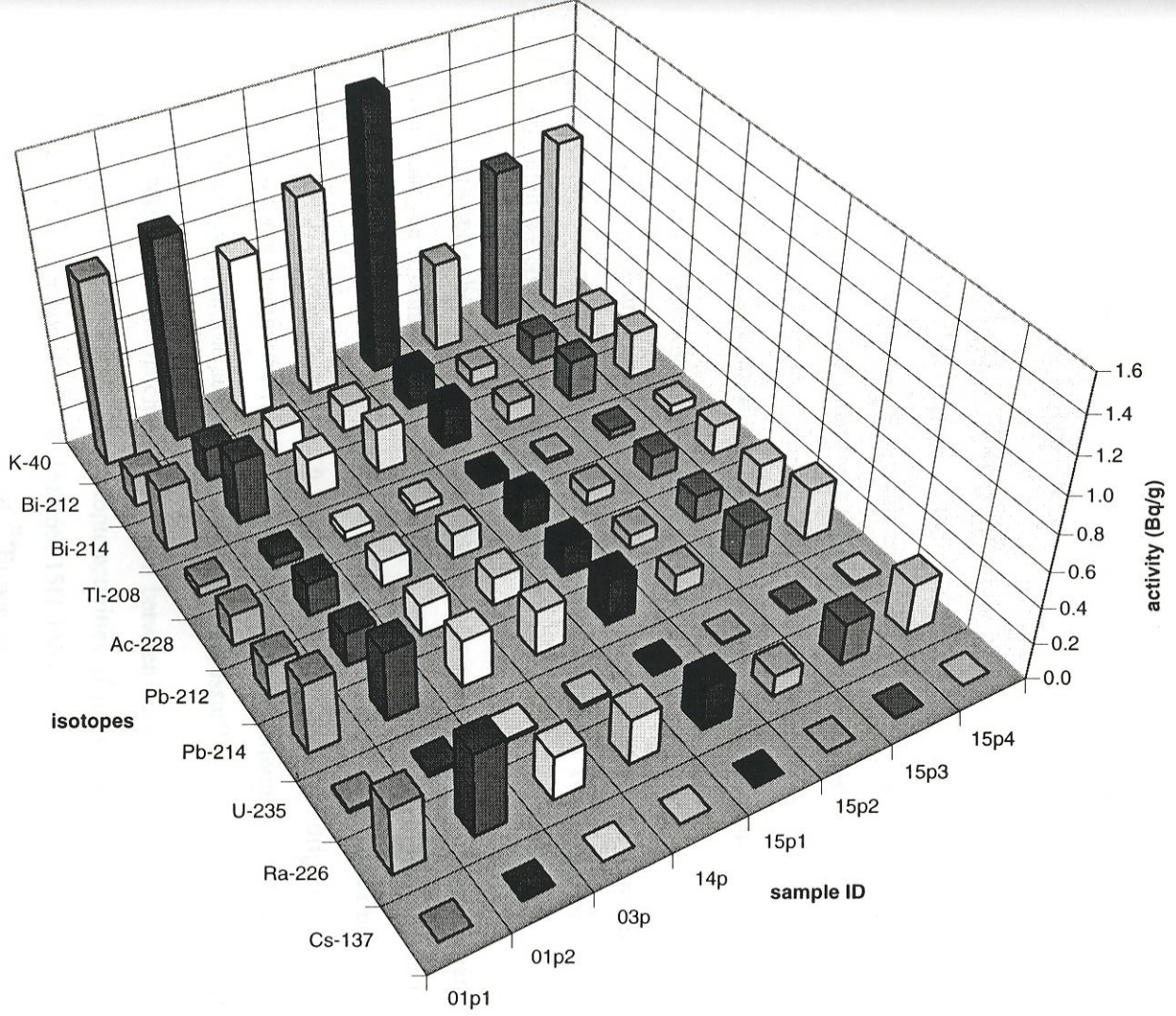

Figure 12: Isotopic Activities of Gamma-Ray Emitters in Commingled Ash

Different burning processes or steam generators have the potential of altering the isotopic composition of the ash. Many removal mechanisms such as isotopic exchange at high temperatures, volatilization of gaseous radon or other nuclides, and electrostatic forces and Brownian motion in airstreams, may have an effect on radionuclide composition. The ability of a radionuclide to sorb to the surfaces of different types of ash also affects the relative magnitudes of activities. Changes in the nuclide composition have an effect on the nuclide conversion between coal and ash. Consequently, if the same coal were to be burned in two different power-generation units, two ashes that were isotopically and radiogenically different could be produced.

Another point to consider is the origin of coal. Many factors contribute to the distribution of radionuclides in the earth's crust, including coal deposits. Different deposits of coal will have different isotopic compositions. In fact, isotopic compositions in the same coal mine will vary. Sampling for this study was done in a somewhat random fashion. In some cases the coal collected from different power stations originated from the same mine, but those samples have different activities. In general, a single coal sample was not necessarily representative of an entire coal mine.

In addition to the properties of combustion and coal, radiation measurements are associated with large amounts of uncertainty, especially when only one sample of a material is available for analysis. Uncertainty in nuclear measurements can be as high as twenty percent. This uncertainty applies throughout the course of this study.

Despite these qualifiers, patterns of isotopic activities can be seen that coincide between some coals and the resultant ashes. For example, set 05-1 (coal 05c1, bottom ash 05b1, and fly ash 05f1) shows the same pattern of isotopic activities. The activities have different absolute magnitudes but similar relative magnitudes, which suggests that 05b1 and 05f1 originated from 05c1, and the burning process did not change the ratios of the radioisotopes. Although the ash of other sets actually originated from the parent coal, the activity patterns do not necessarily indicate a "matched set."

More obvious matches can be seen between bottom ashes and fly ashes of each set of samples than between coal and ash. The isotopic activities of fly ash and bottom ash are compared in Figure 13. With some exceptions (02f/02b, 16f/16b, 15f1/15b1, 14f1/14b1) the same pattern appeared in all sets of fly and bottom ashes. The activity of ^{214}Bi was larger than ^{212}Bi, ^{214}Pb is larger than ^{212}Pb, and, in most cases, the ^{40}K activity was much larger than all other isotopic activities.

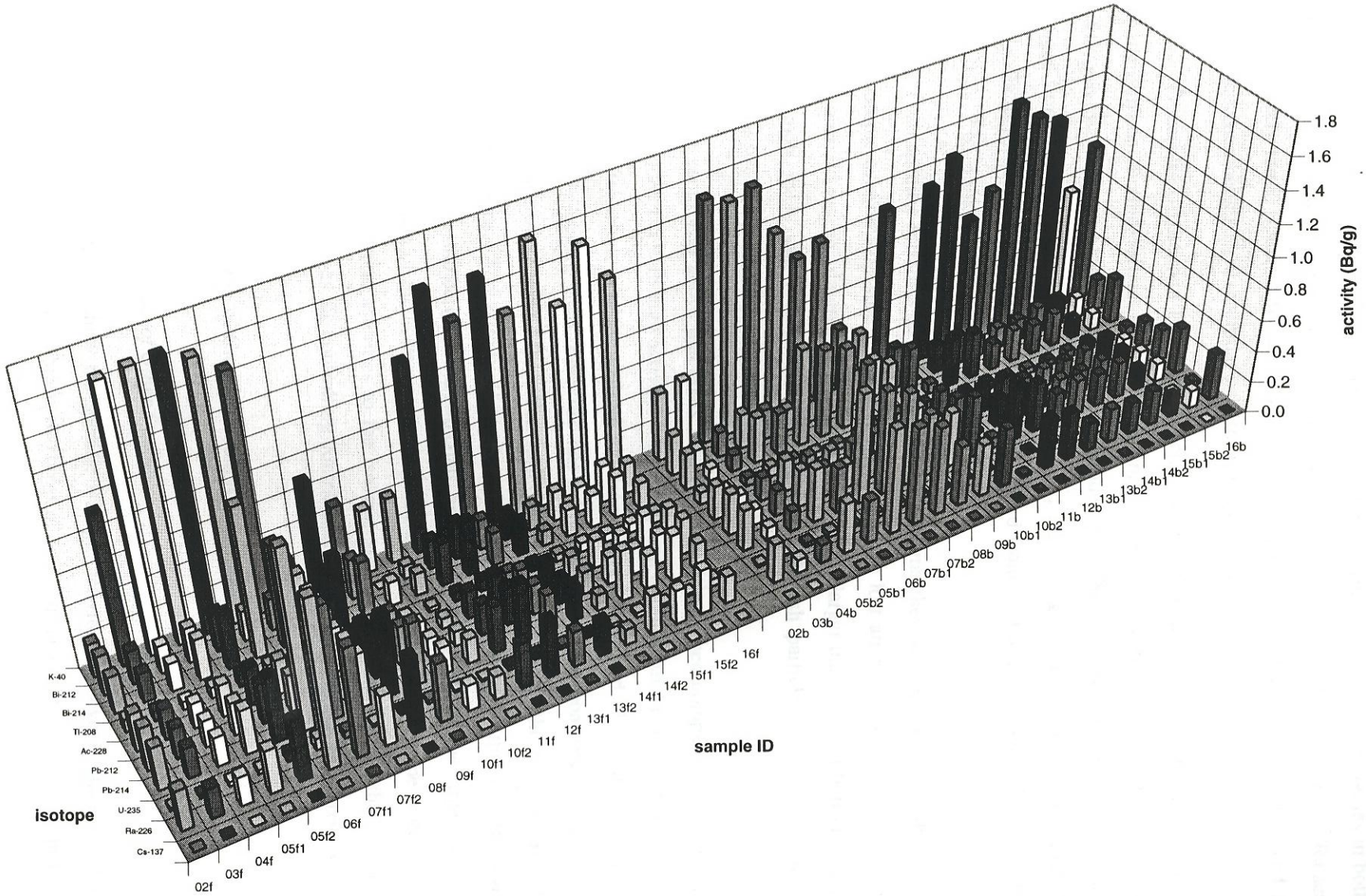

Figure 13: Comparison of Isotopic Activities of Gamma Emitters in Fly Ash and Bottom Ash

Looking at the set 06f1/06b1, the ^{40}K activity was larger than all other activities in both types of ash, and the ^{214}Bi and ^{214}Pb activities were also large in comparison to other activities. ^{226}Ra activity was also relatively large. Other activities have the same relative magnitudes in the bottom and fly ash of power plant number six. These similar patterns suggest "matching" ash samples.

Figures 14, 15, 16, and 17 depict the total specific activities of significant gamma-ray producing isotopes in coal, bottom ash, fly ash, and commingled ash, respectively. These plots include all samples of fly ash involved in the study. Earlier plots included only ESP samples. The observed activities of coal ranged from 0.081 Bq/g to 0.902 Bq/g, and averaged 0.292 Bq/g. Bottom ash ranged from 0.281 to 3.71 Bq/g, fly ash from 1.12 to 5.50 Bq/g, and commingled from 1.13 to 2.99 Bq/g. The ash averages were 2.39 Bq/g, 2.65 Bq/g, and 2.34 Bq/g, respectively.

Total specific activities do not indicate the same matches between coal, fly ash, and bottom ash as the isotopic activities because the relative patterns were lost after combining isotopic activities into absolute sums. In general, the total activities of fly ash were larger than those of bottom ash (Figure 18). This was most likely due to higher surface areas on fly ash particles that provide more sites for radionuclides to sorb, producing a higher activity on fly ash.

It was mentioned above that coals with high activities, when compared to other coals, did not necessarily produce ash with high activity, when compared to other ash. This point can be illustrated by comparing the total activities of the coal/ash sets from plants 06, 11, and 12. As mentioned, coal 11c has the highest activity among coals, coal 12c has a lower activity, and coal 06c has an activity that is roughly the average of 12c and 11c. Comparing the total activities of the resultant ash of each coal yields different results. The activity of bottom ash 12b was greater than that of 11b, which was the opposite of the relationship between the two coals, and the activity of 6b was the largest of all bottom ash samples. The activities of the corresponding fly ash samples followed the same pattern as the bottom ash. By comparing the total activities to the isotopic activities, it can be seen that in most cases the activity of Potassium-40 was the controlling contributor to the total. As the ^{40}K activity varied, the total activity varied in the same manner.

5.2 Comparison of Activities of Coal and Coal Ash to Other Materials

5.2.1 Specific Isotopic Activities of Other Samples

Figures 19, a and b, show the specific isotopic activities of several other materials. Natural construction materials included red brick, two types of clay from central Indiana, two types

