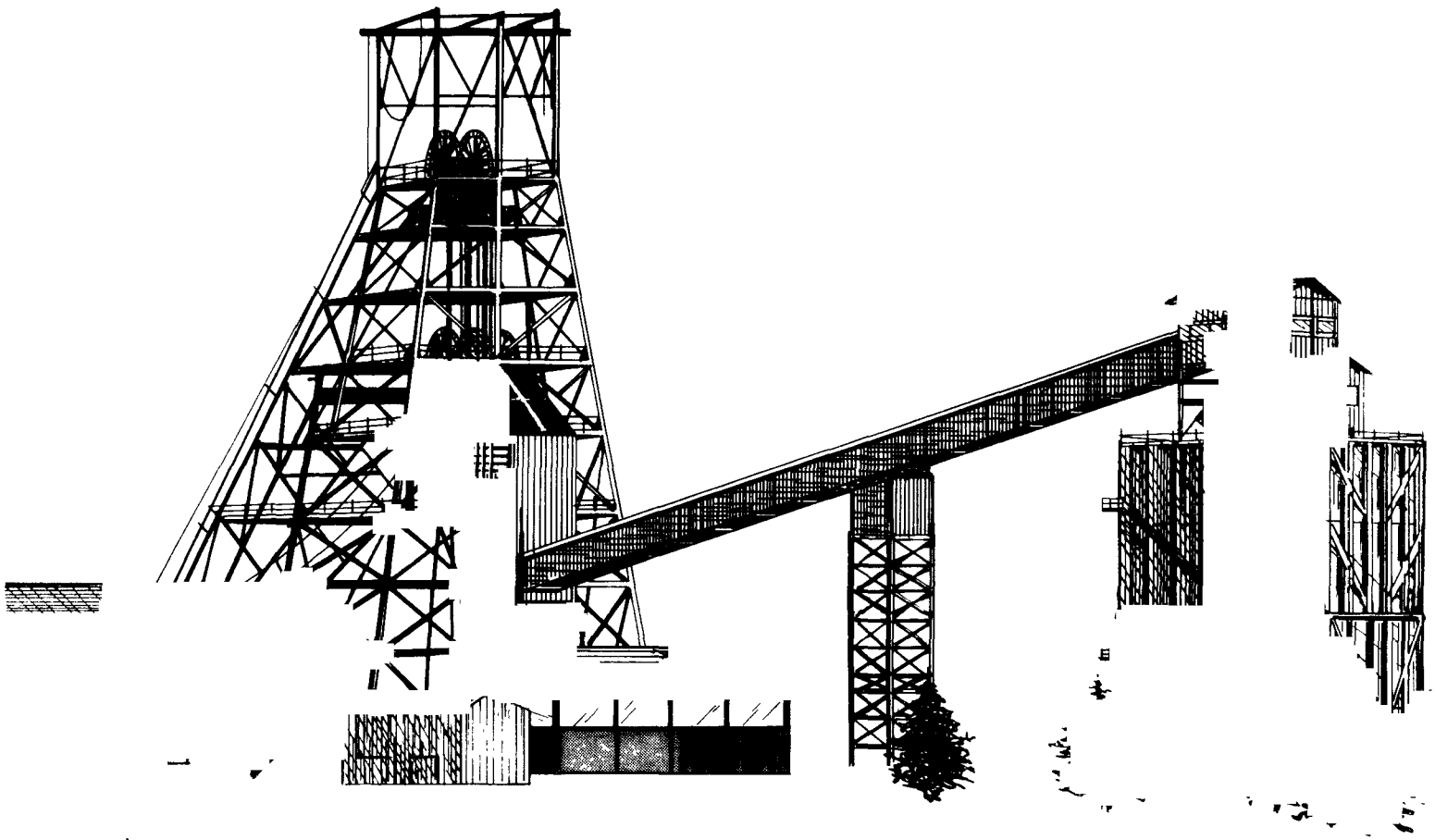
EPA

Emissions Of Naturally Occurring Radioactivity: Fireclay Mine And Refractory Plant



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EMISSIONS OF NATURALLY OCCURRING RADIOACTIVITY: FIRECLAY MINE AND REFRACTORY PLANT

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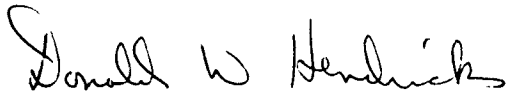
PREFACE

The Office of Radiation Programs of the U.S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and nonionizing radiation, and to promote development of controls necessary to protect the public health and safety. In response to the 1977 amendments to the Clean Air Act the Las Vegas Facility was given the responsibility to collect field data on emissions to the atmosphere of natural radioactivity from operations involved in the mining, milling, and smelting of minerals other than uranium and coal. This report is one of a series which describe an individual facility and the associated radioactivity emissions.

Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.



Donald W. Hendricks
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I BACKGROUND

The Clean Air Act, as amended in August 1977, required the Administrator of the Environmental Protection Agency (EPA) to determine whether emissions of radionuclides into ambient air should be regulated under the Act. In December, 1979, the Administrator listed radionuclides as a hazardous pollutant under Section 112 of the Clean Air Act.

The naturally occurring radionuclides most likely to be emitted in significant quantities are those in the uranium-238 and thorium-232 decay series. These radionuclides and their daughter products occur naturally in widely varying amounts in the soils and rocks that make up the earth's crust. Average values for uranium-238 and thorium-232 in soils are approximately 1.8 ppm (0.6 pCi/g) and 9 ppm (1 pCi/g) respectively (NCRP, 1975).

Almost all operations involving removal and processing of soils and rocks release some of these radionuclides into the air. These releases become potentially important when the materials being handled contain above-average radionuclide concentrations or when processing concentrates the radionuclides significantly above the average amounts in soils and rocks.

Because mining and milling operations involve large quantities of ore, and because there is little information about how these activities release radioactive emissions, EPA, in 1978, began to measure airborne radioactive emissions from various mining, milling, and smelting operations.

Operations were selected for study on the basis of their potential to emit significant quantities of naturally occurring radionuclides to the atmosphere. Some of the factors in the selection included typical mine size, annual U.S. production, measured working levels of radon daughters in underground mines and associated ventilation rates, production rate and process of individual facilities, and previous association with naturally occurring radionuclides. Usually, we chose to look at large facilities in order to get statistically significant results.

These surveys were screening studies designed to identify potentially important sources of emissions of radionuclides into the air. Any such sources can then be studied in detail to determine whether or not a national emission standard for hazardous pollutants is needed under the Clean Air Act.

II INTRODUCTION

Clay mining and manufacturing is a large industry with many mines and mills across the country. Because fire clay has been reported to have high radon daughter working levels in underground mines, (Goodwin, 1978) we studied the fire clay mine and refractory brick plant operated by Globe Refractories, Inc., in Newell, West Virginia.

PEDCo Environmental, under contract with EPA (PEDCo, 1978), conducted the survey and collected samples. Before the survey, representatives of PEDCo Environmental, EPA, and the U.S. Bureau of Mines selected sampling locations. During the week of August 7, 1978, PEDCo Environmental, accompanied by an EPA representative, conducted the sampling and measurement program, collecting effluent and ambient particulate and gas samples as well as information on plant operations. They also installed a meteorological tower for weather measurements. Eberline Instrument Corporation did the radiological analysis of the samples.

III SUMMARY

The survey at Globe Refractories mine and refractory brick plant was the first in a series to determine the quantities of naturally occurring radioactive materials emitted to the atmosphere from mining, milling, benefication, and smelting operations, other than uranium and coal. This plant was selected, in part, because the Mine Safety and Health Administration (MSHA) reported high radon daughter working levels (WL)* in the mine. EPA's WL measurements in the mine were close to those reported

* The working level is defined as any combination of short-lived radon daughter products in 1 liter of air that will result in the ultimate emission of 1.3×10^5 MeV of potential alpha energy. (U.S. Public Health Service, 1957).

by MSHA. EPA's samples of ventilation air, collected just before and just after it reached the working face, measured 0.26 and 0.24 WL, respectively. MSHA had reported 0.3 WL.

Ore samples collected from the mine contained average uranium-234 plus uranium-238 concentrations of 2.3 picocuries/gram (pCi/g) or about 3.7 ppm, about twice the typical value for soil. The high WL measurements are due to the relatively high concentration of natural radioactivity and the relatively low ventilation rate of about two air changes per day. The radon-222 emanation rate from the mine was determined to be 32 curies/year (Ci/y). The total radon-222 release rate measured for the refractory operation was less than 1 Ci/y.

The annual release rate of polonium-210 from the refractory was estimated at 27 microcuries/year (μCi/y). Approximately 26 percent of the polonium-210 in the materials processed through a kiln without emission controls was discharged to the atmosphere. Emissions of polonium-210 from materials processed through two kilns equipped with an ionizing wet scrubber were estimated at 6 percent. Thus the ionizing wet scrubber removed about 77 percent of the polonium-210 which entered it. About 0.11 percent of the uranium in the brick material was emitted from the uncontrolled kiln and about 0.027 percent from the kilns controlled by the scrubber.

IV PLANT OPERATIONS

Globe Refractories, Inc., in Newell, West Virginia, manufactures pouring pit refractories. Figure 1 illustrates the manufacturing processes for the products that Globe manufactures.

The clay used in the manufacture of refractory products is mined underground next to the manufacturing plant (Figure 2). The mine produces 907 Mg (1,000 tons) of clay per day, 231,000 Mg (255,000 tons) per year operating 5 days per week. Two shafts with two reversible fans ventilate the mine; one fan is housed above each shaft. The two fans, each rated at 1,133 m^3/min (40,000 cfm) operate in series, one fan pushes air into the mine while the other draws air out.

