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Emissions Of Naturally Occurring Radioactivity: Monsanto Elemental Phosphorus Plant

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EMISSIONS OF NATURALLY OCCURRING RADIOACTIVITY FROM MONSANTO ELEMENTAL PHOSPHORUS PLANT

by

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FORWARD

The Office of Radiation Programs (ORP) of the U.S. Environmental Protection Agency (EPA) conducts a national program for evaluating exposure of humans to ionizing and nonionizing radiation. The goal of this program is to develop and promote protective controls necessary to ensure 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 describes an individual facility and its associated radioactive emissions.

ORP encourages readers of the report to inform the Director, ORP-Las Vegas Facility, of any omissions or errors. Comments or requests for further information are also invited.

Wayne A. Bliss
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Las Vegas Facility

CONTENTS

	Page
Forward	iii
Figures	vi
Tables	vi
Abbreviations and Symbols	vii
1. Background	1
2. Introduction	3
3. Summary	4
4. Plant Operations	5
Production Process	5
Airborne Emission Sources	7
5. Sample Collection and Analysis	9
Sample Collection	9
Sample Analysis	9
Data Reporting	10
6. Sample Results	11
Process Samples	11
Ambient Air Samples	14
Emission Samples	14
7. Discussion of Results	26
References	27

FIGURES

Number		Page
1	Uranium and Thorium Radioactivity Decay Schemes	2
2	Monsanto Chemical Plant Flow Diagram	6

TABLES

Number		Page
1	Radioactivity Concentrations in Process Materials	12
2	Ambient Radon-222 Concentrations	15
3	Ambient and Stack Particulate Radioactivity Concentrations	16
4	Radon-222 Stack Emissions	18
5	Stack Flow and Particulate Emission Rates	20
6	Particulate Radioactivity Emission Rates	21
7	Particulate Radioactivity Annual Emission Rates	23
8	Aerodynamic Particle Size Distribution	25

LIST OF ABBREVIATIONS AND SYMBOLS

ABBREVIATIONS

Ci	-- curies: 3.7×10^{10} disintegration per second
Ci/y	-- curies per year
kg/h	-- kilograms per hour
km	-- kilometer
mCi/y	-- millicuries (10^{-3}Ci) per year
nCi/m^3	-- nano (10^{-9}Ci) curie per cubic meter
pCi/g	-- picocuries (10^{-12}Ci) per gram
pCi/l	-- picocuries per liter
pCi/m^3	-- picocuries per cubic meter
ppm	-- parts per million
TSP	-- total suspended particulates
μm	-- micrometer: 10^{-6}meters

SYMBOLS

CO	-- carbon monoxide
E-S	-- Engineering Science, Inc.
EIC	-- Eberline Instrument Company
FeP	-- ferrophosphorus

SECTION 1

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 (Figure 1). 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 (1). The radioactivity concentration of each of the daughter products in the two series is approximately equal to that of the uranium-238 or thorium-232 parent.

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 radio active 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 improve the chances of obtaining emission samples with radioactivity contents significantly greater than background.

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.