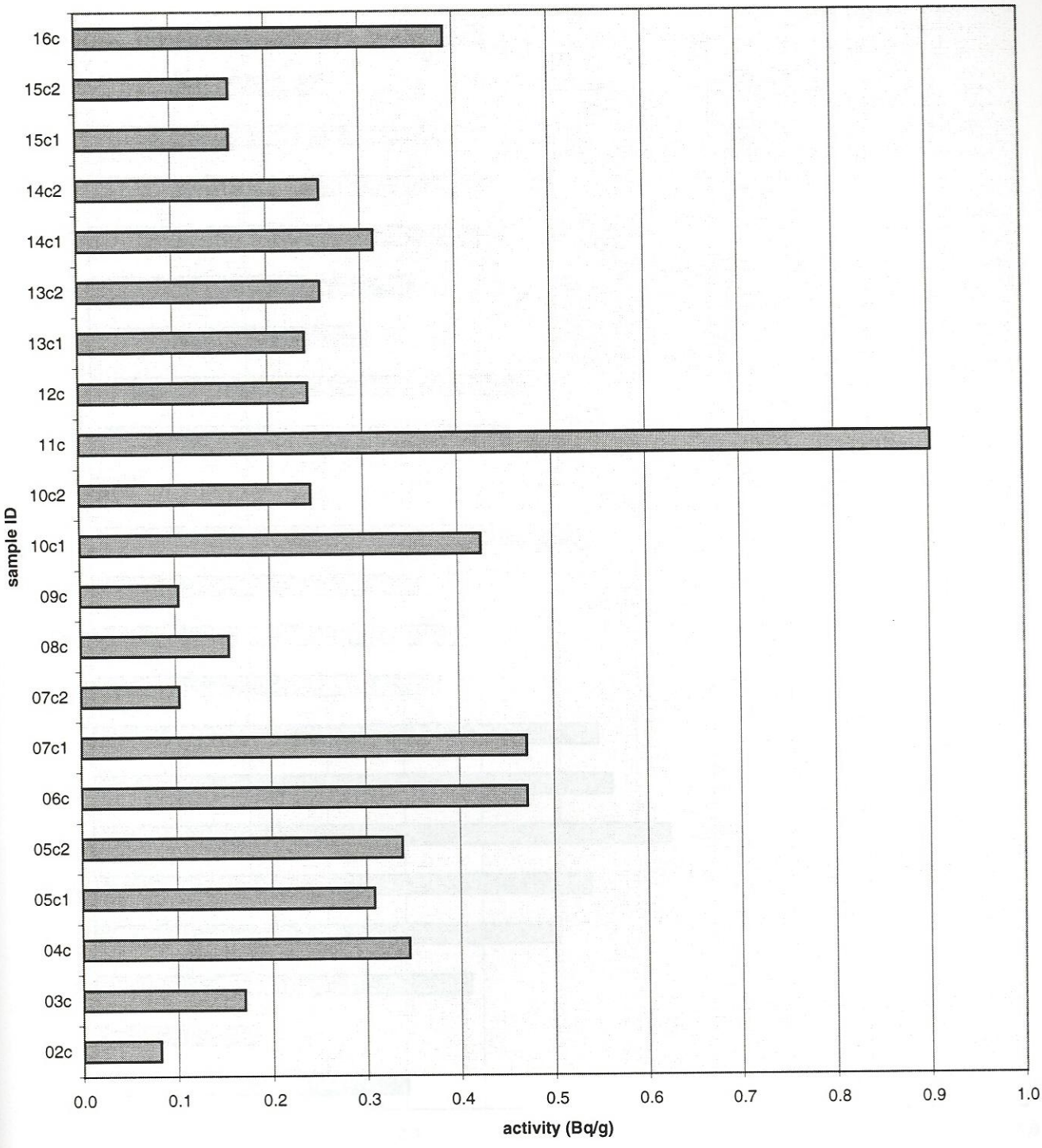

Figure 14: Total Activities of Gamma-ray Emitters in Coal Samples

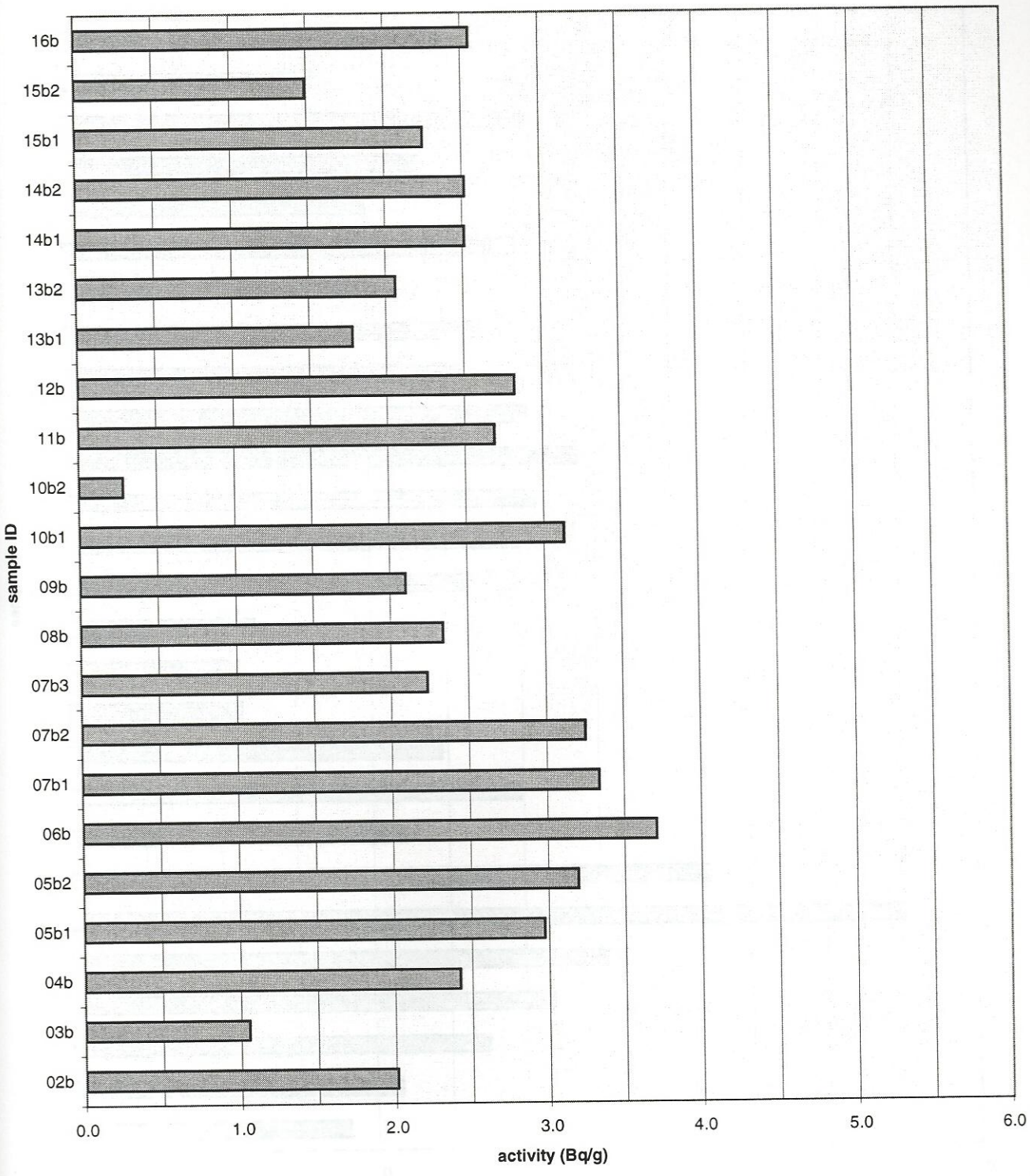

Figure 15: Total Activities of of Gamma-ray Emitters in Bottom Ash Samples

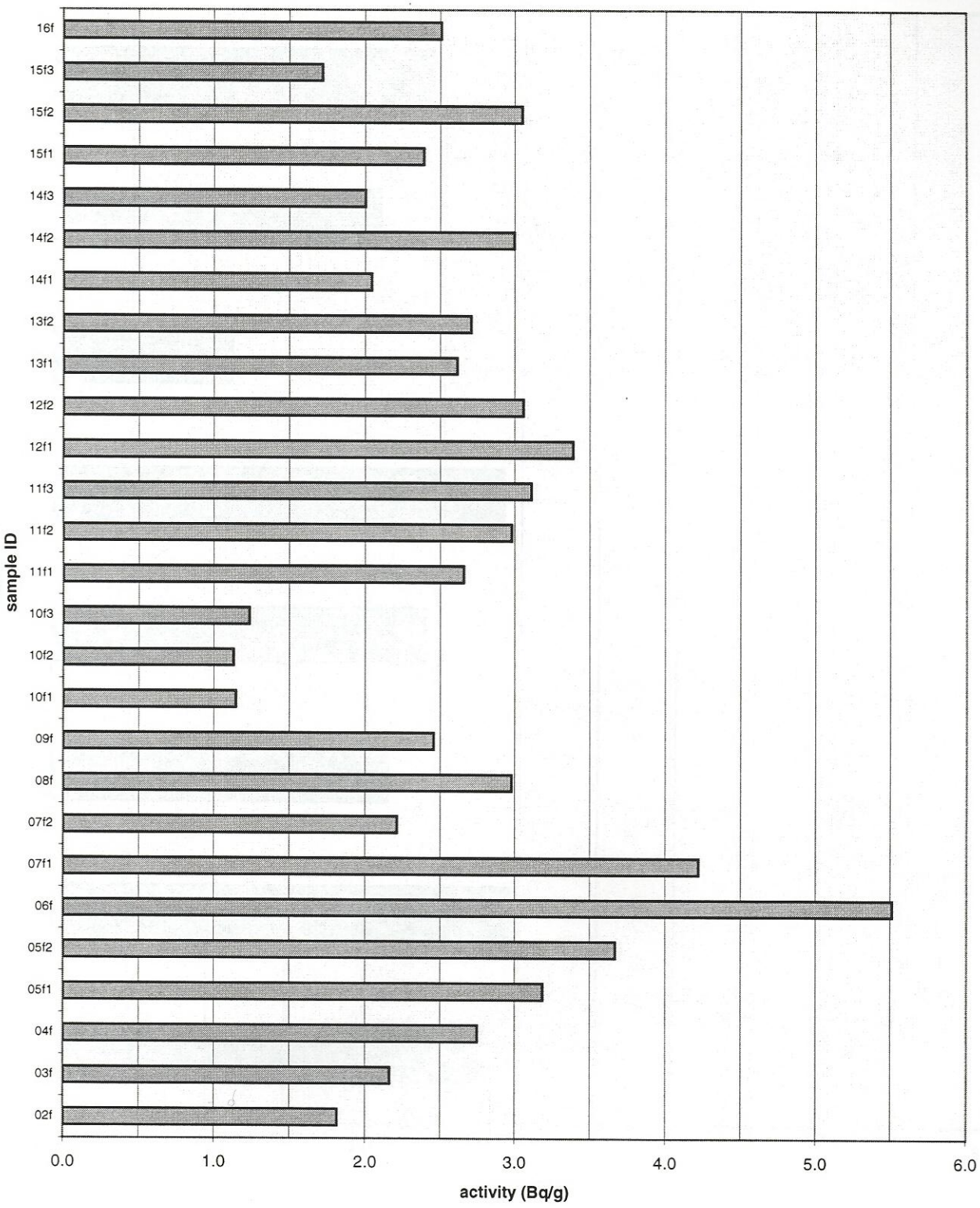

Figure 16: Total Activities of Gamma-ray Emitters in Fly Ash

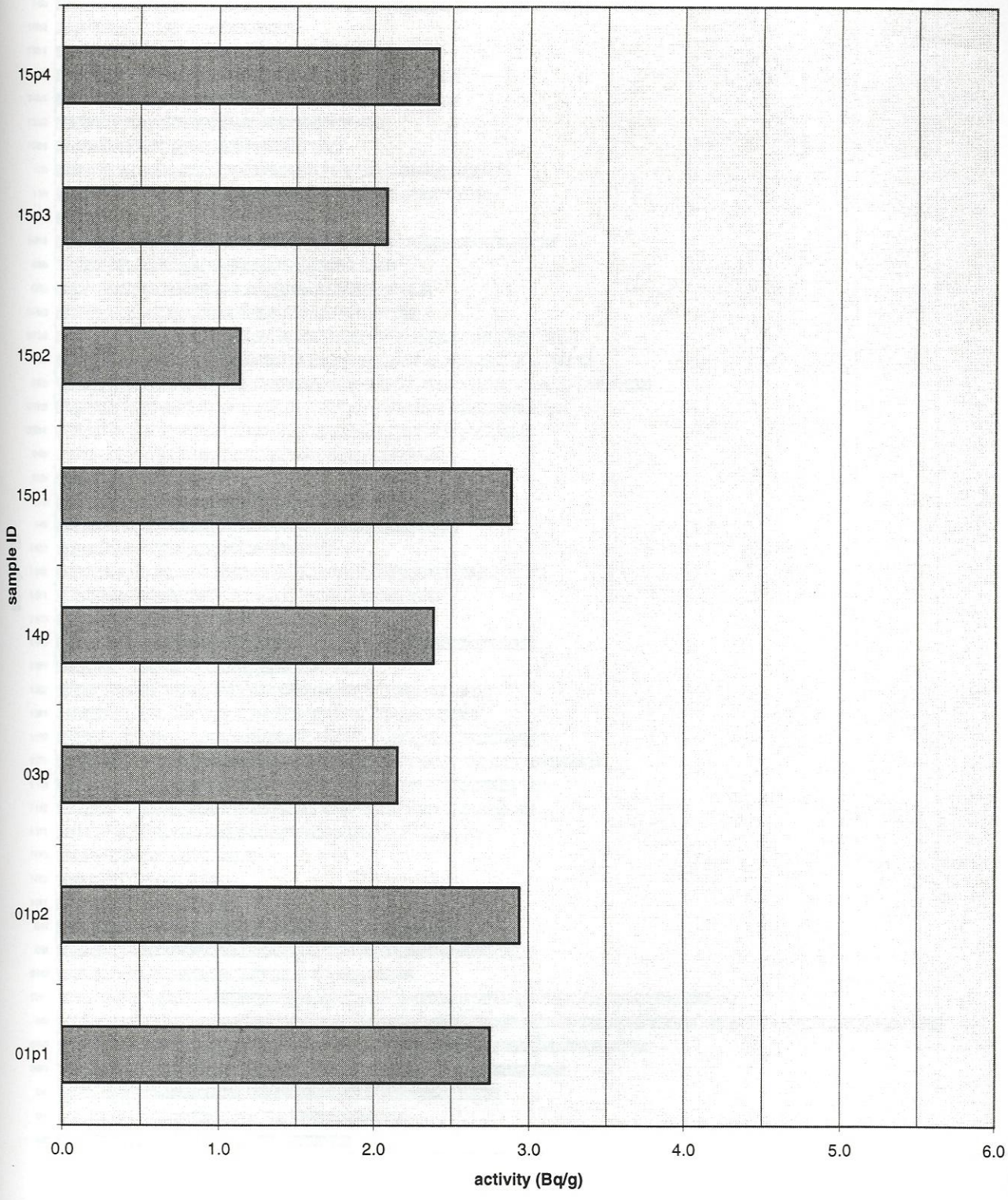

Figure 17: Total Activities of Gamma-ray Emitters in Commingled Ash

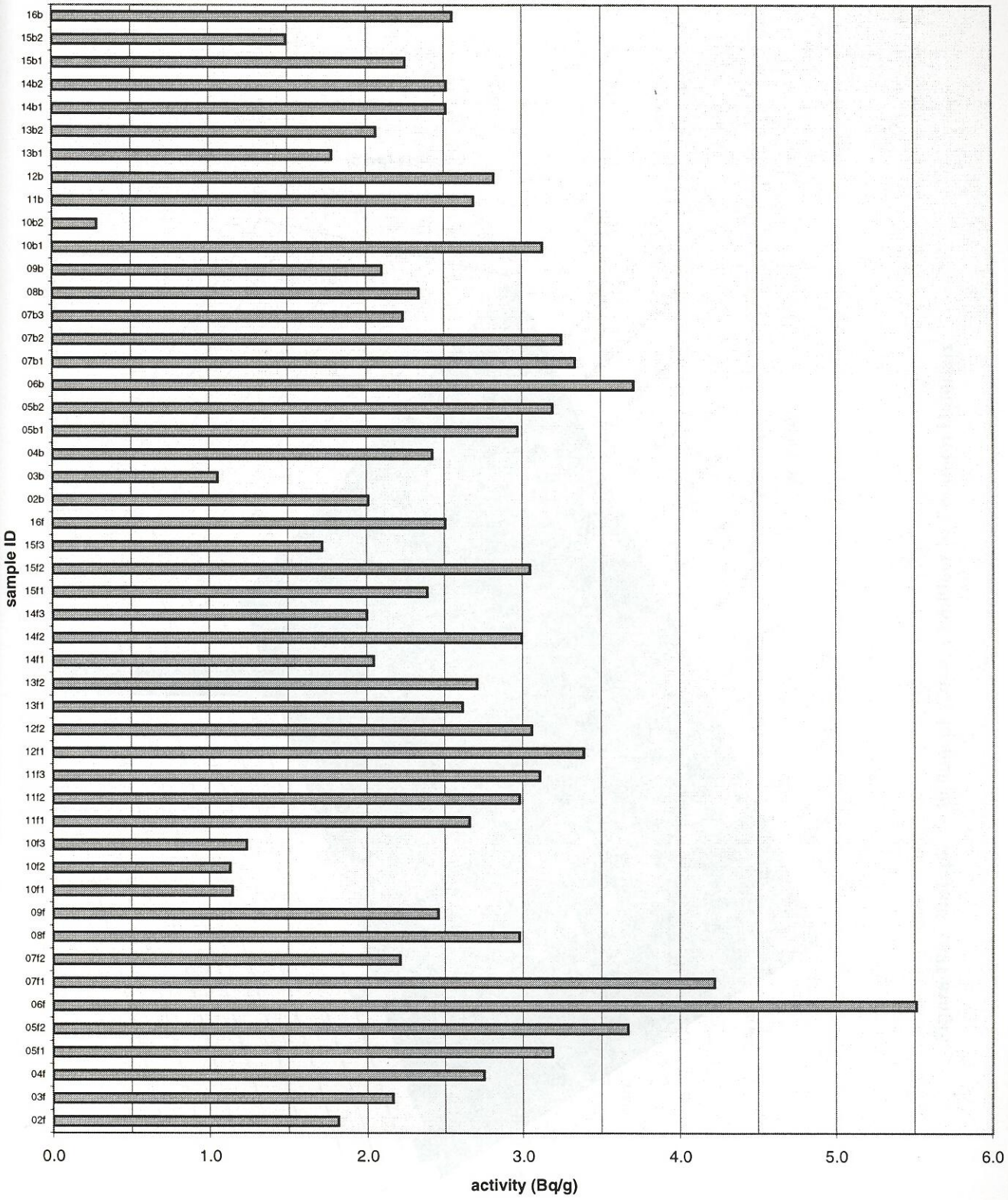

Figure 18: Total Activities of of Gamma-ray Emitters in Bottom and Fly Ash Samples

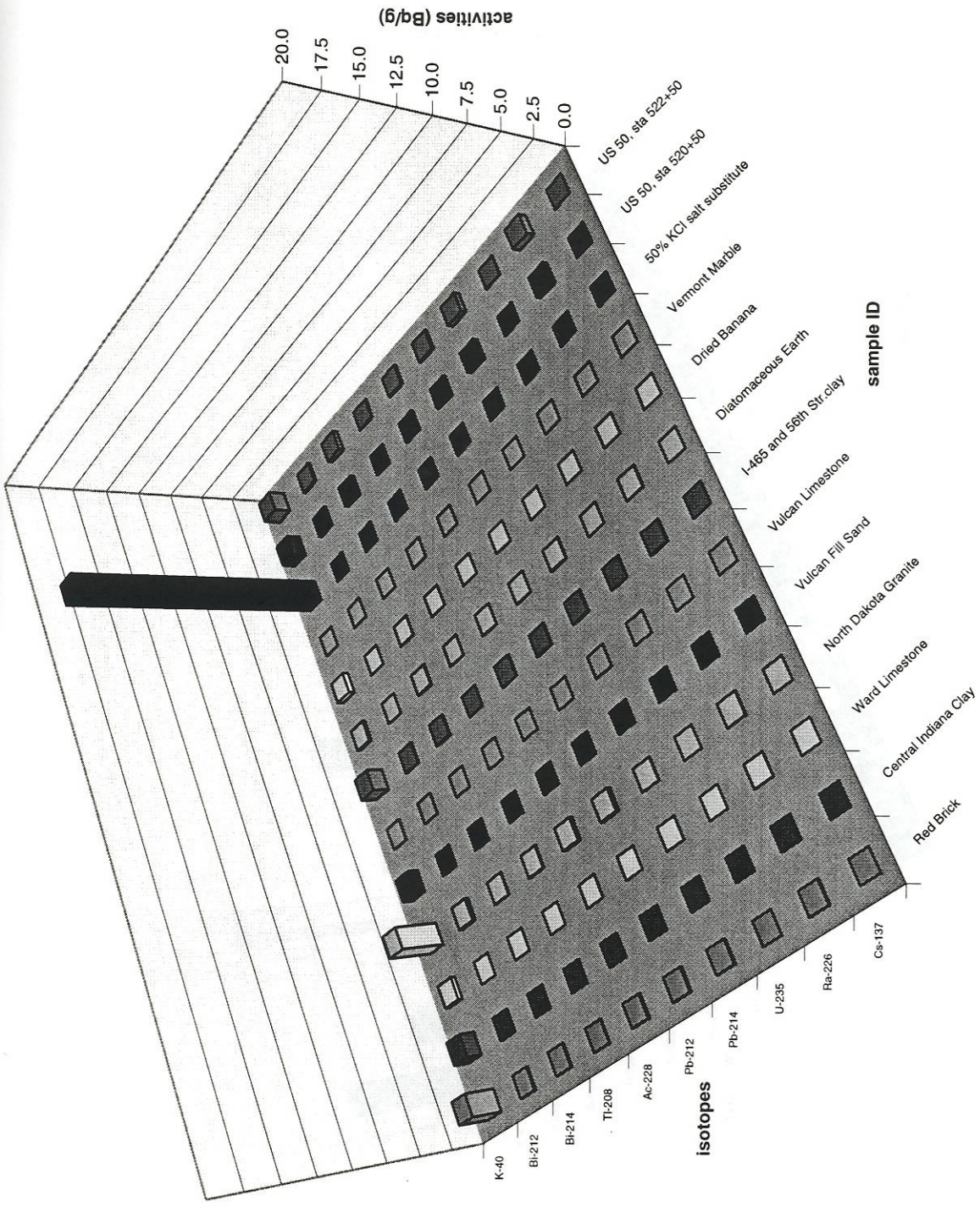

Figure 19a: Isotopic Activities of Gamma Emitters in Common Materials

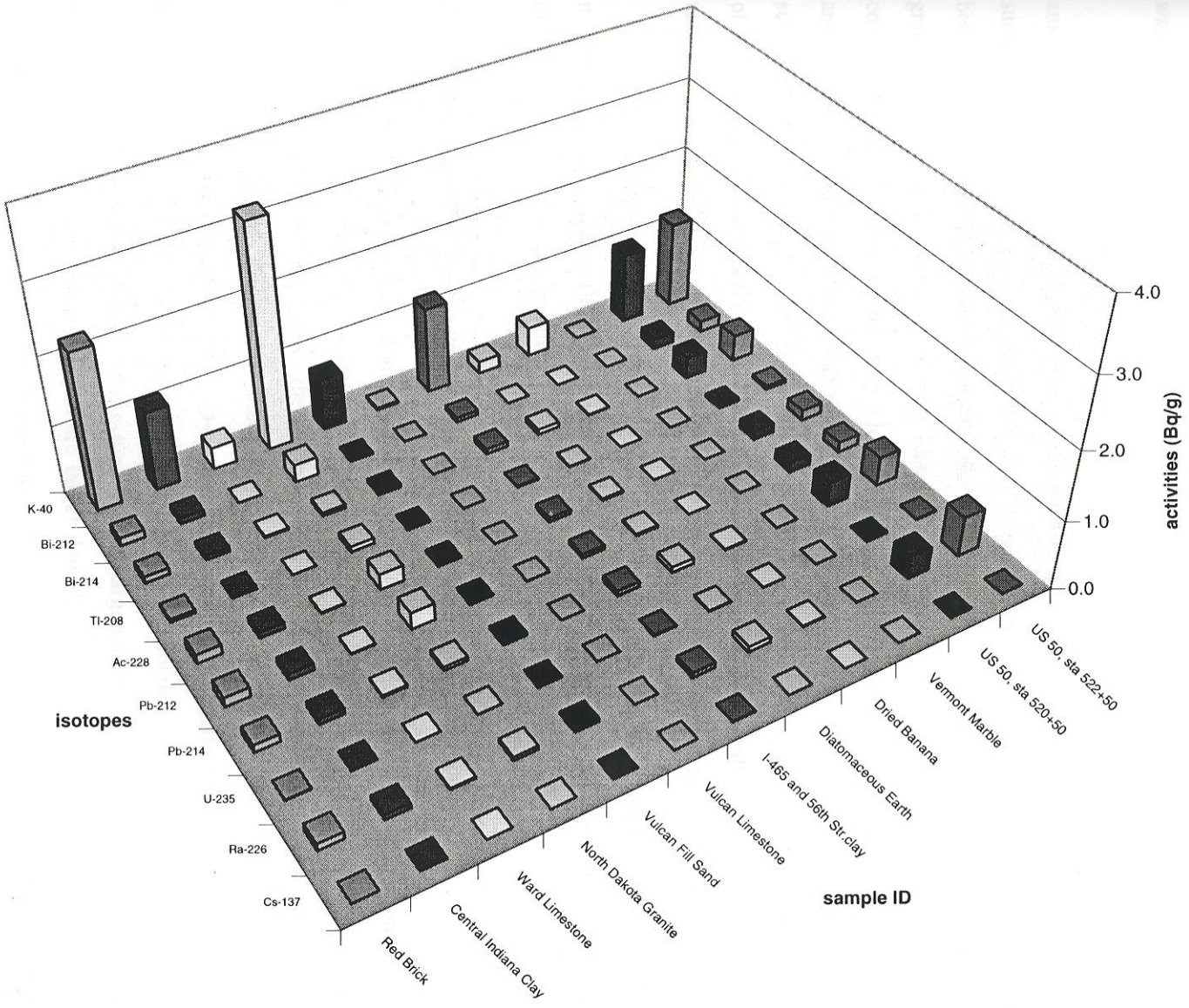

Figure 19b: Isotopic Activities of Gamma Emitters in Common Materials (enlarged without KCl)

of Indiana limestone, fill sand from central Indiana, marble from Vermont, granite from North Dakota, and diatomaceous earth (crushed coral). Two samples of commingled ash from the US 50 highway project in southern Indiana are also included. The sample identified as "50% KCl" was a salt substitute, a replacement for sodium chloride, that contained 50 % NaCl and 50% KCl. A sample of dried bananas was also included.