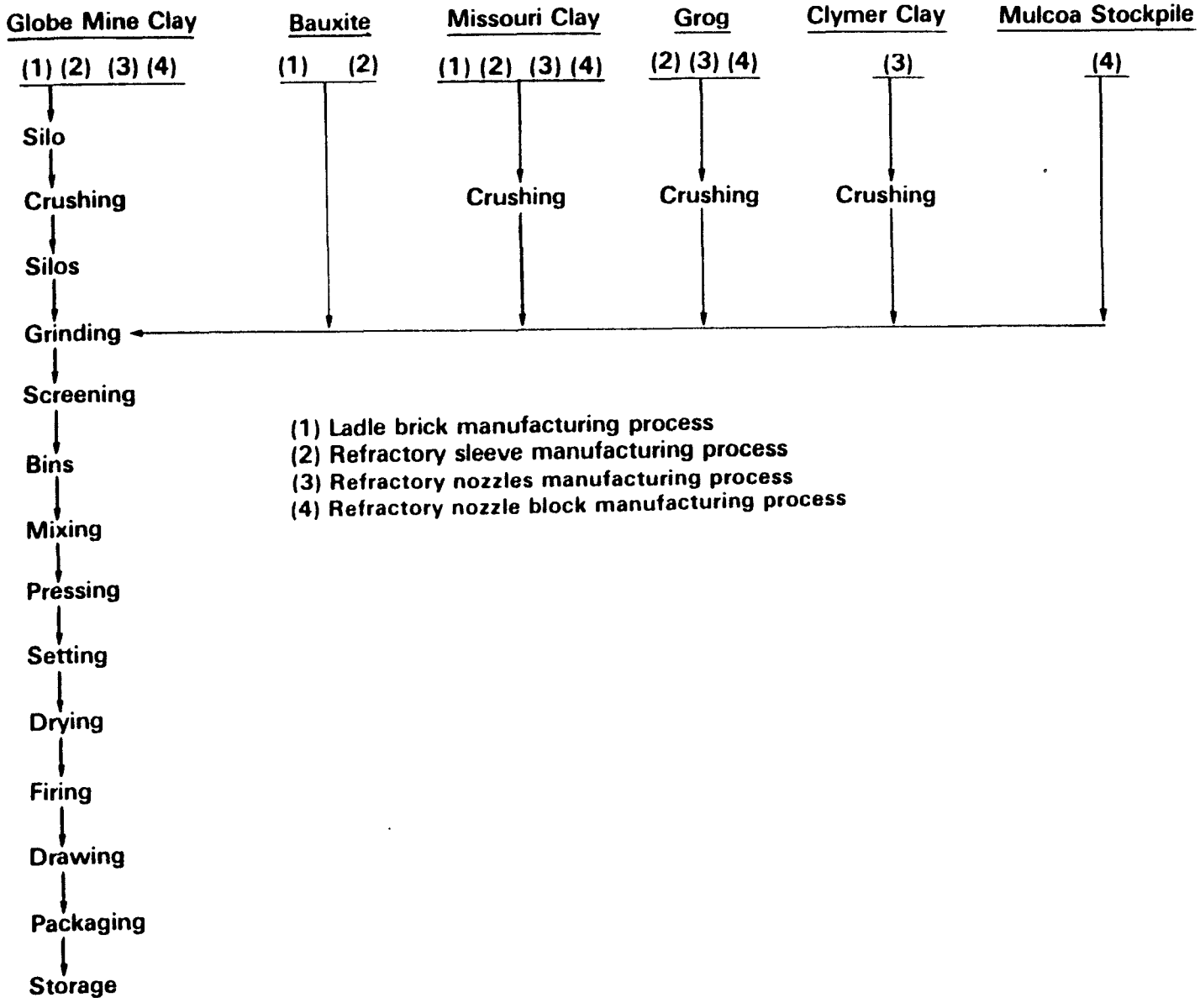

Figure 1. Globe Refractories Manufacturing Processes

The clay is first crushed, then stored until small quantities of other materials are added. Approximately 6,400 to 9,100 Mg (7,000 to 10,000 tons) of bauxite and 11,000 Mg (12,000 tons) of Missouri clay are used per year. The mix varies, depending on the types of bricks produced. The raw mix then goes to a crushing, grinding, and screening operation before being stored again until used. Since these operations are performed on dry materials in an enclosed area, dust is generated. The dust is collected at several pick-up points and passed through two baghouses at the rate of 567 m^3/min (20,000 cfm) each. The cleaned air discharged from the baghouses is recirculated, so there are no direct atmospheric discharges. However, natural ventilation of the building produces some fugitive emissions. This operation processes 23.68 Mg (26.1 tons) per hour.

The bricks are mixed, pressed, and set in part of the same building housing the dryer and kilns. Ventilation is through open ridge-line roof monitors, doors, windows, and wall louvers. Water is added to the dry mix on the second level and the mixture is loaded into presses on the first level.

Before they are fired, the bricks pass through the dryer at the rate of 25.36 Mg (27.95 tons) per hour. Ambient air drawn through the cool-down zone of the kiln preheats and dries the brick. Particulate emissions from the dryer exhaust stacks are very low, and the State requires no control device.

Dried products then pass into one of three kilns, called 4, 5A, and 5B, all heated by natural gas to 1,100°C. Kilns 5A and 5B each process 10.9 Mg (12 tons) per hour. Kiln 4 handles 3.49 Mg (3.85 tons) per hour. At the time of sampling, the exhaust from kilns 4 and 5B passed through an ionizing wet scrubber and discharged through a 45.7-m (150-ft) stack of 1.7-m (5.5-ft) inside diameter. The exhaust from kiln 5A was discharged untreated through a square stack 12.2 meters (40 ft) high by 1.7 meters (5.5 ft) square. At the time of the survey a second scrubber system was being built to serve kiln 5A and was expected to go on-line shortly.

The ionizing wet scrubber controls the opacity of Globe's stack emissions. The opacity problem was caused by ammonium bisulfate, a condensable gas

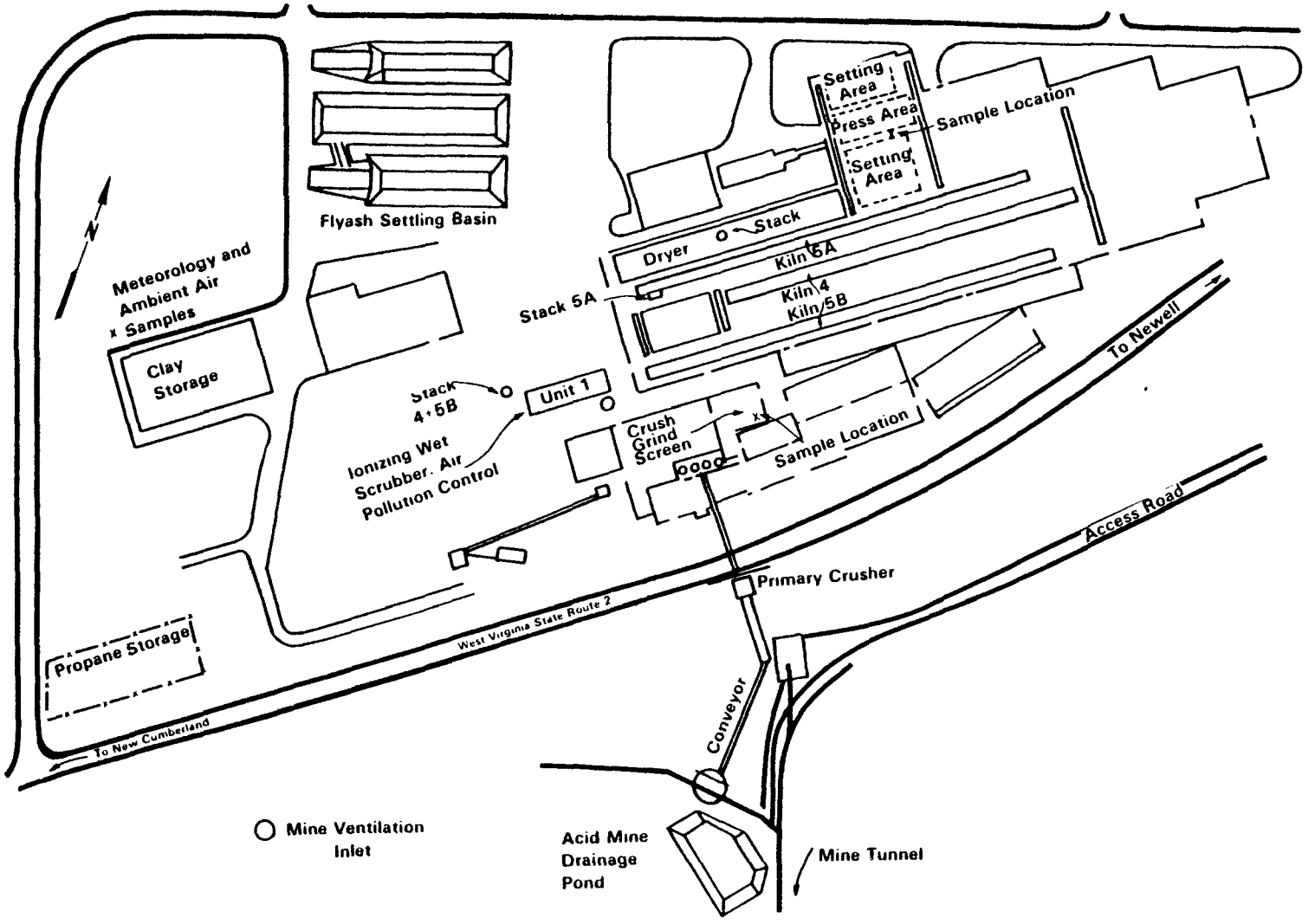

Figure 2. Globe Refractories Inc., Newell, West Virginia

released because of the high concentration (0.7-1.0%) of pyrites in the Lower Kittanning clay deposits. Otherwise, opacity is not a typical problem of brick kiln operations.

Figure 3 is a schematic of the ionizing wet scrubber process. An initial water spray cools kiln exhaust gases which then pass through three packed columns and two ionizing sections. The gases cool from 270°C at the scrubber inlet to 25°C at the outlet. Water is recycled through the flyash settling basin (Figure 2). A small quantity of solid materials is recovered in the scrubber.

Mine drainage and scrubber water is pumped to holding ponds and then to the acid mine drainage pond for treatment prior to discharge into the Ohio River. Because of the relatively low flow rate (0.13 to 0.21 m^3/min) the mine water was not considered a significant source of radon and was not sampled.

The Globe Refractories mine normally operates two shifts per day, 5 days per week from 6:30 A.M. to 9:30 P.M. The first shift prepares clay ore for transport, the second shift conducts drilling and blasting operations. Ore is crushed 20 hours per day, 6 days per week. Other plant operations are continuous.

During the week of August 7, 1978, Globe Refractories produced refractory sleeves in kiln 4. Kilns 5A and 5B produced ladle brick, refractory nozzle block, and some refractory nozzles.