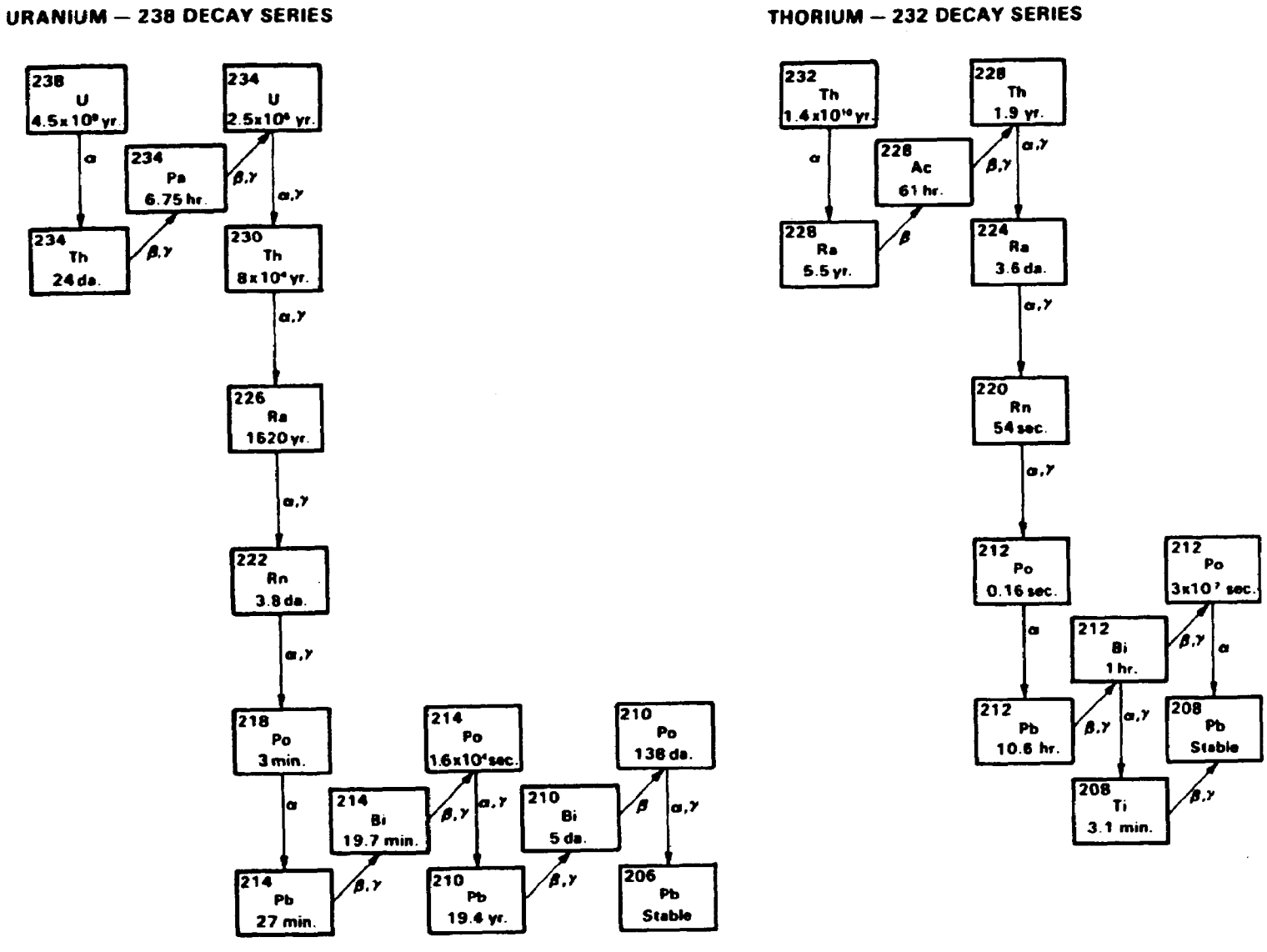

FIGURE 1. URANIUM AND THORIUM RADIOACTIVITY DECAY SERIES

SECTION 2

INTRODUCTION

As early as 1908 it was recognized that phosphate rock contained above normal concentrations of uranium and thorium (2). Those concentrations have been observed in the United States to range from 8 to 399 ppm (2.7 to 133 pCi/g) of uranium-238 and 2 to 19 ppm (0.22 to 2.1 pCi/g) of thorium-232 (3). South Carolina ores had the highest concentrations, while the lowest were found in Tennessee. During the 1950's uranium was recovered as a byproduct of phosphate production.

Since 1974 the EPA has studied various aspects of the radioactivity released to plant environs during benefication and processing of phosphate ores. The EPA had conducted a comprehensive radiological survey of a thermal phosphate (elemental phosphorus) plant in 1974 (4). However, problems with the analysis of lead-210 had greatly reduced the accuracy of the measurements of both lead-210 and polonium-210. Under the added emphasis provided by the Clean Air Act Amendments the decision was made to include at least two elemental phosphorus plants in this series of surveys. The two plants selected were the Stauffer Chemical Company plant in Silver Bow, Montana, and the Monsanto Company plant in Columbia, Tennessee. This report presents the results of the Monsanto facility survey.

Engineering-Science, Inc. (E-S) under contract with the EPA, conducted the survey and collected samples (5). Representatives of E-S and EPA visited the plant before the survey to select sampling locations. During the period December 4-14, 1979, E-S, accompanied by an EPA representative, conducted the sampling and measurement program. E-S collected particulate and gas samples from plant emissions and ambient air as well as information on plant operations. A meteorology station was installed at the ambient air monitoring location.

SECTION 3

SUMMARY

Engineering Science, Inc. (E-S) collected particulate and gaseous emission samples from the major emission points at the Columbia, Tennessee plant of Monsanto. Where more than one stack served a process only one stack was sampled. Ambient particulate and gaseous samples were collected at a site 2 km northwest of the plant.

The thorium decay chain radionuclides, thorium-232 and -228, were found at normal background levels in ore and emission samples. The uranium decay chain radionuclides averaged 3.4 pCi/g in Tennessee ores and 47 pCi/g in ores from Florida.

Kiln exhaust stacks were the major point of release of radioactivity from the plant. Radon emissions from the kilns totaled 9.6 Ci/y. Lead-210 emissions from the kilns were 0.48 Ci/y and polonium-210 emissions were 0.75 Ci/y. These were probably all released primarily from the molten materials in the furnaces and carried to the kilns along with the CO used as fuel.

Except for one source, the non-volatile nuclides, uranium-238 and -234, thorium-230, and radium-226, produced annual releases of these nuclides, averaging 0.24 to 1.5 mCi. The screening plant dust collectors were estimated to release an average of 7.2 mCi/y of each of these nuclides. Possible non-operation of a scrubber on the discharge may be the reason for the higher emissions from this source which accounted for over half of the 13 mCi/y of each nuclide released from all sources at the plant.

The majority of the suspended particulates emitted were found to be above the respirable size range. This probably holds, as well, for the non-volatile radionuclides.

SECTION 4

PLANT OPERATIONS

PRODUCTION PROCESS

The Monsanto Company plant is located approximately 10 km northwest of Columbia, Tennessee. Most of the ore comes from strip mines in the vicinity of the plant, but part of the ore is shipped from Florida. The plant produces elemental phosphorus. Ferrophosphorus (FeP) is an important by-product. At the time of the survey a portion of the slag produced was sold as an aggregate.

Figure 2 is a schematic of the plant operation. Concentrates from the washer and dust collected from the kilns and screening plant are stockpiled in an open, raw materials craneway. From here the materials are sent to one of two stockpiles. As one stockpile is built up - taking about 1 week - the other supplies the kiln. Ores from various sources are initially blended by the stockpiling process in the craneway and are further blended by the kiln feed operation.

