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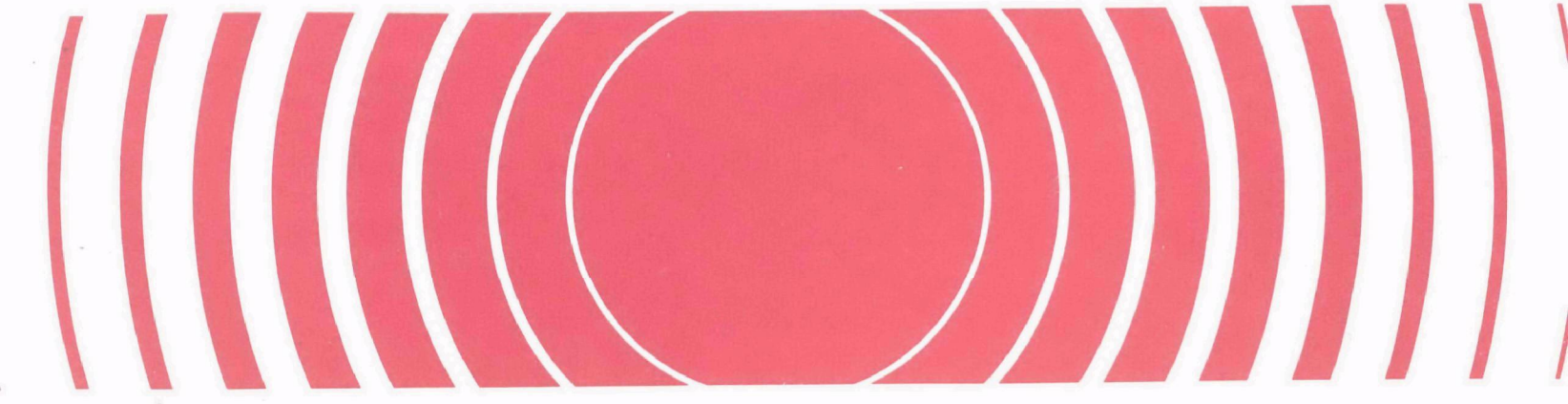
Characterization of Contaminated Soil from the Montclair/Glen Ridge, New Jersey, Superfund Sites



Characterization of Contaminated Soil from the Montclair/Glen Ridge, New Jersey Superfund Sites

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FOREWORD

The Superfund Amendments and Reauthorization Act (SARA) of 1986 and the revision of the National Contingency Plan (NCP) placed new requirements on hazardous waste site cleanup actions. Remedial action at Superfund sites must protect human health and the environment and meet applicable or relevant and appropriate requirements (ARARS) as established by Federal and State standards. The selection of remedies must also be cost-effective and use permanent solutions and treatment technologies or resource recovery technologies to the maximum extent practicable. Treatment methods which permanently and significantly reduce the mobility, toxicity, or volume of hazardous substance are preferred in this Superfund requirement.

The Office of Radiation Programs (ORP), in concert with Superfund requirements, is evaluating the use of physical volume reduction and chemical extraction (VORCE) to remediate radium contaminated soils at the Montclair and Glen Ridge, New Jersey, Superfund sites. The VORCE investigation at these New Jersey sites consists of 1) soil characterization, 2) treatment studies, and 3) technology implementation. The characterization of the soil includes particle size distribution, mineralogical identification, chemical measurements, and radioassay. The soil characterization phase provides important data that paves the way for the treatment and implementation phases that follow. Knowledge of radioactive contaminant distribution as a function of particle size and soil mineralogy is especially fundamental in assessing remedial measures.

This report documents the soil characterization phase conducted at the EPA Eastern Environmental Radiation Facility (EERF) in Montgomery, Alabama. The identification of the spatial distribution of the radium within the soil and its association with specific minerals or materials is shown to relate to the later implementation phase. Procedures developed in this investigation have application to other radioactively contaminated Superfund sites.

The Agency invites all readers of this report to send any comments or suggestions to Mr. Martin P. Halper, Director, Analysis and Support Division, Office of Radiation Programs (ANR-461), U.S. Environmental Protection Agency, Washington, DC 20460.

Richard J. Guimond, Director
Office of Radiation Programs

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I also wish to acknowledge the assistance of Dr. Fred Au of the ORP Las Vegas Facility in supervising contractual services from the University of Nevada and Huffman Laboratories. I also wish to thank Mr. Ray Willingham of the U.S. Army Corps of Engineers, Marietta, Georgia for sedimentation and centrifuge analysis of select samples which enabled completion of a grain size curve and the preparation of silt/clay size fractions for further mineral and radioassay analysis.

I am especially indebted to Mr. R. Jeff Serne and Dr. Robert Erikson of the Battelle Northwest Laboratories for their study of the silt and clay-size fractions with the scanning electron microscope and energy dispersive spectrometer analysis technique and the x-ray diffraction and radioassay of fractions of silt and clay separated by the linear density gradient method. I wish also to thank Mss. Tonya Hudson and Debbie Whittaker of Sanford Cohen and Associates, Incorporated, for their assistance in heavy liquid separations at the Eastern Environmental Radiation Facilities Laboratory. Thanks are also extended to Mss. Virginia Stradford, Bonnie Wyvill, Eleanor Paige, and Phoebe Suber for typing this document.

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ABSTRACT

Volume reduction and treatment techniques are being investigated for possible use in remediation of radium-contaminated soil at the Montclair and Glen Ridge, New Jersey, Superfund sites. The contamination occurs in residential areas situated over former landfills that received various industrial wastes including radium mill tailing wastes in the 1920's. One of the first steps in the investigation was characterization of the soil to determine the most efficient remediation methods. The average radium activity of the contaminated soil is estimated to be in the range of 60-200 pCi/g with higher activity (1000-10,000 pCi/g) occurring in the fine materials.

The characterization procedure included detailed physical sizing, special chemical tests, and radiochemical and mineralogical analysis of 18 size fractions ranging from gravel to clay size on each sample. Special magnetic, heavy liquid, and linear density centrifugation separations were also made on soil fractions to facilitate identification of minerals and materials with high radium activity.

About half of the radium activity is attributed to acid leach radium milling products (radiobarite and amorphous silica). The remainder of the radium is contained in uranium ore minerals (carnotite and uraninite), incinerated radium materials (ferruginous slag), and adsorbtion of radium from solution onto native soil particles (illite, hematite, etc.). An occasional radium vial is also a local anomaly at the sites.

Less than one third of the radium occurs in the soil material greater than 50 sieve (0.15mm) size. This material is comprised essentially of incineration products (coal, coaly slag, white siliceous slag, and ferruginous slag) and native rock particles (sandstone, siltstone, quartzite, etc.). Glass and trash also occur in minor amounts. Approximately 10-15 percent of the radium activity in this fraction is associated with magnetic ferruginous slag particles. The ferruginous slag activity consists of uraninite from incinerated coal ash, incinerated material containing radium, and adsorbed radium. The magnetic properties and coarse particle size of these materials lend themselves to physical separation procedures.

The contaminated soil less than 50 sieve size at both the Glen Ridge and Montclair sites contains more than two thirds of the radium activity which is predominately in the silt and clay fractions comprising about one-fifth of the sample volume. The radiobarite and amorphous silica contain the major portion of the radium activity. Uranium ore minerals occur mainly in the sand and upper silt size and comprise about 10 percent of the radium activity. Background minerals (zircon, monazite, etc.) and adsorbed radium on illite and other clay minerals comprise another 5-15 percent of the radium activity.

1. INTRODUCTION

The passage of the 1986 Superfund Amendments and Reauthorization Act (SARA) and the revision of the National Contingency Plan (NCP) placed new requirements on hazardous waste site cleanup, emphasizing treatment over traditional remediation approaches. In concert with these requirements for the cleanup of sites on the National Priority List (NPL), the Environmental Protection Agency's (EPA) Office of Radiation Programs (ORP) is evaluating the use of physical volume reduction and chemical extraction to remediate radium contaminated soils at the Montclair and Glen Ridge, New Jersey, Superfund sites. These residential sites are located over former landfill areas that received radium contaminated tailings and other radium industrial wastes in the 1920's.

The Volume Reduction/Chemical Extraction (VORCE) project initiated as the remediation study of the Montclair and Glen Ridge Superfund sites consists of 1) soil characterization, 2) treatment studies, and 3) technology implementation. The characterization of the soil includes physical separations, mineralogical identification, chemical measurements, and radioassay. The soil characterization phase provides important data that paves the way for the treatment and implementation phases that follow. Knowledge of radioactive contaminant distribution as a function of particle size and soil mineralogy is especially fundamental in assessing remedial measures.

This report documents the soil characterization phase conducted at the EPA Eastern Environmental Radiation Facility (EERF) in Montgomery, Alabama; the Battelle Pacific Northwest Laboratory (PNL), Richland, Washington; the EPA Las Vegas Facility (LVF); the University of Nevada at Las Vegas, Nevada; and the Army Corps of Engineers (COE), Marietta, Georgia.

2. SITE INVESTIGATIONS AND GEOLOGY

Residential areas of Montclair, Glen Ridge, and West Orange, New Jersey, covering approximately 50, 45, and 9 acres, respectively, contain an estimated 300,000 cubic yards of soil contaminated with high levels of gamma radiation and radon gas. Radioactive contamination is associated with the natural soil to depths up to 20 feet. The contaminated areas are former landfills that received a variety of waste materials. The waste material was randomly dumped and subsequently covered with a layer of top soil up to 3-feet thick to provide land suitable

for housing construction. In 1979, an aerial gamma survey by the New Jersey Department of Environmental Protection identified the sites of elevated gamma radiation.

Studies suggested that the radium was discarded tailing waste from extraction of radium from uranium ore (carnotite from Colorado Plateau) by the acid leach process from mills located in New Jersey. Field borings by Camp Dresser and McKee, Inc. (CDM 885) found "hot spots" of these White Sands tailings as thin layers in the reddish brown to grey colored admixture of coal, ash, soil, construction material, and industrial wastes found in various proportions in the former landfill areas that constituted the contaminated soil sites. Olsson in a cursory examination of the White Sands at West Orange, NJ, site, cited the presence of yellow carnotite and barite (coprecipitate in acid leach process) in these sands (OL 86). The production of radium in New Jersey terminated in 1926 and after 1922, Belgian Congo (Zaire) uranium ore (uraninite) replaced the Colorado Plateau uranium ore (carnotite) as the world's primary source of radium (La 84). While these early investigations did not identify uraninite at these sites, this mineral is of common occurrence in the landfill; however, the source of the uraninite may not necessarily be related to a foreign source, as will be discussed in Section 5.2.3.1.

The only information from earlier investigations that related radioactivity to particle size was on the thin bands of tailing sands designated as hot spots. This information provided focus on sand-sized and finer materials as the most troublesome source of radioactivity. If the radium milling waste and raw ore constitute the major source of contamination, it is not surprising that the sand size or smaller material played the major role since the ore was crushed and passed through number 16 sieve (1.18mm) prior to the acid leach treatment (d'A 21). The lesser radioactivity of larger materials is therefore related to accumulation of radium residues in incinerated slags or from materials that adsorbed radium.

In a reconnaissance visit to the Montclair/Glen Ridge, NJ, sites, observations were made of the glacial deposits hosting the contaminated landfill areas. Laboratory analysis of the glacial material outside of the contaminated area in nearby Nishuane Park in Montclair, NJ, revealed that the fines (fine sand, silt, and clay) consist of approximately 70 percent quartz, 15 percent feldspar, 5 percent mafic mineral (hornblende, etc.), 5 percent mica/illite, 5 percent chlorite,

and trace amounts of kaolinite and montmorillonite (Ne 87). The rock particles in the glacial material arepredominantly smooth, rounded quartzite and subangular, brownish-red sandstone and siltstone; minor other varieties are granite, basalt, and varieties of metamorphic rock.

The former landfills are contained in surficial host beds of unconsolidated Pleistocene glacial deposits that range in thickness from 28 to 84 feet above bedrock. The glacial deposits at Montclair are poorly sorted glacial till (boulder clay) while well sorted stratified drift of more uniform texture occurs at the Glen Ridge site.

The bedrock underlying the glacial deposits consists of alternating strata of sedimentary rock units of the Brunswick formation of Triassic age. The sedimentary rock units are red beds of sandstone and siltstone and local conglomerate that dip gently to the northwest. Rounded to subangular gravel-size particles of these sedimentary units are well represented in the coarser materials of the glacial deposits that overlay the bedrock.

The average activity of the Montclair and Glen Ridge contaminated soils is estimated at 64.3 pCi/g (NJEPD 87). These soils are located in the unsaturated zone above the water table at all the sites. The radium 226 activity in the groundwater beneath the contaminated soil is 2.3 pCi/L, at all but one location. These are within the maximum limit of 5.0 pCi/L dictated by the Safe Drinking Water (SDW) standard, however, the 11 pCi/L of radium 226 at one Montcliar site does exceed the SDW standard.