V SAMPLING LOCATIONS AND PROCEDURES

A. Site Selection

During their presurvey visit to the Globe site, PEDCo and EPA personnel selected sampling locations and specified types of samples to be collected. Sampling locations selected were:

1. Mine ventilation inlet.
2. Mine ventilation exhaust.
3. Crushing, grinding, and screening building.

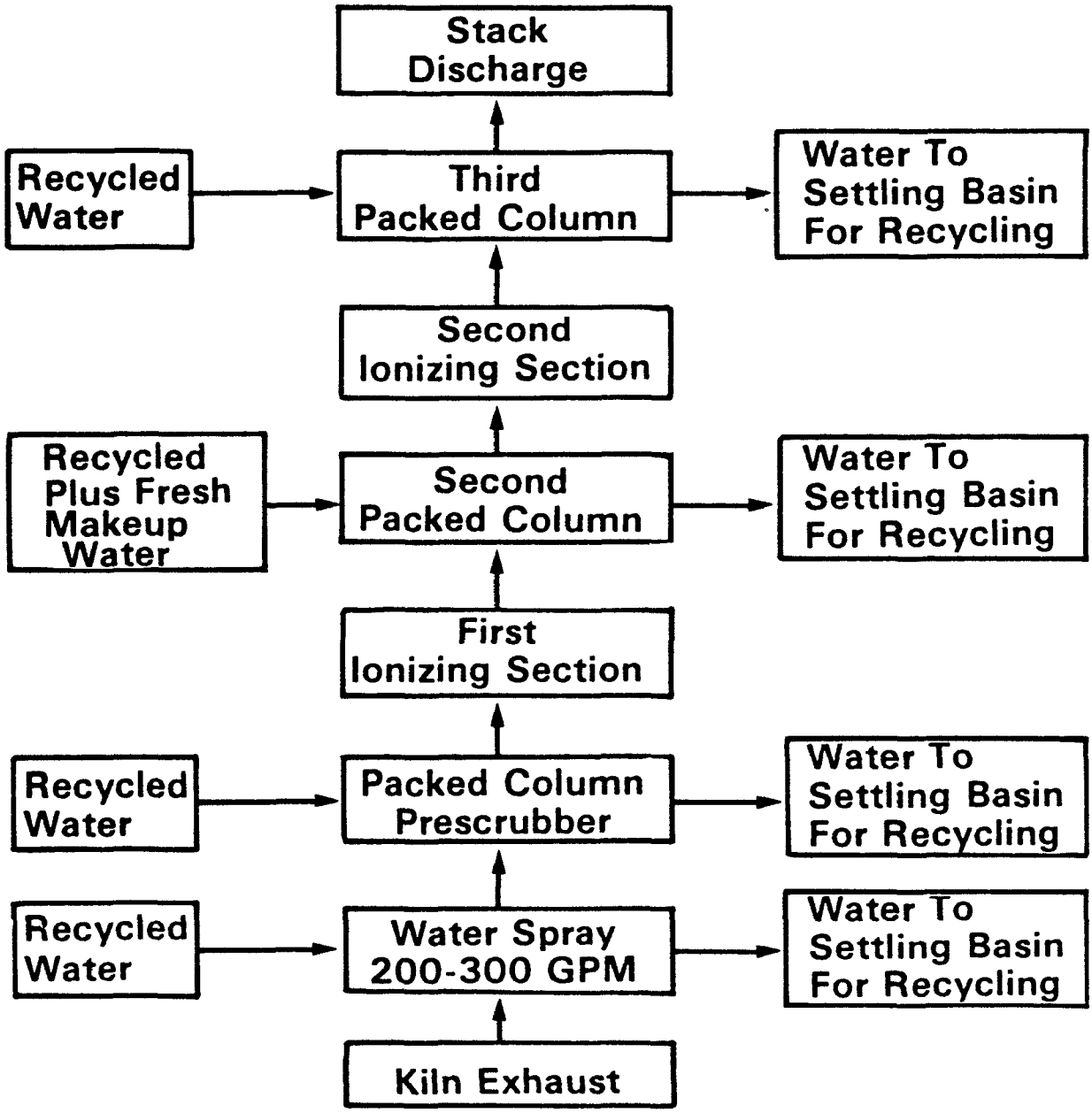

Figure 3. Ionizing Wet Scrubber Flow Schematic

4. Mixing, pressing, and setting building.
5. Dryer exhaust stack.
6. Uncontrolled kiln (5A) exhaust.
7. Controlled kiln exhaust (4 and 5B).
8. Background sample at location upwind from plant.

Major process steps and sampling points are related schematically in Figure 4.

B. Mine Sampling Locations

Three-to-four-hour gas samples for radon-222 analysis were collected at the mine ventilation inlet and the mine ventilation exhaust located 2.3 km south-southeast of the ventilation inlet (Figure 2). Exhaust air discharges through a 91-m (300-ft) drilled shaft. A horizontally mounted exhaust fan in a small building atop the shaft forces the air through a turning vane section to a vertical discharge 2.4 m (8 ft) square, 2.4 m above the surface. Gas samples for radon-222 analysis and high volume size-fractionated samples were collected in the room atop the exhaust shaft just ahead of the exhaust fan. Gross particulate emission samples were collected at the vertical discharge.

The EPA Project Officer made radon daughter WL measurements at three locations in the mine, using a portable, battery-operated air sampler to collect samples. The filters were immediately counted on a portable alpha counter and the results analyzed using the Thomas modification of the Tsivoglou method for determining radon daughters in air (Thomas, 1971). Samples were collected from the ventilation air before it arrived at the mine's working area, after it left the working area, and at the bottom of the exhaust shaft.

C. Plant Sample Locations

Gas samples of air in the crushing, grinding, and screening building were collected for radon-222 analysis. The samples were collected above the storage hoppers near an upper level window through which much of the room air discharged.

Air samples for radon-222 analysis were also collected from the mixing,

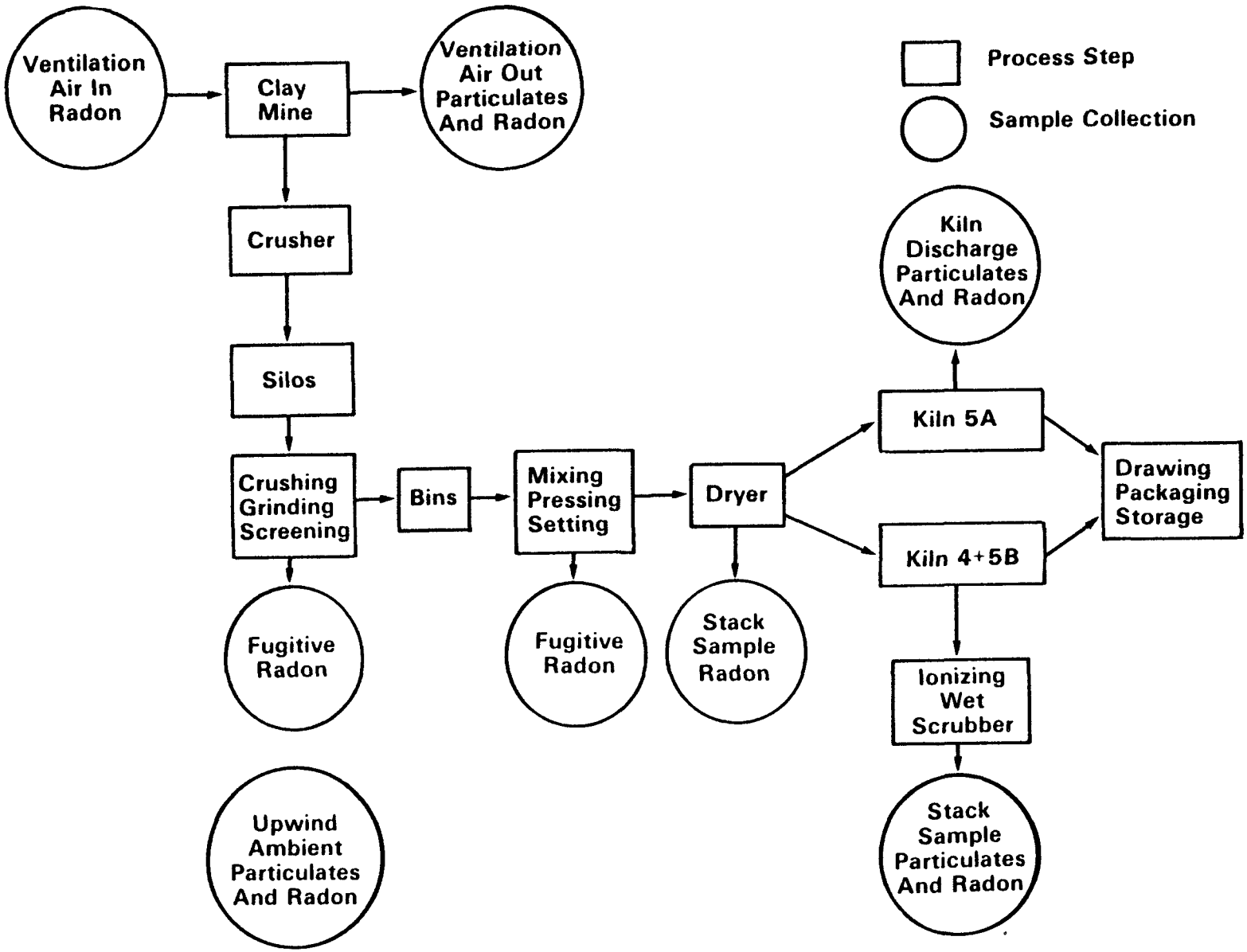

Figure 4. Process Steps and Sampling Points at Globe Refractories

pressing, and setting area. Samples were collected from the third level of the building below the open roof ventilator (ridgeline roof monitor).

Time-integrated gas samples for radon-222 analysis were collected from the dryer exhaust stack above the dryer.

Gas samples, gross particulate emission samples, and size-fractionated particulate samples were collected from the kiln stack serving kiln 5A and from the exhaust stack of the ionizing wet scrubber serving kilns 4 and 5B.

Ambient samples of particulates and gas were collected at the west end of the plant property in an area normally upwind from the emission points. The meteorology tower was also installed here.

D. Sampling Techniques

Whenever possible, surveyors collected samples using EPA reference methods (40 CFR 60). Stack sampling points were selected according to EPA Method 1. Stack gas velocity and volumetric flow rate were determined by EPA Method 2. Gas samples for radon analysis were collected using EPA Method 3. Particulate emissions and stack samples for particle size distribution were collected following EPA Method 5.

A Rader Hi-Volume Sampler was used to sample mine particulate emissions isokinetically. Size-fractionated samples of mine particulate emissions were collected using a high volume air sampler with a Sierra high volume cascade impactor head sampling at 1.13 m^3/min (40 cfm).

Ambient airborne particulates were collected according to the EPA reference method for determining airborne particulates (40 CFR 50).

E. Sample Analysis

Following EPA Reference Method 5, PEDCo made mass determinations on size-fractionated particulate samples from the ventilation exhaust.

Eberline Instrument Corporation (EIC) performed the radiological analyses. They reported all results as radioactivity concentration plus or minus twice the standard counting error. If the radioactivity concentration was less than twice the standard error a lower limit of detection (LLD) has been reported. The LLD is defined (Harley, 1977) as the smallest concentration of radioactive material sampled that has a 95 percent probability of being validly detected.

Whole air samples were analysed for radon in 1.3-liter chambers coated internally with a zinc-sulfide phosphor. Alpha particles striking the phosphor cause scintillations which are detected by a photomultiplier tube optically coupled to a window in one end of the chamber. The LLD reported as of time of collection, varied from 0.11 picocuries/liter (pCi/l) to 0.21 pCi/l depending on the background count rate of individual chambers and the time between collection and analysis.