Natural materials have a large range of activities. The marble sample contained no measurable amounts of gamma radiation, while the granite sample was the most radioactive of the natural materials surveyed. The limestone sample from a Vulcan Materials-owned quarry contained only ^{40}K. Red brick, both clays, fill sand, diatomaceous earth, the limestone sample from the quarry of Ward Stone and granite contained trace amounts of most isotopes. Clay was the only material in the entire study that contained ^{137}Cs, because of the very close proximity to the surface of the earth, where significant amounts of radioactive fallout from nuclear tests collects, and its ability to sorb cesium in the soil subsurface. The activity of ^{40}K in the salt substitute was several times larger than the specific activities of other samples, isotopic and total. Since bananas are a source of potassium, they naturally contain ^{40}K, and trace amounts of other radionuclides were present because plants tend to uptake some heavy metals.

The activities of ^{226}Ra, ^{214}Pb, and ^{214}Bi in the US 50 samples were larger than those of all other materials. ^{40}K activities in these ash samples were greater than those of all other materials except KCl, one clay sample, brick, and granite.

5.2.2 Comparison of Isotopic Activities of Natural Materials to Coal and Ash

Figures 20 through 23 (a and b) compare the isotopic activities of gamma-ray emitting isotopes of each group of samples to natural materials. The activity of KCl dwarfs the activity of any other sample in the study. In most cases the activities of the natural materials were larger than those of coal, except for ^{40}K. Figure 20a displays the isotopic activities of natural samples and coal in greater detail without KCl. The samples of brick, clay, granite, diatomaceous earth, fill sand, and construction ash have larger activities than most coals, while limestone and marble have smaller activities. Coal 11c appears to distinguish itself from other coals. It has a large contribution of ^{226}Ra, ^{214}Pb, and ^{214}Bi. With the exception of granite, these isotopes are larger than the analogous activities for the natural samples. Almost all ash samples display larger isotopic activities than the other samples. The exception to this was, of course, ^{40}K.

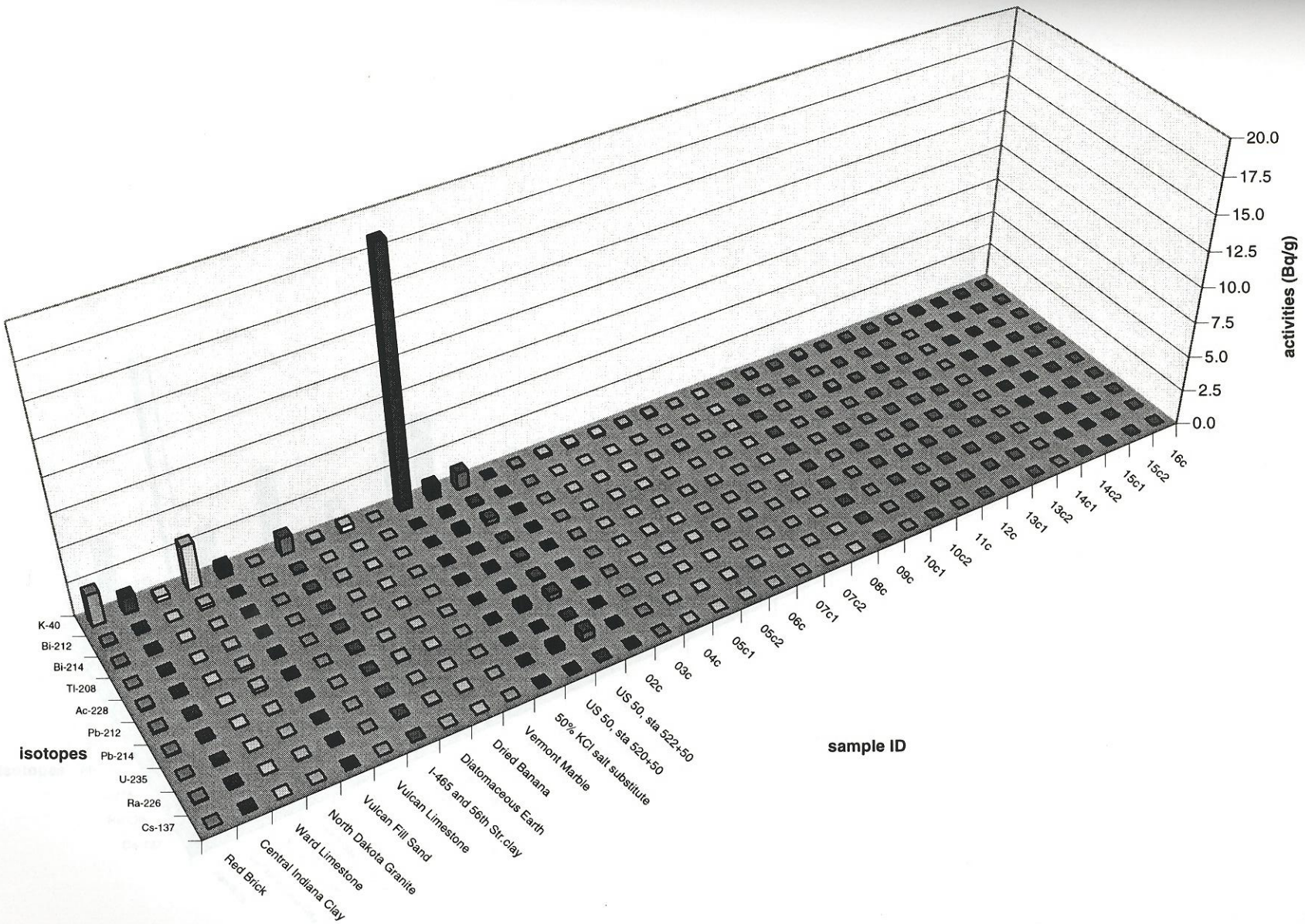

Figure 20a: Comparison of Isotopic Activities of Common Materials to Coal

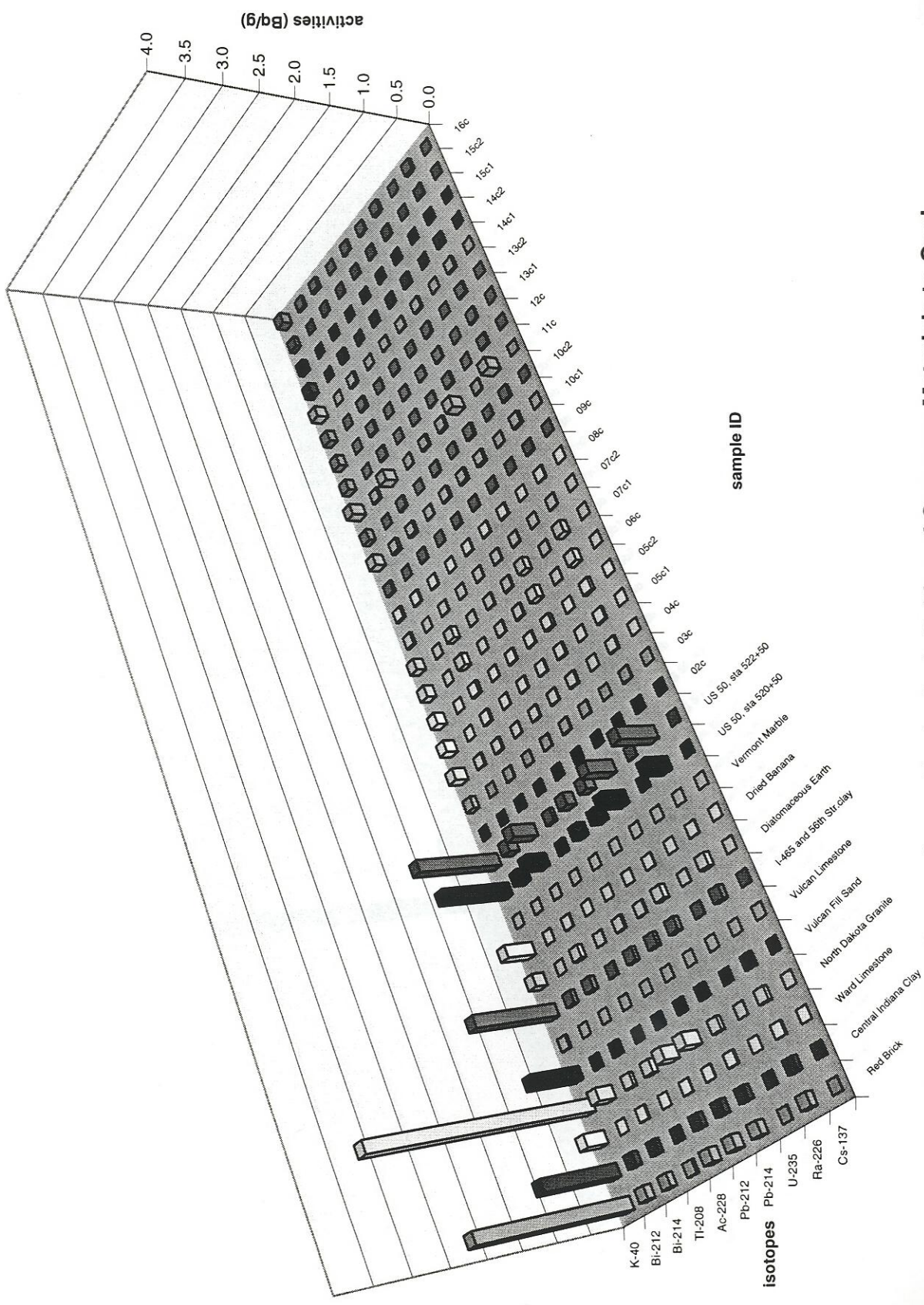

Figure 20b: Comparison of Isotopic Activities of Common Materials to Coal (enlarged without KCl)

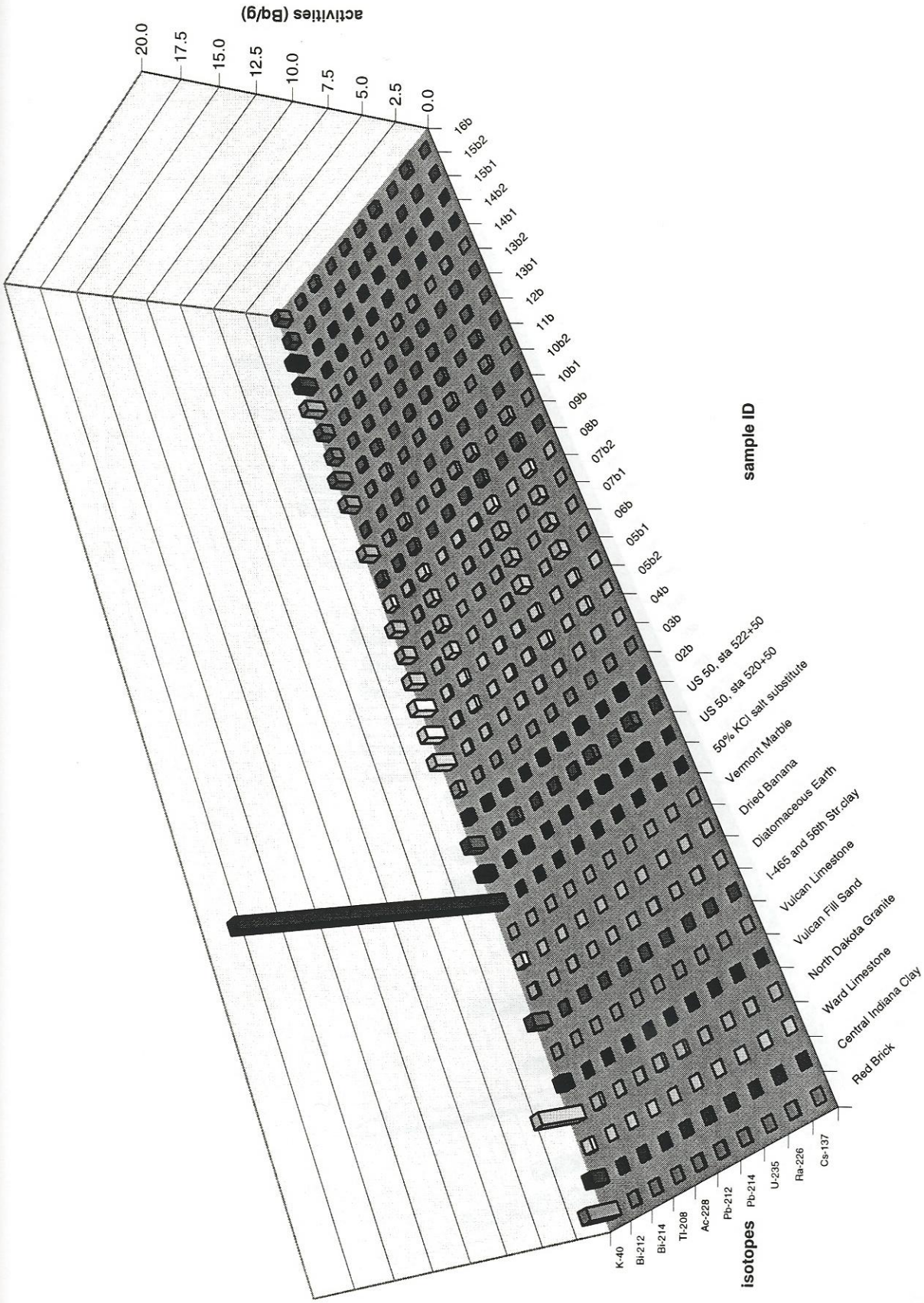

Figure 21a: Comparison of Isotopic Activities of Common Materials to Bottom Ash

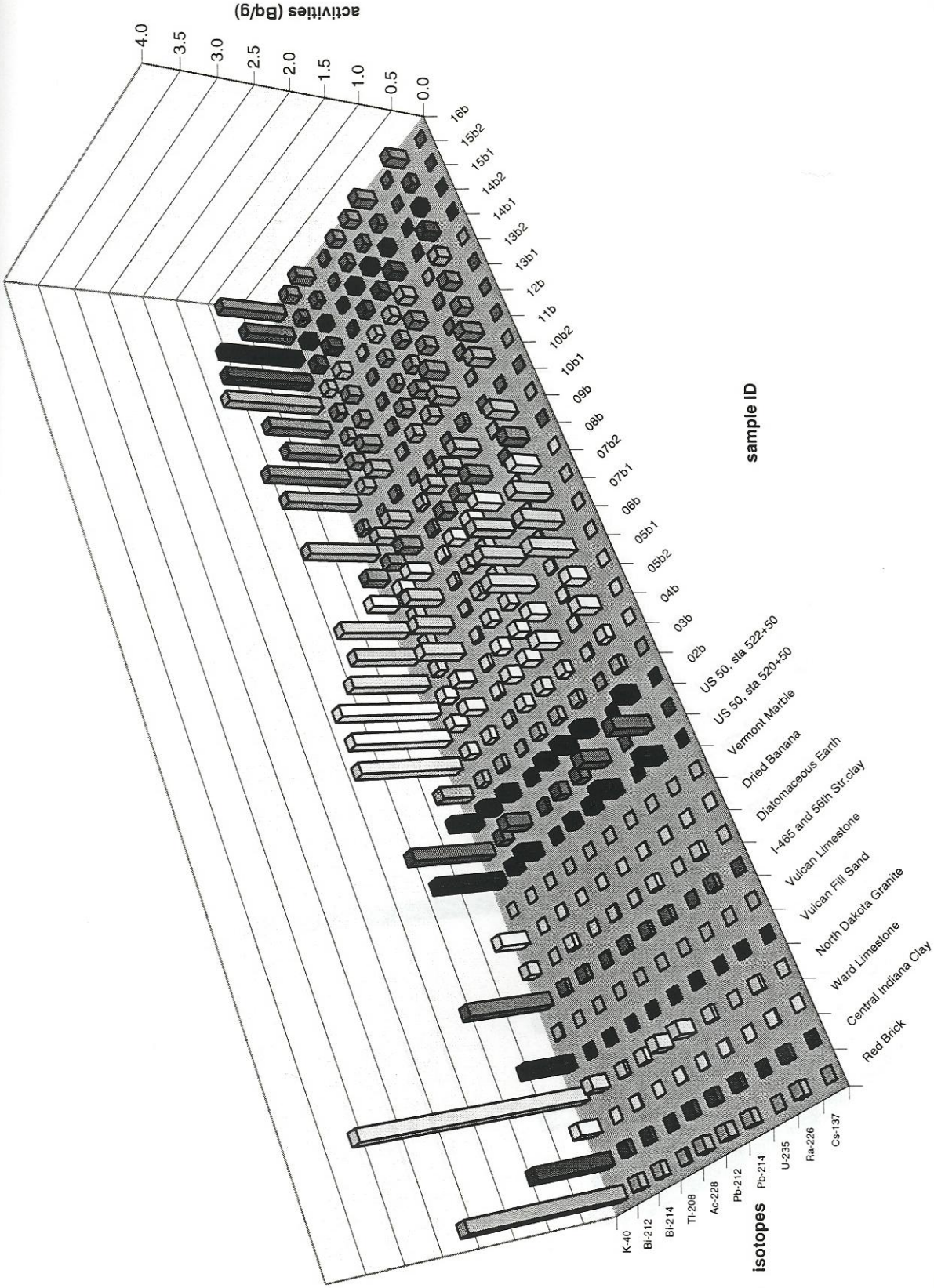

Figure 21b: Comparison of Isotopic Activities of Common Materials to Bottom Ash (enlarged without KCl)