3. CHARACTERIZATION PROCEDURE

The majority of the soil characterization testing phase was conducted at the EPA's Eastern Environmental Radiation Facility (EERF) Laboratory in Montgomery, Alabama, using representative samples acquired and processed from the Montclair and Glen Ridge sites. A small quantity of high radium activity tailings sands from a location in the Montclair site was also tested. This sample is referred to in this report as "White Sands." The representative samples were oven dried at 60^{o}C prior to size separations into fractions by sieving, sedimentation,

centrifugation, and heavy liquid methods. The upper limit of 60°C in the oven drying stage was fixed so as to maintain the clay mineral structure which might otherwise be lost at higher temperatures, e.g., montmorillonite loses some interlayer water at higher temperatures.

The testing methods used to characterize the contaminated soil and the size fractions used in this initial phase of the VORCE project are listed in Figure 1. Initial sizing and radioassay of all size fractions were performed at the EERF laboratory prior to sending representative fractions to other laboratories for special tests. All heavy mineral and magnetic separations and detailed radioassay and petrographic microscope examinations of gravel and sand-size soil fractions were conducted at the EERF laboratory; however, special testing of silt and clay-size fractions as well as ancillary testing of bulk samples (chemical and cursory X-ray diffraction analysis) was conducted by other laboratories (identified in the discussion of testing methods.)

3.1 PHYSICAL METHODS

The physical testing included screening, sieving, sedimentation, and centrifugation techniques as well as special density separations employing heavy liquids. The purpose was to obtain a texture grain-size distribution curve and 18 gradational soil fractions on each sample for various testing procedures designed to provide data for the treatment studies and technology implementation phases.

3.1.1 Screening and Sieving

Dry screening, using a Gilson Screener (Model TM-4), was performed at the EERF laboratory on approximately 2-kg samples of Montclair and Glen Ridge soil. The nest of screens included: 1 inch, 1/2 inch, 1/4 inch, and No. 4 sieve (4.75 mm). The weight percent of gravel on each screen was determined for the gravel-size (+ No. 4 sieve) material (Figure 2). The minus number 4 size soil was saved for wet sieving.

Representative 100-gram portions of minus number 4 sieve size samples were wet sieved in a Brinkman Table Top Vibrator Screener using sieve numbers 10 (2 mm), 16 (1.18 mm), 50 (300 microns), 100 (150 microns, 140 (106 microns) and 200 (75 microns). The cumulative weight, in relation to total sample, was determined for each sieve size and recorded (Figure 2).

Sieve No.	Size(mm)	Soil Size	Sizing Method	Separation Method	Analytical Methods
4	25.00 12.50 4.75	Gravel	Gilson Mechanical Screener		Gamma Spectroscopy Alpha Spectroscopy Magnetic Properties
10 16 50 60 100 140 200	2.00 1.18 .30 .25 .15 .106 .075	Sand	Brinkman Vibrator Screener	Bromoform and Tetrabromoethane Sink Float Method (heavy mineral concentration)	Gamma Spectroscopy Alpha Spectroscopy Petrographic Microscopy Chemistry Magnetic Properties
270 400	.050 .038 .015 .005 .002	Silt	Sedimentation	Heavy Liquid Linear Density Method (high activity separation)	Gamma Spectroscopy Alpha Spectroscopy X-Ray Diffraction Scanning/Transmission Electron Microscope w/X-Ray Analyzer
	.0005 -.0005	Clay	Centrifugation		

Figure 1. Laboratory methods for characterization of radium contaminated soils

	Sieve Size	Microns	Weight Percent Retained	
			Montclair	Glen Ridge
Gravel	1 inch		5	4
	1/2 inch		8	9
	1/4 inch		9	13
	No. 4	4750	4	9
Sand	No. 10	2000	6	10
	No. 16	1180	3	3
	No. 50	300	20	17
	No. 60	250	6	4
	No. 100	150	8	5
	No. 140	106	7	3
	No. 200	75	3	3
Silt		53	6	4
		38	3	3
		15	6	4
		5	4	5
		2	1	2
Clay		0.5	0.5	1
		Pan	0.5	1

Figure 2. Weight percent of Montclair and Glen Ridge contaminated soil retained by selected sieves.

The minus number 200 sieve size material was collected for several runs and representative Montclair, Glen Ridge, and White Sands samples were collected, radioassayed and delivered to the Army of Corps of Engineers (COE), Marietta, Georgia, laboratory for separation into 5 silt-size and 2 clay-size fractions.

3.1.2 Sedimentation and Centrifuge Separations

The representative silt and clay-size fractions of the Montclair, Glen Ridge, and White Sands samples were separated into the following fractions by sedimentation and centrifuge techniques: silt-size into -75/+50 microns, -50/+38 microns, -38/+15 microns, -15/+5 microns, and -5/+2 microns; clay-size into -2/+0.5 microns and -0.5 microns. The cumulative fractions from several runs were combined to obtain sufficient sample material for the mineral identification tests and radioassay analyses. The weight percent was obtained on the larger silt-sizes by weighing on a top-loading balance. The percentage distributions of the finest fractions were obtained by the pipet method (Fo 74). The weight distribution is listed in Figure 2.

The COE prepared texture grain-size distribution curves from pipet analysis (silt and clay) data and screen and sieve (gravel and sand) data provided by EERF. These grain-size curves and their construction provide the means of proportioning weight distribution for any fractional size consideration.

3.1.3 Density Separations

Since high density uranium minerals and radium bearing barite (radiobarite) could comprise a relatively high percentage of the radioactive contamination, the sand and upper silt-size soil was separated into a heavy (greater than 2.9 specific gravity) and light fraction by simple sink-float techniques. The heavy liquids used for the separations at the EERF laboratory were bromoform and tetrabromoethene.

Density separation of silt and clay-size particles is possible by linear density gradient separations using heavy liquids and centrifugation techniques, such as described by Mattigod and Ervin (Ma 83). The silt and clay-size fractions

sent to the PNL, Richland, Washington, laboratory were prepared for linear density gradient separations using continuous mixtures of heavy liquid (tetrabromethane) and a less dense liquid (absolute ethanol and polyvinylpyrolidonone). The separation of the soil minerals was then achieved by centrifugation until three isopycnic bands were formed in a series in the centrifuge tubes (Er 88).

The sink-float density separation of sand-size soil fractions provided positive identification of mineral constituents using a Nikon Research Polarizing Microscope at the EERF laboratory. Linear density separations of the silt and clay-size particles at the PNL laboratory enabled X-ray diffraction of the separate density bands and microprobe fluoresence of particles to facilitate mineral identification.

3.2 MINERAL IDENTIFICATION METHODS

The goal of mineral identification is to provide data to assess the association of radionuclides with specific minerals or materials comprising the contaminated soil. Radionuclides occur in the White Sands and in the complex admixture of native soil, industrial waste, and unprocessed uranium ore as components of mineral structures and adsorbates on particle surfaces. Thus, in addition to isolation and concentration by gradation fractions and density separations, a combination of instrumentation and chemical methods for mineral identification and soil characterization is required (Figure 1). These are addressed in the following paragraphs.

3.2.1 Radioassay and Chemical Methods

The EERF laboratory radioassayed each size fraction of the Montclair, Glen Ridge, and White Sands samples as an initial step in determining size ranges in which the radium 226 activity was concentrated. The radium 226 activity levels were obtained by gamma-ray spectroscopy using nigh purity germanium detectors (Li 84). Radium 226 was identified and measured using the 186 MeV photopeak. Since only very small quantities of uranium 235 were found in the samples (0.1 percent), interference by a 185 MeV photopeak was not a significant consideration (Ri 88). The measured radium 226 activity will be discussed in Section 4.

The uranium isotopes and their daughters were estimated on the Glen Ridge and White Sands silt/clay size linear density gradient bands by counting gamma and X-rays on a high-resolution intrinsic germanium diode coupled to a Nuclear Data ND 6620 Nuclear Analyzer. Details of this procedure are contained in the PNL report by Erikson and Serne (ER 89). The significance of this data will be discussed in a later section.

The EERF laboratory also performed radiochemical analysis of uranium 238, radium 226, thorium 230, and thorium 232 on select gravel particles and the light and heavy density sand-size fractions of the Montclair, Glen Ridge, and White Sands samples. The radium 226 was measured by gamma-ray spectroscopy and the other radioisotopes by radiochemical methods (Li 84). The thorium was separated by ion-exchange chromatography and counted by alpha spectroscopsy using thorium 234 as a tracer to determine the chemical yield. Uranium was extracted from the thorium analysis mixture into triisoctylamine (TIOA), stripped from the extract with nitric acid, coprecipitated with lanthanum fluoride carrier, and counted by alpha spectroscopy using uranium 232 as a tracer to determine the chemical yield (Li 84). These results and additional chemical analysis of vanadium, iron, and barium, by Galbraith Laboratories, Inc., are reported in appropriate sections of this report.

Specific chemical tests were performed on selected fractions of the Montclair, Glen Ridge, and White Sands samples. These tests, conducted by Huffman Laboratories, Inc. were for identification of chemical constituents that might interfere with volume reduction and chemical extraction processes. Of special consideration are the values of barium and vanadium which are used as chemical signatures in the assessment of radiobarite and carnotite. The results of these tests will be discussed in a later section.

3.2.2 Petrographic Polarizing Microscope and Microscopic Methods

The gravel and sand-size materials were initially identified by visual examination and a binocular microscope. The Nikon Research Polarizing Microscope, Model OPTIPHOT-POL, was used to examine the light and heavy minerals in the size range from -50 sieve size (300 microns) to +270 sieve size (53 microns), using area statistical counts on 300 grains of each sand fraction. Opaque minerals were examined using reflected light. Select size fractions were also subjected to chemical

assay for those elements diagnostic to specific minerals of interest, e.g., Ba is diagnositic to radiobarite of the radium extraction tailings sands and vanadium is a signature of carnotite (hydrated potassium uranium vanadate) from the Colorado Plateau uranium ore. X-ray diffraction and magnetic separations were also used to support identification of opaque minerals and some of the industrial gravel slag particles.

3.2.3 X-Ray Diffraction

Initial x-ray diffraction of select gravel particles and all size fractions less than number 4 sieve size, was performed at the University of Nevada (Las Vegas) on a Philips (Norelco) x-ray diffraction unit, Model 12206/53. Sedimented powder slides were subjected to a scan between 4 degrees and 50 degrees 2 theta to obtain qualitative cursory information on major mineral phases. X-ray diffractograms of some of the fine sand and silt fractions contained identifiable peaks of carnotite. In the White Sands, barite was identified as the most abundant of the heavy minerals (greater than 2.9 specific gravity) in the sand-size material. These initial x-ray diffraction scans, nowever, did little more than show the complex nature of the contaminated soils and the need for petrographic microscopic analysis in the sand-size material and use of linear density gradient separations for x-ray diffraction of silt and clay-size materials.

The soil mineralogy of the linear density separated silt and clay fractions prepared by the PNL Richland laboratory was determined using a Philips ADP-3520 x-ray diffractometer equipped with a graphite monochrometer. Each sample was scanned between 4 and 65 degrees 2-theta using Cu-K alpha radiation at 40 kV, 20mA. All diffractograms were obtained on soil samples oriented on glass slides using absolute ethanol.

3.2.4 Scanning Electron Microscopy/Energy Dispersive Energy Analyzer

Silt and clay-size soil fractions having the highest radionuclide concentrations were further characterized by the PNL Richland laboratory to determine the associations of the radionuclides with various soil minerals. Samples were mounted on brass stubs and were carbon coated. A JEOL JSM-25S III scanning electron microscope equipped with a Tracor Northern TN-2000 energy dispersive x-ray spectrometer (EDS) was used at an accelerating potential of 30 kV.

3.2.5 Magnetic Separation

The sand-size heavy mineral fractions of the Montclair and Glen Ridge contaminated soils were separated into magnetic and nonmagnetic fractions by a standard laboratory hand-held magnet with a steel needle attachment. The magnetic separates were weighed on a table top balance and representative portions of the magnetic and nonmagnetic fractions assayed for U-238, Th-230, Th-232, Ra-226, Ba, V, and Fe.

4. RESULTS OF RADIUM 226 ANALYSIS ON SOIL FRACTIONS

The radium 226 activity level of the bulk soil samples was 814 and 182 pCi/g respectively for the Glen Ridge and Montclair sites and 3400 pCi/g for the White Sands sample. This activity level is significantly higher than the average reported for the Montclair and Glen Ridge contaminated soils (NJEDP 87); however, the more highly contaminated soils were selected intentionally in order to more effectively evaluate contaminating source materials.

The radium 226 activity levels for gravel sized materials and the individual fractions of sand, silt, and clay-sized materials are listed in Table 1. In all samples, the concentratiaon of radium 226 is greatest in the fine fractions. The Glen Ridge silt/clay fractions comprise approximately 20 percent of the contaminated soil; however, they contain 57 percent of the radium with highest values of greater than 3,000 pCi/g in the fine silt and clay-size materials. The Montclair samples contain less radium activity than the Glen Ridge samples, but the percentage distribution of radium is generally similar. The Montclair gravel size contains less slag and more rock particles than the Glen Ridge gravel-size materials.

The White Sands are a recognized radioactive anomaly in the contaminated soil and this material has been cited as probable tailing waste from the extraction of radium by the acid leach milling process (O1 86). The radium 226 activity levels of this material of predominantly sand-silt size is several times that found in comparable Glen Ridge soil fractions and as much as an order of magnitude greater than similar sizes of Montclair soil fractions (Table 1). The highest radium 226 activity in the sand, silt, and clay size fractions of the White Sand is respectively 1,913, 17,620, and 21,800 pCi/g.