Airborne particulates on filters and process samples were analyzed by completely dissolving the samples and separating the elements of interest using radiochemical techniques. Analysts counted the separated elements, U, Th, and Po, on alpha spectrometers for isotopic quantitation. Lead-210 was separated; bismuth-210 was allowed to ingrow and was separated from the lead-210 and then was counted on a beta counter to determine the lead-210. Radon gas that emanated from the separated radium-226 was collected and counted, as the whole air radon samples had been counted, to determine the radium activity.

VI SAMPLE RESULTS

A. Process Samples

Samples of clay were collected from the top, middle, and bottom of the layers being mined. One sample of the product leaving the crushing, grinding, and screening operation, a sample of green brick before firing, and a sample of fired brick were also collected. Analytical results indicated that concentrations of elements of the uranium-238 decay chain increase with depth in the ore bed. No statistically significant differences were found between the results for the last three process samples. Results are shown in Table 1.

Table 1. PROCESS SAMPLE RADIONUCLIDE CONTENTS

Sample	Radionuclide Concentrations (pCi/g)[a]								
	U-234	U-235	U-238	Th-228	Th-230	Th-232	Ra-226	Pb-210	Po-210
Ore-Vein Top	0.94 ± 0.24	< 0.03	0.88 ± 0.23	2.0 ± 1.1	1.2 ± 0.66	2.1 ± 1.1	1.0 ± 0.27	1.5 ± 0.76	1.2 ± 0.54
Ore-Vein Middle	1.2 ± 0.25	< 0.08	1.3 ± 0.26	2.0 ± 1.0	1.7 ± 0.87	1.8 ± 0.91	1.4 ± 0.35	2.3 ± 1.4	1.1 ± 0.52
Ore-Vein Bottom	1.6 ± 0.33	< 0.05	1.6 ± 0.33	1.9 ± 0.99	1.6 ± 0.84	2.0 ± 1.0	1.9 ± 0.46	1.8 ± 0.86	0.84 ± 0.49
Crush, Grind Screen Prod.	1.4 ± 0.23	< 0.07	1.2 ± 0.21	1.6 ± 0.20	1.6 ± 0.20	1.7 ± 0.20	2.0 ± 0.49	1.9 ± 0.93	0.96 ± 0.51
Green Brick	0.84 ± 0.18	< 0.10	0.93 ± 0.19	2.6 ± 1.3	2.0 ± 1.0	2.5 ± 1.3	1.3 ± 0.33	1.3 ± 1.0	1.0 ± 0.51
Fired Brick	0.96 ± 0.18	< 0.08	1.0 ± 0.19	2.7 ± 1.4	1.7 ± 0.90	2.7 ± 1.3	1.1 ± 0.29	2.3 ± 0.91	1.6 ± 0.59

a. Uncertainties given with results are twice the standard deviation based on counting results only.

Table 2. AMBIENT RADON CONCENTRATIONS AT GLOBE REFRACTORIES DURING August 7-10, 1978

Date	Time	Radon Concentration (pCi/l)[a] Upwind	Mine Inlet	Average[b]
8/7	1444-1844		<0.11	
	1505-1900	0.12 ± 0.10		<0.12
8/8	0925-1405		0.25 ± 0.12	0.25 ± 0.12
	1418-1822		0.14 ± 0.12	
	1425-1645	<0.14		<0.14
8/9	0954-1402		0.26 ± 0.10[c]	
			0.42 ± 0.09[c]	
	1015-1400	0.42 ± 0.09[c]		
		<0.13[c]		0.31 ± 0.22
	1404-1818		0.53 ± 0.18	0.53 ± 0.18
8/10	1125-1530	<0.12		
	1130-1510		<0.21	<0.17

a. Uncertainties given for individual results are twice the standard deviation based on counting results only. Uncertainties for averages are twice the standard error of the mean.

b. For averaging purposes, less than detectable (<) values were assumed to represent the ambient concentration with a 100% uncertainty at two standard deviations.

c. Duplicate analyses.

B. Background Samples

Prevailing winds during the time of year samples were collected tend to be upriver along the Ohio River valley, or from the southwest. Meteorological results, shown in daily wind roses in Figures 5 to 8, showed that this condition prevailed during sampling and confirmed that the ambient station was usually upwind of the plant. Table 2 shows that ambient radon concentrations varied from less than 0.11 pCi/l to 0.53 ± 0.18 pCi/l. Because of the variability of ambient radon concentrations and the fact that the samples do not represent continuous coverage, no overall average background is calculated. Rather, the ambient concentrations from similar sampling periods at the ambient station and mine inlet were averaged to obtain a background which could be subtracted from stack concentrations measured during the same periods.

Airborne particulates collected on air filters at the upwind background sampling site were also analyzed. The analytical procedure requires that the filter and collected particulates be completely dissolved. Since the filter contains trace amounts of naturally occurring radioactivity, it was necessary to consider how it contributes to the gross activity of the filter samples. Eadie and Bernhardt (1976) conducted a study of the radioactivity content of the various filters used at the Las Vegas Facility of the Office of Radiation Programs. This study showed that Microsorban polystyrene fiber filters generally have lower radioactivity than the glass fiber or cellulose filters used. Thus, Microsorban filters were used where possible to reduce the filters effect on the sample results and their estimated contribution was subtracted from the measurements. Table 3 gives the calculated average concentration of the radionuclides measured over each sampling period. No isotopes of thorium were detected on any ambient air sample.

C. Emission Samples

1. *Mine Emission Samples*

Three radon samples were collected at the mine exhaust during 3 to 4 hour periods representing each of the mine's two working shifts. Radon concentrations, shown in Table 4, varied from 15 to 34 pCi/l. Ambient radon concentrations did not measurably affect the mine discharge concentrations.