It is necessary to form the finely divided ore into larger, stable agglomerates for proper operation of the reduction furnaces. This is accomplished by passing the ore through three rotary kilns. The kilns raise the ore temperature to its incipient melting point and the tumbling action forms it into the desired nodular form. The hot nodules pass through coolers and are conveyed by a series of conveyors to the nodule storage craneway. A clamshell bucket on an overhead crane moves the nodules to storage bins from which they pass through the screening plant on their way to the burden scales and on to the furnace stocking system. At the burden scales coke is added to serve as the reducing agent and silica and lime are added, as needed, to achieve the necessary balance of calcium and silicon to form a proper slag. The approximate reaction in the furnace is:

$$2\ Ca_3(PO_4)_2 + 10\ C + 6\ SiO_2 \rightarrow P_4 + 10\ CO + 6\ CaSiO_3$$

In addition, the iron naturally present in ore reacts with some of the phosphorus to produce FeP. The blended furnace feed enters the furnaces continually from the top and progresses downward until reaching the molten layer on the bottom. Phosphorus and carbon monoxide (CO) are driven off as gases and are vented near the top of the furnace. The slag and FeP which are continually collecting in the furnace are periodically "tapped off." Molten slag is carried by bucket to a dump. Front-end loaders transfer the cooled slag to a storage pile. At the time of the survey, slag was being sold from storage for use as aggregate.

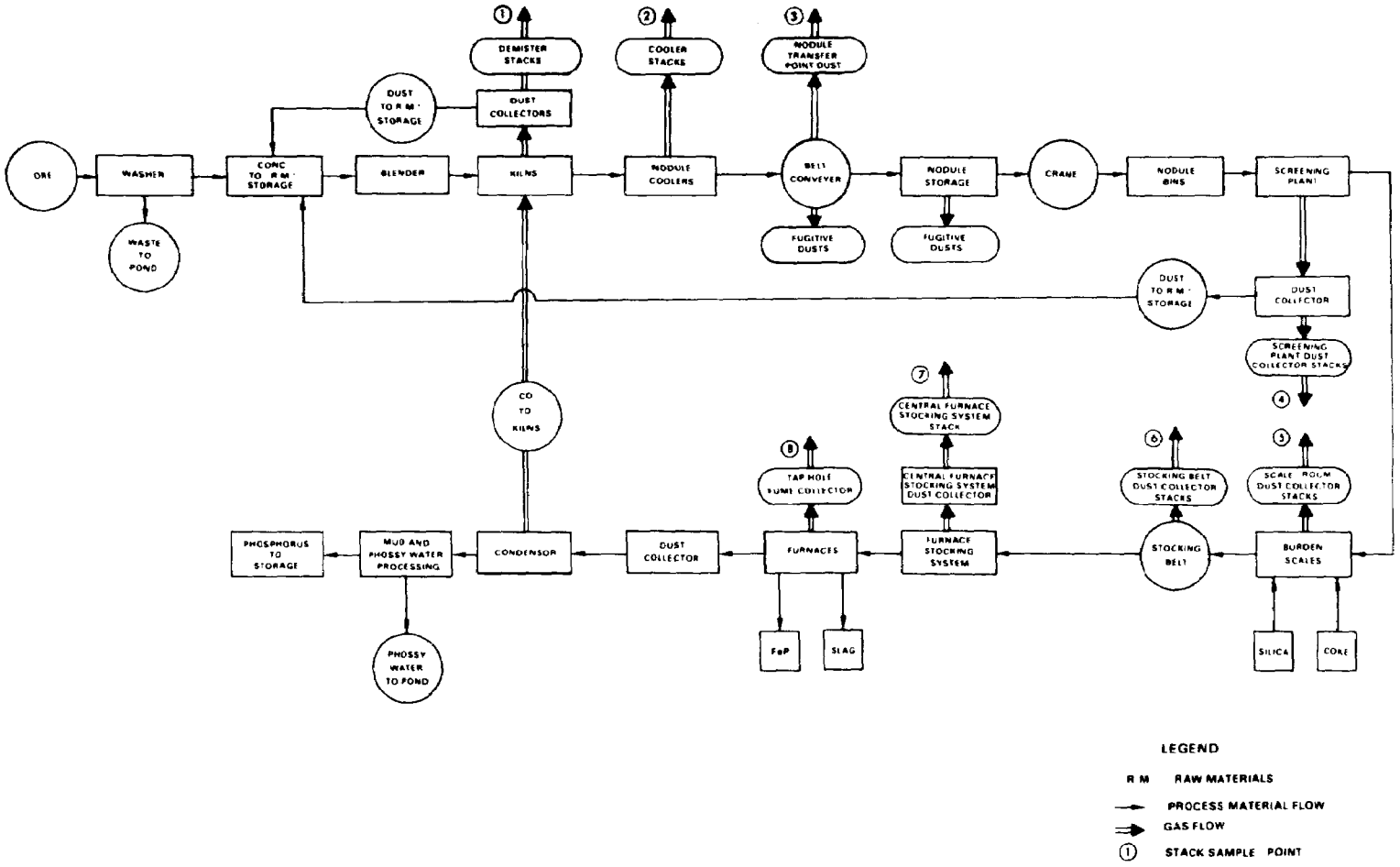

FIGURE 2. MONSANTO CHEMICAL PLANT FLOW DIAGRAM

Furnace off-gases pass through dust collectors then through water spray condensors. Phosphorus is cooled to the molten state in the condensors. The mix of phosphorus and water - phossy water - and mud go to a processing system where phosphorus is separated and piped to storage. The clean off-gases leaving the condensors contain a high concentration of CO and are used as fuel in the kilns to supplement the coal fuel.

AIRBORNE EMISSION SOURCES

The Monsanto emission inventory includes 54 controlled and uncontrolled sources. Thirty-one sources treat emissions from coke handling, boilers, short-term intermittent operations, or emergency release emissions. Open sources, such as craneways and open-belt conveyors are not easily sampled. Eight stacks were selected as representing the important emission sources based on reported mass emission rates or process involved. These sources are shown in Figure 2, keyed to the following descriptions.

Kilns (1)

The three kilns discharge emissions through individual dust collection chambers and spray scrubbers. Lime is added to the scrubber spray to aid in removal of fluorine and sulfur dioxide. Entrained water in the exhaust gases is removed in two demister stacks. One demister stack serves emissions from kilns 1 and 2. The other demister stack serves kiln 3. The 5.4-m diameter stack discharges about 35 m above ground level. An existing Monsanto sampling system was used to collect samples from the top of the stack serving kiln 3.