Table 1. Radium 226 activity and percentages on gravel, sand, and silt/clay sizes. Analysis by EERF.

Size Sieve	Microns	White Sands	Glen Ridge			Montclair		
		pCi/g	Wgt %	pCi/g	%Ra	Wgt %	pCi/g	%Ra
-1 1/2/+4	4750	-	35	346	16	26	44	8
-4/+10	2000	296	10	307		6	26	
-10/+16	1180	641	3	268		3	39	
-16/+50	300	993	17	500	27	20	100	33
-50/+100	150	980	9	472		14	113	
-100/+140	106	1,036	3	498		7	138	
140/+200	75	1,913	3	677		3	170	
	50	8,656	4	1,047		6	304	
	38	14,490	3	953		3	240	
	15	9,525	4	1,164		6	360	
	5	15,170	5	3,034		4	430	
	2	17,620	2	3,039		1	644	
					57			59
	0.5	21,800	1	576		0.5	1,113	
	-0.5	3,725	1	3,301		0.5	264	

Notes: 1. The average Ra 226 activity on a representative White Sands, Glen Ridge, and Montclair sample is respectively 3400, 814, and 182 pCi/g.

The source of the radium activity in the soil fractions and the association of radium 226 with specific minerals or materials in the soil fractions is one of the major considerations in the sections that follow.

5. MINERAL COMPOSITION OF CONTAMINATED SOIL

The contaminated soils at the Montclair and Glen Ridge sites are heterogeneous mixtures of native soil, industrial wastes, mining waste, and household waste materials. These materials are characterized in Table 2 with the average composition computed as the sum of the weighted averages of the 18 soil fractions tested for each sample. The average mineral composition for each size class (gravel, sand, silt, and clay-size) is also listed in Table 2. The chemical compositions listed in Table 3 were used to help assess the percentages of some of the minerals.

The composition of the coarser gravel-size materials stands in sharp contrast to the more homogeneous fine sand, silt, and clay-size particles that contain the highest activity levels of radium 226. In order to better relate the activity levels of Ra-226, U-238, and Th-230 to the mineral composition, radiochemical and chemical data, each size class will be treated separately.

5.1 GRAVEL-SIZED MATERIAL

The gravel sized materials are those retained between the No. 4 sieve-size (4.75mm) and 3 inch sieve size as specified in ASTM D643-78. The ASTM standard is one of the several grain-size scales described that is an acceptable standard currently in use in the United States (Di 82). Since the boulders were removed from the samples to facilitate mixing, the gravel-size material is the largest size class of the samples tested. The gravel-sized materials, with boulders removed, comprise 35 percent of the Glen Ridge and 26 percent of the Montclair contaminated soil. A typical gravel-size fraction is shown for the Montclair sample in Plate 1. The radium 226 activity on particle types and the fractional percentage distribution of particles within the gravel size material is

TABLE 2. MINERAL AND MATERIAL COMPOSITION OF MONTCLAIR/GLEN RIDGE

SIZE CLASS	GRAVEL		SAND		SILT		CLAY		AVERAGE %	
SITE LOCATION	G/R	Mtclr	G/R	Mtclr	G/R	Mtclr	G/R	Mtclr	G/R	Mtclr
WEIGHT PERCENT	35	26	45	53	18	20	2	1		
FURNACE-FIRED MATERIALS										
ANTHRACITE COAL	12	13	2	3	—	—	—	—	5	5
COALY SLAG	44	22	14	19	—	—	—	—	22	16
WHITE SLAG	20	5	18	12	—	—	—	—	15	8
FERRUGINOUS SLAG	7	5	2	1	—	—	—	—	3	2
GLASS/TRASH ROCK PARTICLES	5	3	—	—	—	—	—	—	2	1
SANDSTONE/SILTSTONE	6	20	3	2	2	—	—	—	4	6
QUARTZITE	5	25	5	3	3	—	—	—	4	8
GRANITE/BASALT	1	7	—	—	—	—	—	—	1	2
MINERALS										
QUARTZ	—	—	51	52	63	73	15	15	35	43
FELDSPAR	—	—	2	2	1	2	1	1	1	2
RADIOACTIVE[1]										
AMORPHOUS SILICA	—	—	0.1	0.1	20	10	20	10	4.1	2.2
RADIOBARITE	—	—	0.3	0.2	0.5	0.2	0.5	0.2	0.2	0.1
CARNOTITE	—	—	0.7	0.2	0.9	0.2	0.9	0.2	0.5	0.1
URANINITE	—	—	0.8	0.3	0.1	0.1	0.1	0.1	0.2	0.1
ZIRCON	—	—	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
MONAZITE	—	—	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
OTHER RADIOACTIVE[2]	—	—	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
CLAY MINERALS										
ILLITE	—	—	0.1	0.1	8	8	40	45	1.2	1.4
KAOLINITE/CHLORITE	—	—	0.1	0.1	5	5	22	22	0.7	1.1
HEAVY MINERALS										
MAGNETITE	—	—	0.4	0.3	0.2	0.1	0.1	0.1	0.2	0.2
HEMATITE	—	—	0.3	0.2	0.1	0.1	0.1	0.1	0.2	0.1
HORNBLENDE	—	—	0.1	0.2	0.1	0.1	0.1	0.1	0.1	0.1
OTHER[3]	—	—	0.4	1.0	0.1	0.5	0.1	0.1	0.2	0.6

[1] Radioactive minerals percentage based on petrographic microscope, XRD, SEM, EDX, and radiochemical analysis of density separated fractions.

[2] Other radioactive minerals include tyuyamunite, autunite, thorite, and gummite.

[3] Other heavy minerals, in order of decreasing abundance: ilmenite/leucoxene, garnet, rutile, tourmaline, epidote, [redacted], mullite, sillimanite, staurolite, and kyanite.

TABLE 3. CHEMICAL COMPOSITION OF MONTCLAIR AND GLEN RIDGE SOIL SAMPLES

CHEMICAL COMPOSITION		WHITE SANDS (TAILINGS)				GLEN RIDGE					MONTCLAIR		
		MINUS NO. 4 TOTAL SAMPLE	PLUS 50 (-10) SIEVE	PLUS 100 (-50) SIEVE	MINUS 200 TOTAL SAMPLE	MINUS NO. 4 TOTAL SAMPLE	PLUS 50 (-10) SIEVE	PLUS 100 (-50) SIEVE	MINUS 200 TOTAL SAMPLE	MINUS NO. 4 TOTAL SAMPLE	PLUS 50 (-10) SIEVE	PLUS 100 (-50) SIEVE	MINUS 200 TOTAL SAMPLE
SULFUR	% —	0.09	0.07	0.04	0.10	0.11	0.013	0.04	0.08	0.06	0.06	0.02	0.04
CARBONATE C	% —	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
TOTAL CARBON	% —	0.95	0.40	0.04	0.67	7.72	10.25	1.87	1.76	4.70	5.66	0.77	1.36
ORGANIC C	% —	0.95	0.40	0.04	0.67	7.72	10.25	1.87	1.76	4.70	5.66	0.77	1.36
ALUMINUM	% —	1.36	1.67	0.52	1.39	6.72	7.05	4.47	7.82	5.44	4.55	2.94	6.77
BARIUM	% —	0.30	0.30	0.12	0.38	0.14	0.16	0.08	0.24	0.09	0.07	0.05	0.11
CALCIUM	% —	0.07	0.06	<0.01	0.05	0.32	0.04	0.15	0.29	0.36	0.34	0.10	0.31
IRON	% —	0.45	0.65	0.07	0.30	3.84	4.22	2.06	2.11	3.49	3.78	1.69	2.58
FE2 IRON	PPM	8.4-	4.8	1.2-	3.4-	23.0-	29.0-	13.0	23.0-	22.0-	24.0-	12.0	21.0-
FE3 IRON	% —	0.45	0.65	0.07	0.30	3.84	4.22	2.06	2.11	3.49	3.78	1.69	2.58
LEAD	% —	0.016-	0.023	0.012	0.007	0.028	0.056	0.056	0.048	0.041	0.066	0.031	0.049
MAGNESIUM	% —	0.08-	0.14	0.03	0.06	0.30	0.36	0.24	0.36	0.45	0.28	0.18	0.34
MANGANESE	% —	0.006-	0.006	0.002	0.008	0.034	0.036	0.024	0.023	0.046	0.051	0.022	0.046
POTASSIUM	% —	0.22-	0.30	0.04	0.35	0.96	1.13	0.66	1.26	1.00	0.87	0.63	1.42
SILICON	% —	42.4	43.7	46.0	42.1	31.4-	30.1-	39.3	32.1	35.1	36.3	39.7	34.9-
SODIUM	% —	0.29-	0.16	0.02	0.23	0.51	0.38	0.31	0.47	0.63	0.39	0.41	0.79
TITANIUM	% —	0.14-	0.20	0.13	0.32	0.61	0.66	0.39	0.86	0.52	0.39	0.32	0.82
VANADIUM	% —	0.069-	0.083	0.025	0.070	0.082	0.101	0.061	0.105	0.021	0.021	0.011	0.02
T-PHOS	% —	0.04	0.02	0.01	0.04	0.05	0.10	0.04	0.10	0.04	0.03	0.02	0.05
SULFIDE-S	% —	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01

Analysis by Huffman Laboratories (Hu88).

depicted graphically in Figures 3 and 4. Furnace fired coaly material (coal, coaly slag, white slag, and ferruginous slag) comprises most of the Glen Ridge (83 percent) and a considerable portion of the Montclair (45 percent) gravel-sized materials. Rock particles comprise more than half of the Montclair gravel (52 percent) and a substantial portion of the Glen Ridge (12 percent) material. Glass and other varieties of discarded household waste (leather, ceramic material, etc.) comprise a few percent of the gravel-sized material at both sites.

The radium 226 activity of the Glen Ridge gravel materials averages 346 pCi/g and represents approximately 16 percent of the radioactivity of the sample tested. The radium-226 activity at the Montclair site averages 44 pCi/g, approximating 8 percent of the radioactivity in the sample (Table 1). Measurements of radium 226 activity on random gravel-size particles by EERF found the highest readings in the slag particles with ferruginous slag containing the highest radium 226 activity (Table 4 and Figure 3).

5.1.1 Furnace Fired Material

The furnace fired material, which comprises 83 percent of the Glen Ridge and 43 percent of the Montclair gravel-sized material, contains the highest radium 226 activity levels. The furnace fired constituents consist of (a) unburned anthracite coal, (b) interlayered coaly slag, (c) white slag and (d) brownish-red ferruginous slag. (Figure 4).

5.1.1.1 Antracite Coal. Unburned, black subangular, anthracite coal occurs in generally similar amounts at both the Montclair and Glen Ridge sites. Anthracite coal from the Appalacian coal fields of Pennsylvania deposits was in abundant supply in the 1920's; however, this variety of coal has since been depleted. The coal ranges from 5 to 68 pCi/g radium 226 (Figure 3) for several particles tested at EERF (Ri 89). Approximately 20 percent of the radium 226 is in secular equilibrium in the coal particles assuming no process has separated the parent uranium from the radium.