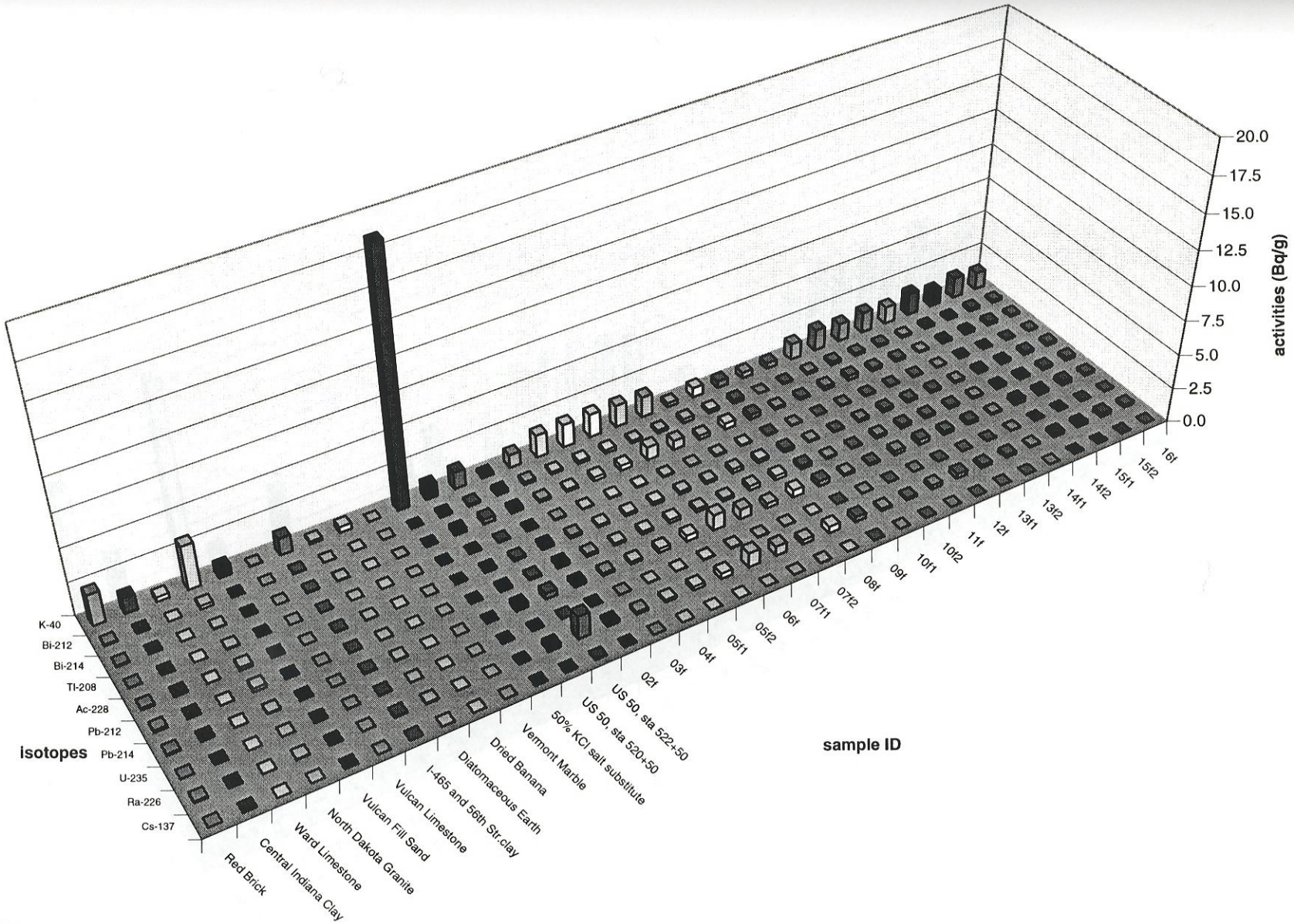

Figure 22a: Comparison of Isotopic Activities of Common Materials to Fly Ash

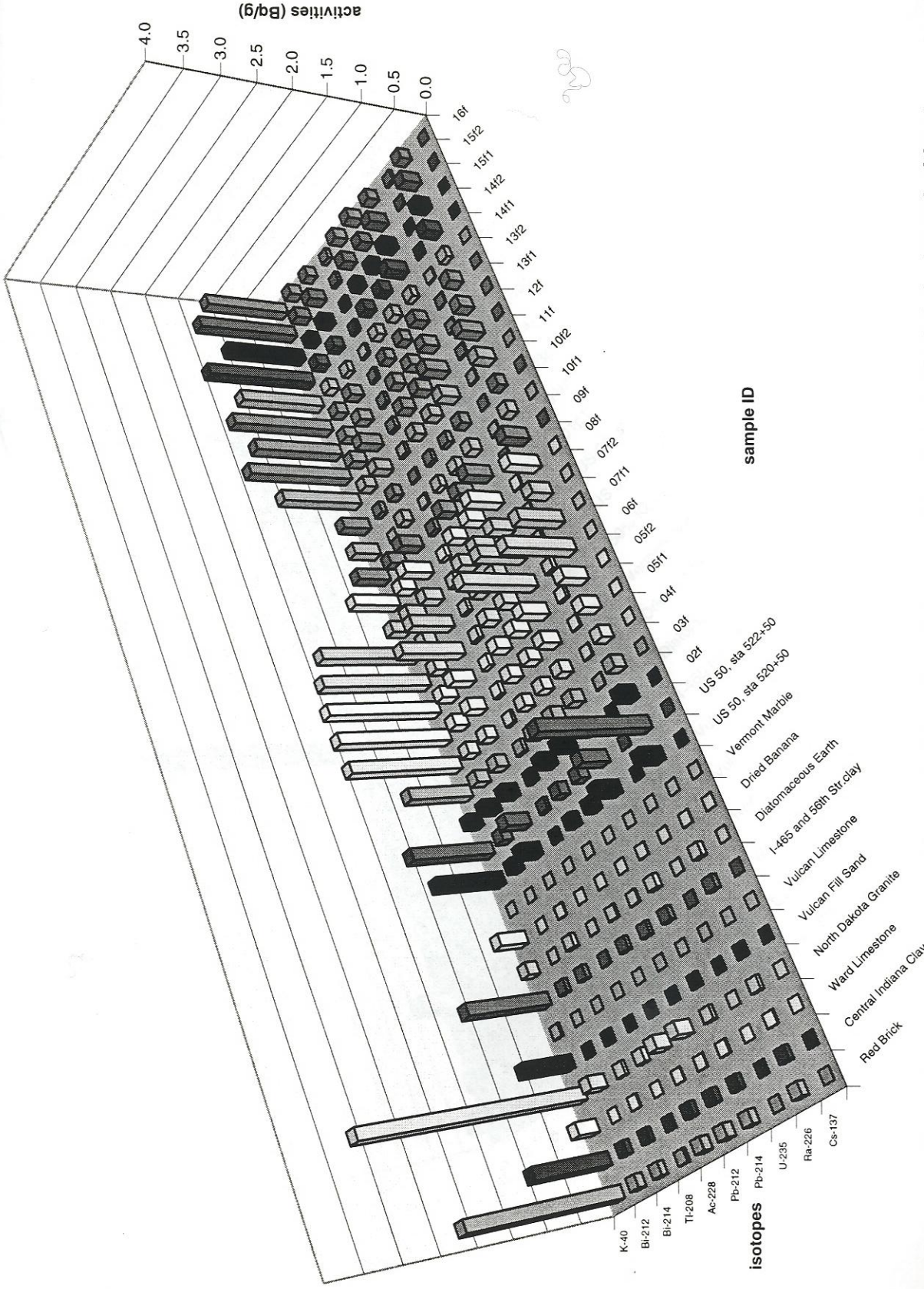

Figure 22b: Comparison of Isotopic Activities of Common Materials to Fly Ash (enlarged without KCl)

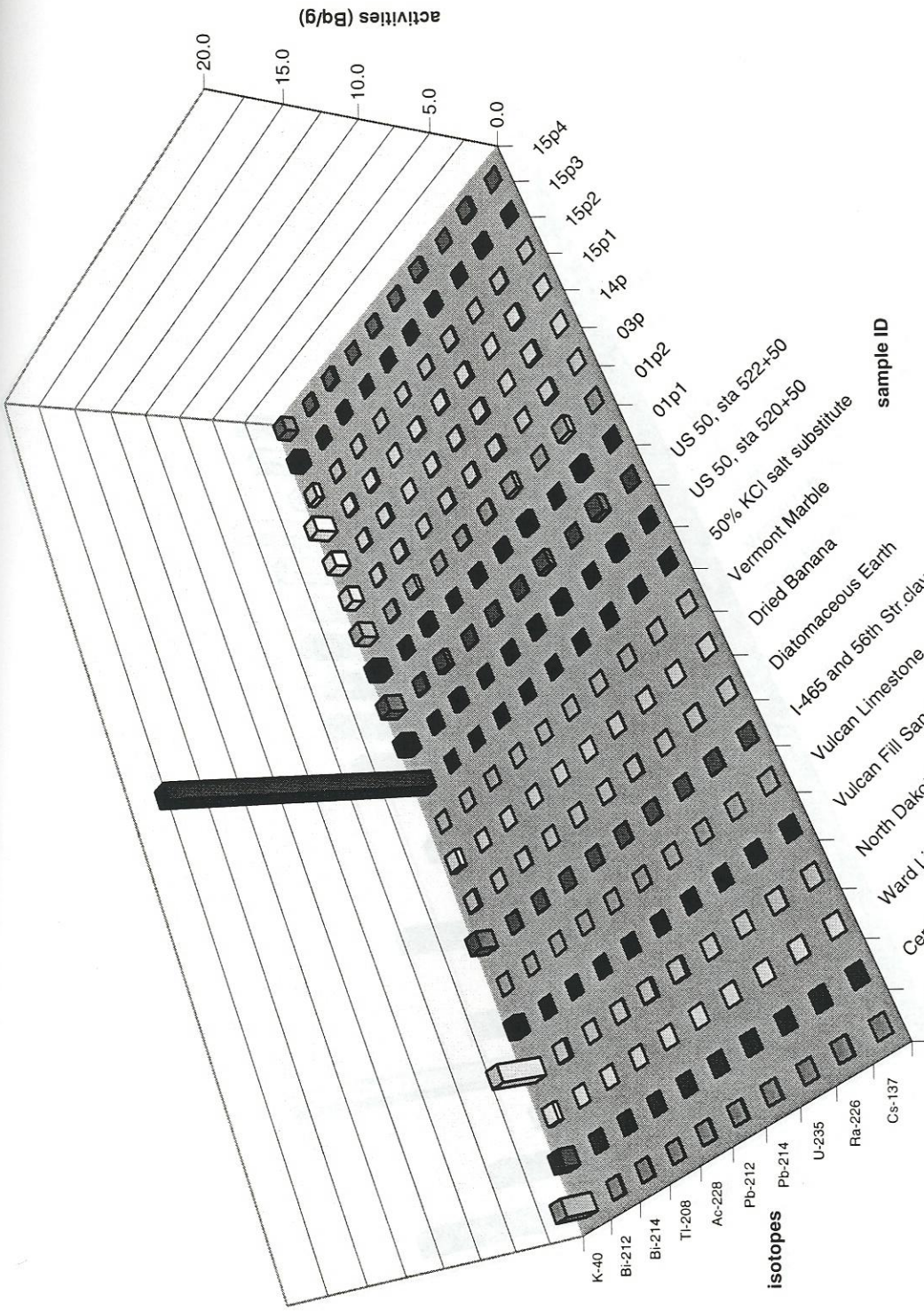

Figure 23a: Comparison of Isotopic Activities of Common Materials to Commingled Ash

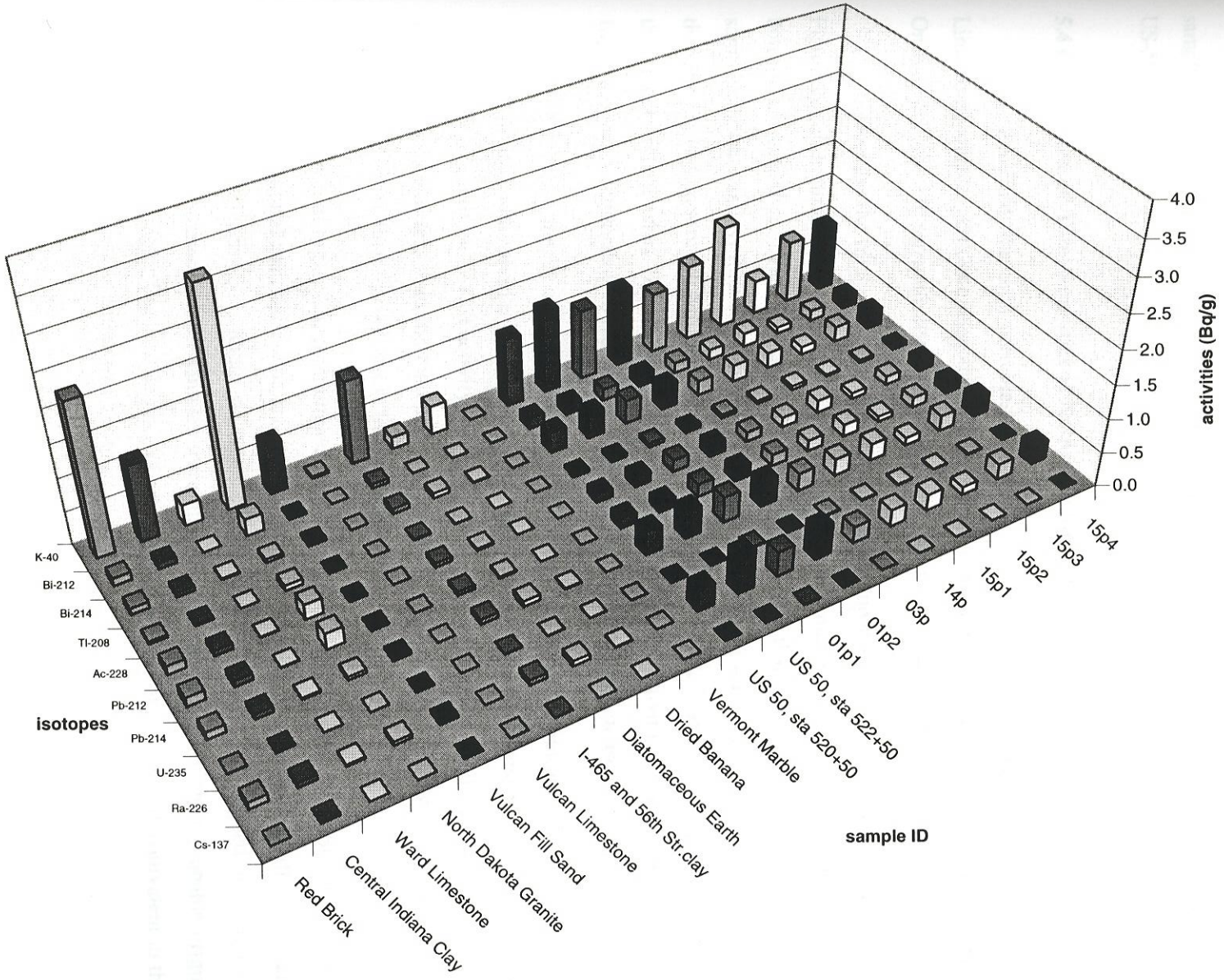

Figure 23b: Comparison of Isotopic Activities of Common Materials to Commingled Ash (enlarged without KCl)

5.3 Total Activities of Natural Samples

Total activities of natural samples are in Figure 24. Salt substitute has the largest activity of all samples surveyed, despite having only one isotope contributing to the total. Clay, brick, granite, and the US-50 construction ash have the largest activities of the remaining samples.

5.4 Comparison of Total Activities of Natural Samples to Coal and Ash

As expected, the total activities of coal were less than those of natural samples (Figure 25). Limestone, marble, and diatomaceous earth are the only materials that have total activities less than coal. Once again, coal 11c separates itself from the group, having the highest total activity among coals.

Generally, the total activities of bottom ash are comparable to those of the natural materials. In Figure 26, all natural samples, except Vulcan limestone and marble, were contained within the range of bottom ash activities. The total activity of granite was less than the activities of all 22 bottom ash samples, brick was less than four bottom ashes, and KCl activity was more than a factor of five greater than the highest bottom ash activity, 06b. Traditional fill materials, limestone and sand, were less active than most bottom ashes. These results suggest that bottom ash, activities ranging from 0.281 to 3.71 Bq/g, was nearly equivalent to many of the building materials currently in use.

The results for fly ash were somewhat different than those of bottom ash. In Figure 27, five natural samples, both samples of limestone, marble, diatomaceous earth, and fill sand, fell outside the range of fly ash activities. KCl activity was about three times larger than the highest fly ash activity, and granite activity was less than the total activities of two fly ash samples. These results indicate that fly ash was more active than many construction materials.

As shown in Figure 28, commingled ash was approximately equivalent to bottom ash in terms of comparison to natural materials. The activities of granite and brick are greater than all commingled ash activities, and KCl activity is about six times greater than the largest commingled ash activity. The activities of the construction ash samples were also comparable because of their probable origin from commingled ash storage. The total activity of commingled ash was approximately equivalent to the total activity of traditional construction materials.