Nodule Coolers (2)

Each nodule cooler has an emission control system and discharge stack. Emissions are controlled by jet eductors. The exhaust stack on the nodule cooler at kiln 3 was sampled. Samples were collected on both inlet and exhaust of the jet eductor to determine the fraction removed. The 4.4-m diameter stack discharges 25 m above ground.

Nodule Transfer Points (3)

Particulate emissions from the screening plant operations are controlled by three "Type R" rotoclones composed of nine "clones" each. "Type R" rotoclones combine water scrubbing with multiclone separation. For purposes of this survey the screening plant dust collectors were identified by emission source. One dust collector served the nodule transfer points on the pan conveyors. The others controlled emissions from other screening plant points. Nodules from the coolers are diverted at several points to other conveyors for delivery to multiple entires into the nodule storage craneway. Dusts generated at these transfer points are collected by hooding systems and treated for removal of particulates by a "Type R" rotoclone. The 1-m diameter stack discharges at a height of 29 m.

Screening Plant Dust Collector (4)

Two dust collectors control emissions from the screening plant other than the nodule transfer points. The two "Type R" rotoclone dust collectors and their emissions are essentially identical so only one source was sampled. Test results indicated that the water to the rotoclone was off during testing. This has been corrected by Monsanto. The 1-m diameter stack discharges at a height of 29 m.

South Scale Room Dust Collector (5)

Nodules, coke, and silica are weighed and fed into the furnace stocking system by means of weighing belt conveyors. Two dust control systems are in use in this area. The south scale room Luhr baghouse dust collector controls emissions from the area where nodules are weighed. Because of the dissimilarity of the emissions this was considered to be a single stack source. The other stack was not sampled. The 0.9-m diameter stack exhausts at a height of 13 m.

Stocking System Belt Dust Collector (6)

The stocking system belt conveyor moves the blended furnace feed from the scale room to the furnace stocking system. The conveyor and transfer points are hooded and particulate emissions are controlled by two similar systems. One stack was selected as representative of the two stacks. Particulate emissions are controlled by a Luhr baghouse dust collector. The 0.75-m diameter stack discharges at a height of 21 m.

Central Furnace Stocking System (7)

The stocking system belt conveyor transfers furnace feed to the central furnace stocking system. From there, three pivoting belt conveyors stock the six furnace stocking bins. Emissions from the central furnace stocking system are controlled by a Buell bag dust collector. The 0.9-m diameter stack exhausts at a height of 48 m.

Furnace Taphole Fume Collector (8)

Each of the six furnaces has a hood, emission control system, and stack. Slag is tapped from each furnace for 20 to 25 minutes with approximately 20 minutes between taps. Ferrophosphorus is tapped once or twice daily. Fumes in the stack effluent are controlled by a venturi scrubber. The stacks are 1.1 m in diameter and discharge at a height of 38 m.

Other Sampling Points

In addition to the eight controlled sources sampled, high-volume air samplers were operated near the slag dump and adjacent to the Florida ore rail car dump and stockpile. The Florida ore pile is located close to the slag storage pile.

SECTION 5

SAMPLE COLLECTION AND ANALYSIS

SAMPLE COLLECTION

Most stack samples were collected using EPA reference methods for stationary sources (6). Stack sampling points were selected according to EPA Method 1, Sample and Velocity Traverses for Stationary Sources. Stack gas velocity and volumetric flow rate were determined by EPA Method 2, Determination of Stack Gas Velocity and Volumetric Flow Rate (Type S pilot tube). Gas samples for radon-222 analysis were collected using EPA Method 3, Gas Analysis for Carbon Dioxide, Oxygen, Excess Air, and Dry Molecular Weight. Total suspended particulates (TSP) in ducts and exhaust stacks were determined using EPA Method 5, Determination of Particulate Emissions from Stationary Sources, or EPA Method 17, Determination of Particulate Emissions from Stationary Sources (In-stack Filtration Method). High volume ambient TSP samples were collected in accordance with the Reference Method for the Determination of Suspended Particulates in the Atmosphere (7).

E-S collected Method 5 TSP samples on 7.6-cm (3-inch) diameter glass fiber filters and Method 17 TSP samples on 5- by 12.7-cm (2- by 5-inch) glass fiber filters. Ambient TSP samples were collected on 20.3- by 25.4-cm (8- by 10-inch) Microsorban polystyrene fiber filters. Stack gas and ambient whole air samples for radon analysis were collected in 30-liter Tedlar bags.

E-S sampled each of the emission points described in Section 4-B. They collected two to four samples each of TSP and gas samples from each point. Two samples were collected simultaneously on the south scale room dust collector stack as part of the quality assurance program. Two size-fractionated samples were collected from each of the sources except the kiln demister, nodule cooler, and furnace taphole fume scrubber.

Samples of process materials were collected to relate radionuclide emission rates to the radioactivity of the material handled at each emission point and to permit a radionuclide balance through the process.

SAMPLE ANALYSIS

E-S made mass determinations on TSP samples before forwarding them to Eberline Instrument Corporation (EIC) for radiochemical analysis. Stack and

ambient gas samples were shipped to EIC for arrival within 2 days of collection.

EIC analyzed process and TSP samples by dissolving the samples and separating the elements of interest by chemical techniques. The separated uranium and thorium fractions were counted on alpha spectrometers for individual isotopic quantitation. An alpha scintillation counter measured the polonium-210. Lead was separated and set aside for about 2 weeks to allow for ingrowth of bismuth-210 from lead-210. After the ingrowth period the bismuth-210 was separated from the lead and was counted on a beta counter to quantitate lead-210. Radium was separated and enclosed as a solution in a sealed tube to allow for ingrowth of radon from radium-226. After 3 weeks of ingrowth the radon gas was evolved and collected in an alpha scintillation cell to be counted. Stack and ambient gas samples were transferred to alpha scintillation cells and counted for radon.