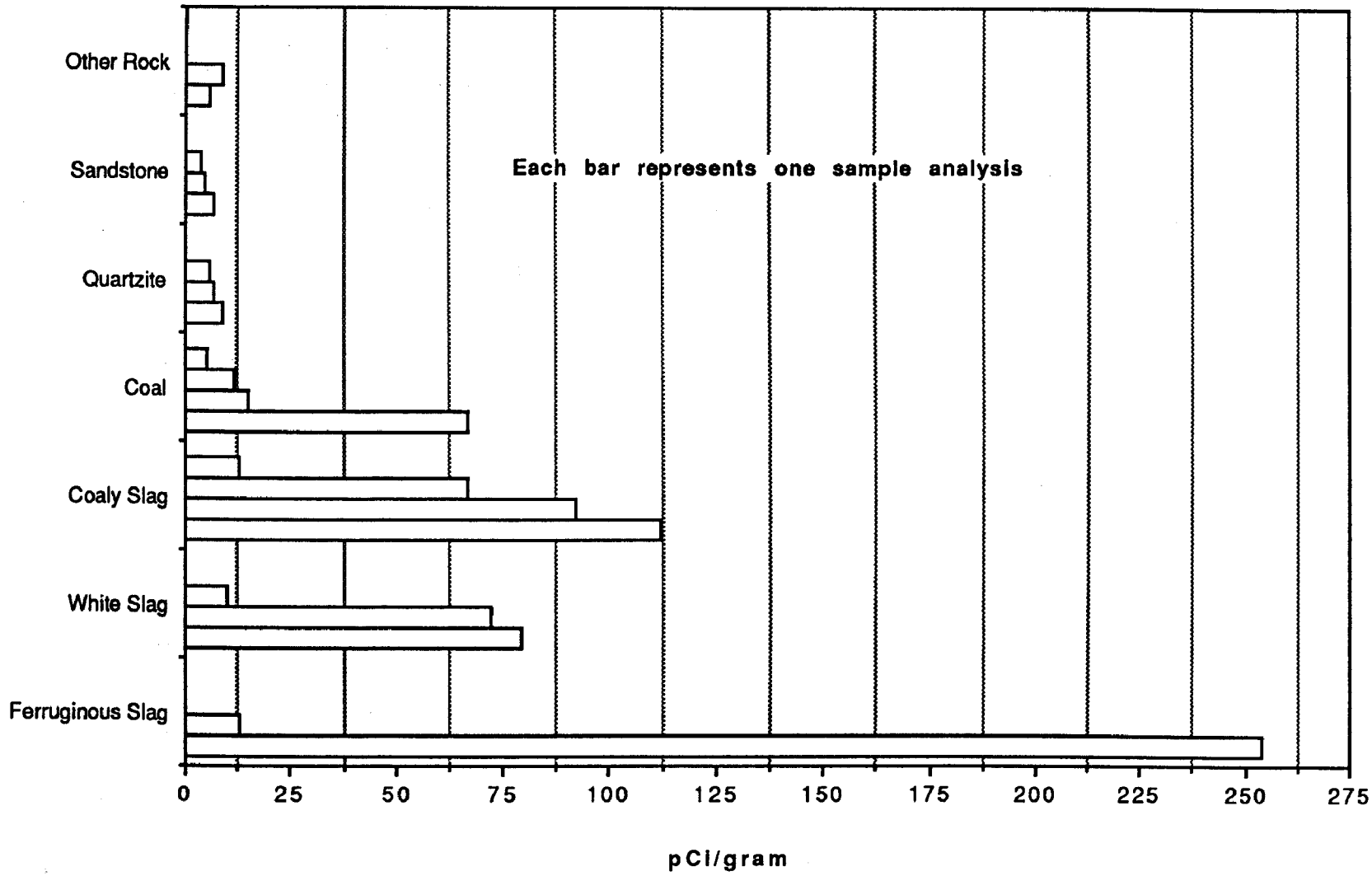

Figure 3. Radium 226 Activity on Washed Gravel-Size Particles

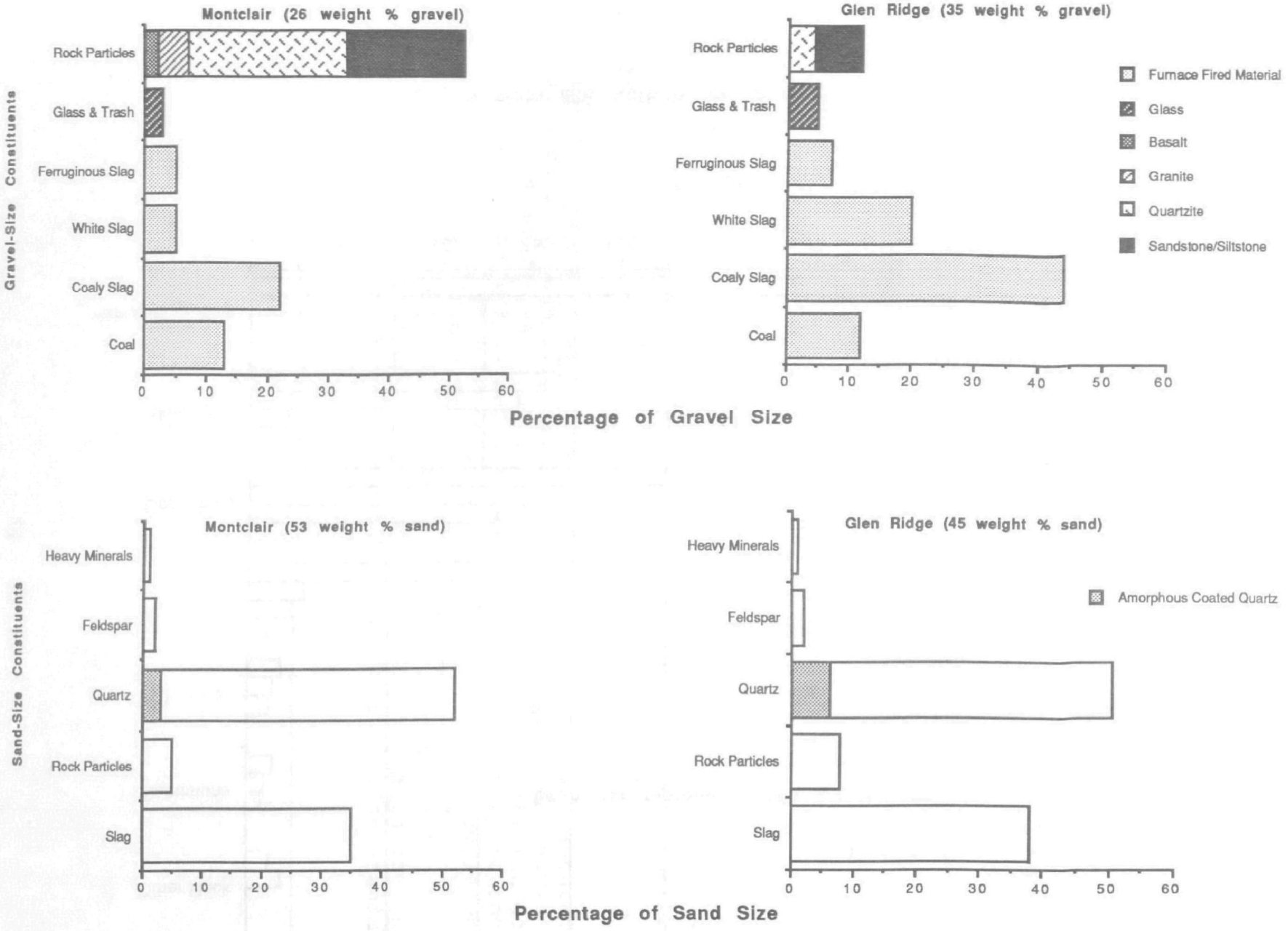

Figure 4. Composition of Gravel and Sand Size Materials in Montclair and Glen Ridge Contaminated Soils

The National Coal Resource Data System of the United States Geological Survey lists the relative amount of U and Th in coal in the United States (Fi 88) as follows: (1 pCi/g U = approximately 3 ppm U)

	Geometric Mean (PPM)	Maximum Value (PPM)	Number of Tests
		ALL U.S. COAL	
U	2.59	2674	6532
Th	3.12	92	7001
		APPALACHIAN COAL	
U	1.7	63.4	4213
Th	2.9	47.8	4361
		ANTHRACITE COAL	
U	1.4	25.2	47
Th	5.2	14.4	40

The uranium in the coal particle tested at EERF from the Montclair and Glen Ridge sites exceeds the uranium in the average coal in the three categories cited above. Extreme uranium values occur in some of the lignite coal deposits of the Dakotas and Texas. The lignite coal of the Dakotas, mined for its uranium content, was ashed to concentrate the uranium ore prior to milling in order to reduce the hauling cost (Fi 88). Apparently, burning the coal does not affect the uranium which in the ash is concentrated as much as 10 times or more. Thus, a considerable portion of radioactivity in the gravel is from the uranium originally present in the coal from the Appalachian coal fields. The radium activity of uranium mineral, however, is probably not more than 4.5 percent of the total radium activity in the samples.

5.1.1.2 Coaly Slag. The coaly slag comprises approximately 44 percent of the Glen Ridge gravel and 22 percent of the Montclair gravel-sized materials. The coaly slag is mottled in appearance and consists of partially burned coal, in various stages of decomposition to slag. The radium 226 activity on several

particles ranges from 12 to 110 pCi/g (Figure 3). Approximately 15 percent of the radium 226 is in secular equilibrium with uranium.

5.1.1.3 White Slag. White flat-shaped siliceous slag averages 20 percent of the Glen Ridge and 5 percent of the Montclair gravel-size materials. X-ray diffraction analysis of the white slag reveals a composition of mullite, quartz, cristobalite, and an amorphous silica-rich glass. The mullite ($Al_6 Si_2 O_{13}$) is a refractory material resulting from heating of high aluminous silicate minerals such as clay minerals. The amorphous glassy material appears as a broad welt on the x-ray diffractogram between 20 and 30 degrees 2 theta. The high siliceous composition and fine grain size (large surface area) are factors that would favor high adsorption of radium 226. The initial radium 226 activity of 71.9 pCi/g in the white slags and relatively low uranium (0.98 pCi/g) tend to support the view that more than 90 percent of the radium 226 on this highly siliceous material may be a result of adsorption or perhaps radium paint residue from burned rags containing radium paint.

Nirdosh (1984) in studies of radium in Elliot Lake uranium ore concludes that surface adsorption may play a major role in retention of radium by solids when the radium content of the solids is low (Ni 84). The Langmuir theory of surface adsorption is also favored over leaching in very dilute solutions and the adsorbed trace impurities are difficult to wash off completely.

5.1.1.4 Ferruginous Slag. Reddish-brown ferruginous slag comprises between 5 and 7 percent of the gravel and consists of hematite and other metals that probably accumulate as a heavy residue in a coal- fired furnace. Pyrite (FeS_2) is a common occurrence in coal and the oxidation of pyrite to hematite (Fe_2O_3) or magnetite ($Fe_3 O_4$) in the furnace probably comprises the bulk of the accumulation of metals formed on the slag at the bottom of the furnace. The relatively high magnetic content of this material makes all of the particles weak to strongly magnetic and much of this material can be removed with a magnet.

The highest radium 226 activity occurs in the ferruginous slag (Table 4). Secular equilibrium between radium and uranium, however, is limited to about 15 percent of the radium activity.

The remaining 85 percent of the radium 226 activity is believed to be a result of radium painted residues from incinerated clean-up rags or materials from the paint and adsorption of the radium from solution.

A special test on representative soil samples from the Montclair and Glen Ridge sites was conducted to determine the feasibility of magnetic separation of gravel-sized materials including also sand-size materials larger than the plus 50 sieve size. The tests conducted at EERF found more than twice the radium 226 associated with the magnetic fraction per unit volume. It seems feasible to expect removal of approximately 10 percent of the radium activity by magnetic means on these furnace fired ferruginous slag particles.

5.1.2 Rock Particles

Rock particles comprise approximately 52 percent of the Montclair and 12 percent of the Glen Ridge gravel-size material (Figure 4). The smooth, rounded, dense, tan, quartzite particles constitute about 50 percent and 30 percent respectively of the Montclair and Glen Ridge rock particles. The quartzite is transported glacial drift from the Pleistocene materials that blanket the bedrock. The red-brown, smooth-to-rough surfaced, subangular, dense sandstone and siltstone particles present in generally similar proportions as the quartzite were probably derived from the underlying bedrock (Brunswick Formation). Lesser amounts of granite, gneiss, basalt and minor other rock varieties comprise the remainder of the rock particles.

The radium 226 activity associated with the rock particles ranges from 3.7 to 9.0 pCi/g, and approximately one-third of the radium is in secular equilibrium. The other two-thirds of the radium 226 is attributed to adsorption of the radium from solution.

5.1.3 Glass and Trash

Glass and other debris typical of materials discarded into dumpsites comprise about 5 percent of the Glen Ridge and 3 percent of the Montclair gravel-size materials. Radium 226 activity values on these materials may have local high anomalies in organic-rich materials. Virtually all of the radium 226 is a result of adsorption of the radium from solution.

5.2 SAND-SIZE MATERIALS

The sand-size materials are those particles retained between the number 4 sieve (4.75mm) and number 200 sieve (0.05mm) (ASTM D643-78). Seven fractions of the sand-size soil were used to characterize the sand material; the sieve sizes used included the number 10, 16, 50, 60, 100, 140, and 200. The sand-size materials of the soil samples comprise 53 percent of the Montclair and 45 percent of the Glen Ridge materials. White Sands tailings from a near-surface strata at the Montclair site constitute a high radium 226 activity anomaly for comparison with similar materials dispersed in the radium contaminated soils.

The sand-size materials listed in Table 2 average 35-38 percent slag particles, 5-8 percent rock particles, and the major portion consists of homogeneous minerals (quartz, feldspar, and minor radioactive and other heavy minerals). The slag and rock particles occur only in the plus 100 sieve size or larger materials with virtually homogeneous minerals below that size. The major composition change occurs at about the 50 sieve size.

5.2.1 Quartz and Quartz Films

Quartz is the dominant material (80-95 percent) in the fine sand-size fractions (100, 140, and 200 sieve sizes). Some of the quartz grains contain an amorphous coating which is more abundant on the quartz grains on the white sands (8 percent) as compared to Glen Ridge (5 percent) and Montclair (3 percent) samples (Figure 4). These coatings are believed to be (a) predominantly white amorphous silica released as a precipitate from the acid-leach process, and (b) minor amounts of soft, yellow, radioactive carnotite adhering to quartz from unprocessed sandstone uranium ore. Both types of films on quartz were observed in reflected and transmitted light with the petrographic microscope. The radium 226 activity on the light (less than 2.9 specific gravity) slag, feldspar, and quartz (including quartz coatings) accounts for 36 to 69 percent of the minus 50 and plus 270 sieve size material respectively at the Montclair and Glen Ridge sites (Table 5).