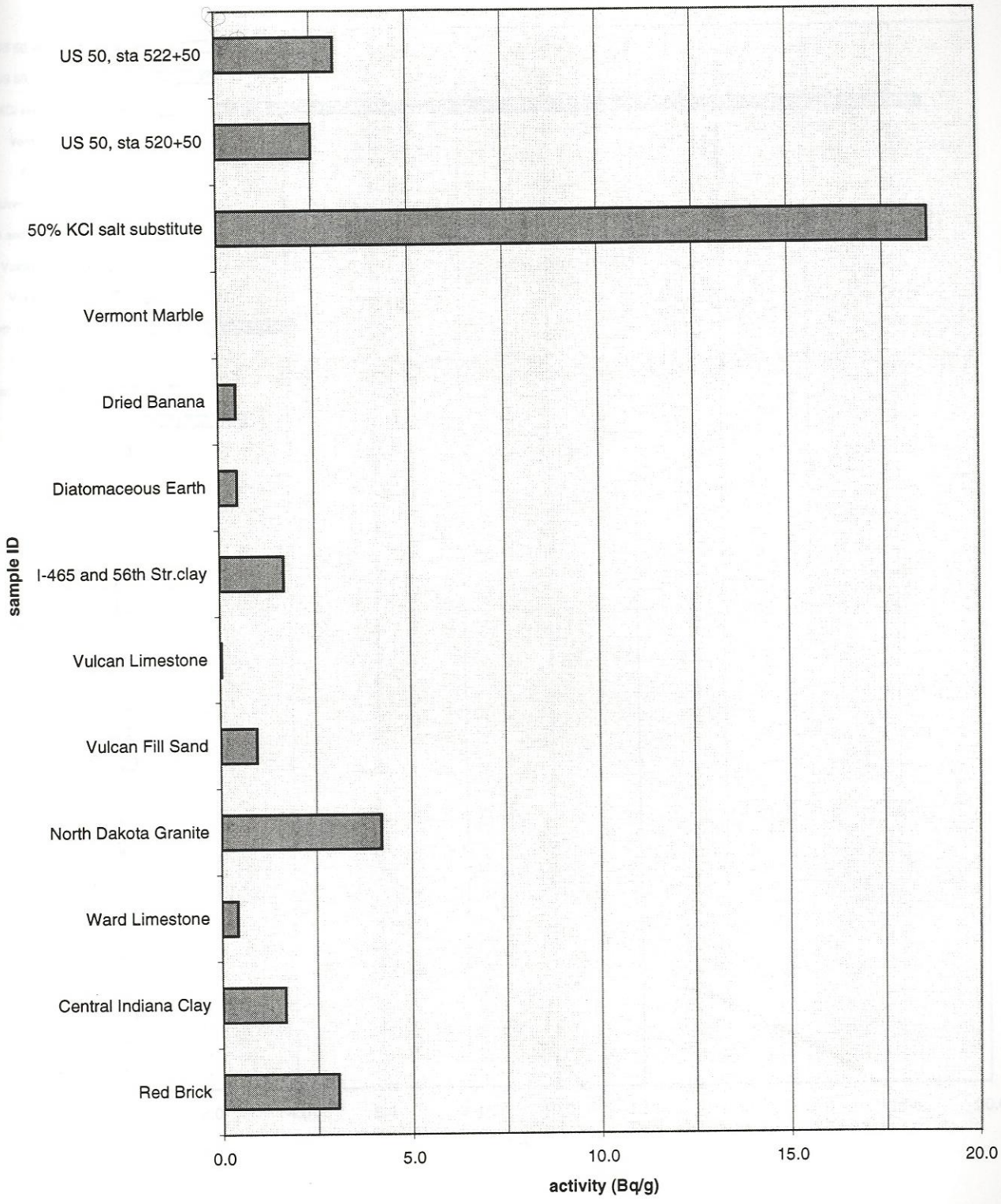

Figure 24: Total Activities of of Gamma-ray Emitters in Common Samples

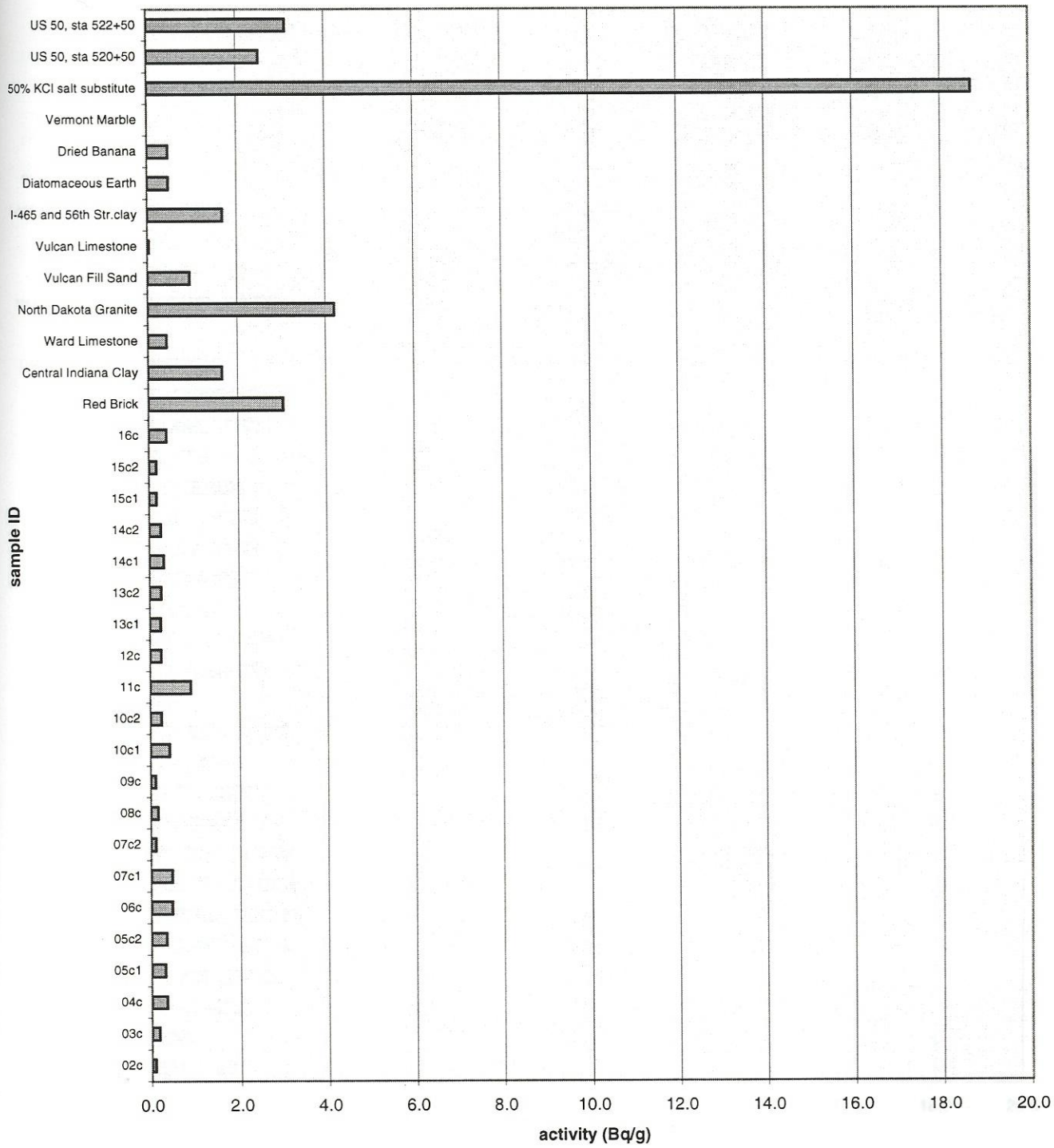

Figure 25: Comparison of Total Activities of Coal and Common Materials

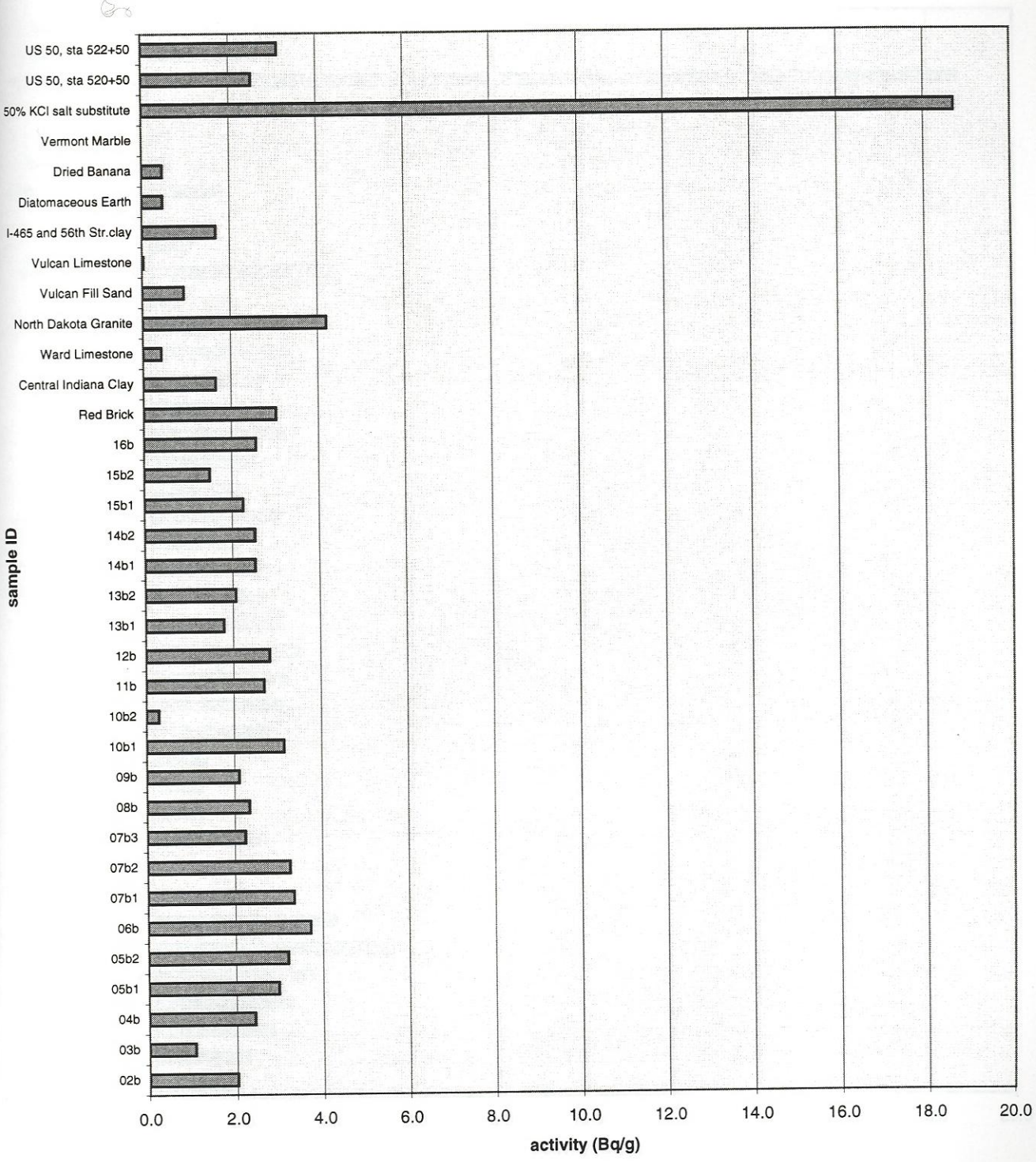

Figure 26: Comparison of Total Activities of Bottom Ash and Common Materials

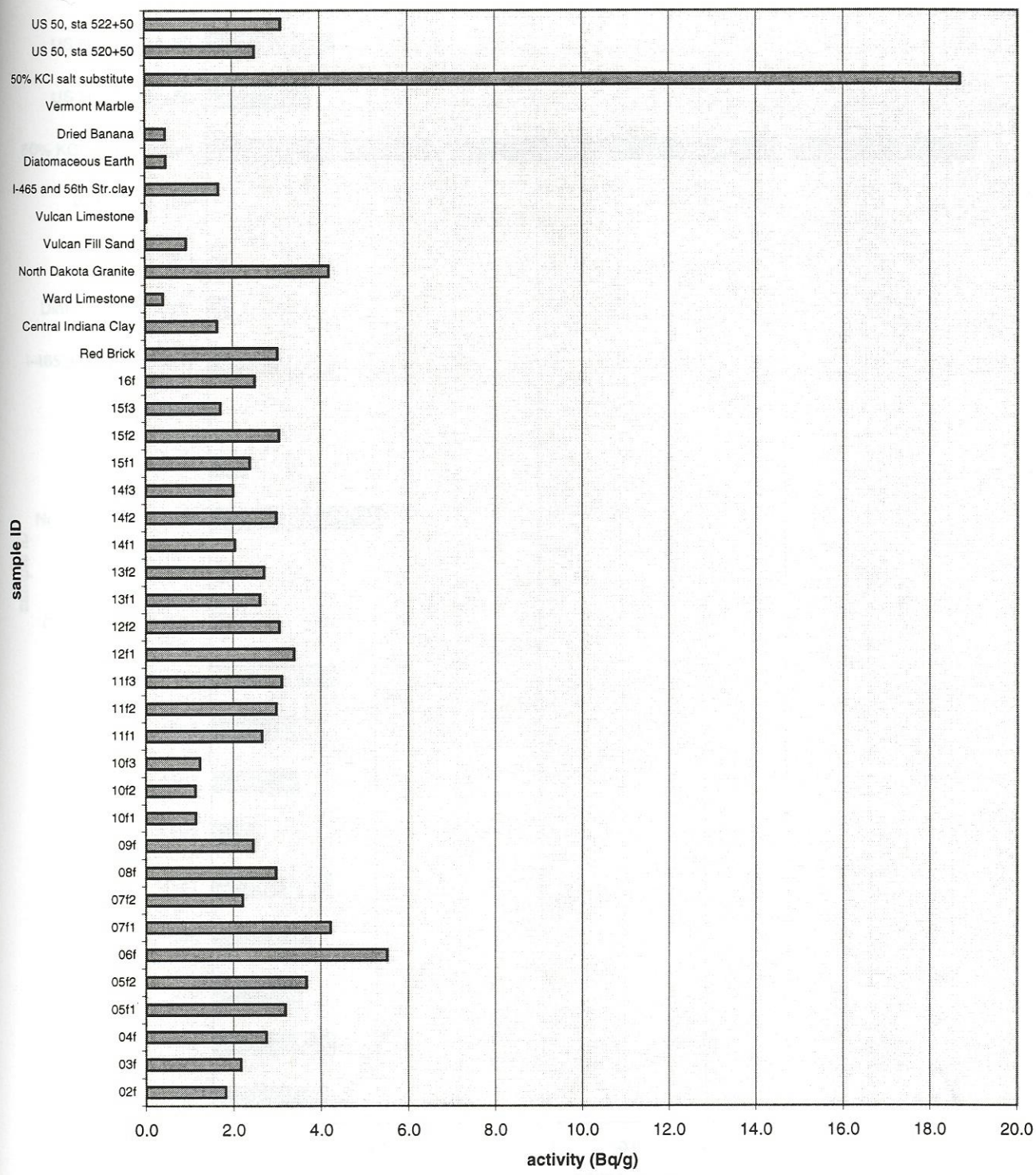

Figure 27: Comparison of Total Activities of Fly Ash and Common Materials

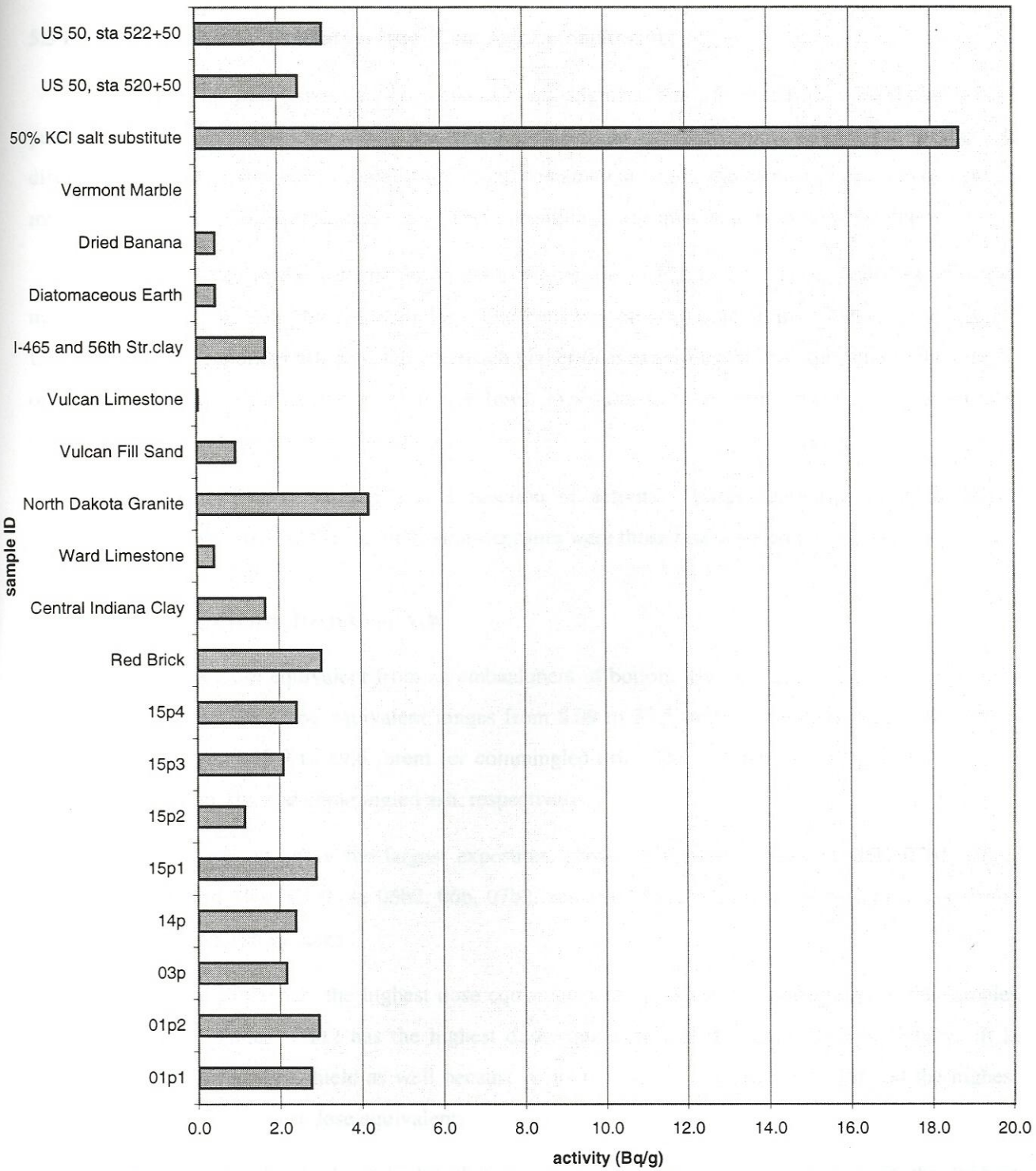

Figure 28: Comparison of Total Activities of Commingled Ash and Common Materials

5.5 Exposure to Gamma Radiation from Coal Ash Embankments

Exposure for people working on a coal-ash embankment was calculated for a 2000 hour work-year of occupational exposure, which was much greater than exposures expected for the public. A distance of one meter, the approximate height of the human torso, above the surface of an embankment of material was used for exposure calculations. The embankment was infinite in both area and depth.

Densities used in the calculation of the exposure are in Figure 29. These densities were the measured densities of one-liter test samples. The densities of ash range from 0.302 to 1.693 g/cm^3. Under actual circumstances ash densities are much higher than experimental densities, due to the effects of ash compaction. A larger density results in lower exposures because more shielding is present in a fixed volume to attenuate more gamma rays.

Along with density, exposure is a function of activity. Larger activities result in larger exposures. Activities used in the calculation of exposure were those that were presented above.

5.6 Total Dose Equivalent from Coal Ash

The annual dose equivalent from an embankment of bottom, fly, or commingled ash is given in Figures 30, 31, and 32. Dose equivalent ranges from 8.09 to 37.5 mrem for bottom ash, 7.90 to 59.1 mrem for fly ash, and 10.9 to 39.6 mrem for commingled ash. The averages are 18.1, 25.9, and 20.9 mrem/yr for bottom, fly, and commingled ash, respectively.

Bottom ashes that have the largest exposures, above 20 mrem/yr are 05b2, 06b, 07b1, 07b2, 10b1, 11b, 12b, and 16b. Of these 05b2, 06b, 07b2, and 10b1 have densities and activities among the highest of all bottom ash samples.

In the case of fly ash, the highest dose equivalents, above 30 mrem, correspond to the samples with the lowest densities. 12f2 has the highest dose equivalent and the second-lowest density. It is apparent that 12f2 does not shield as well because of its low density. Conversely, 16f had the highest density and the fourth-lowest dose equivalent.

For commingled ash the samples that correspond to the highest densities and the highest activities have the lowest dose equivalents. 01p1 produces the second-lowest dose equivalent with the second-highest density and third-highest activity.