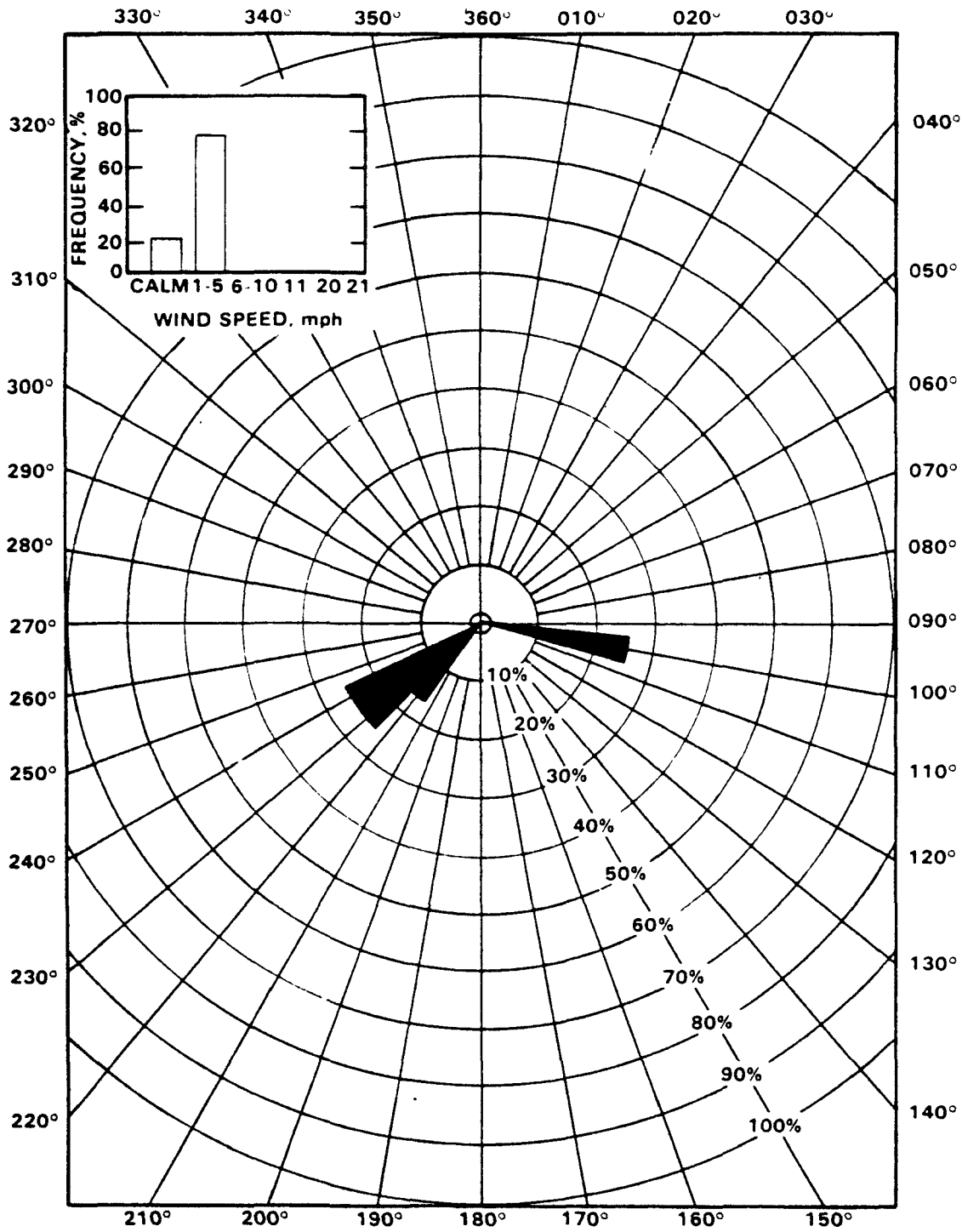

Figure 5. Wind Rose for 8/7/78

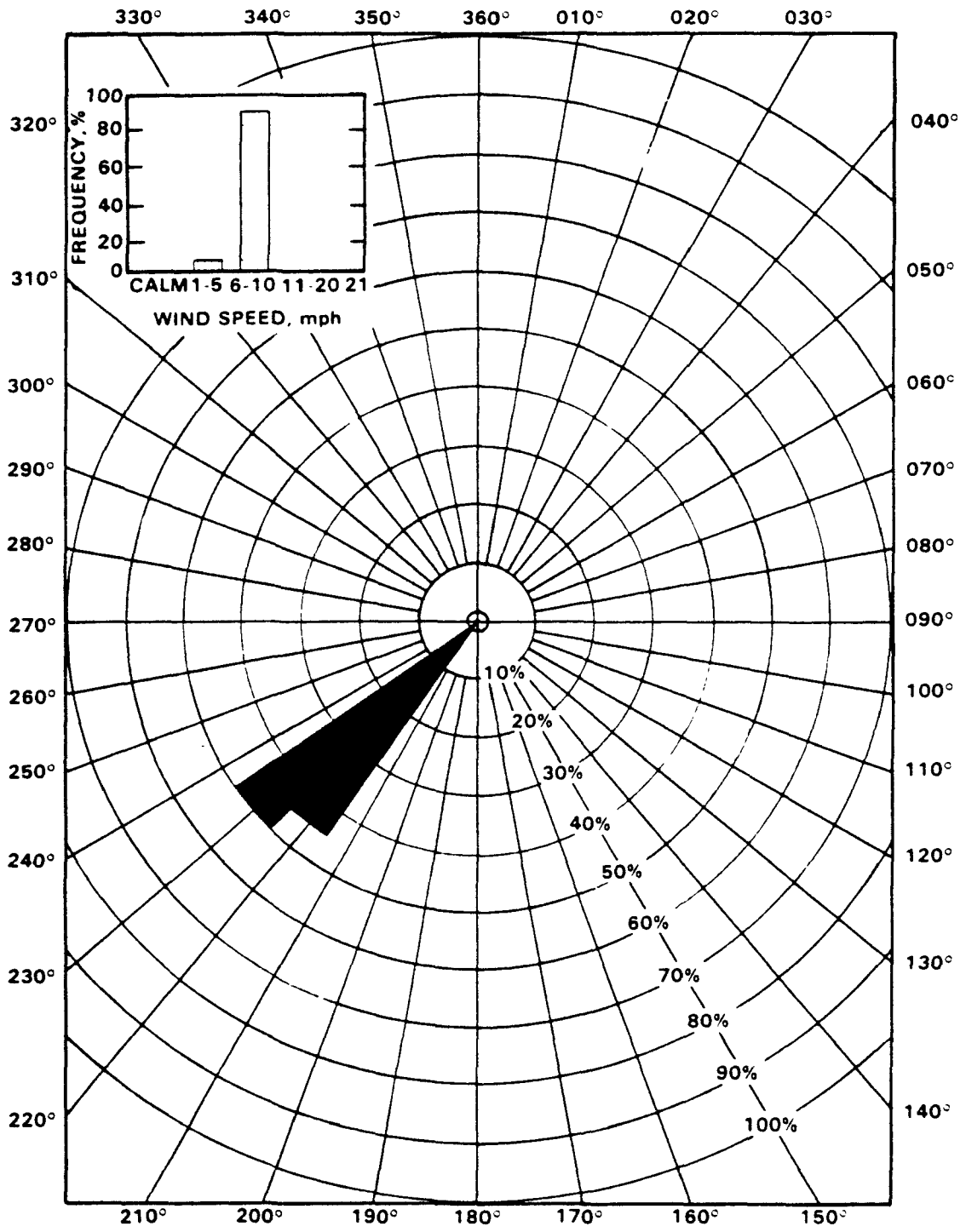

Figure 6. Wind Rose for 8/8/78

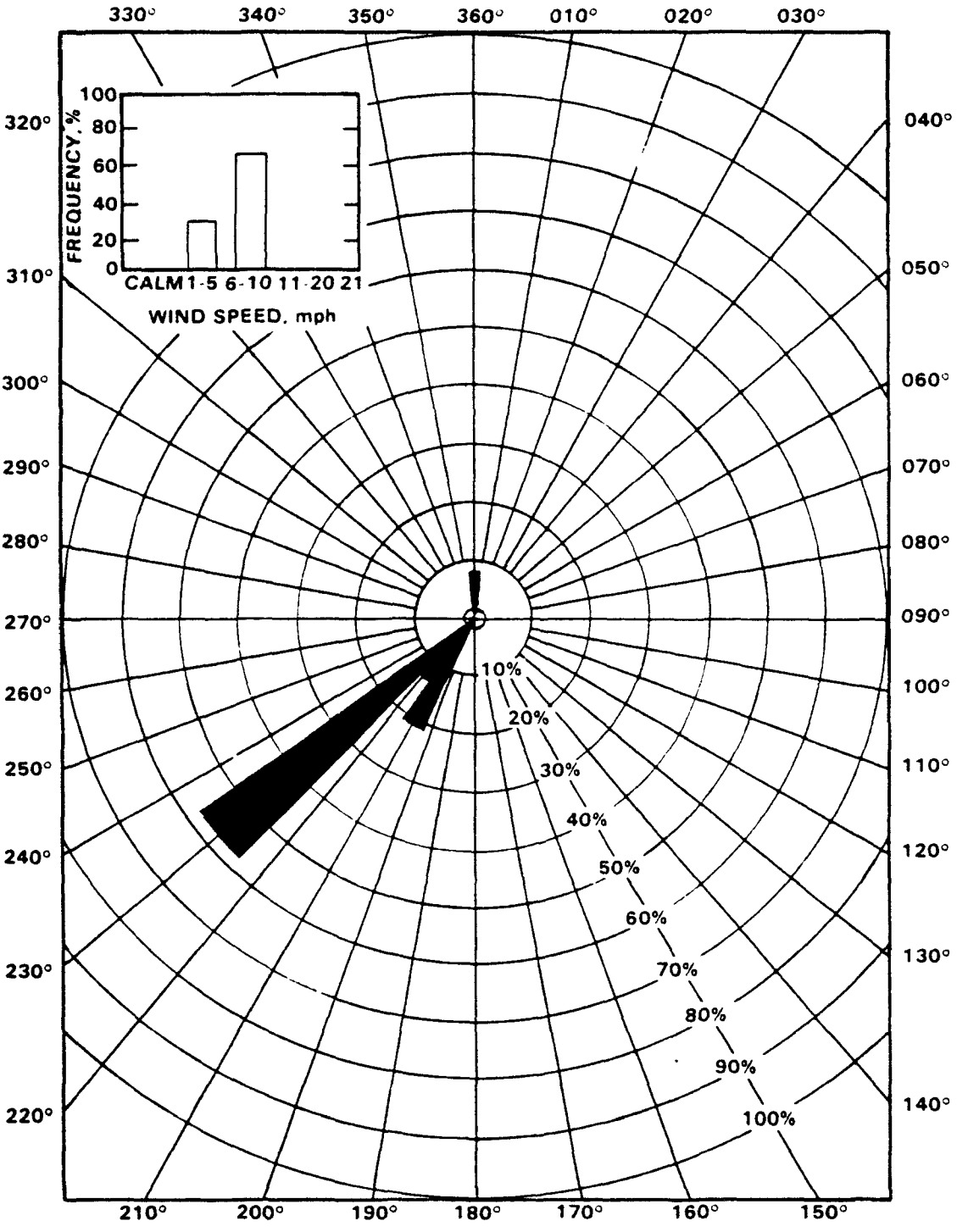

Figure 7. Wind Rose for 8/9/78

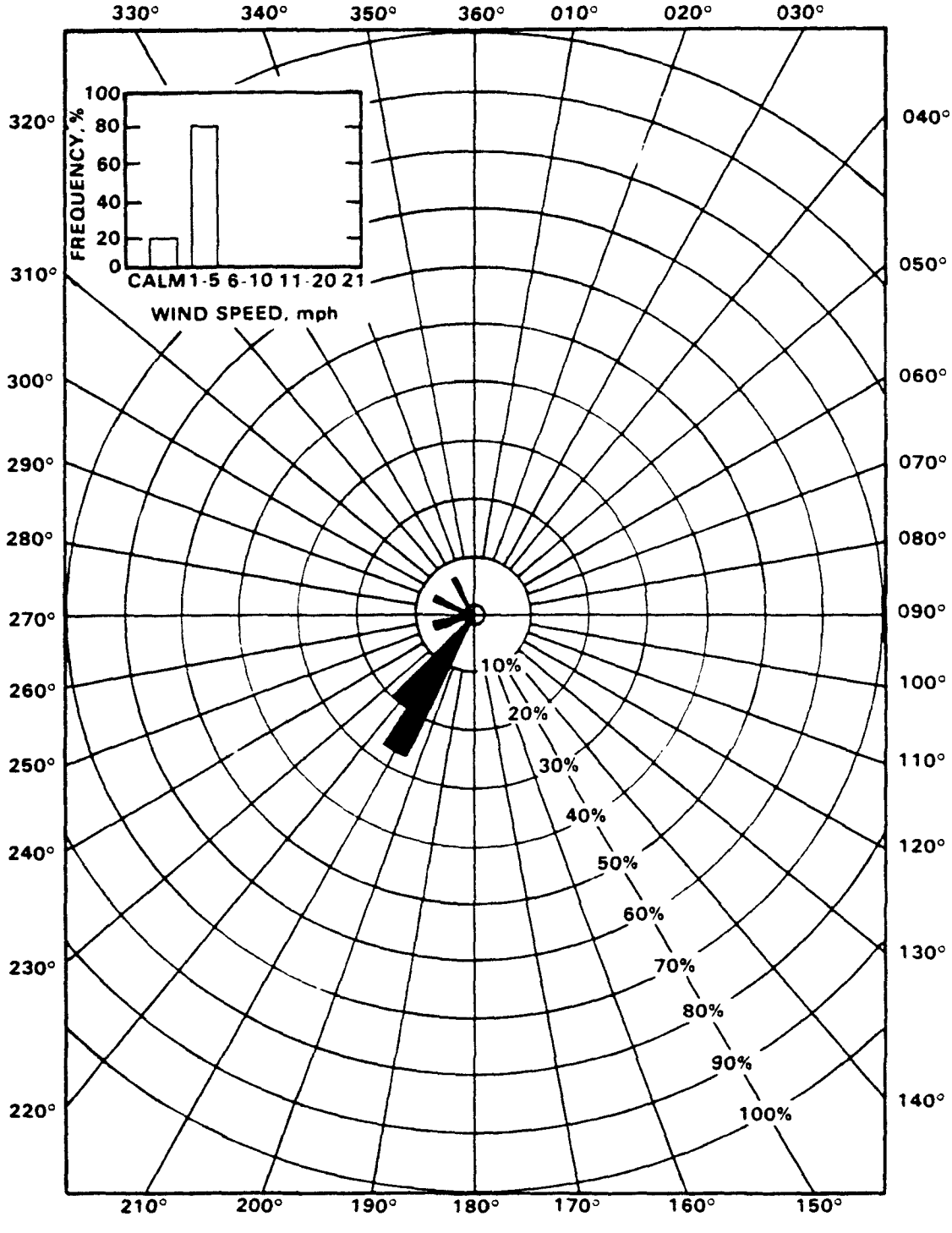

Figure 8. Wind Rose for 8/10/78

Table 3. PARTICULATE RADIOACTIVITY CONCENTRATIONS AT GLOBE REFRACTORIES INC.