The adsorptive properties of quartz for most radionuclides are negligible as indicated by measured sorption distribution coefficients (Kd). Measured Kd values for radium on quartz reported by Fordham, however, are 1900 mL/g, suggesting that

radium has a strong attraction for solids (Fo 73). Nirdosh et al (1987) in an extensive investigation of radium adsorption on quartz found that radium, unlike other alkaline-earth cations, adsorbs on quartz at pH 1 and the order of magnitude of adsorption at pH 1 is comparable to that at pH 10 (Ni 87). The nature of the radium adsorption is believed to be "specific" adsorption and the radium ions are strongly bonded to the solid quartz surface by chemical or Van der Waal forces and are essentially nonhydrated. Other alkaline earth elements (such as Ca) are bonded by weak electrostatic forces and are hydrated; as such, they can be easily desorbed when the electrostatic attraction between the adsorbant and adsorbate is weakened. Radium by contrast is strongly adsorbed but not easily desorbed (Ni 87).

5.2.2 Feldspar

Feldspar comprises from 3 to 5 percent of the fine sand fraction. Both K-feldspar and Na, Ca, feldspar are present in generally similar proportions in the Montclair and Glen Ridge samples but are negligible in the White Sands sample. The radium 226 distribution coefficient measured on albite (Na-rich plagioclase feldspar) has been reported as 20,000 ml/g (Pi 86). This high adsorption is probably due to cation exchange of Ra for Na and/or Ca in this otherwise tightly bonded alumnosilicate mineral structure.

5.2.3 Heavy Minerals

The heavy minerals (greater than 2.9 specific gravity) comprise from less than 1 to 3 percent of the fine sand and upper-silt size fractions. The heavy minerals of the White Sands separated by bromoform contain barite (radiobarite) as the principal constituent (Figure 5). The heavy minerals of the White Sand fractions average 0.5 percent of the sand with minerals composition of the heavies averaging 55 percent barite, 17 percent zircon, 16 percent black opaques (hematite, magnetite, ilmenite and uraninite), 3 percent rutile, 2 percent carnotite, and 7 percent minor others (tourmaline, monazite, garnet, hornblende, pyroxene, etc.). The major portion of the heavy minerals consists of residuals from the radium extraction acid leach process (radiobarite) and uranium ore and associated gaugue minerals. However, some have infiltrated into the White Sands from host media (hornblende magnetite, hematite, etc).

TABLE 4. RADIOCHEMICAL ANALYSIS OF MONTCLAIR/GLEN RIDGE GRAVEL PARTICLES. ANALYSIS BY EERF.

GRAVEL-SIZE PARTICLES	LOCATION	U-238 pCi/g	Ra-226 pCi/g	Th-230 pCi/g	Th-232 pCi/g	Fe ppm
FURNACE FIRED COALY SLAG	R	2.77	12.91	4.40	2.51	—
	M	4.13	10.41	19.18	2.24	—
	G	3.49	42.06	2.84	0.63	—
	G	7.39	78.87	31.89	4.04	—
FERRUGINOUS SLAG	M	6.07	13.10	2.75	1.61	55,300
	M	3.92	68.57	3.38	3.02	—
	R	2.71	6.19	3.32	2.12	—
	G	40.63	253.90	63.59	0.47	—
	G	16.83	126.80	50.14	3.03	—
WHITE SLAG	G	0.98	71.90	14.95	3.38	930
	G	10.20	91.57	13.72	2.07	—
COAL	M	1.66	5.51	0.70	0.54	2,000
	R	4.43	20.22	1.70	0.94	—
	M	6.49	18.95	1.76	0.38	—
	G	8.55	66.73	7.30	0.63	—
ROCK PARTICLES SANDSTONE	G	1.16	3.70	1.11	1.10	17,900
	R	0.86	6.74	0.76	0.86	—
	R	0.54	8.99	2.41	0.41	—
	M	1.83	5.42	0.82	0.92	—
QUARTZITE	M	1.20	6.80	1.28	1.01	18,900
	M	0.64	5.73	0.51	0.47	—
OTHER ROCK	M	3.58	9.18	3.80	0.69	—
TRASH WOOD (PLASTIC)	R	3.00	19.22	6.09	0.99	—
LEATHER	M	17.88	72.50	21.28	0.53	—
GLASS (POTTERY)	G	2.11	15.94	7.63	0.54	—

Note: 1. M = Montclair soil; R = Representative soil;
G = Glen Ridge soil.
2. Fe analysis by Galbraith Laboratories

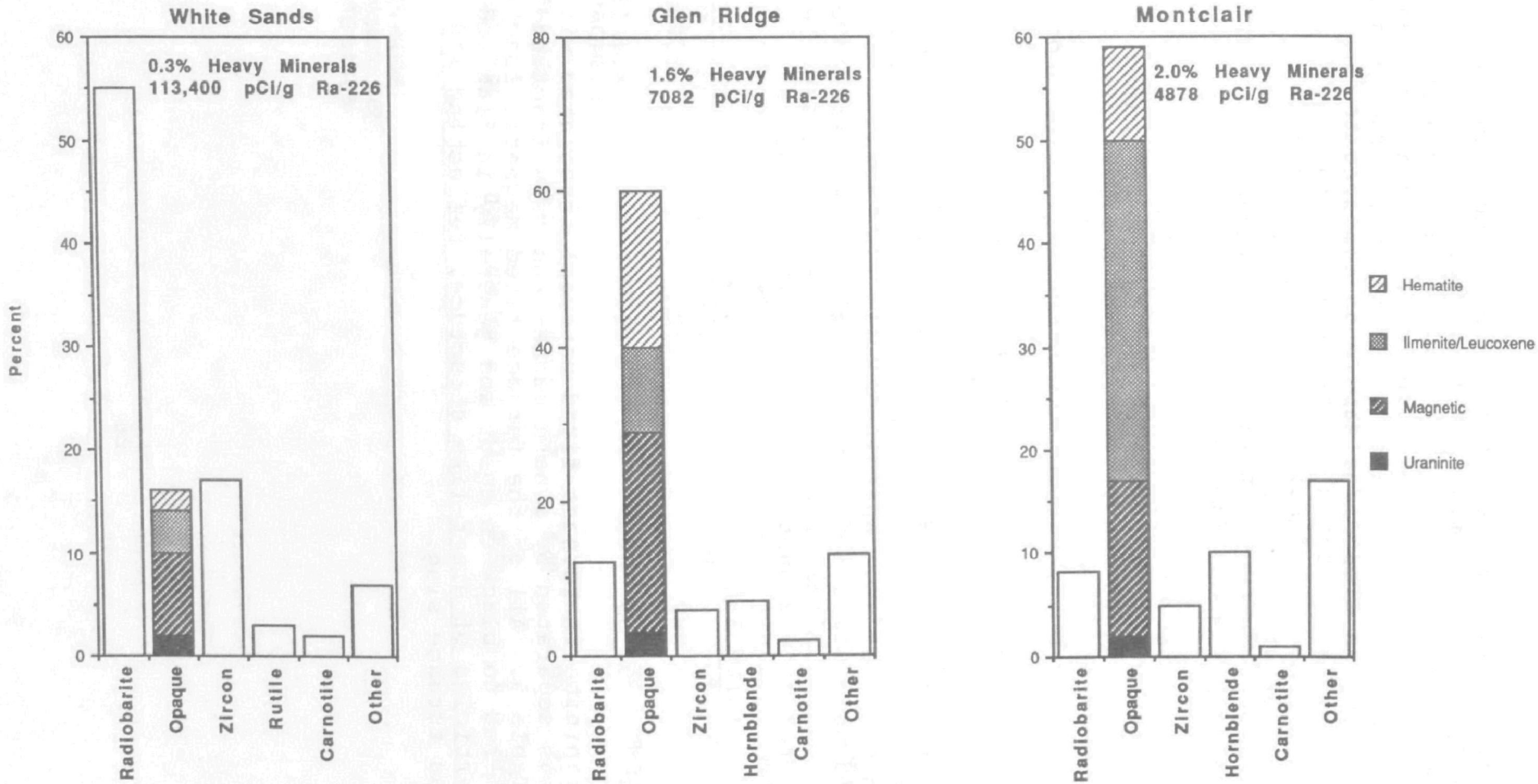

Figure 5. Comparison of Heavy Mineral Suite in -50/+270 Sieve Size White Sands, Glen Ridge, and Montclair Material

The average heavy mineral fractions of the Glen Ridge and Montclair samples contain minor representation of the heavy minerals common to the tailings as well as major minerals common to the glacial fills and furnace-fired metallic opaque materials (uraninite, magnetite, and hematite). The greater radioactivity in the Glen Ridge sample as compared to the Montclair sample is reflected in the heavy minerals. The Glen Ridge heavy mineral fraction contains both greater concentrations of uraninite from furnace fired materials and somewhat larger proportions of radiobarite and carnotite from the unprocessed ore and acid leach process.

5.2.3.1 Radioactive Minerals. The radioactive minerals in the White Sands are restricted to carnotite and minor uraninite and tyuyamunite. The latter was identified by x-ray fluorescence probe of silt-sized material (Er 88). The carnotite is readily observed under the petrographic microscope as yellow, smooth, elongated, discrete particles and as yellowish films on quartz grains. Since vanadium is abundant in carnotite and tyuyamunite, but impoverished in other materials at the New Jersey sites, this element is used as a chemical signature to calculate the amount of the uranyl vanadate minerals in the contaminated soils. The percent of vanadium for representative material in the less than number 4 sieve size of the contaminated soil samples from the Montclair and Glen Ridge sites reported by Huffman Laboratory (Table 3) is listed below with the approximate amount of carnotite it represents.

	White Sands	Glen Ridge	Montclair
Vanadium %	0.069	0.082	0.021
Carnotite %	0. 59	0. 70	0. 18

The uraninite in furnace fired material associated with magnetite was separated by a hand magnet and this fraction is listed in Table 5. All of the furnace fired magnetic fraction is brownish-red to black in color and generally in flat and elongate particles of sheet-like structure, reflecting its formation as furnace slag.

TABLE 5. RADIOCHEMICAL ANALYSIS OF MONTCLAIR/GLEN RIDGE SAND-SIZE MATERIAL BETWEEN –50 AND +270 SIEVE SIZE. ANALYIS BY EERF.

SAND (–50/+270 SIZE)	Wt%	Ra-226 pCi/g	% Ra	U-238 pCi/g	% U	Th 230 pCi/g	Th 232 pCi/g	Fe ppm
MTCLR LIGHT (–2.9)	98.0	43.03	36	8.75	90	39.89	0.60	476
MTCLR HEAVY (+2.9) MAGNETIC	0.4	190.70	1	19.28	1	74.89	1.86	6,800
MTCLR HEAVY (+2.9) NON-MAGNETIC	1.6	4,687.00	63	54.51	9	470.02	16.08	7,700
G/R LIGHT (–2.9)	98.4	181.10	69	28.44	22	194.80	1.95	365
G/R HEAVY (+2.9) MAGNETIC	0.3	1184.00	1	190.40	4	432.25	2.84	5,200
G/R HEAVY (+2.9) NON-MAGNETIC	1.3	5898.00	30	728.49	74	862.15	23.01	7,000
W/S LIGHT (–2.9)	99.7	806.5	57	2.49	99	12.02	0.02	24
W/S HEAVY (+2.9)	0.3	204,700	43	9.62	1	47.01	0.43	398

G/R = GLEN RIDGE
MTCLR = MONTCLAIR
W/S = WHITE SANDS

A larger fraction of the uraninite occurs in the nonmagnetic heavy mineral fraction with about half of the uraninite similar in appearance to the furnace material and the rest clearly naturally occurring uraninite. The latter consists of pitch black, equidimensioned, high lustered, botryoidal or "bubbly" textured particles. The highest thorium 232 and uranium 238 also occurs in the nonmagnetic heavy mineral fraction (Table 5). While thorium 232 occurs in monazite, which is present in similar amounts in all the heavy mineral fractions from the sites, an anamolous amount occurs in the Glen Ridge heavy mineral fraction. Uraninite from high temperature vein deposits is known to contain thorium 232 in variable amounts in solid solution with uranium 238. Thus the natural high temperature uraninite occurring in the Glen Ridge nonmagnetic heavy mineral fraction appears to correlate with the anomalous thorium 232 (23.01 pCi/g) reported in Table 5.

The Belgian Congo uranium ore, reportedly used for most of the world uranium supply after 1923 (La 84), is a high temperature uraninite. However, the high temperature uraninite in the Glen Ridge sample lacks some of the characteristics of that deposit and could have originated in the Colorado Plateau deposits. Garrels and Larsen (1959) in a geochemical study of uranium ores of the Colorado Plateau, reported that the classic low temperature, oxidized, sedimentary, carnotite deposits sometime occur in proximity to veins or "pipes" that are of hydrothermal origin (Ga 59). According to Frondel (1958), oxidized deposits of uranium and vanadium have been known in the Colorado Plateau since 1898, but uraninite was not recognized as a valuable uranium ore mineral in the Colorado Plateau area until 1948-49. Thus, in the 1920's, when the carnotite was used for radium production in New Jersey, any uraninite that occurred with the carnotite was probably treated as a gangue mineral rather than an ore mineral.