DATA REPORTING

The radioactivity reported for each sample, is the net radioactivity plus or minus twice the standard deviation due to counting statistics. Net radioactivity is the gross sample radioactivity minus the counting equipment background and minus either a) for filter samples, an average value for the radioactivity content of a blank filter, or b) for stack radon samples, the ambient radon concentration. The standard deviation is based only on the random variations inherent in radioactivity counting and is propagated through the various steps to the final result. This random variation, plus the variable radioactivity content of individual filters, occasionally results in a net radioactivity of less than zero. Of course, there is no negative radioactivity. In these cases, as with all others, the net result must be considered along with the standard deviation. Averages of multiple samples from a source are given with a standard deviation due to counting statistics. This uncertainty is propagated from the sample standard deviations.

SECTION 6

SAMPLE RESULTS

PROCESS SAMPLES

Process sample radioactivity concentrations are given in Table 1. Ore from the six Tennessee sources had similar radioactivity contents. The average concentration of uranium decay chain radionuclides was 3.4 pCi/g. Tailings solids averaged 3.0 pCi/g. The washed ore kiln feed averaged 4.0 pCi/g in the 1st shift sample and 6.6 pCi/g in the 2nd shift sample. This indicates that the uranium and its decay products are more closely associated with the phosphate ore than with the barren fines. As expected, Florida ore had a higher radioactivity content with uranium decay chain nuclides averaging 47 pCi/g. Thorium decay chain nuclide concentrations of about 1 pCi/g in Tennessee ores and 0.5 pCi/g in kiln feed were in the range expected for normal rock. Thorium-232 and -228 were at normal concentrations in ore and were generally not found at levels significantly different from background in emission samples. Therefore, the remainder of the sample results discussion will include only uranium decay chain nuclides. Thorium-232 and -228 results are included in the data tables, however.

Nodule samples were collected from the conveyor belt and from the craneway storage area at the same time as the 1st shift kiln feed samples. Nodules from the conveyor had an average radionuclide concentration of 7.4 pCi/g, not including the more volatile polonium-210. The craneway composite sample averaged 3.5 pCi/g for the same nuclides. The differences in concentrations in the kiln feed and nodule samples is probably due to some inhomogeneity resulting from blending of Tennessee and Florida ores. Although blending provides large scale homogeneity sufficient for production, it apparently is not complete on the small scale of a sample prepared for analysis.

Coal, coke, and silica all had normal, background concentrations of radioactivity. Slag and FeP had approximately the same concentrations of uranium, however, about 90 percent of the total uranium was in slag due to the relative amounts produced. Thorium from both decay chains was found primarily in slag, as was radium. Lead-210 and polonium-210 were found in higher concentrations in the FeP (3.9 ± 0.5 and 1.0 ± 0.9 pCi/g) than in slag (1.5 ± 0.5 and <0.8 pCi/g). Most of the lead-210 and polonium-210 originally in the ore, however, is volatilized in the kiln. The kiln emission control systems show high concentrations of these isotopes in their discharge water.