Sample Location	Date/Time Collected	Radioactivity Concentrations (fCi/m^3)[a]								
		U-234	U-235	U-238	Th-228	Th-230	Th-232	Ra-226	Pb-210	Po-210
Ambient Air	8/7 1200 8/8 1200	0.11 ± 0.08	<0.07 (0.003)	<0.07	<0.05	<0.05	<0.05	d	<5	<10
Ambient Air	8/8 1200 8/8 1830	0.86 ± 0.41	<0.2 (0.040)	1.0± 0.5	<0.2	<0.2	<0.2	d	<20	<30
Ambient Air	8/9 1200 8/10 1200	0.18 ± 0.10	<0.05 (0.007)	0.17 ± 0.10	<0.1	<0.2	<0.1	d	<6	<7
Kiln 5A	8/8 1600 8/8 1700	190 ± 90	<60 (7.8)	150 ± 80	<40	480± 170	<40	0	<5,000	49,000 ± 16,000
Kiln 5AC	8/9 1055 8/9 1210	300 ± 120	<100 (12)	240 ± 110	<40	<40	<40	<500	<5,000	44,000 ± 14,000
Kiln 5AC	8/9 1057 8/9 1211	<100	<60 (4.7)	<100	<9	<170	<9	0	<7,000	53,000 ± 19,000
Kilns 4/5B Scrubber	8/9 1050 8/9 1240	84 ± 42	<40 (3.9)	89 ± 42	<40	<70	<40	<120	<3,000	18,000 8,100
Kilns 4/5B Scrubber	8/9 1500 8/9 1652	66 ± 41	<10 (3.1)	71 ± 41	<30	95± 71	<30	<70	<3,000	15,000 ± 7,900
Mine Exhaust	8/7 1610 8/7 1647	<4	<3 (0.25)	7.3 ± 6.6	<6	<2	<6	160 ± 20	<300	<200
Mine Exhaust	8/8 1015 8/8 1115	<7	<7 (0.19)	<6	<6	<6	<6	<20	<700	<600

a. Concentrations plus or minus twice the standard deviation. Standard deviation of concentration propagated from standard deviation of sample count and standard error of the mean of multiple blank analyses. Samples with net count rates below the lower limit of detection (LLD) for the counting system are reported as less than (<) the concentration calculated from the LLD.
b. Concentrations in parentheses calcualted from average of U-234 and U-238 and theoretical isotopic ratios in nature.
c. Duplicate samples.
d. Blank sample analytical results too variable to evaluate.

Table 4. RADON EMISSION SAMPLES AT GLOBE REFRACTORIES

LOCATION	DATE	TIME ON	TIME OFF	RADON CONCENTRATIONS (pCi/l)[a] GROSS	NET	AVERAGE	ANNUAL EMMISSION (Curies)
Mine Exhaust	8/7	1522	1815	34 ± 0.7	34 ± 0.7		
	8/8	0945	1357	26 ± 0.6	26 ± 0.6		
	8/8	1359	1809	27 ± 0.4	27 ± 0.4		
	8/9	1010	1412	15 ± 5.4[c]	15 ± 5.4		
	8/9	1418	1805	28 ± 0.4	28 ± 0.4		
	8/10	1145	1530	23 ± 0.4	23 ± 0.4	26 ± 5.1	16 ± 3.1
Kilns 4 and 5B (Scrubber)	8/7	1500	1900	0.32 ± 0.11	0.21 ± 0.19		
	8/8	1415	1800	0.34 ± 0.14	0.20 ± 0.18		
	8/9	1015	1400	0.26 ± 0.20[c]	<0.26		
	8/10	1120	1350	0.18 ± 0.12	<0.22	<0.22	<0.11
Kiln 5A (No Control)	8/7	1504	1904	0.33 ± 0.12	0.22 ± 0.19		
	8/8	1350	1750	0.44 ± 0.13	0.30 ± 0.19		
	8/9	1000	1400	0.26 ± 0.24[c]	<0.26		
	8/10	1128	1530	0.32 ± 0.16	<0.38	<0.29	<0.15
Dryer	8/7	1500	1905	<0.19	<0.19		
	8/8	1400	1750	<0.12	<0.12		
	8/9	1027	1400	0.38 ± 0.06[c]	0.04 ± 0.15		
	8/10	1119	1540	<0.16	<0.16	<0.13	
Crushing[b] Grinding, Screening	8/7	1500	1905	<0.12	<0.12		
	8/8	1350	1800	<0.14	<0.14		
	8/9	1019	1355	<0.09[c]	<0.09		
	8/10	1135	1545	<0.12	<0.12	<0.12	
Mixing[b] Pressing, Setting	8/7	1505	1900	0.29 ± 0.11	0.17 ± 0.16		
	8/8	1405	1811	0.26 ± 0.14	<0.32		
	8/9	1034	1405	0.64 ± 0.10[c]	0.33 ± 0.10		
	8/10	1123	1527	<0.16	<0.16	<0.25	<0.67

a. Uncertainties given for individual results are twice the standard deviation based on counting results only. Uncertainties for net results are propagated from the individual values involved. Uncertainties for averages are twice the standard error of the mean.

b. Concentrations measured inside buildings.

c. Average of duplicate analyses.

However the concentration varied with the shifts. During the first shift, radon concentrations varied from 15 to 26 pCi/l net, averaging 21 pCi/l. During the second shift, concentrations varied from 27 to 34 pCi/l, averaging 30 pCi/l. The overall average was 26 pCi/l.

Three airborne particulate samples were collected in the mine on August 8 from 1055 to 1230 hours to measure radon daughter working levels. At the same time, a radon sample was being collected at the mine exhaust. A sample taken upwind of the working face, just before the ventilation air reached the working face, measured 0.26 WL. A sample from downwind of the working face measured 0.24 WL. A sample collected at the bottom of the 91-m (300-ft) shaft, which carries mine air upward to the exhaust fan, measured 0.029 WL. Most of the dust visible downwind of the working face had been lost before the air reached the bottom of the exhaust shaft, about 500 m away. The rough tunnel surfaces appeared to serve as impaction surfaces, removing airborne particulates between the working area and exhaust shaft. This may also explain the reduction in WL measured at the bottom of the exhaust shaft.

Gross isotopic activities of the high volume air samples collected at the mine discharge did not differ significantly from the blank filter analyses. Only the uranium-238 and radium-226 on the filter collected August 7 had net results that were greater than the 2-sigma error terms. Visual examination of the high volume cascade impactor sample sets collected with the high volume air samples indicated that the airborne particulates consisted primarily of diesel exhaust (PEDCo, 1978). Radioactivity concentrations were too low to permit radionuclide analysis on the size fractionated samples of mine exhaust.

2. Plant Emission Samples

Radon-222 concentrations above ambient were measured in the effluents from both kiln exhaust stacks and in the mixing, pressing, and setting area. No radon concentrations different from ambient were measured in either the dryer exhaust or in fugitive emissions from the crushing, grinding, and screening building. Particulate radioactivity was measured in the two kiln exhaust stacks.

Air flow through the mixing, pressing, and setting area was estimated by measuring air velocities at all openings with inflow as well as the size of the openings. During sampling the ventilation rate in this area was estimated at 5,100 cubic meters per minute (181,000 cfm). Stack air flow measurements made during collection of the Method 5 particulate samples are summarized in Table 5.

The calculated annual radon-222 emission rates were: kiln 5A, <0.15 Ci; kilns 4 and 5B, <0.11 Ci; mixing, pressing, and setting area, <0.67 Ci.

Particulate stack emission samples, shown in Table 3, contained measurable concentrations of uranium-234 and -238, thorium-230, and polonium-210. Uranium-234 and -238 concentrations in the uncontrolled exhaust from kiln 5A ranged from <0.1 to 0.3 pCi/m^3. The concentrations measured in the ionizing wet scrubber exhaust on kilns 4 and 5B ranged from 0.066 to 0.089 pCi/m^3. Thorium-230 concentrations measured in the kiln 5A emissions ranged from <0.04 to 0.48 pCi/m^3. Those in the scrubber exhaust were <0.07 and 0.095 pCi/m^3. Polonium-210 concentrations in the kiln 5A emissions were 44 to 53 pCi/m^3, while those in the scrubber exhaust from kilns 4 and 5B were 15 and 18 pCi/m^3.

The concentrations in Table 3 were multiplied by the flow rates in Table 5 to obtain the annual emission rates based on each sample shown in Table 6. The results for each source were averaged and are presented in Table 7 as the average annual release rate in µCi/y.

Mass distributions by aerodynamic particle size of the particulate emissions from the mine and kiln stacks are shown in Figures 9 to 12. Figure 9 shows the particle size distribution in mine ventilation emissions during the first shift when the major activity is loading and hauling clay ore from the mine. The distribution would extrapolate to a geometric median diameter of 0.06 µm with a geometric standard deviation of 5. Due to the relatively small size of particles emitted and the color of the backup filter, the particulate emissions from the mine seem to result primarily from diesel exhaust.

Table 5. STACK FLOW MEASUREMENTS

Sample Location	Date	Time	Flow Rate (sm^3/min)*	Temperature (°C)	Stack Height(m)
Kiln 5A	8/8	1600-1700	984	272	12.2
Kiln 5A	8/9	1055-1210	1001	266	12.2
Kiln 5A	8/9	1057-1211	938	264	12.2
Kiln 4+5B	8/9	1050-1240	916	24.4	45.7
Kiln 4+5B	8/9	1500-1652	922	26.1	45.7

* Flow rate at standard conditions = 20°C, 760 mm Hg.

Table 6. ANNUAL PARTICULATE RADIOACTIVITY RELEASE RATE DETERMINED FROM EACH SAMPLE AT GLOBE REFRACTORIES

Release Point	Date Time Collected	Radioactivity Release Rate (μCi/y)[a]								
		U-234	U-235	U-238	Th-228	Th-230	Th-232	Ra-226	Pb-210	Po-210
Kiln 5A	8/8 1600 8/8 1700	110 ± 50	<30	85 ± 44	<20	260 ± 95	<20	0	<3,000	27,000 ± 8,800
Kiln 5A[b]	8/9 1055 8/9 1210	170 ± 68	<5	140 ± 62	<20	<20	<20	<540	<3,000	25,000 ± 7,900
Kiln 5A[b]	8/9 1057 8/9 1211	<110	<30	<120	<6	<100	<6	0	<3,000	29,000 ± 10,000
Kilns 4+5B	8/9 1050 8/9 1240	43 ± 22	<20	44 ± 22	<20	<30	<20	<60	<2,000	9,100 ± 4,100
Kilns 4+5B	8/9 1500 3/9 1652	34 ± 21	<5	34 ± 21	<20	51 ± 34	<20	<30	<2,000	7,600 ± 4,100
Mine Exhaust	8/7 1610 8/7 1647	<3	<2	4.4 ± 4.0	<4	<1	<4	96 ± 12	<200	<200
Mine Exhaust	8/8 1015 8/8 1115	<5	<5	<4	<4	<4	<4	<20	<500	<200

a. Uncertainties given with results are twice the standard deviation based on counting results only.

b. Duplicate samples.