Radiochemical studies of minus 4 sieve size materials at EERF and special studies on silt and clay fractions by Battelle Northwest Laboratory indicate that the uranium 238 occurring in the contaminated soils would account for only 10 percent of the progenies. Thus, approximately 10 percent of the radioactivity at the sites is a result of the uranium minerals carnotite and uraninite and very minor other uranium bearing minerals. The relative proportion of canotite may be approximated from vanadium assay and the uraninite from uranium assay of tne magnetic and nonmagnetic heavy fractions.

5.2.3.2 Radiobarite. Radiobarite (radium barium sulfate) comprises the major portion of the White Sands heavy mineral fraction which averages approximately 0.3 percent of the sand fraction. When viewed under the petrographic microscope, the barite grains are white to yellowish color, anhedral in shape, and have surfaces ranging from smooth and clear to rough, mottled, and pitted. The radiobarite grains were probably smooth and unblemished when they first formed as a coprecipitate during the acid-leach stage of the radium milling process. The radiobarite subsequently incorporated into mill tailings and removed to burial in the Montclair and Glen Ridge landfills may have encountered local anaerobic microenvironments resulting from contact with organics or other redox governing factors. Investigations of uranium mill tailings have disclosed possible reducing conditions in a large uranium pile of the Western United States that is in proximity to uranium mills. Sulfate-reducing bacteria acting on sulfate compounds in these uranium mill tailing piles may have occurred in localized anaerobic microenvironments and may have posed possible localized sulfide-forming conditions (La 86 and Sh 84). The pitted surfaces of the radiobarite in the Montclair and Glen Ridge samples may similiarly be a result of such localized anaerobic microenvironment activity which resulted in pyrite (FeS_2) being formed on radiobarite surfaces. The pyrite would have since been oxidized and removed resulting in the pitted surfaces of the radiobarite. There is probably a correlation between radium 226 activity and degree of surface alteration of the radiobarite. Since some of the radium 226 was probably released in the alteration process, the unblemished radiobarite would contain higher radium 226 activity levels.

The radiobarite is more dispersed in the average Glen Ridge and Montclair contaminated soil than in the White Sands. Since barium is negligible in the glacial till, barium may be used as a signature for the abundance of radiobarite formed as a coprecipitate in the radium extraction acid leach process. The percent of barium in the minus number 4 sieve size material (Table 3) is listed below with the amount of radiobarite it represents.

	White Sands	Glen Ridge	Montclair
Barium %	0.38	0.24	0.11
Radiobarite %	0.76	0.48	0.22

The 0.08 and 0.04 percent barium reported respectively for the average Glen Ridge and Montclair sites by the New Jersey Environmental Protection Department (NJEPD 87) would appear to correlate with the samples used in this investigation, since higher activity samples were used in this characterization study.

5.2.3.3 Natural Background Minerals. Zircon and monazite, two slightly radioactive heavy minerals that contain minor amounts of uranium, radium, and thorium, are common background minerals in most natural soils. Both of these minerals are highly resistant to weathering and hence occur in many deposits. Zircon ($ZrSiO_4$) comprises 17 percent of the White Sands heavy mineral fraction and 5 percent of the Montclair and Glen Ridge sand-size heavy mineral fractions whereas monazite ($CePO_4$) comprises generally less than 1 percent of the heavy mineral fractions. As much as 3 percent uranium and 13 percent thorium have been reported in some zircons (He 58). Monazite contains variable amounts of ThO_2 (up to 30 percent) and occasionally contains some uranium. Both monazite and zircon are the chief contributors to the uranium and thorium content of the natural background radiation.

5.2.3.4 Other Heavy Minerals. Other heavy minerals, exclusive of the radioactive minerals previously cited, that occur in both the Glen Ridge and Montclair soils include: magnetite, hematite, hornblende, illmenite/ leucoxene, garnet, rutile, tourmaline, epidote, staurolite, mullite, sillimanite, staurolite and kyanite (Table 2). Magnetite, hematite, and mullite are heavy minerals formed from furnace-fired materials. The remaining heavy minerals are rock forming minerals that most abundantly originate from the host glacial tills. A few highly resistant minerals (zircon, monazite and rutite) occur with the tailing sands.

The most significant mineral as regards retention of radium 226 is probably hematite. Some of the hematite is an oxidation product of magnetite from furnace-fired material and other waste debris while a lesser amount is from glacial till (red sandstone and siltstone, etc). This mineral is highly adsorptive of radium in the process of formation in the fresh state but less adsorptive when fully formed. According to Krishnaswami et al (1982), freshly precipitated ferric hydroxide had a measured radium Kd of 28,000 mL/g which is the most effective adsorbent known for radium (Ki 82). The highest activity for radium 226 found in the ferruginous slag (524 pCi/g) appears to corroborate this view (Ri 89).

5.3 SILT AND CLAY-SIZE MATERIAL

The silt size-material comprises the soil particles between 0.074mm and 0.002 mm size. Clay-size materials are all particles less than 0.002 mm. The boundary between the silt and clay-size is in accordance with the soil scientist classification (USDA 75). The weight percents of the silt (18-20 percent) and clay-size (1-2 percent) fractions are listed in Table 2. The estimates of mineral composition of the silt and clay-size materials (Table 2) are based on: 1) x-ray diffraction (XRD) analysis (bulk soil fractions and linear density fractions), and 2) chemical composition. Two of the x-ray diffractograms of the light (2.10-2.25 g/cm^3), medium (2.25-2.71 g/cm^3) and heavy (2.71-2.96 g/cm^3), density fractions of the 10 to 20 micron silt fraction of the Glen Ridge contaminated soil are depicted in Figure 6.

The most abundant mineral in the silt-size material is quartz, comprising approximately two thirds of the sample. Amorphous silica and clay minerals (illite, chlorite, and kaolinite) comprise respectively 10-20 percent and 5-8 percent of the silt-size material (Table 2). Chemical analysis of barium and vanadium in the bulk silt and clay-size fractions were used to compute barite and carnotite for the Montclair and Glen Ridge fines. The results were: 0.2 percent barite and 0.2 percent carnotite for Montclair; 0.5 percent barite and 0.9 percent carnotite for Glen Ridge (Table 2). Minor amounts of hematite, feldspar, cristobalite and mullite are also disclosed in the X-ray diffractograms in the density fractions of the Glen Ridge silt (Figure 6).

The clay-size materials comprise one to two percent of the samples and are comprised of amorphous silica, illite, chlorite, kaolinite, and quartz (Table 2).

Of special interest is the percentage of radium associated with the linear density layers of the 20 to 45 micron, 10 to 20 micron, and 2 to 10 micron-size fractions of the Glen Ridge soil samples (Table 6). In all three of these size fractions, the amorphous silica predominates in the light density band (2.0 to 2.35 specific gravity). The amorphous silica in the light density bands contains between 25 to 31 percent of the radium-226 activity in the silt/clay fractions. The identification of the x-ray amorphous material as amorphous silica was provided by the scanning electron microscope and x-ray fluoresence probe (Er 88).

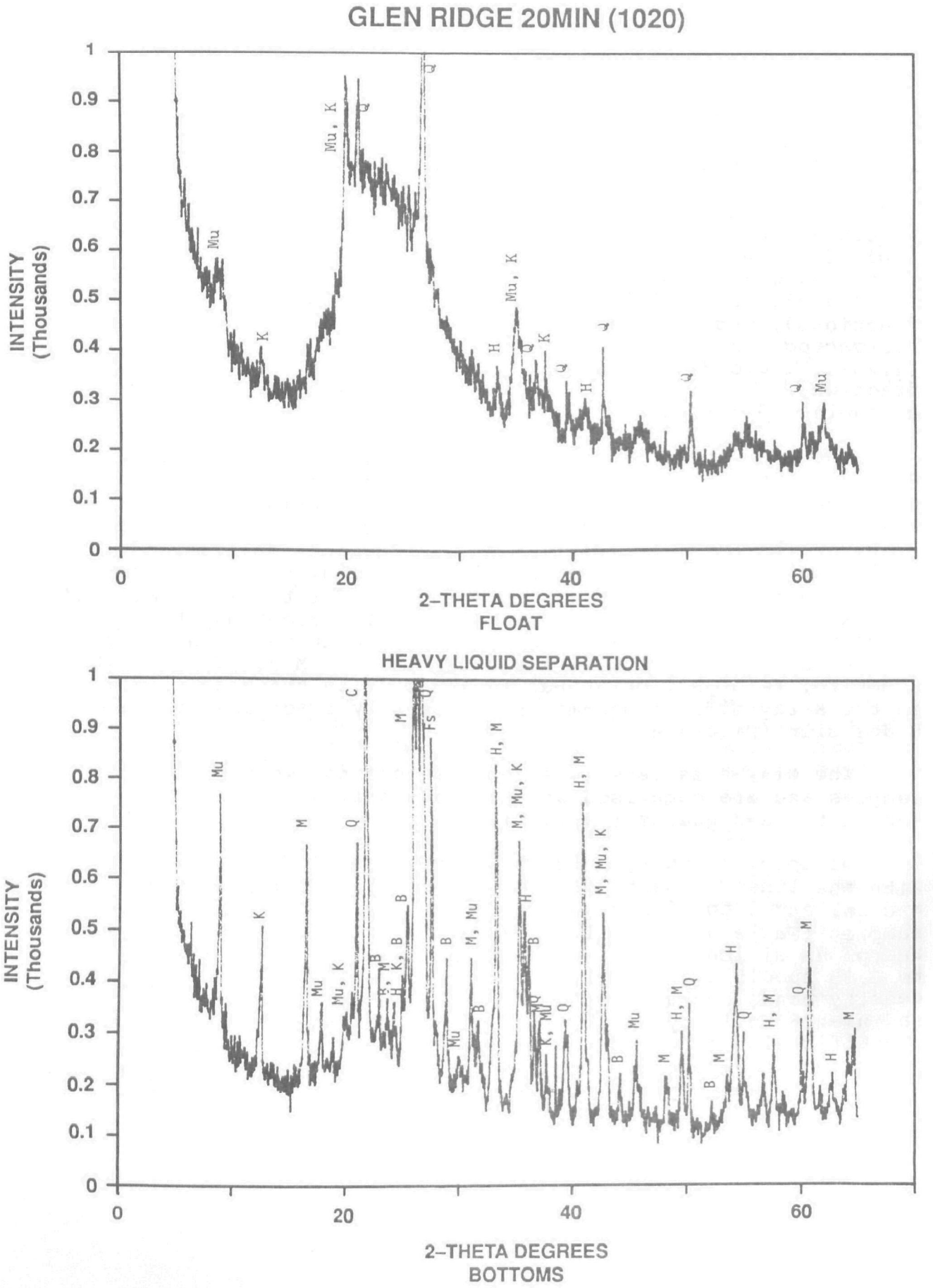

Figure 6. X-ray diffractograms of Glen Ridge 10 to 20 micron-size light and heavy density fractions

The amorphous silica concentration decreases in the middle density fraction of the Glen Ridge silt fractions where quartz, kaolinite, illite/muscovite, feldspar, and barite are identified. Mullite, hematite, and cristabolite from the furnace-fired slag material also occur in this density fraction.

Barite (radiobarite) clearly dominates the heavy density fraction (2.62 to 2.69 specific gravity) in the silt samples (Table 6). The highest concentration of hematite also occurs in this fraction. Minor quantities of quartz, illite, kaolinite, feldspar, mullite, and cristabolite are also present. Although no specific uranium mineral peak was observed by XRD in any of the density bands because concentrations were not sufficient to exceed background scatter, positive analysis was made of a uranium mineral with the x-ray fluorescence probe (SEM/EDS). The mineral was an uranyl vanadate (carnotite or tyuyamunite). The use of autoradiography techniques provides positive information on the association of specific radionuclides with specific minerals. The alpha tracks emitted by the radionuclide enable focus on specific target minerals which are identified using SEM/EDS techniques.

Based on x-ray diffraction analysis of the Glen Ridge linear density gradient bands and the measurement of radium 226 activity associated with these bands (Table 6), it is estimated that within the silt-clay size soil fractions the percentage of Ra 226 is proportioned as follows:

Radium 226 - Glen Ridge Silt/Clay Size

30%	Amorphous Silica (coprecipitate of acid leach process
47%	Radiobarite (coprecipitate of acid leach process))
23%	Hematite, illite, kaolinite, feldspar, mullite, cristabolite and quartz (adsorbate from solution).

The minerals in the above listing identified as adsorbable from solution are listed in order of high to low Kd values reported in the literature (Fo 73, On 81, Kr 82 and Pi 86). Onishi and others cite a direct correlation of cation exchange capacity with adsorption and ion exchange as probably the principal radium adsorption mechanism (On 81).

The thorium 230 activity is subordinate to the activity levels in the gravel and sand-sized soil fractions (Tables 4 and 5). However, in the silt and clay-sized fractions, PNL measured thorium at higher activity levels than radium 226 (Er 88). This

TABLE 6. MINERAL COMPOSITION AND RADIUM 226 ACTIVITY OF SILT-SIZE (–45 MICRON +2 MICRON) SOIL FROM GLEN RIDGE SITE. ANALYSIS BY PNL.