TABLE 1. RADIOACTIVITY CONCENTRATIONS IN PROCESS MATERIALS

Sample	Radionuclide Concentrations (pCi/g)[a]							
	U-238	U-234	Th-230	Ra-226	Pb-210	Po-210	Th-232	Th-228
Ore-McEvens Muck	2.6 ± 0.4	2.5 ± 0.4	4.2 ± 1.5	2.8 ± 0.8	4.1 ± 0.8	4.3 ± 1.0	1.4 ± 0.7	1.4 ± 0.7
Ore-Paisley Muck	2.2 ± 0.3	2.3 ± 0.3	1.8 ± 0.7	2.9 ± 0.9	5.5 ± 0.8	4.1 ± 1.0	0.20 ± 0.18	0.72 ± 0.37
Ore-Henson's Muck	2.7 ± 0.3	2.7 ± 0.3	2.6 ± 1.0	3.1 ± 0.9	2.0 ± 0.7	2.8 ± 0.9	0.89 ± 0.51	0.95 ± 0.53
Ore-Kincaid Tract	3.2 ± 0.4	3.5 ± 0.5	6.1 ± 2.5	3.4 ± 1.0	3.9 ± 0.8	5.0 ± 1.1	1.6 ± 0.9	1.5 ± 0.8
Ore-Gilbert's	2.7 ± 0.4	2.5 ± 0.4	2.2 ± 0.9	3.0 ± 0.9	5.4 ± 0.8	3.3 ± 0.9	0.58 ± 0.38	0.58 ± 0.38
Ore-Jones Tract	2.5 ± 0.3	2.8 ± 0.4	4.8 ± 1.6	3.2 ± 1.0	4.5 ± 0.8	5.3 ± 1.1	0.23 ± 0.24	1.2 ± 0.6
Average	2.7 ± 0.3	2.7 ± 0.4	3.6 ± 1.7	3.1 ± 0.2	4.2 ± 1.3	4.1 ± 1 0	0.82 ± 0.6	1.1 ± 0.4
Ore Washer Tailings Water (pCi/l)	1.2 ± 0.4	0.69 ± 0.29	2.4 ± 1.1	1.8 ± 0.5	26 ± 22	<11	0.12 ± 0.17	0.12 ± 0.17
Ore Washer Tailings Suspended Solids	3.1 ± 0.4	2.8 ± 0.4	1.3 ± 0.6	3.7 ± 1.1	3.6 ± 0.8	3.6 ± 0.9	0.77 ± 0.45	0.11 ± 0.16
Ore-Florida Rock[b]	39 ± 3	40 ± 3	54 ± 10	50 ± 11	44 ± 1	57 ± 2	0.31 ± 0.20	0.44 ± 0.23
Ore-Florida Stockpile	46 ± 5	47 ± 5	21 ± 7	49 ± 15	56 ± 2	62 ± 3	-0.18 ± 0.13	-0.19 ± 0.22
Florida Ore Average	43 ± 5	44 ± 5	38 ± 23	50 ± 1	50 ± 8	60 ± 4	0.07 ± 0.35	0.13 ± 0.45
Kiln 3 Feed, 1st Shift[b]	3.5 ± 0.3	3.7 ± 0.3	4.1 ± 0.9	3.8 ± 0.8	4.0 ± 0.5	4.8 ± 0.7	0.57 ± 0.25	0.40 ± 0.20
Kiln 3 Feed, 2nd Shift[b]	6.3 ± 0.5	6.5 ± 0.5	7.4 ± 1.6	6.8 ± 1.5	5.0 ± 0.5	7.4 ± 0.9	0.49 ± 0.25	0.68 ± 0.29
Kiln 3 Feed Average	4.9 ± 2.0	5.1 ± 2.0	5.8 ± 2.3	5.3 ± 2.1	4.5 ± 0.7	6.1 ± 1.8	0.53 ± 0.06	0.54 ± 0.20
Nodules[b]	7.5 ± 0.6	7.7 ± 0.6	7.1 ± 1.6	7.7 ± 1.6	7.2 ± 0.6	3.3 ± 1.5	0.57 ± 0.27	0.62 ± 0.30
Craneway Nodules Composite[b]	2.8 ± 0.3	2.9 ± 0.3	4.0 ± 0.9	3.1 ± 0.6	4.6 ± 0.6	3.6 ± 1.4	0.69 ± 0.30	0.72 ± 0.30
Coal - Kiln Fuel	0.49 ± 0.15	0.34 ± 0.12	0.46 ± 0.35	0.39 ± 0.12	1.2 ± 0.6	1.1 ± 1.0	0.23 ± 0.24	0.23 ± 0.24
Silca[b]	0.32 ± 0.1	0.38 ± 0.07	1.1 ± 0.4	0.35 ± 0.08	0.99 ± 0.47	<0.8	0.26 ± 0.15	0.65 ± 0.47
Coke	0.42 ± 0.10	0.47 ± 0.11	0.56 ± 0.27	0.51 ± 0.15	0.91 ± 0.63	<0.8	0.26 ± 0.18	0.32 ± 0.20
Slag[b]	11 ± 1	11 ± 1	22 ± 4	12 ± 3	1.5 ± 0.5	<0.8	0.87 ± 0.33	1.1 ± 0.5

(continued)

TABLE 1(continued)

Sample	Radionuclide Concentrations (pCi/g)[a]							
	U-238	U-234	Th-230	Ra-226	Pb-210	Po-210	Th-232	Th-228
Ferrophosphorus[b]	9.7 ± 1.4	9.8 ± 1.4	0.05 ± 0.07	0.19 ± 0.04	3.9 ± 0.5	1.0 ± 0.9	0.12 ± 0.3	0.21 ± 0.14
Kiln 3 Spray Lime Slurry Dissolved Solids (pCi/l)	2.3 ± 0.8	3.5 ± 0.9	1.1 ± 0.9	4.7 ± 1.4	3.1 ± 6.8	<5	1.4 ± 1.0	1.4 ± 1.0
Kiln 3 Spray Lime Slurry Suspended Solids (pCi/l)	36 ± 13	39 ± 14	28 ± 19	45 ± 13	-20 ± 90	<40	17 ± 15	26 ± 20
Kiln 3 Spray Lime Slurry Total Solids (pCi/l)	38 ± 13	43 ± 14	28 ± 19	50 ± 13	-17 ± 90	<45	18 ± 15	27 ± 20
Kiln 3 Spray Water Dissolved Solids (pCi/l)	1.7 ± 1.0	1.0 ± 1.1	0.56 ± 0.46	1.0 ± 0.5	1.5 ± 12	<5	0.09 ± 19	0.09 ± 0.19
Kiln 3 Spray Water Suspended Solids (pCi/l)	3.1 ± 1.3	3.5 ± 1.4	2.0 ± 1.3	4.0 ± 1.2	130 ± 20	130 ± 35	1.5 ± 1.2	1.5 ± 1.2
Kiln 3 Spray Water Total Solids (pCi/l)	4.8 ± 1.6	5.5 ± 1.8	2.6 ± 1.4	5.0 ± 1.3	130 ± 23	130 ± 35	1.6 ± 1.2	1.6 ± 1.2
Kiln 3 Spray Discharge Dissolved Solids (pCi/l)	1.5 ± 0.8	2.1 ± 0.9	0.82 ± 0.68	0.78 ± 0.23	4.9 ± 6.8	<5	0.14 ± 0.27	0.14 ± 0.27
Kiln 3 Spray Discharge Suspended Solids (pCi/l)	54 ± 9	55 ± 9	24 ± 10	59 ± 18	990 ± 60	1700 ± 170	6.5 ± 4.7	5.2 ± 4.0
Kiln 3 Spray Discharge Total Solids (pCi/l)	56 ± 9	57 ± 9	25 ± 10	60 ± 18	990 ± 60	1700 ± 170	6.6 ± 4.7	5.3 ± 4.0

a) Picocuries (10^{-12} curies) per gram plus or minus twice the standard deviation based on counting statistics. Units in picocuries per liter (pCi/l) where indicated in sample description.

b) The results are derived from duplicate analyses.