Table 7. AVERAGE ANNUAL PARTICULATE RADIOACTIVITY RELEASE RATE AT GLOBE REFRACTORIES

Release Point	Annual Release Rate (μCi/y)								
	U-234	U-235	U-238	Th-228	Th-230	Th-232	Ra-226	Pb-210	Po-210
Kiln 5A (Uncontrolled)	<130	<20 (<4.6)[a]	<115	<15	<130	<15	<180	<3,000	27,000
Kilns 4+5B (Scrubber)	39	<20 (1.8)[a]	40	<20	<40	<20	<50	<2,000	8,400
Mine Exhaust	<4	<4 (<0.18)[a]	<4	<4	<3	<4	<60	<350	<200

a. Values in parentheses are release rates calculated from average of U-234 and U-238 and theoretical isotopic ratios in nature.

Figure 10 shows the particle size distribution of mine emissions during the second shift when drilling and blasting are the major mining activities. The geometric median diameter of this sample is 0.48 μm with a geometric standard deviation of 6.7.

Figure 11 shows the particle size distribution of the emissions from the uncontrolled stack of kiln 5A. The geometric median diameter is 14 μm with a geometric standard deviation of 5.8.

The size distribution of particulates from the ionizing wet scrubber serving kilns 4 and 5B, shown in Figure 12, demonstrates a geometric median diameter of 4.3 μm with a geometric standard deviation of 2.8.

We did no radionuclide analysis on the individual stages of the size-fractionated emission samples because we believed the levels were too low to get reliable results. However, we have been able to perform such analyses on similar samples from other facilities. The results from those samples show that most of the polonium-210 is associated with the smaller particles. This would be expected due to the evolution of polonium-210 as a vapor from the fired brick.

VII POPULATION DISTRIBUTION

Globe Refractories is located in a rural area of low population density. Homes are scattered around the mine exhaust, with the nearest about 150 m northeast. A small riverside resort with several permanent residents is about 200 m north northeast of the plant. A light industrial area is adjacent to the southwest plant boundary.

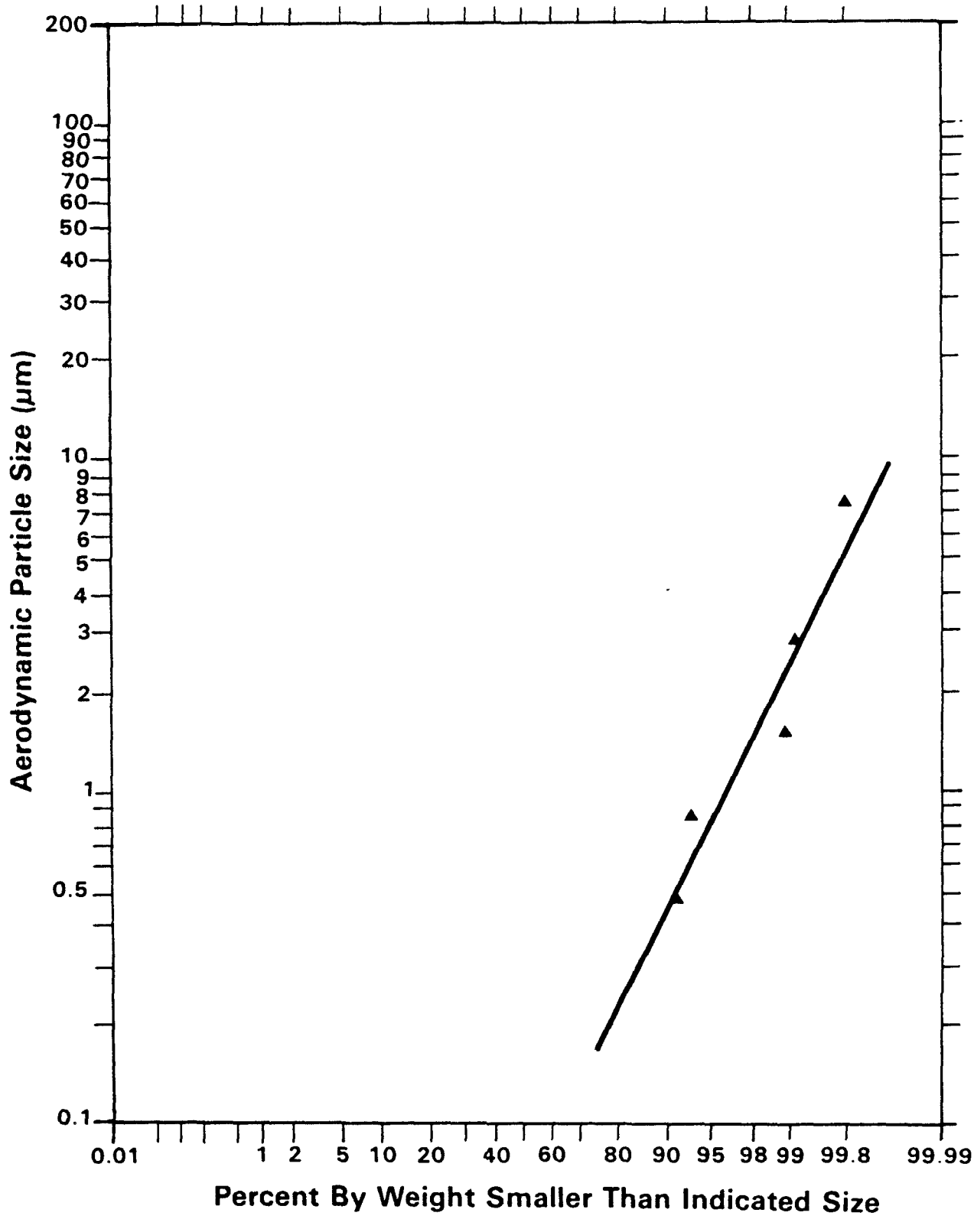

Figure 9. Particle Size Distribution for Mine Ventilation Exhaust-First Shift

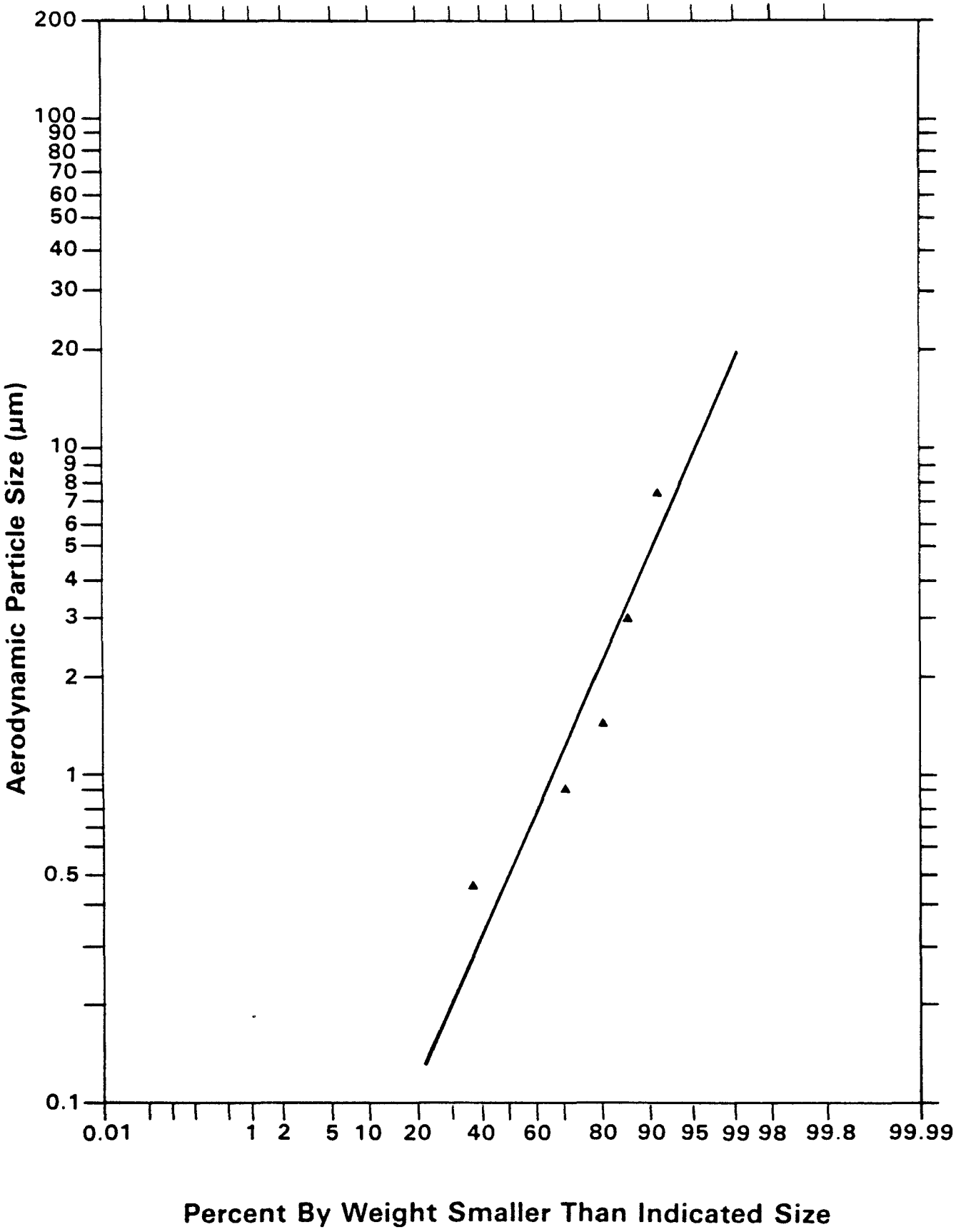

Figure 10. Particle Size Distribution for Mine Ventilation Exhaust - Second Shift