Wt%	DENSITY	% FRACTION	Ra-226 ACTIVITY	Ra-226 CONTENT	% Ra	MINERAL COMPOSITION[1,2]
			GLEN RIDGE 20–45 MICRON SIZE			
	Light 2.10 – 2.35	29.22	2,620 pCi/g	766 pCi	30.94	Major: AS;Q Minor: M/I, K, M, F
7.2	Medium 2.35 – 2.71	48.26	1,400 pCi/g	676 pCi	27.30	Major: Q, I, K, F, C, M Minor: AS, B, H
	Heavy 2.71 – 2.96	22.52	4,590 pCi/g	1,034 pCi	41.76	Major: B, H, M, Q Minor: K, M/I, F, C
			GLEN RIDGE 10–20 MICRON SIZE			
	Light 2.10 – 2.25	32.30	1,640 pCi/g	530 pCi	25.21	Major: AS, Q, M/I Minor: K, H
11.7	Medium 2.25 – 2.71	55.69	1,040 pCi/g	579 pCi	27.55	Major: Q, M/I, K, F, C Minor: M, B, H, AS
	Heavy 2.71 – 2.96	12.01	8,270 pCi/g	993 pCi	47.24	Major: B, H, M Minor: Q, M/I, K, F, C
			GLEN RIDGE 2–10 MICRON SIZE			
	Light 2.10 – 2.27	31.68	2,010 pCi/g	637 pCi	25.82	Major: AS, Q Minor: M/I, K, M, H
3.6	Medium 2.27 – 2.62	45.01	1,450 pCi/g	653 pCi	26.47	Major: AS, Q, K, M/I F, M, C Minor: H, B
	Heavy 2.62 – 2.96	23.31	5,050 pCi/g	1,770 pCi	47.71	Major: B, H, M Minor: Q, M/I, F, C

Notes: 1. Mineral composition based on x-ray diffraction (XRD) analysis linear density gradient bands and x-ray fluorescence of x-ray amorphous material.

2. Amorphous Silica = AS; Quartz = Q; Mica/Illite = M/I; Kaolinite = K; Feldspar = F, Mullite = M; Barite (Radiobarite) = B; Cristabolite = C; Hematite = H.

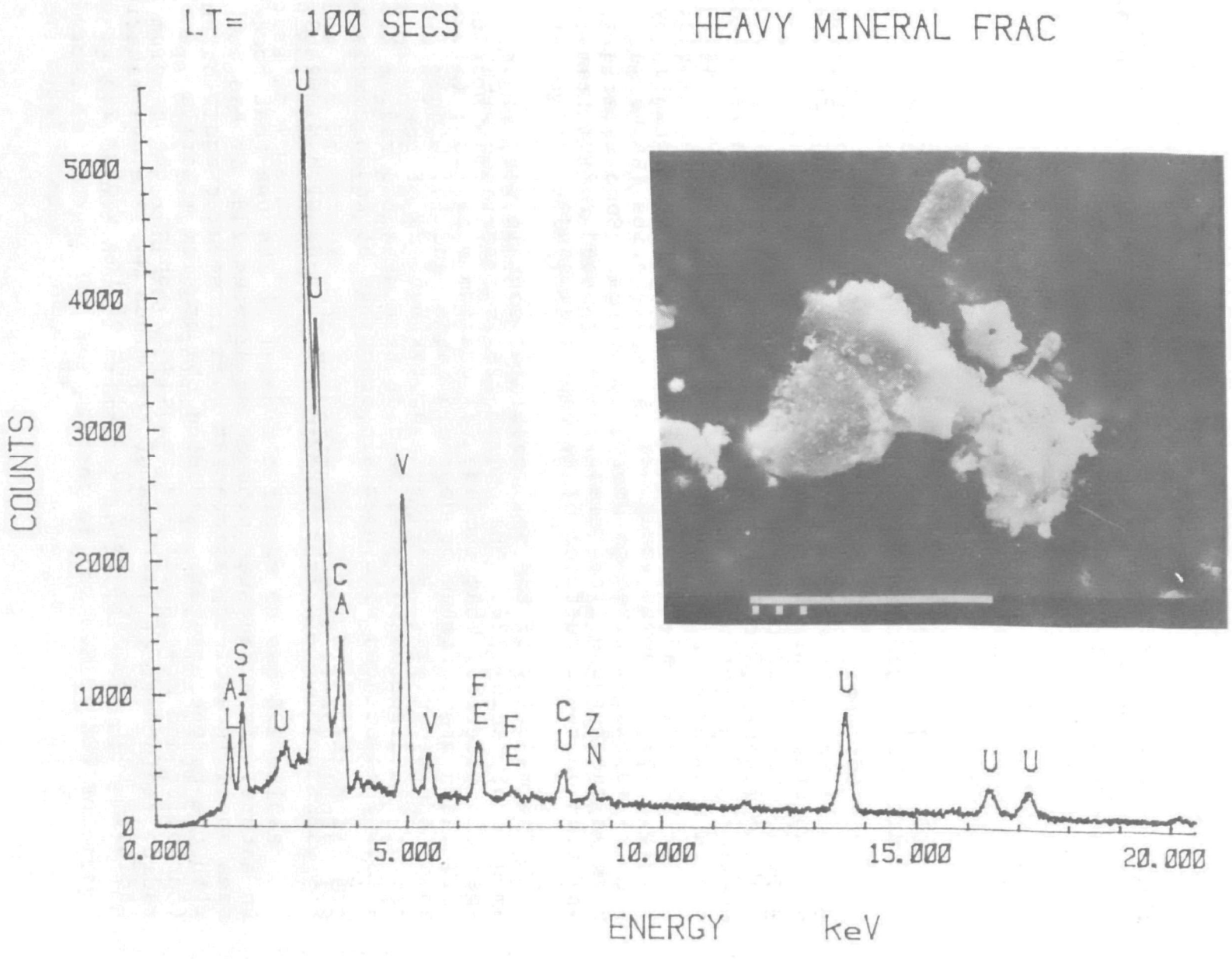

Figure 7. SEM photomicrograph and EDX spectrum for carnotite/tyuyamunite in the heavy mineral fraction of the Glen Ridge soil. The length of the bar is 100 microns.

relative increase of thorium 230 associated with the finer material could possibly be related to the greater abundance of amorphous silica that occurs in the fine fractions. In studies of migration of radium and thorium from natural uranium deposits, Airey (1982) reports that radium tends to be associated with clay/quartz, whereas uranium and thorium tend to be associated with the iron phase of the weathering profile (Ai 82). Hence, it is possible that the highest activity levels of thorium 230 to radium 226 in the light density fractions of the silt is a consequence of thorium attachment to amorphous silica during the acid leach process. The generally high percentage of radium 226 in the higher density fractions may be the result of adsorption to hematite particles.

6. ASSESSMENT OF PERCENTAGE OF RADIUM 226 DISTRIBUTION ON SOIL MATERIALS

The percentage of radium 226 associated with specific soil materials was determined by fractional separations of minerals and materials by sieving, heavy liquid separations, magnetic separations, radiochemical analyses, and chemical analyses. The uranium minerals were identified by petrographic analysis and chemical assay for uranium. The radium 226 occurs both as in growth progeny of uranium 238 in uranium minerals and as radium separated from uranium. The radium separated from uranium by the acid leach process occurs as (a) radiobarite ($Ba,Ra\ SO_4$) and (b) as radium fixed to amorphous silica. Some materials, such as the furnace fired slags, have adsorbed radium from solution on their surfaces or by cation exchange.

The majority of the soil particles contain very minor amounts of radium 226. The radium 226 is associated with specific materials that include (a) uranium minerals ; (b) acid-leach precipitates or coprecipitates, and (c) highly adsorbent materials. Reasonable approximations of the radium 226 distribution will be made from measured radium 226 activity associated with specific fractions of known composition.

6.1 RADIUM 226 IN SECULAR EQUILIBRIUM WITH URANIUM MINERALS

Radium 226 has no specific mineral of its own that occurs in nature. As a daughter product of uranium 238, it has the same specific activity as uranium when in secular equilibrium with the parent uranium. Assuming the uranium minerals (carnotite and uraninite) are relatively intact, the uranium and radium radiochemical analyses of the minus No. 4 sieve material indicate that approximately 10 percent of the radium 226 is in

secular equilibrium with the uranium 238 (Ri 88). Measurements of the silt/clay fractions also indicate a similar percent of secular equilibrium materials (Er 89). The gravel-size material, however, contains 15 percent to 33 percent of the radium in secular equilibrium with uranium. This difference is not surprising since the gravel is comprised largely of rock particles and furnace-fired slag and is free of radium solids of sand-size and smaller size formed from the acid leach process or which occur as uranium ore minerals.

The uranium minerals, for calculation purposes, used to assess the radium 226 activity distribution include (a) the furnace fired slag uranium minerals from the Appalachian coal fields and (b) the uranium ore minerals associated in the tailings sands from the radium processing plant. The former is uraninite associated with the larger soil particles and the latter is carnotite and uraninite in essentially equal amounts that occur in the finer soil materials.

A major composition change occurs at the plus 50 sieve size. Practically all the furnace-fired slag and rock particles occur above this size (Table 2). The percentage of radium 226 activity in the plus 50 sieve size for the Glen Ridge soil approximates 30 percent. Special tests conducted at EERF on plus 50 sieve materials on Montclair and Glen Ridge soils found that magnetic particles have more than twice the radium activity per unit volume. It is estimated that about 10 percent of the radium activity on the plus 50 sieve size particles could be removed by simple magnetic separation.

In the assessment of the percentage of radium 226 activity associated with uranium ore minerals, the carnotite and uraninite are found to be essentially equal. The percentage of radium 226 activity on the minus 50 sieve size soil fraction from Montclair and Glen Ridge is respectively 83 and 70 percent. Thus, if the carnotite and uraninite in this material is considered equal in distribution for the 10 percent of the radium 226 in secular equilibrium, the percent of radium 226 for each of these minerals in the total Glen Ridge and Montclair samples is 4 percent (Table 7).

6.2 RADIUM 226 IN NATURAL BACKGROUND MINERALS

The radioactive background minerals that are ubiquitous in the Colorado Plateau ore minerals and the glacial tills of New Jersey include zircon and monazite. These two minerals and several very minor others (gummite, autunite, thorite, etc.) are estimated to account for one percent of the radium 226 activity in the total sample (Table 7).

Table 7. Distribution of radium 226 on specific materials of Montclair and Glen Ridge

Materials	Percent Radium Distribution Glen Ridge	Montclair(1)	Weight Percent Glen Ridge	Montclair
Radium in Secular Equilibrium in Uranium Minerals				
Uraninite (ashed)	5	4	0.2	0.1
Uraninite (ore)	4	4	0.2	0.1
Carnotite (ore)	4	4	0.5	0.1
Zircon/Monazite	1	1	0.1	0.1
Acid-Leach Radium in Tailings Precipitates and Coprecipitates				
Radiobarite	36	46	0.2	0.1
Amorphous Silica	26	35	4.1	2.2
Adsorption of Radium on particles (2)				
Magnetic Ferruginous Slag			3.0	2.0
Other Furnace-Fired Slag			42.0	29.0
Illite/Mica			1.2	1.4
Feldspar	24	6	1.0	2.0
Hematite			0.2	0.1
Kaolinite/Chlorite			0.7	1.1
Rock Particles			9.0	16.0
Quartz			35.0	43.0
Other Materials			2.6	1.7

1. Estimated values of Montclair samples for silt/clay fractions (lack linear density gradient measurements).

2. Materials are listed in order of highest degree of adsorption to lowest degree of adsorption from top to bottom.

3. Magnetic ferruginous slag and other furnace slag contains the major occurrence of radium 226 in the gravel and upper sand-size fractions as uraninite ash and as high adsorption on iron and other slag surface materials.

6.3 RADIUM 226 ASSOCIATED WITH RADIOBARITE

Radiobarite in the contaminated soil is a product of the acid leaching process which removed uranium and thorium from the ore mineral to isolate radium for industry. It was precipitated with barium from solution as barium (radium) sulfate and the uranium and thorium remained in solution for removal by

decantation. The radiobarite is restricted to the sand and silt-size materials in the contaminated soil. The barite is readily concentrated by heavy liquids and is associated with the nonmagnetic heavy mineral fraction (Table 5).

The barite in the nonmagnetic heavy liquid fraction is associated with uranium minerals and other heavy minerals, generally low in radium 226 activity. The radium not in secular equilibrium with uranium is assessed as radium affixed to radiobarite for calculation purposes. Since hematite and some background radioactive minerals (zircon and monazite) may account for a small fraction of the radium 226 activity, the radium 226 activity assigned to radiobarite is a maximum figure.

In the Glen Ridge sample, 30 percent of the radium 226 activity in the sand-size material is affiliated with the non-magnetic heavy mineral fraction. Since 12 percent is required for secular equilibrium, 88 percent is used in the calculation that finds 26.4 percent radium 226 assigned to the radiobarite in the sand-size fraction. Since the sand-size fraction is 30 percent of total radium activity in the sample, the radiobarite from the sand-size fraction is 8.7 percent of the total radium 226 in the sample.

The radium 226 activity in the radiobarite of the Glen Ridge silt/clay size linear density bands is 47 percent Er 89). Since the radium 226 activity of the silt/clay size fraction comprises 59 percent of the total sample, the maximum radium 226 associated with the radiobarite is 59 x 47 or 28 percent. The sum of the percentage of radium 226 activity for radiobarite in both the sand and silt/clay size fraction of the Glen Ridge sample is 36 percent (80 + 28 percent) (Table 7).