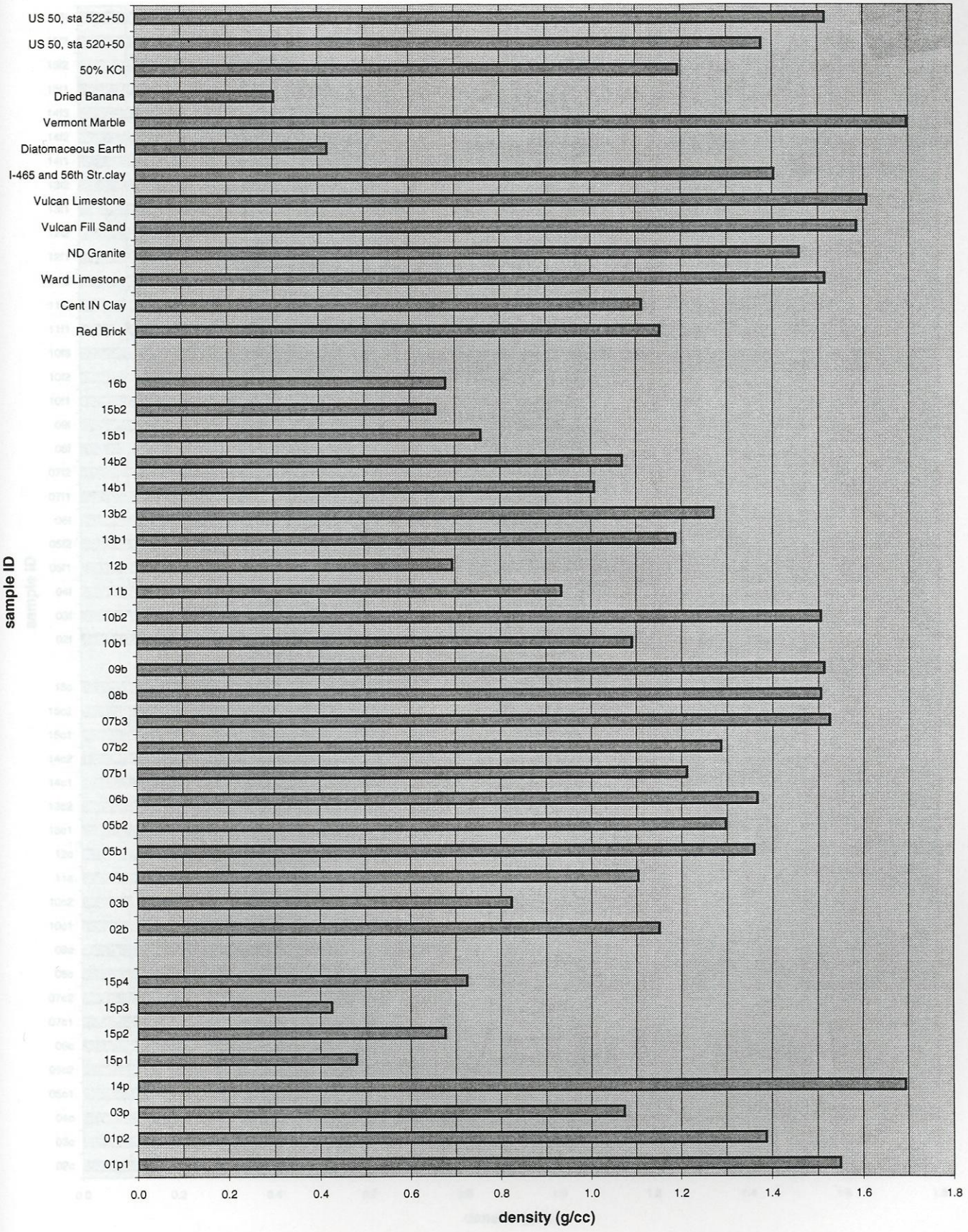

Figure 29: Densities of 1-liter Samples

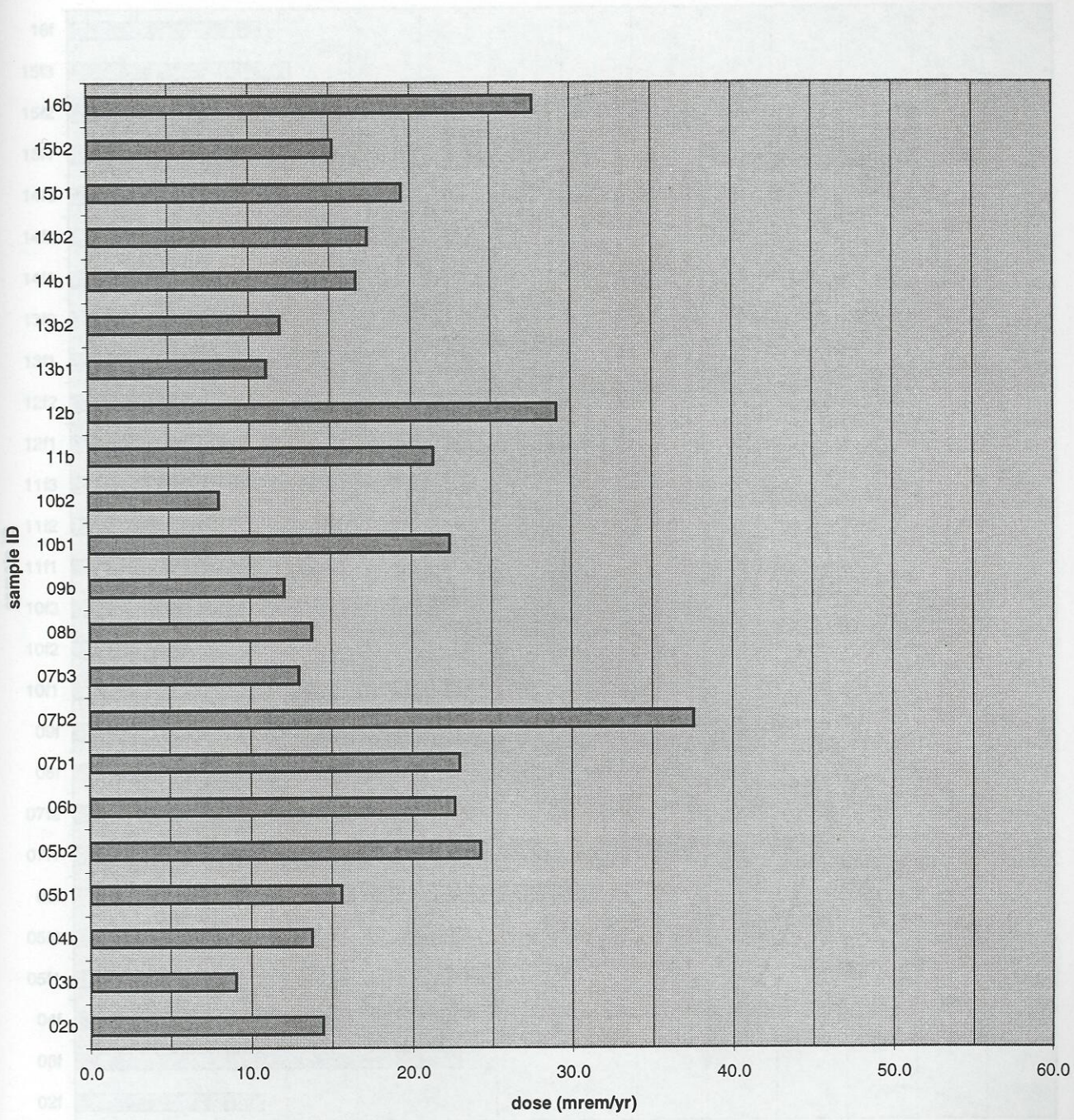

Figure 30: Dose Equivalent Rate of Gamma Rays from Bottom Ash Samples

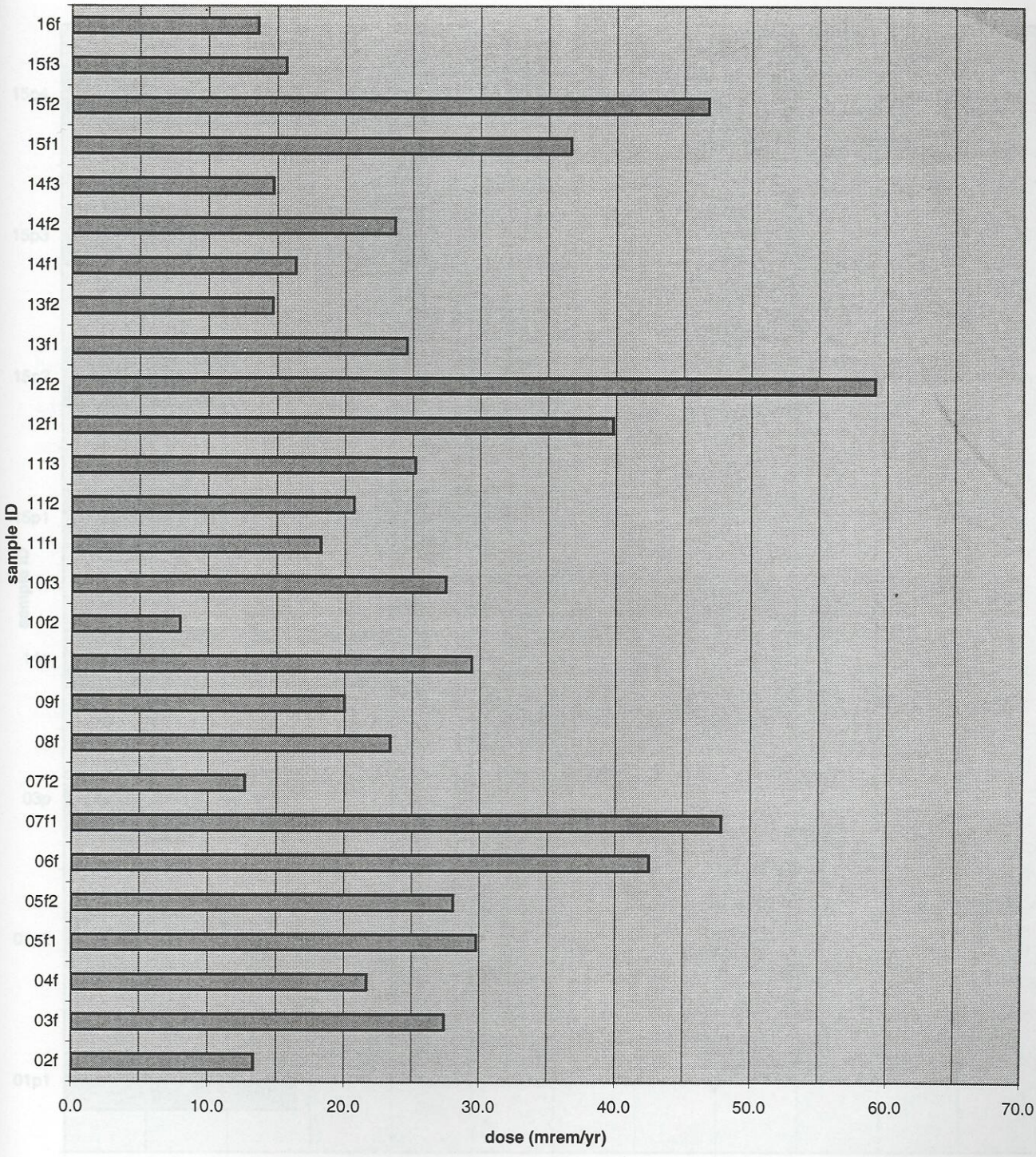

Figure 31: Dose Equivalent Rate of Gamma Rays from Fly Ash Samples

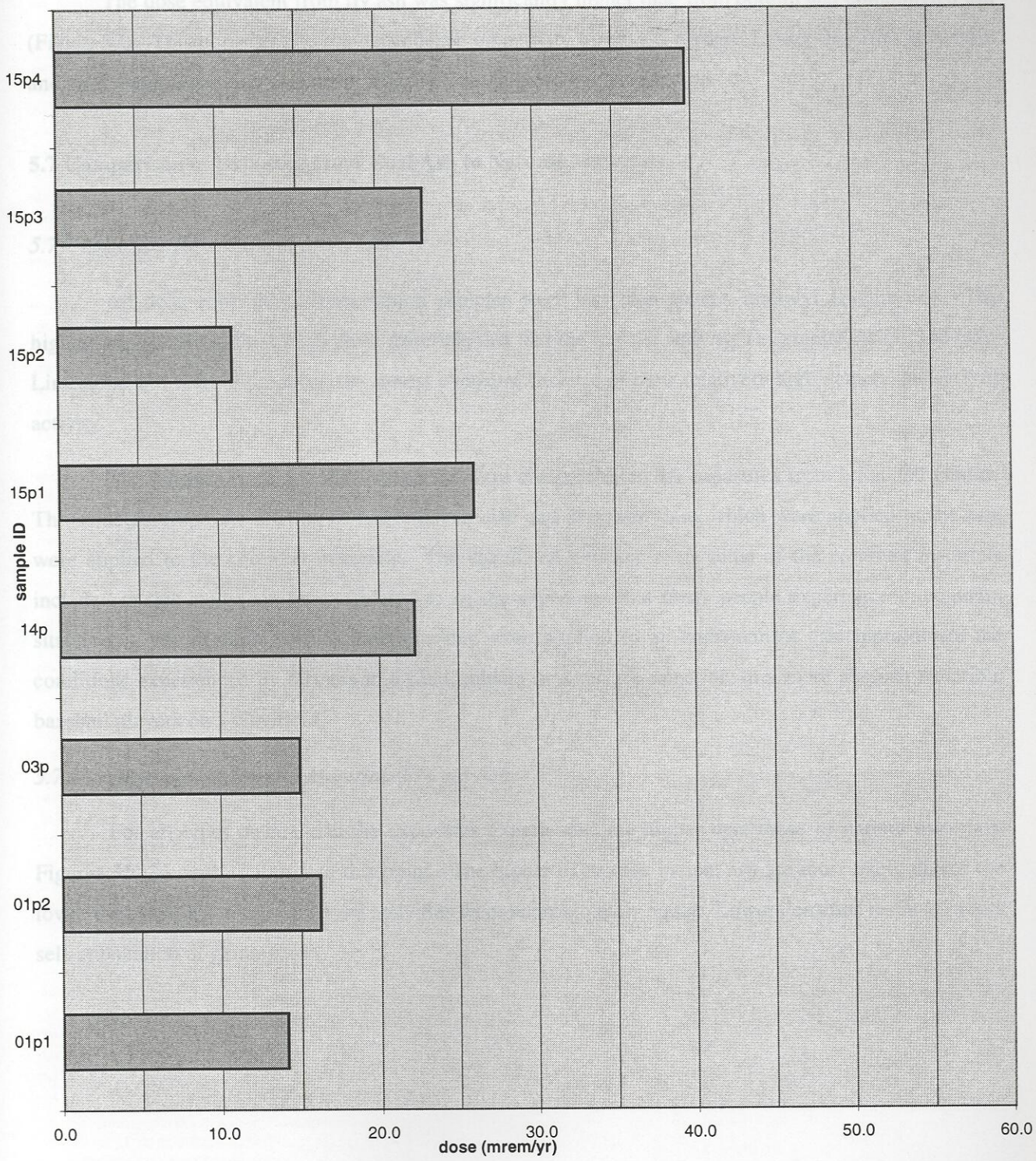

Figure 32: Dose Equivalent Rate of Gamma Rays from Commingled Ash Samples

The dose equivalent from fly ash was significantly higher than from bottom and commingled ash (Figure 33). This correlated to the dependence on activity discussed earlier. Larger densities in bottom and commingled ash may also result in more shielding of gamma radiation.

5.7 Comparison of Exposure from Coal Ash to Natural Materials

5.7.1 Exposure from Natural Materials

All dose equivalents from natural samples were less than sixteen mrem/yr (Figure 34). The highest exposures resulted from those materials that had the highest activity, i.e. granite, brick, and clay. Limestone and marble produced the lowest exposure because of their relatively high density and lack of activity.

The samples from US 50 construction were comparable to the exposures from brick and granite. The same assumptions relating to embankment size and exposure time, which were applied to the ash, were applied to the common materials. The calculated exposures for some of the common materials included in this study can be considered to be the exposures that many people experience in common situations. For example, the assumptions that were applied to an embankment can approximate the conditions experienced by farmers in a field, miners in a surface mine, or, in a more abstract situation, baseball players on a clay field.

5.7.2 Comparison of Natural Materials to Coal Ash

For all types of coal ash the exposures experienced are higher than those of natural materials. Figures 35, 36, and 37 illustrate this point. The higher exposures in coal ash are most likely due to the lower densities of the ash, since the activities were approximately equal. Lower densities resulted in less self-attenuation of gamma rays.