Waste water from phossy water processing and kiln spray chamber discharges is recycled through a pond, which serves as a settling pond for suspended solids. This water is used to make lime slurry and spray for the kiln spray chambers. Uranium-234 and -238, thorium-230, and radium-226 concentrations in the spray water averaged 4.5 pCi/l. Lead-210 and polonium-210 concentrations were 130 pCi/l. Lime normally has uranium-234 and -238, thorium-230, and radium-226 concentrations of 0.3 to 0.5 pCi/g and very low concentrations of lead-210 and polonium-210 which are volatilized during the calcining process. This natural radioactivity of lime is reflected in the lime slurry which averaged 40 pCi/l for the non-volatile radionuclides. There is no obvious reason for the difference in lead-210 and polonium-210 concentrations between the kiln spray water and lime slurry samples. Radioactivity concentrations of the kiln spray chamber discharge demonstrate the removal of radioactivity at that point. The average non-volatile radionuclide content was 50 pCi/l, or about 38 pCi/l above the weighted concentration expected from spray water and slurry alone. Lead-210, with 990 ± 60 pCi/l, and polonium-210, with 1,700 ± 170 pCi/l, were the major radionuclides in the discharge.

AMBIENT AIR SAMPLES

Airborne radon concentrations measured at the ambient station are shown in Table 2. Ambient radon concentrations typically vary from less then 0.1 to about 1 nCi/m^3, depending on time of day, season, and meteorology (1). All ambient radon measurements were within the expected normal range.

Ambient airborne particulate radioactivity concentrations are reported in Table 3. All results are within the range expected for normal ambient air (1). Meteorological measurements at the ambient station show that the station was downwind of the plant less than 15 percent of the time during collection of the first particulate sample reported and less than 5 percent of the time for the second sample reported. No effect of plant emissions is apparent on either sample.

EMISSION SAMPLES

Radon

Of four sources sampled for radon only the kiln demister was found to release radon in quantities signficantly different than background. The net concentration above ambient, shown in Table 4, averaged 1.8 ± 0.3 nCi/m^3. On this basis, the total radon released annually from the three kilns was projected to be 9.6 Ci.

TABLE 2. AMBIENT RADON-222 CONCENTRATIONS

Date	Time On - Off	Radon-222 Concentration (nCi/m^3)[a]
12/4	1330 - 1615	0.14 ± 0.06
12/5	1130 - 1330	0.14 ± 0.06
12/6	1030 - 1300	0.08 ± 0.04
12/6	1300 - 1540	0.17 ± 0.04
12/10	1130 - 1400	0.15 ± 0.06
12/10	1400 - 1600	0.16 ± 0.05
12/11	1030 - 1420	0.09 ± 0.04
12/11	1430 - 1600	0.08 ± 0.03
12/12	0915 - 1215	0.60 ± 0.07
12/12	1425 - 1708	0.34 ± 0.05
12/13	0800 - 1115	0.26 ± 0.05
12/13	1115 - 1630	0.27 ± 0.07

a) Nanocuries (10^{-9} curies) per cubic meter plus or minus twice the standard deviation based on counting statistics.

Total Suspended Particulates

Each of the processes sampled generates large quantities of airborne particulates, primarily from comminution of materials during handling. It would be expected, then, that particulate emissions would reflect the radioactivity of the process materials. This was found to be the case, within the limits of sample variability and analytical accuracy, for the non-volatile radionuclides for all sources, with the possible exception of the furnace taphole fume collector. The fume collector emits fumes resulting mainly from the small amounts of phosphorus released and which are very low in radioactivity Stack exhaust gas flow rates and TSP concentrations are given in Table 5.

Kiln Emissions

As shown in Table 3, uranium chain nuclides were in approximate equilibrium, with concentrations of about 0.4 pCi/m^3, except for lead-210 and polonium-210. Polonium was partially volatilized, as shown by its depletion in nodules, and both lead and polonium were nearly completely volatilized in the furnaces and returned to the kiln with the CO fuel. Most of these two nuclides was removed in the spray chamber and demister, but they were still measured at concentrations of 33 to 120 pCi/m^3 for lead-210 and 64 to 280 pCi/m^3 for polonium-210. Emission rates determined from individual samples