The Montclair sample lacks the linear density gradient measurements; however, if it is assumed to be generally similar in proportion to the Glen Ridge sample, the radiobarite possibly contains 46 percent of the radium 226 activity in the Montclair sample.

Thus, while radiobarite comprises but 0.2 weight percent of the Glen Ridge sample (Table 2), it comprises more than a third of the radium 226 activity. The radiobarite in the Montclair site comprises 0.1 weight percent of the sample but could possibly contain nearly half of the radium 226 activity.

6.4 RADIUM 226 ASSOCIATED WITH AMORPHOUS SILICA

Amorphous silica was observed on the quartz grains of the sand fractions with more representation in the White Sands. The Glen Ridge sand-size light fractions, free of the heavy minerals containing radiobarite and uranium minerals, contains 69 percent of the radium 226 activity (Table 5). The only other material in competition for this relatively high radium activity is feldspar and quartz with moderate adsorption capabilities and some carnotite adhering to some of the quartz grains. The latter may be determined by assigning the amount necessary for secular equilibrium; i.e., 28.44 pCi/g Ra 226 to match the 28.44 pCi/g U 238. Thus, 16 percent of the radium 226 is attributed to carnotite, leaving 84 percent for amorphous silica and adsorbed radium. The percent of radium activity in the sand approximates 9 percent for the total sample.

In the silt/clay fraction, the linear density band of light material contains 30 percent of the radium activity (Table 5). Since the silt/clay Glen Ridge fractions comprise 57 percent of the radium 226 activity, the amount of radium 226 on the amorphous silica is calculated as 17.1 percent. Thus, the sum of Ra 226 percentages for the sand and silt/clay size of the Glen Ridge sample represented by the amorphous silica is 26 percent (Table 7).

Similar calculations for the Montclair sample, assuming parallel conditions for the silt/clay fractions at Glen Ridge, indicate that 35 percent of the radium 226 may be a result of the amorphous silica and radium adsorbed onto quartz and feldspar. Since the activity level of Ra 226 is 4 to 5 times greater on the Glen Ridge sample, the Montclair sample may have a higher percent of the radium as adsorption onto particle surfaces than occurs on the Glen Ridge sample. Thus, the amorphous silica values are maximum values of radium 226 association since they include adsorbed radium.

6.5 ADSORBED RADIUM 226 ASSOCIATED WITH SOIL

There are no site specific measurements of the radium equilibrium distribution coefficient (Kd). The Kd is commonly used to determine the degree of adsorption of an ion by an

adsorbent from a solution. The few measurements in the literature range from 1700 mL/g for quartz to 28,000 mL/g for ferric hydroxide; some intermediate values include 20,000 mL/g for muscovite, 20,000 mL/g for albite, 6,500 mL/g for montmorillonite, and 1900 mL/g for kaolinite (NJEPD 87). Since illite, the most abundant clay mineral at the Montclair and Glen Ridge sites, has a mineral structure similar to muscovite, the adsorption in the silt/clay fraction could be significant. On the other hand, considerable adsorption is also apparent from measured values of radium 226 on the furnace-fired slag materials (Table 4). This assumption is depicted graphically in Figure 8.

An approximation of the percentage of radium activity resulting from adsorption is calculated by difference from the sum of the Ra 226 percentages assessed for the uranium minerals and acid leach products. The result is 24 percent for the Glen Ridge sample and 6 percent for Montclair (Table 7).

The assessment of the percent radium distribution for the Glen Ridge sample is depicted in Figure 8.

7. DISCUSSION

The methodology used in this investigation has provided an assessment of the percentage of radium 226 activity associated with specific materials of known size and distribution. The magnetic ferruginous slag and other furnace-fired slag material comprises the major distribution of radium activity in the gravel and larger sand-sized materials. The major radium activity in the Glen Ridge sample occurs in the fine sand and silt/clay size fractions as acid leach-derived radiobarite and amorphous silica. Some of the radium 226 activity of the Glen Ridge sample is associated with uranium ore minerals and uraninite coal ash. The remainder is attributed to furnace fired radium paint residue and adsorption of radium on surfaces of materials. This methodology has application to other radium-contaminated soils which exist at several Superfund sites.

Magnetic ferruginous slag contains furnace-fired uraninite, radium paint residue, and adsorbed Ra 226. It is expected that as much as 25 percent (the +50 sieve size) of the radium 226 activity might be removed from material by a simple magnet applied to the washed material.

In any chemical treatment to remove radium, it is well to consider the following:

o Amorphous silica probably contains both Ra 226 and Th 230 in a weak bond. This material contains approximately 26 percent of the radium 226 activity on the Glen Ridge sample in the sand-size (as coatings on quartz) and in the silt-size as discrete particles.

o Carnotite occurs as coatings on quartz sand and as discrete particles in the sand and silt size. This soft, yellow mineral is readily soluble in weak HCl and comprises about 4 percent of the radium activity in both samples.

o Uraninite occurs as coal-fired slag and could be removed with the magnetic ferruginous slag with a magnet. As much as 4.5 percent of the radium activity occurs in furnace-fired slag with most associated with the magnetic slag. A black, equidimensional shaped, hard, dense, uraninite occurs in the fine sand and upper silt size of both the Montclair and Glen Ridge samples as discrete particles. With a density of approximately 10, these heavy projectile-like particles will with agitation sink through layers of soil to bottom positions in separation pans for removal. This same mineral is, however, insoluble in weak HCl.

o Radiobarite occurs as dense particles of sand and silt size and is relatively insoluble. It comprises more than a third of the radium activity of the Glen Ridge soil.

o Radium 226 adsorbed on solid surfaces is probably difficult to remove without strong agitation or solvent extraction. This is based on relatively high radium equilibrium distribution coefficient (Kd) values reported in the literature.

o Extraction tests conducted on contaminated soils of average 75 pCi/g from the Montclair and Glen Ridge sites (using standard RCRA method) found radium 226 releases of 10 pCi/L (NJEPD 87). Ground water beneath these sites contains 2.3 pCi/L Ra 226. This suggests that relatively weak acid solutions might be effective in some of the radium removal.

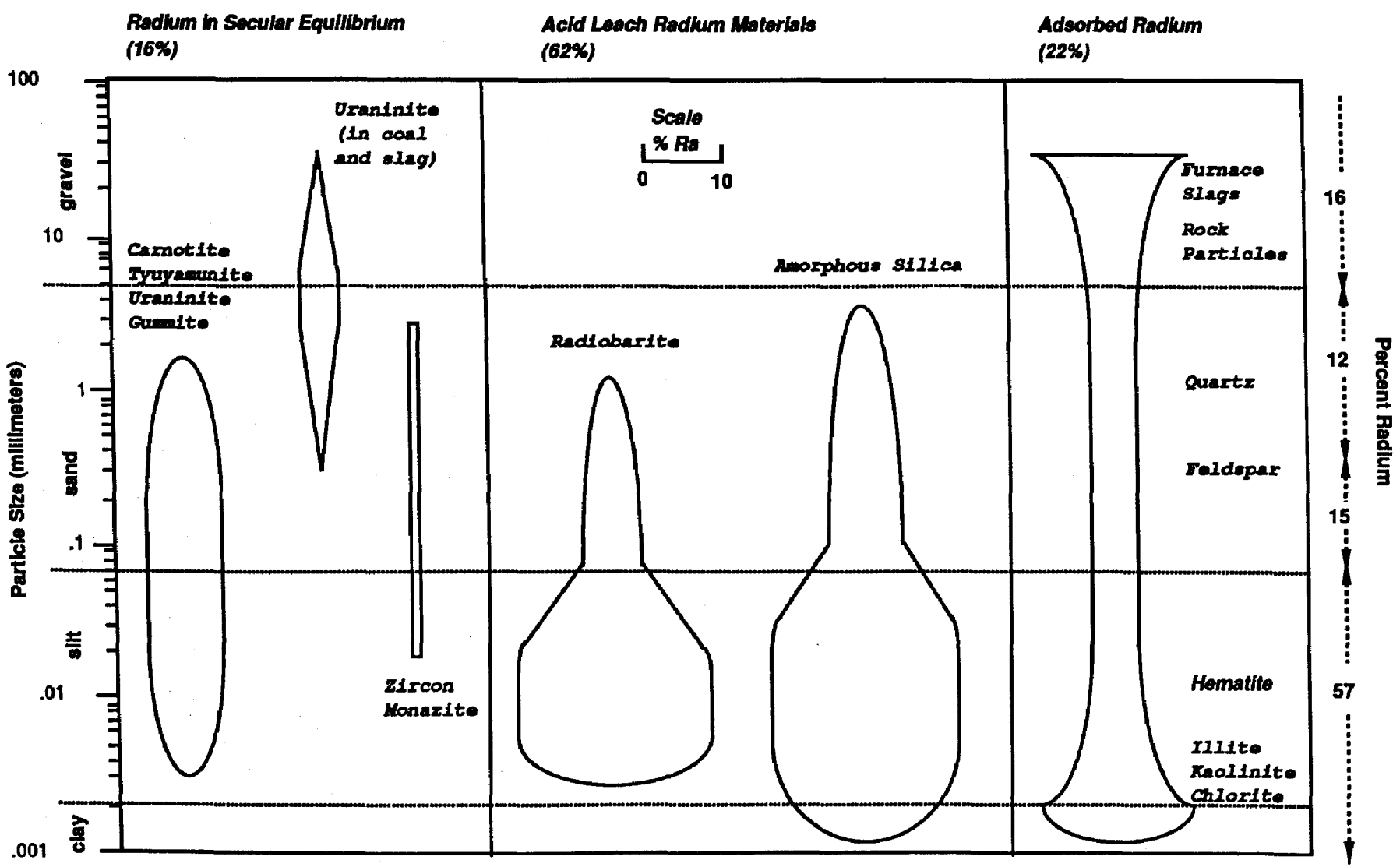

Figure 8. Relationship of Particle Size and Mineral Composition to Percent Radium Distribution in Glen Ridge Soil

8. SUMMARY

The radium contaminated soils at the Montclair and Glen Ridge, New Jersey, Superfund sites were characterized as a first step in a remedial investigation to determine the physical sizing, mineral composition, and radium distribution in the minerals and materials comprising the soil. A grain size distribution curve was constructed on each soil and 18 size fractions of each soil were obtained for complete mineral and radiochemical analysis. In addition, soil fractions were further segregated by magnetic separation with a hand-held magnet and by heavy mineral separations using sink float methods for sand-size materials and linear density centrifugation methods for the silt and clay size fractions. Chemical analysis was also performed on aggregate and fractionated samples to provide chemical signatures on diagnostic minerals and to correlate with mineral determinations.

The characterization process found that uranium ore minerals and the precipitates and coprecipitates from the acid leach process used to obtain radium were the chief contaminants in the soil with most of this high radium contaminated material confined to the fine particles (fine-sand to clay-size particles). Incinerated slag particles in the coarse particles (gravel and coarse sand-size) were also contaminated to a lesser degree with radium. Some of the radium of the furnace fired slag was a result of ashing of coal containing uraninite; however, a larger more significant amount was probably from (a) burning of radium contaminated materials that also became part of the slag and (b) adsorption of radium on slag surfaces after being placed in the landfill. Some of the radium also occurs in natural background minerals (zircon, monazite, etc.) and adsorbed on host mineral surfaces in the landfill.

The acid-leach radium in tailings materials was found associated with radiobarite (Ba Ra SO_4) and amorphous silica. The radiobarite comprises 36 percent on the Glen Ridge and 46 percent of the Montclair soil. Amorphous silica, occurring as amorphous coatings on quartz grains in sand-size and as discrete particles in silt and clay-size, comprises 26 percent of the Glen Ridge and 35 percent of the Montclair soil. Whereas the radiobarite is relatively insoluble, the radium and thorium associated with amorphous silica is more readily solubilized.

The uranium ore minerals were identifed by mineral analysis and correlated with secular equilibrium (balance Ra activity with U values). The uraninite from coal ash, in slag particles,

averages 5 percent radium activity in the Glen Ridge and 4 percent radium activity in the Montclair samples. Uraninite and carnotite occurring as discrete ore minerals average 4 percent radium activity for both the Glen Ridge and Montclair samples. Zircon and monazite, limited to microscopic identification, approximate 1 percent of the radium activity in both the Glen Ridge and Montclair samples.

The magnetic ferruginous slag in the coarse particle size fractions contain the ashed uraninite from coal and any radium introduced by incineration of radium contaminated materials. This material also contains some adsorbed radium from solution. It is estimated from radiochemical measurements of magnetic separations of the plus number 50 sieve size (0.15 mm) that 10 percent of the radium 226 activity is associated with this fraction. This information is especially significant in volume reduction remedial measures for the coarse particles in the soil.

The remainder of the radium activity in the soil is associated with radium adsorbing to particle surfaces. The chief adsorbates are illite, chlorite, kaolinite, and hematite; however, radium also adsorbs to feldspar and quartz. The amount of adsorbed radium probably comprises from 5 to 10 percent of the radium activity.

The information made available by the soil characterization has application to volume reduction and treatment processes in remedial investigation procedures.

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