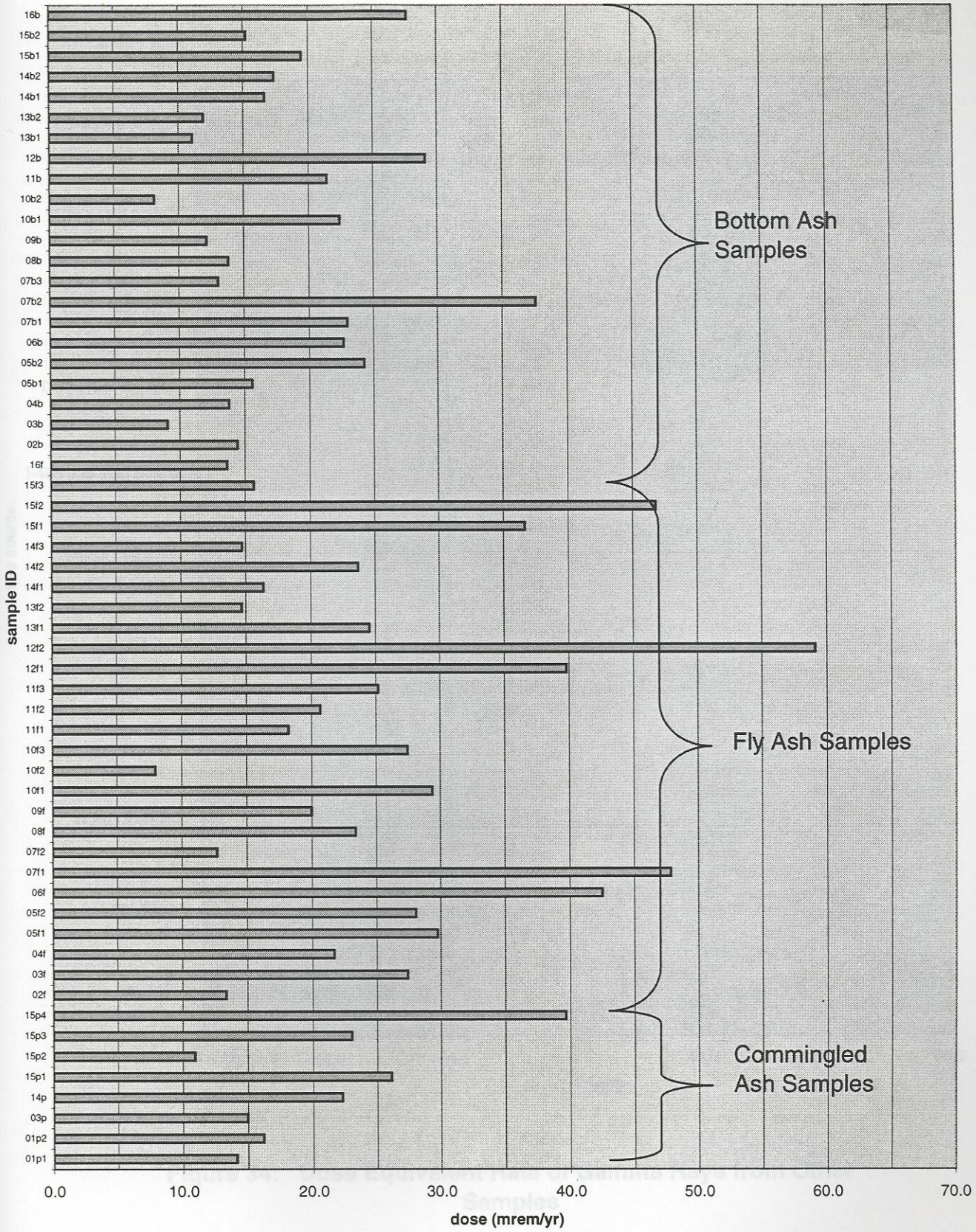

Figure 33: Dose Equivalent Rate of Gamma Rays from All Ash Samples

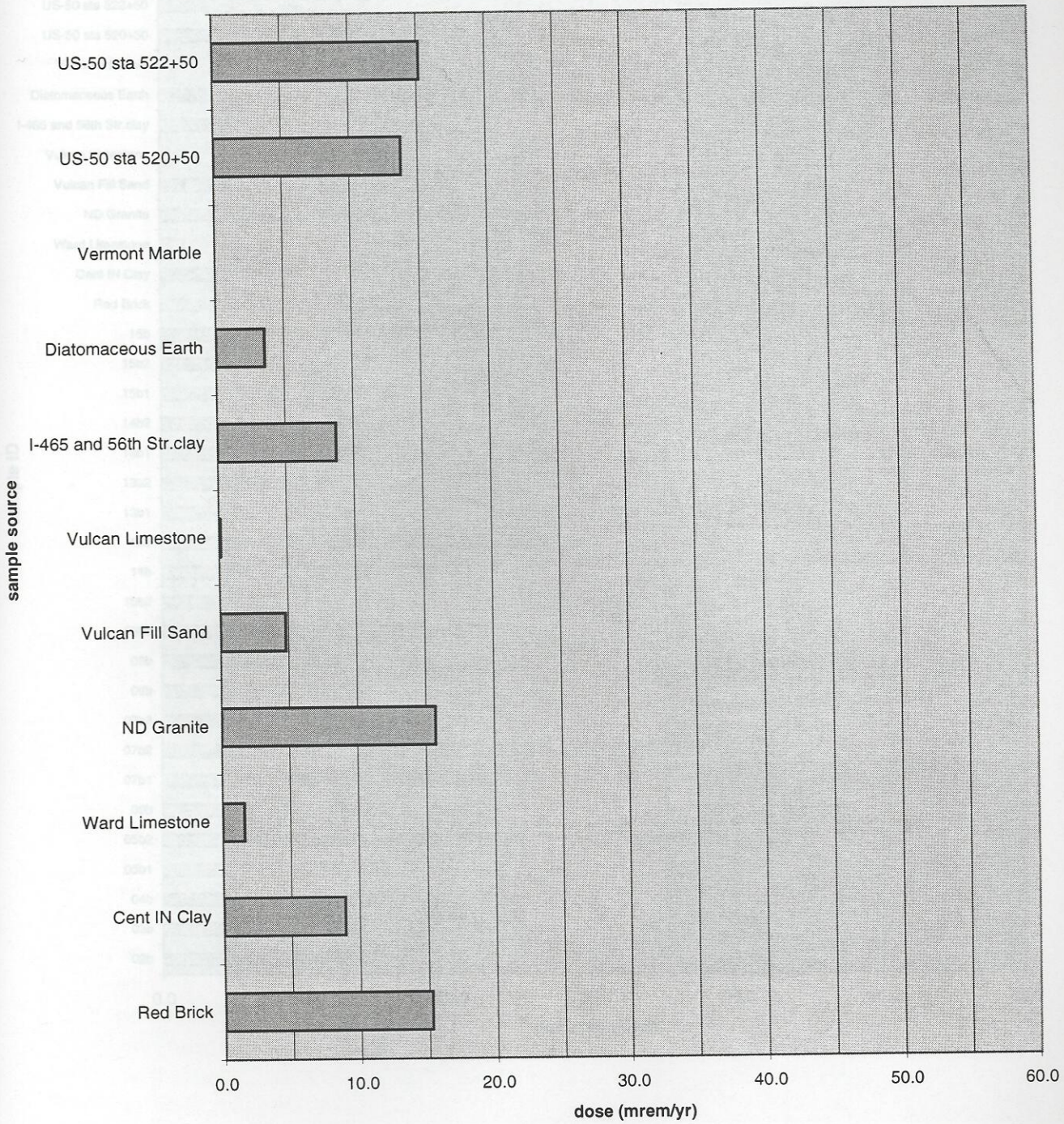

Figure 34: Dose Equivalent Rate of Gamma Rays from Other Samples

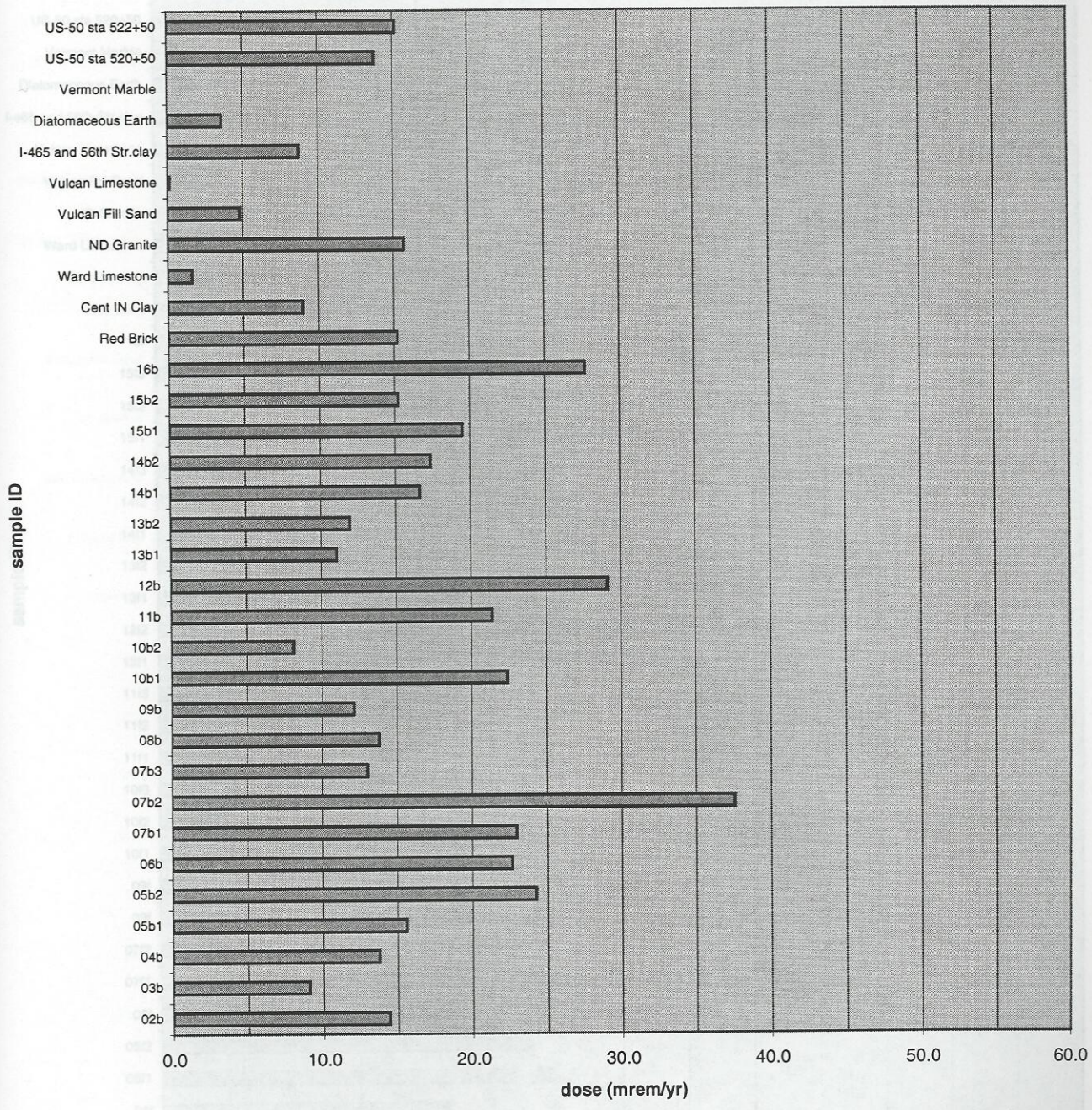

Figure 35: Comparison of Dose Equivalent Rate from Bottom Ash and Other Samples

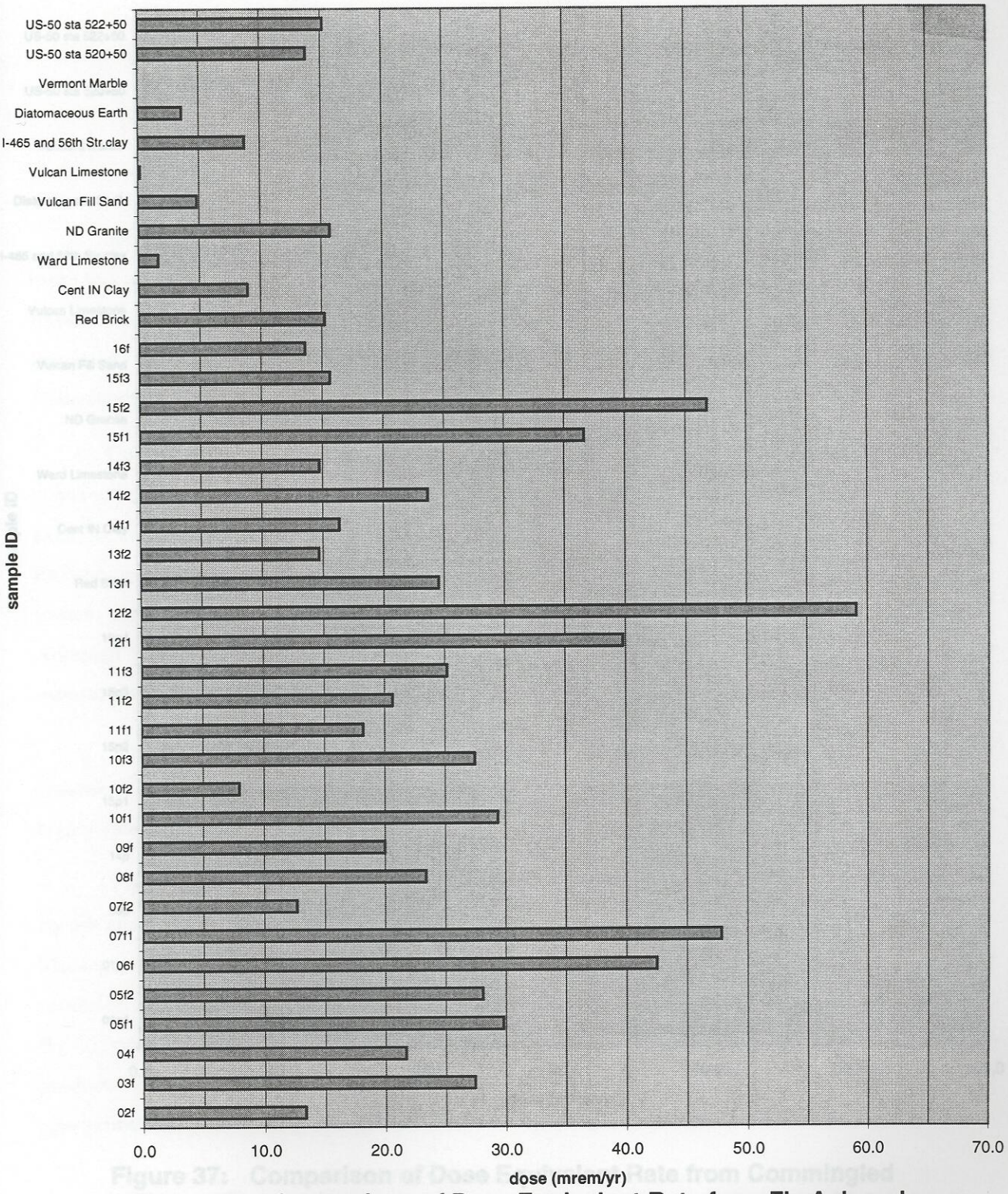

Figure 36: Comparison of Dose Equivalent Rate from Fly Ash and Other Samples

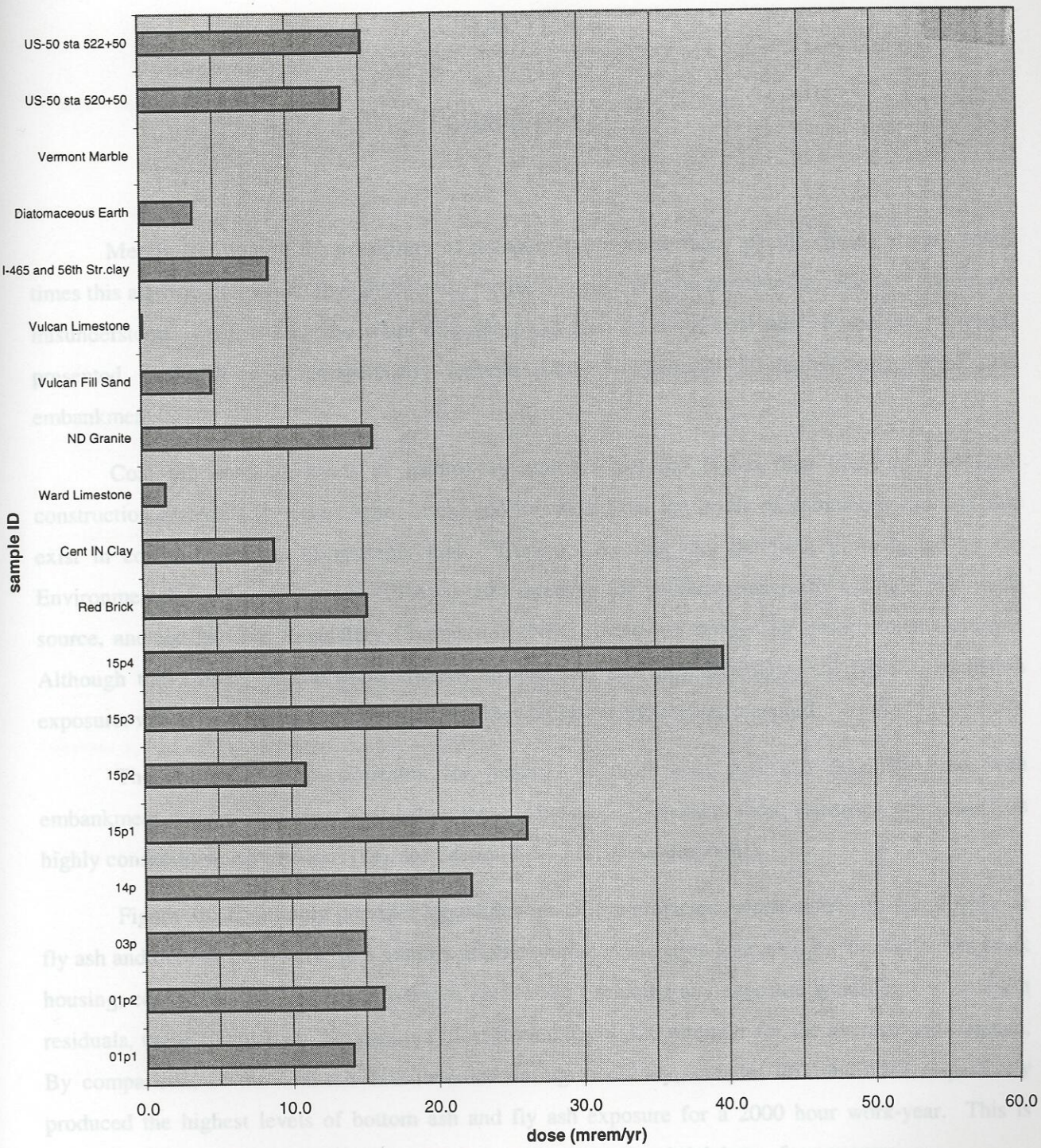

Figure 37: Comparison of Dose Equivalent Rate from Commingled Ash and Other Samples

CHAPTER 6

Conclusions

Merely mentioning the possibility of radiation exposure to many people causes alarm. Many times this alarm is premature, though, because the facts about radiation exposure in certain situations are misunderstood. Such is the case when discussing radiation levels in coal ash. Based on the results presented, coal ash is a radiogenically feasible alternative material in the construction of road embankments.

Coal ash contains levels of gamma-ray emitters that are higher than those of traditional construction materials (i.e. clay, sand, brick, and limestone) but the levels of gamma-ray emitters that exist in coal ash are not exceedingly high. Consider the fact that the limit promulgated by the Environmental Protection Agency (EPA) is 100 mrem/yr for public exposure to a single man-made source, and the Nuclear Regulatory Commission (NRC) limit is 5 rem/yr for occupational exposure. Although these limits do not apply directly to coal ash and they encompass all types of radiation exposure, which have not been addressed here, they represent an accepted standard.

The exposure levels projected for gamma radiation with coal ash residuals used with embankment construction were well below these standards. Even then, these estimates were based on highly conservative assumptions (i.e., see Section 4.5.1 Model Assumptions).

Figure 38 accordingly provides a qualification of the projected 'worst-case' emission levels for fly ash and bottom ash relative to a number of other common radiation sources (i.e., inhaled radon, brick housing, cosmic sources, terrestrial sources, etc.). Even excluding any exposure whatsoever to coal ash residuals, these cumulative, 'background' dosages are about 320 mrem/yr for the average adult human. By comparison, of the coal-ash data recorded during this study, samples 7b2 and 12f2 respectively produced the highest levels of bottom ash and fly ash exposure for a 2000 hour work-year. This is approximately equal to the dose equivalent received by living in a brick home for one year.

Given these quantities, it can be seen that exposures from coal ash embankments are relatively low. In turn, these residuals would only contribute small amounts of exposure each year when used as an alternative material in road construction.

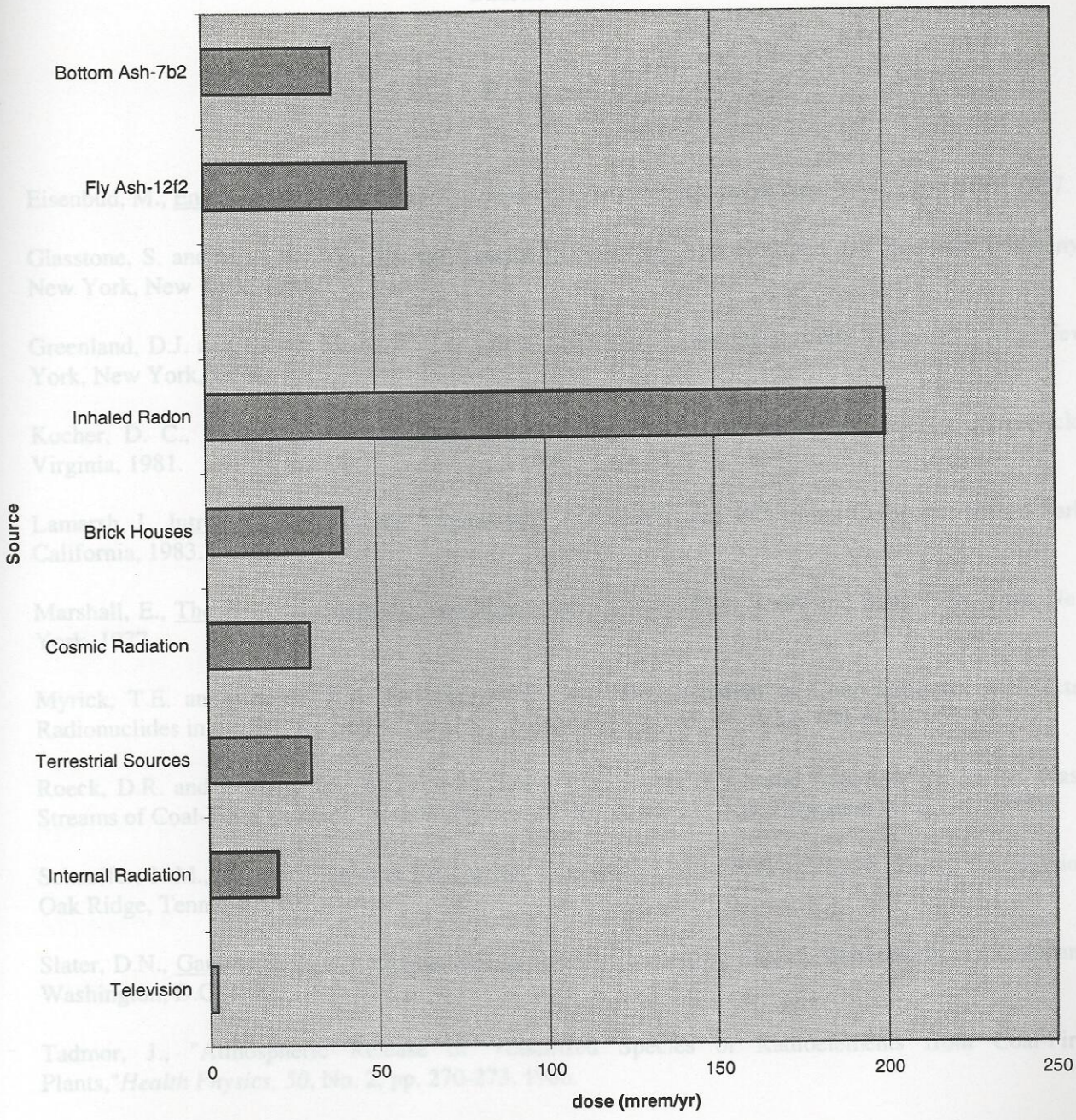

Figure 38: Comparative Dose Equivalent from Common Sources (population average) [Source: Lamarsh, 1983]

CHAPTER 7

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Appendices

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Appendix A

Isotopic Activities (Bq/g) of Gamma-ray Emitting Isotopes

(Complete Samples and Common Materials)

Commingled Ash

	K-40	Bi-212	Bi-214	Tl-208	Ac-228	Pb-212	Pb-214	U-235	Ra-226	Cs-137
01p1	1.033	0.171	0.350	0.057	0.174	0.190	0.385	0.017	0.370	0.000
01p2	1.132	0.181	0.355	0.056	0.180	0.182	0.378	0.014	0.464	0.000
03p	0.895	0.157	0.219	0.052	0.144	0.171	0.261	0.015	0.241	0.000
14p	1.122	0.149	0.248	0.048	0.144	0.158	0.248	0.017	0.250	0.000
15p1	1.532	0.201	0.232	0.057	0.186	0.174	0.248	0.014	0.240	0.000
15p2	0.503	0.092	0.114	0.024	0.081	0.079	0.114	0.009	0.117	0.000
15p3	0.915	0.153	0.226	0.044	0.130	0.146	0.226	0.013	0.228	0.000
15p4	0.974	0.192	0.264	0.055	0.166	0.177	0.289	0.015	0.279	0.000