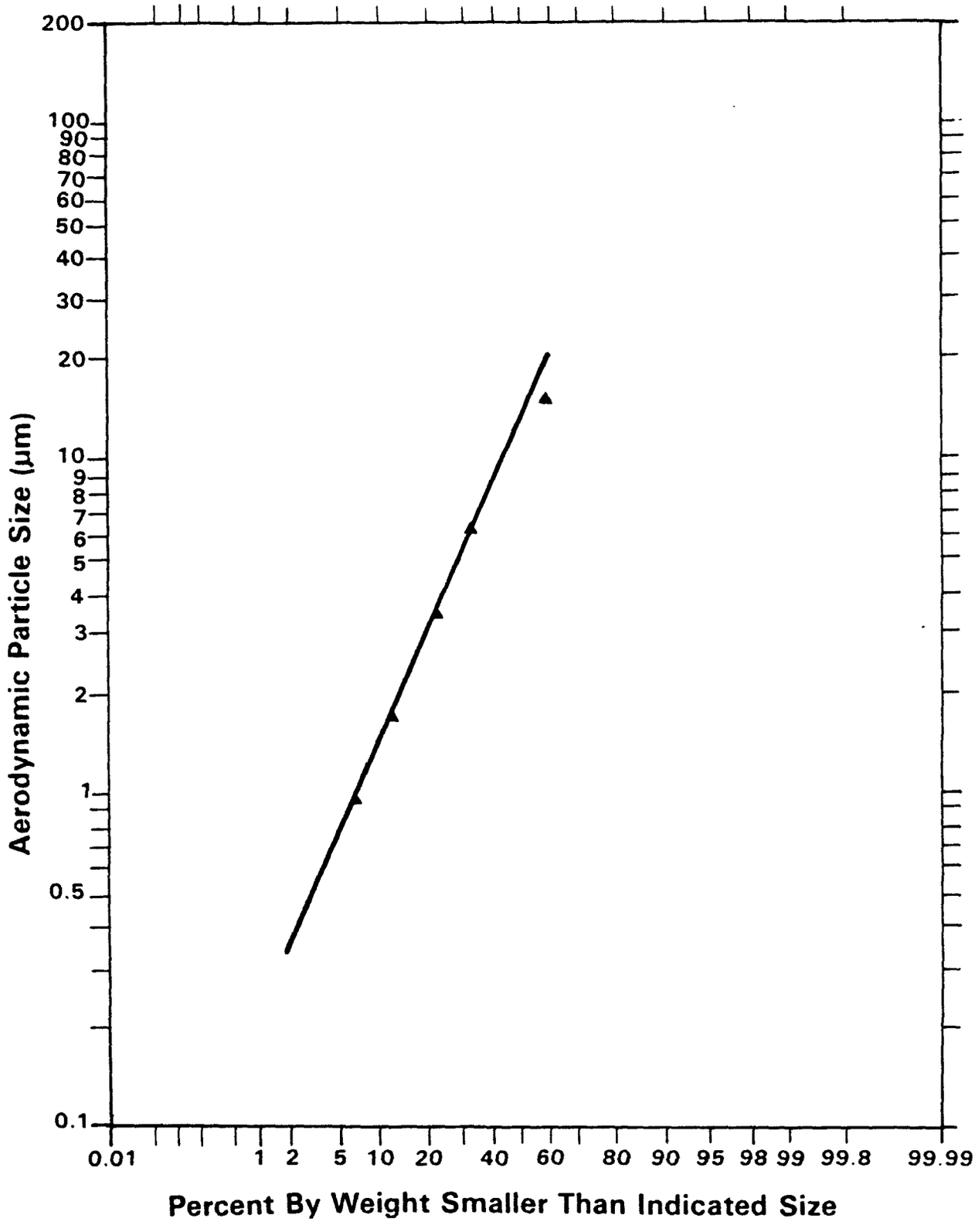

Figure 11. Particle Size Distribution for Uncontrolled Kiln Outlet

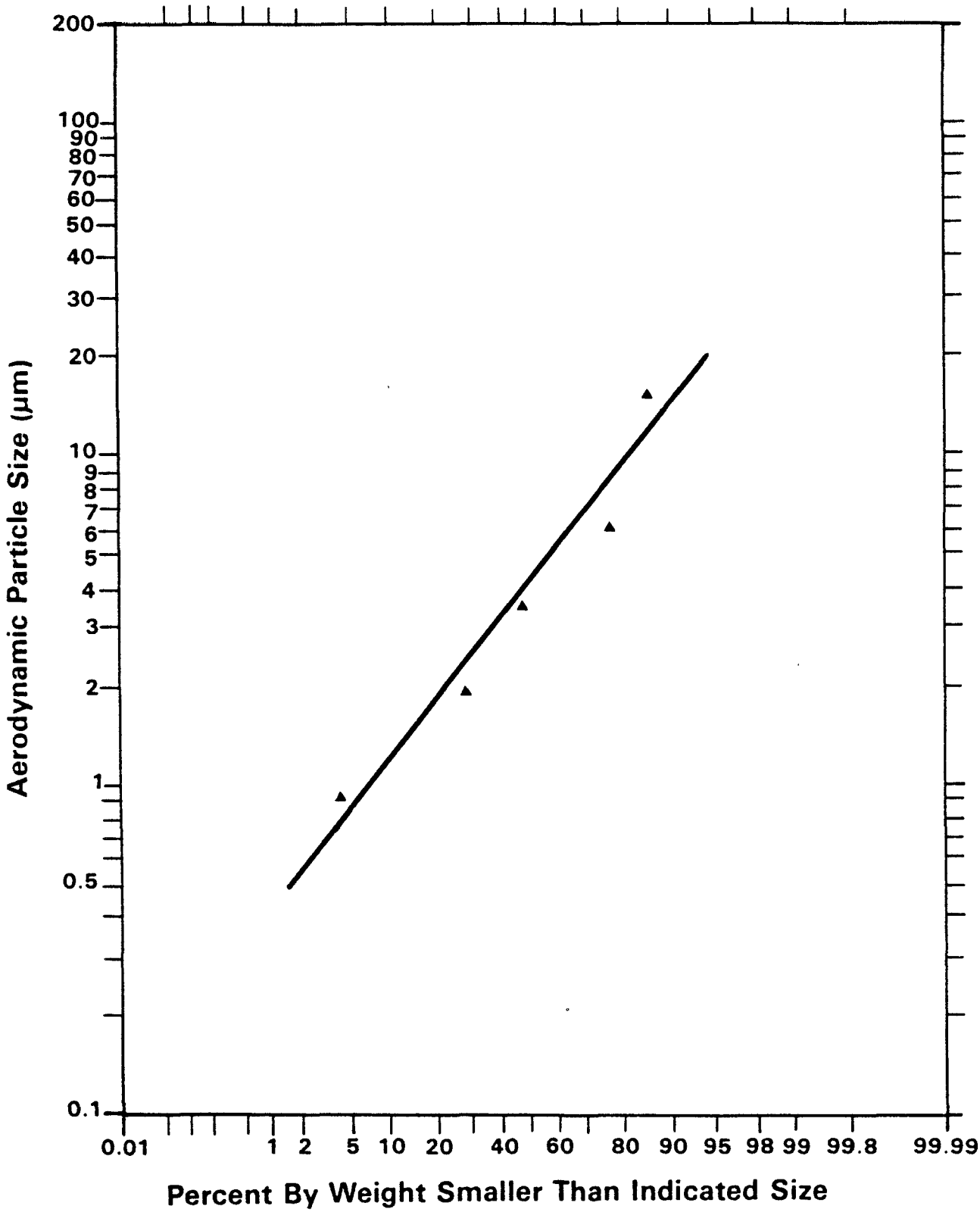

Figure 12. Particle Size Distribution for Scrubber Outlet

VIII DISCUSSION OF RESULTS

Gross concentrations of radon-222 in the mixing, pressing, and setting area and in the kiln exhausts ranged from ambient to approximately three times ambient. The major point of release, however, occurred at the mine ventilation exhaust where concentrations of radon-222 were two orders of magnitude greater than ambient. The calculated annual release of 16 curies from the mine ventilation exhaust accounts for more than 95 percent of the total measured radon-222 release from the combined mine and plant operation. In contrast, the particulate radioactivity release rate from the mine accounted for less than 2 percent of the total particulate radioactivity from the combined operations.

A map of the mine was used to estimate the surface area and volume subject to ventilation. The actual contribution to the radon exhaust of any area of the mine must be estimated because the ventilation air does not travel uniformly through the mine. The total surface area subject to ventilation was estimated at 0.98 km^2; the ventilated volume at 7.9×10^5 m^3.

Assuming uniform ventilation through the mine at a rate of 1133 m^3/min (40,000 cfm), the air is exchanged once every 700 minutes, or approximately twice daily. The working area of the mine during the survey was near the exhaust end of the ventilation flow. Except for the radon picked up from the working face, the concentration of radon at the work area should have been essentially the same as it was at the exhaust.

The majority of the particulate radioactivity released from the kilns was due to polonium-210. The effluent from kiln 5A, released directly to the atmosphere, discharges 27 mCi per year. An additional 8.4 mCi per year is discharged from the ionizing wet scrubber that handles the effluents from kilns 4 and 5B, yielding an annual total of about 35 millicuries (mCi). The effluents also contain about 150 μCi per year each of uranium-234, uranium-238, and thorium-230, and an estimated 6.4 μCi of uranium-235.

The average concentration of polonium-210 in the ore and process materials was 1.1 pCi/g. Given the kiln's annual production rates, it was determined that 26 percent of the polonium-210 included in the material processed through kiln 5A is released to the atmosphere. In addition, 6 percent of the polonium-210 passing through kilns 4 and 5B is discharged through the ionizing wet scrubber. Thus, the ionizing wet scrubber cuts the polonium-210 emissions by 77 percent.

The total uranium-234 plus uranium-238 concentration in the material through the kilns was 2.3 pCi/g. Kiln 5A's release of about 245 μCi/yr for the two isotopes constitutes about 0.11 percent of that processed. In contrast, the release fraction through the ionizing wet scrubber on kilns 4 and 5B was 0.027 percent. Thus the ionizing wet scrubber cuts uranium emissions by 75 percent, about the same as for polonium-210.

Measured release fractions of uranium-234 and -235, thorium-230, and radium-226 from the kilns were approximately 1 percent of the polonium-210 release fractions. The kiln's operating temperature of 1,100°C appears to readily vaporize polonium, which has a boiling point, in its elemental form, of 1,040°C. The other naturally occurring radionuclides are much less volatile at that temperature, but some vapor does seem to be given off.

The sample of natural gas fuel for the kilns contained 1.8 pCi of radon-222 per liter. Kiln 5A consumed 17 m^3/min (36,000 cubic feet per hour) of fuel, releasing 31 nCi/min or 0.016 Ci/yr. That amount would be about 10 percent of the estimated amount of less than 0.15 Ci/yr released from the kiln. Kilns 4 and 5B burn 23.8 m^3/min (50,500 cfh). The 42.9 nanocuries/minute that the fuel provides would total 0.023 Ci/yr, or about 20 percent of the less than 0.11 Ci/yr released from the ionizing wet scrubber. Although these figures suggest a 50 percent removal by the scrubber, the difference is not statistically significant, and the results should not be used to imply a removal efficiency.

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16. ABSTRACT

Atmospheric emissions of naturally occurring radioactivity were measured at a fireclay mine and the associated plant that produces refractory brick products. The only significant radioactive emission from the mine was radon-222. An analysis of the ore radioactivity and surface area of the mine indicated that the radon released is comparable to that from any similar surface area of similar radioactivity. The major particulate radioactivity from the refractory operation was polonium-210, released as the brick was fired. Approximately 26 percent of the polonium-210 in green brick was driven off in the kilns.

17. KEY WORDS AND DOCUMENT ANALYSIS

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