Fly Ash

	K-40	Bi-212	Bi-214	Tl-208	Ac-228	Pb-212	Pb-214	U-235	Ra-226	Cs-137
02f	0.210	0.247	0.232	0.081	0.236	0.270	0.269	0.016	0.252	0.000
03f	0.946	0.208	0.180	0.060	0.189	0.190	0.187	0.017	0.185	0.000
04f	1.652	0.171	0.174	0.048	0.154	0.164	0.180	0.025	0.179	0.000
05f1	1.671	0.216	0.259	0.065	0.199	0.204	0.278	0.024	0.270	0.000
05f2	1.674	0.244	0.350	0.075	0.233	0.251	0.408	0.049	0.383	0.000
06f	1.609	0.189	0.949	0.067	0.215	0.235	1.126	0.069	1.050	0.000
07f1	1.480	0.198	0.651	0.065	0.188	0.198	0.711	0.042	0.686	0.000
07f2	0.273	0.288	0.286	0.092	0.275	0.320	0.341	0.017	0.316	0.000
08f	0.699	0.241	0.437	0.071	0.240	0.273	0.513	0.025	0.479	0.000
09f	0.487	0.253	0.343	0.078	0.241	0.263	0.397	0.020	0.372	0.000
10f1	0.400	0.000	0.148	0.032	0.101	0.116	0.170	0.016	0.160	0.000
10f2	0.417	0.082	0.137	0.026	0.062	0.091	0.154	0.013	0.147	0.000
11f	1.188	0.183	0.265	0.055	0.171	0.196	0.292	0.024	0.281	0.000
12f	1.549	0.234	0.341	0.069	0.213	0.231	0.367	0.027	0.356	0.000
13f1	1.320	0.190	0.207	0.057	0.178	0.189	0.233	0.019	0.221	0.000
13f2	1.515	0.169	0.208	0.048	0.160	0.161	0.215	0.018	0.213	0.000
14f1	1.256	0.150	0.095	0.044	0.134	0.141	0.100	0.024	0.099	0.000
14f2	1.627	0.190	0.171	0.056	0.176	0.174	0.322	0.028	0.250	0.000
15f1	1.196	0.160	0.204	0.052	0.152	0.166	0.224	0.018	0.216	0.000
15f2	1.501	0.235	0.264	0.058	0.211	0.192	0.291	0.018	0.279	0.000
16f	1.263	0.192	0.162	0.070	0.199	0.228	0.175	0.045	0.171	0.000

Bottom Ash

	K-40	Bi-212	Bi-214	Tl-208	Ac-228	Pb-212	Pb-214	U-235	Ra-226	Cs-137
02b	0.439	0.255	0.231	0.077	0.217	0.264	0.263	0.014	0.249	0.000
03b	0.467	0.104	0.077	0.031	0.095	0.101	0.086	0.009	0.083	0.000
04b	1.523	0.161	0.105	0.050	0.150	0.163	0.125	0.025	0.117	0.000
05b2	1.462	0.225	0.296	0.065	0.205	0.236	0.345	0.033	0.324	0.000
05b1	1.499	0.197	0.265	0.057	0.173	0.195	0.288	0.017	0.279	0.000
06b	1.179	0.142	0.625	0.044	0.152	0.153	0.708	0.031	0.673	0.000
07b1	0.974	0.137	0.566	0.044	0.141	0.159	0.664	0.032	0.619	0.000
07b2	1.023	0.158	0.521	0.056	0.151	0.158	0.594	0.028	0.562	0.000
08b	0.417	0.230	0.361	0.073	0.223	0.231	0.401	0.018	0.383	0.000
09b	0.331	0.247	0.311	0.069	0.202	0.231	0.353	0.019	0.334	0.000
10b1	1.073	0.273	0.355	0.082	0.252	0.298	0.396	0.020	0.379	0.000
10b2	0.086	0.000	0.061	0.006	0.020	0.022	0.070	0.000	0.016	0.000
11b	1.113	0.179	0.308	0.048	0.161	0.185	0.346	0.020	0.329	0.000
12b	1.248	0.192	0.294	0.057	0.173	0.209	0.316	0.023	0.307	0.000
13b1	0.815	0.122	0.170	0.041	0.116	0.132	0.191	0.012	0.182	0.000
13b2	0.953	0.139	0.208	0.039	0.126	0.143	0.223	0.014	0.217	0.000
14b1	1.445	0.142	0.190	0.040	0.123	0.144	0.210	0.016	0.201	0.000
14b2	1.319	0.170	0.213	0.048	0.138	0.158	0.230	0.013	0.224	0.000
15b1	1.253	0.180	0.128	0.057	0.163	0.176	0.139	0.019	0.134	0.000
15b2	0.756	0.132	0.109	0.036	0.106	0.109	0.116	0.012	0.113	0.000
16b	0.999	0.205	0.293	0.052	0.174	0.202	0.299	0.028	0.300	0.000

Coal

	K-40	Bi-212	Bi-214	Tl-208	Ac-228	Pb-212	Pb-214	U-235	Ra-226	Cs-137
02c	0.000	0.000	0.015	0.005	0.016	0.013	0.015	0.002	0.015	0.000
03c	0.086	0.000	0.016	0.004	0.012	0.017	0.016	0.002	0.016	0.000
04c	0.195	0.024	0.026	0.006	0.016	0.018	0.030	0.003	0.028	0.000
05c1	0.170	0.000	0.030	0.006	0.021	0.021	0.029	0.003	0.029	0.000
05c2	0.145	0.000	0.047	0.006	0.022	0.019	0.046	0.005	0.047	0.000
06c	0.143	0.000	0.092	0.005	0.016	0.020	0.094	0.006	0.094	0.000
07c1	0.143	0.000	0.092	0.005	0.016	0.020	0.094	0.006	0.094	0.000
07c2	0.037	0.000	0.012	0.004	0.013	0.011	0.011	0.002	0.012	0.000
08c	0.041	0.023	0.015	0.004	0.016	0.017	0.020	0.003	0.018	0.000
09c	0.037	0.000	0.012	0.004	0.013	0.011	0.011	0.002	0.012	0.000
10c1	0.153	0.048	0.043	0.013	0.038	0.041	0.041	0.004	0.042	0.000
10c2	0.105	0.000	0.030	0.005	0.020	0.014	0.034	0.003	0.032	0.000
11c	0.210	0.038	0.183	0.010	0.034	0.036	0.190	0.013	0.187	0.000
12c	0.124	0.000	0.023	0.005	0.015	0.017	0.029	0.003	0.026	0.000
13c1	0.119	0.000	0.034	0.004	0.015	0.015	0.022	0.001	0.028	0.000
13c2	0.122	0.000	0.037	0.005	0.015	0.015	0.027	0.003	0.033	0.000
14c1	0.168	0.000	0.040	0.004	0.015	0.016	0.032	0.003	0.036	0.000
14c2	0.122	0.000	0.035	0.004	0.016	0.014	0.030	0.002	0.033	0.000
15c1	0.074	0.000	0.017	0.005	0.014	0.015	0.017	0.004	0.017	0.000
15c2	0.074	0.000	0.017	0.005	0.014	0.015	0.017	0.004	0.017	0.000
16c	0.145	0.036	0.051	0.008	0.024	0.024	0.047	0.006	0.049	0.000

Common Materials

	K-40	Bi-212	Bi-214	Tl-208	Ac-228	Pb-212	Pb-214	U-235	Ra-226	Cs-137
Red Brick	2.290	0.114	0.093	0.040	0.130	0.134	0.109	0.008	0.102	0.000
Central Indiana Clay	1.142	0.084	0.062	0.025	0.079	0.088	0.076	0.008	0.070	0.012
Ward Limestone	0.312	0.000	0.022	0.000	0.013	0.012	0.026	0.002	0.024	0.000
North Dakota Granite	3.244	0.251	0.050	0.076	0.224	0.237	0.054	0.004	0.052	0.000
Vulcan Fill Sand	0.745	0.024	0.040	0.008	0.025	0.022	0.037	0.003	0.039	0.000
Vulcan Limestone	0.029	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000
I-465 and 56th Str.clay	1.244	0.064	0.075	0.018	0.060	0.057	0.075	0.006	0.075	0.003
Diatomaceous Earth	0.174	0.000	0.072	0.007	0.031	0.025	0.080	0.007	0.077	0.000
Dried Banana	0.422	0.000	0.013	0.000	0.000	0.000	0.013	0.003	0.013	0.000
Vermont Marble	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
50% KCl salt substitute	18.692	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
US 50, sta 520+50	1.015	0.156	0.308	0.038	0.127	0.151	0.358	0.018	0.336	0.000
US 50, sta 522+50	1.228	0.156	0.372	0.052	0.149	0.148	0.417	0.011	0.576	0.000

Appendix B

Total Activities (Bq/g) of Gamma-ray Emitting Isotopes *(All Samples)*

Commingled Ash		Fly Ash		Bottom Ash		Coal		Common Materials	
01p1	2.75	02f	1.81	02b	2.01	02c	0.08	Red Brick	3.02
01p2	2.94	03f	2.16	03b	1.05	03c	0.17	Central Indiana Clay	1.65
03p	2.15	04f	2.75	04b	2.42	04c	0.35	Ward Limestone	0.41
14p	2.38	05f1	3.19	05b1	2.97	05c1	0.31	North Dakota Granite	4.19
15p1	2.88	05f2	3.67	05b2	3.19	05c2	0.34	Vulcan Fill Sand	0.94
15p2	1.13	06f	5.51	06b	3.71	06c	0.47	Vulcan Limestone	0.03
15p3	2.08	07f1	4.22	07b1	3.34	07c1	0.47	I-465 and 56th Str.clay	1.68
15p4	2.41	07f2	2.21	07b2	3.25	07c2	0.10	Diatomaceous Earth	0.47
		08f	2.98	07b3	2.23	08c	0.16	Dried Banana	0.46
		09f	2.45	08b	2.34	09c	0.10	Vermont Marble	0.00
		10f1	1.14	09b	2.10	10c1	0.42	50% KCl salt substitute	18.69
		10f2	1.13	10b1	3.13	10c2	0.24	US 50, sta 520+50	2.51
		10f3	1.23	10b2	0.28	11c	0.90	US 50, sta 522+50	3.11
		11f1	2.66	11b	2.69	12c	0.24		
		11f2	2.98	12b	2.82	13c1	0.24		
		11f3	3.11	13b1	1.78	13c2	0.26		
		12f1	3.39	13b2	2.06	14c1	0.31		
		12f2	3.06	14b1	2.51	14c2	0.26		
		13f1	2.61	14b2	2.51	15c1	0.16		
		13f2	2.71	15b1	2.25	15c2	0.16		
		14f1	2.04	15b2	1.49	16c	0.39		
		14f2	2.99	16b	2.55				
		14f3	2.00						
		15f1	2.39						
		15f2	3.05						
		15f3	1.72						
		16f	2.50						

Appendix C

Densities (g/cm^3) of 1-Liter Test Samples

Commingled Ash		Fly Ash		Bottom Ash		Coal		Common Materials	
01p1	1.549	02f	1.196	02b	1.150	02c	0.589	Red Brick	1.153
01p2	1.387	03f	0.546	03b	0.822	03c	0.638	Cent IN Clay	1.112
03p	1.073	04f	0.817	04b	1.103	04c	0.831	Ward Limestone	1.514
14p	1.693	05f1	0.730	05b1	1.358	05c1	0.798	ND Granite	1.458
15p1	0.479	05f2	0.936	05b2	1.296	05c2	0.819	Vulcan Fill Sand	1.584
15p2	0.676	06f	1.075	06b	1.365	06c	0.784	Vulcan Limestone	1.607
15p3	0.425	07f1	0.696	07b1	1.211	07c1	0.784	I-465 and 56th Str.clay	1.403
15p4	0.724	07f2	1.138	07b2	1.286	07c2	0.767	Diatomaceous Earth	0.420
		08f	1.060	07b3	1.524	08c	0.728	Vermont Marble	1.696
		09f	1.044	08b	1.505	09c	0.767	Dried Banana	0.303
		10f1	0.260	09b	1.512	10c1	0.730	50% KCl	1.193
		10f2	1.130	10b1	1.090	10c2	0.740	US 50, sta 520+50	1.375
		10f3	0.308	10b2	1.505	11c	0.775	US 50, sta 522+50	1.515
		11f1	1.069	11b	0.934	12c	0.802		
		11f2	1.138	12b	0.693	13c1	0.752		
		11f3	1.272	13b1	1.186	13c2	0.753		
		12f1	0.595	13b2	1.270	14c1	0.770		
		12f2	0.302	14b1	1.007	14c2	0.789		
		13f1	0.726	14b2	1.069	15c1	0.721		
		13f2	0.615	15b1	0.756	15c2	0.721		
		14f1	0.782	15b2	0.657	16c	0.725		
		14f2	0.625	16b	0.679				
		14f3	0.915						
		15f1	0.413						
		15f2	0.447						
		15f3	0.416						
		16f	1.283						

Appendix D

Dose Equivalent (mrem/yr) from an Embankment Composed of Gamma-ray Emitting Materials

Commingled Ash		Fly Ash		Bottom Ash		Common Materials	
01p1	14.1	02f	13.3	02b	14.4	Red Brick	15.2
01p2	16.2	03f	27.4	03b	9.0	Cent IN Clay	8.9
03p	14.9	04f	21.6	04b	13.8	Ward Limestone	1.6
14p	22.2	05f1	29.8	05b1	15.6	ND Granite	15.7
15p1	26.1	05f2	28.0	05b2	24.2	Vulcan Fill Sand	4.8
15p2	10.9	06f	42.5	06b	22.6	Vulcan Limestone	0.1
15p3	23.0	07f1	47.8	07b1	22.9	I-465 and 56th Str.clay	8.7
15p4	39.6	07f2	12.7	07b2	37.5	Diatomaceous Earth	3.6
		08f	23.3	07b3	13.0	Vermont Marble	0.0
		09f	20.0	08b	13.8	US-50 sta 520+50	13.8
		10f1	29.4	09b	12.1	US-50 sta 522+50	15.1
		10f2	7.9	10b1	22.4		
		10f3	27.4	10b2	8.1		
		11f1	18.2	11b	21.4		
		11f2	20.6	12b	29.1		
		11f3	25.2	13b1	11.0		
		12f1	39.8	13b2	11.9		
		12f2	59.1	14b1	16.6		
		13f1	24.5	14b2	17.3		
		13f2	14.7	15b1	19.5		
		14f1	16.3	15b2	15.2		
		14f2	23.6	16b	27.7		
		14f3	14.7				
		15f1	36.6				
		15f2	46.8				
		15f3	15.6				
		16f	13.6				

Appendix E

Dose Equivalent (mrem/yr) from Common Sources

Television	2
Internal Radiation	20
Terrestrial Sources	30
Cosmic Radiation	30
Brick Houses	40
Inhaled Radon	200

Appendix F
Key Definitions Listing

1. **Activity:** Rate of decay of radionuclides.

2. **Bequerel:** The metric unit of activity is called a Bequerel (Bq), which corresponds to one particle (i.e., nucleus, alpha particle, beta particle, or photon) emitted per second.

3. **Curie & picoCurie:** The English unit of activity is called a Curie (Ci). Units of Curies are considerably larger than Bequerels. $3.7x10^{10}$ Bq is equivalent to one Curie. A picoCurie (pCi), a unit most used to describe very low levels or radiation, is 10^{-12} Curies.

4. **rad:** Radiation Absorbed Dose is a measure of the amount of energy that is deposited per unit mass of an incident material.

5. **rem:** Radiation Equivalent Man is a measure of the amount of biological effect on a human caused by radiation exposure. It is the number of rads multiplied by the relative biological effectiveness (RBE).

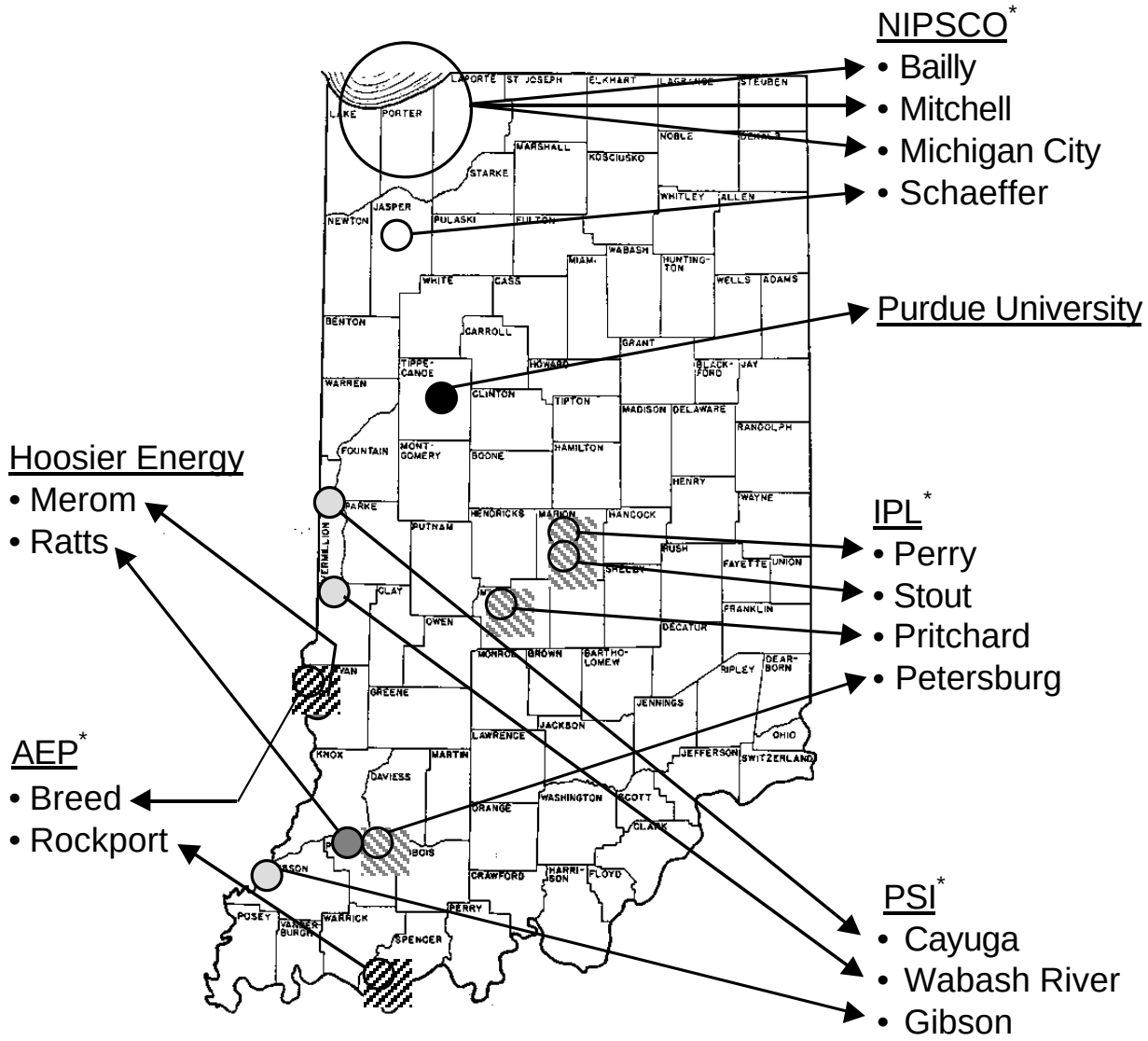
6. **intensity:** The intensity of a gamma ray refers to the average number of times it is released during a nuclear decay. The intensity of a 1460 keV gamma ray from the decay of Potassium-40 is 0.1067, meaning that this particular gamma ray is emitted by 10.67% of all Potassium-40 decays.

7. **absorption/attenuation coefficient:** An attenuation coefficient is the probability that a particle will pass unimpeded through a shield with a thickness of one centimeter. An absorption coefficient is the probability that a particle deposits energy into a shield that is one centimeter thick. Both of these values have units of inverse centimeters.

8. **spectrum:** Energy spectrums provide a representation of the energy and number of photons being emitted by a given material. Each radionuclide contributes part of a composite spectrum displayed by a multi-channel analyzer. Each specific radionuclide can be identified by its unique contribution to this spectrum.

Appendix G
Sampling Locations within the State of Indiana (16 sites)

INDOT - Purdue Coal Ash Radiation Evaluation
Power Plant Coal Ash Sampling Locations



AEP American Electric Power
PSI Public Service Indiana
IPL Indianapolis Power & Light
NIPSCO Northern Indiana Power Supply Company