TABLE 3. AMBIENT AND STACK PARTICULATE RADIOACTIVITY CONCENTRATIONS

Source	Collected	Radioactivity Concentration (pCi/m^3)[a]							
		U-238	U-234	Th-230	Ra-226	Pb-210	Po-210	Th-232	Th-228
Ambient Station	1600 - 12/06 1830 - 12/07	0.00005 ± 0.00011	-0.00001 ± 0.00016	0.00015 ± 0.00055	-0.00024 ± 0.00056	0.024 ± 0.020	0.010 ± 0.019	-0.00009 ± 0.00029	-0.00006 ± 0.00023
Ambient Station	0910 - 12/12 1030 - 12/14	0.00007 ± 0.00008	0.0 ± 0.0001	0.0002 ± 0.0004	0.00024 ± 0.00032	0.0081 ± 0.0019	0.006 ± 0.003	-0.00004 ± 0.00018	-0.00003 ± 0.00015
Florida Ore, Slag Storage	1440 - 12/07 1915 - 12/08	0.0022 ± 0.0006	0.0053 ± 0.0005	0.0086 ± 0.0016	0.0038 ± 0.0009	0.0032 ± 0.0030	0.0078 ± 0.0042	0.0 ± 0.00036	0.00003 ± 0.00030
Slag Dump	1200 - 12/10 0800 - 12/12	0.00047 ± 0.00016	0.00071 ± 0.00020	0.00077 ± 0.00056	0.00001 ± 0.00035	0.017 ± 0.002	0.0066 ± 0.0038	-0.00005 ± 0.00019	-0.00003 ± 0.00015
Slag Dump	0800 - 12/12 1220 - 12/12	0.0021 ± 0.0010	0.0021 ± 0.0013	0.0025 ± 0.0040	0.0023 ± 0.0038	-0.013 ± 0.018	0.011 ± 0.017	0.0005 ± 0.0024	0.0007 ± 0.0021
No. 3 Kiln Demister[b]	1340 - 1540 12/06	0.39 ± 0.18	0.42 ± 0.21	0.38 ± 0.51	0.40 ± 0.288	110 ± 8	280 ± 15	-0.35 ± 0.32	0.33 ± 0.31
No. 3 Kiln Demister[b]	1720 - 1920 12/06	0.43 ± 0.16	0.90 ± 0.24	0.21 ± 0.40	0.47 ± 0.27	33 ± 3	64 ± 9	0.02 ± 0.25	0.04 ± 0.24
No. 3 Kiln Demister[b]	0952 - 1152 12/07	0.34 ± 0.15	0.41 ± 0.20	0.22 ± 0.39	0.26 ± 0.22	120 ± 5	67 ± 8	0.05 ± 0.24	0.07 ± 0.24
No. 3 Nodule Cooler	1338 - 1643 12/11	0.38 ± 0.34	0.19 ± 0.39	0.09 ± 0.88	-0.12 ± 0.52	-0.2 ± 4.9	0.7 ± 2.1	-0.17 ± 0.53	-0.10 ± 0.51
No. 3 Nodule Cooler	1012 - 1243 12/14	0.42 ± 0.37	0.35 ± 0.45	0.6 ± 1.1	-0.05 ± 0.55	-1.6 ± 5.3	-0.4 ± 2.0	-0.14 ± 0.58	-0.07 ± 0.56
Nodule Transfer Point Exhaust	0922 - 1034 12/06	1.9 ± 0.4	2.0 ± 0.5	2.7 ± 1.3	1.8 ± 0.7	-2.7 ± 5.0	0.6 ± 1.7	0.10 ± 0.56	0.01 ± 0.50
Nodule Transfer Point Exhaust	1208 - 1320 12/06	3.2 ± 0.6	3.5 ± 0.8	5.6 ± 2.4	4.3 ± 1.5	1.1 ± 8.4	2.1 ± 3.2	-0.05 ± 0.85	-1.2 ± 1.0
Nodule Transfer Point Exhaust	1505 - 1617 12/06	0.93 ± 0.350.	0.78 ± 0.41	0.50 ± 0.89	2.0 ± 3.1	-1.0 ± 4.8	0.01 ± 1.7	-0.02 ± 0.52	-1.8 ± 0.9
Screening Plant Dust Collector	1057 - 1240 12/07	3.4 ± 0.4	4.1 ± 0.7	3.7 ± 1.6	4.0 ± 1.4	-3.3 ± 5.3	1.4 ± 2.2	-0.05 ± 0.57	-1.1 ± 0.8
Screening Plant Dust Collector	1408 - 1520 12/07	13 ± 1	13 ± 1	15 ± 5	13 ± 4	9.0 ± 5.3	1.6 ± 5.1	1.6 ± 1.5	1.9 ± 1.6
Screening Plant Dust Collector	0933 - 1045 12/10	1.4 ± 0.5	1.9 ± 1.3	1.0 ± 0.6	0.9 ± 5.5	0.5 ± 2.1	0.5 ± 2.1	0.13 ± 0.63	0.11 ± 0.59
South Scale Room Dust Collector	0913 - 1133 12/04	1.1 ± 0.4	1.6 ± 0.5	1.7 ± 1.0	1.1 ± 0.6	-0.5 ± 3.2	-0.03 ± 1.7	-0.16 ± 0.48	-0.10 ± 0.46
South Scale Room Dust Collector	1522 - 1820 12/04	2.0 ± 0.5	2.2 ± 0.6	2.7 ± 1.3	2.3 ± 1.0	0.7 ± 5.8	-1.0 ± 2.1	-0.10 ± 0.64	-0.03 ± 0.59
South Scale Room Dust Collector	1524 - 1822 12/04	1.4 ± 0.4	1.8 ± 0.5	1.4 ± 0.9	1.2 ± 0.6	-3.5 ± 4.9	0.3 ± 1.8	0.13 ± 0.52	0.19 ± 0.50
South Scale Room Dust Collector	0928 - 1202 12/05	2.0 ± 0.5	1.8 ± 0.6	1.0 ± 1.2	2.0 ± 0.9	-1.3 ± 5.7	-0.4 ± 2.3	-0.21 ± 0.64	-0.13 ± 0.55

(continued)

TABLE 3. (Continued)

Source	Collected	Radioactivity Concentration (pCi/m^3)[a]							
		U-238	U-234	Th-230	Ra-226	Pb-210	Po-210	Th-232	Th-228
Stocking System Dust Collector	1040 - 1309 12/04	0.80 ± 0.29	0.97 ± 0.34	1.4 ± 1.5	0.57 ± 0.43	-0.8 ± 3.5	-0.2 ± 1.4	0.02 ± 0.40	0.04 ± 0.37
Stocking System Dust Collector	1453 - 1705 12/04	0.95 ± 0.31	0.87 ± 0.36	0.97 ± 0.71	0.81 ± 0.46	0.1 ± 2.6	-0.7 ± 1.3	-0.02 ± 0.39	0.03 ± 0.38
Stocking System Dust Collector	0905 - 1117 12/05	0.66 ± 0.28	0.98 ± 0.33	0.55 ± 0.67	0.67 ± 0.43	0.7 ± 4.3	-0.4 ± 1.3	0.06 ± 0.40	0.11 ± 0.39
Central Furnace Stocking System	1351 - 1609 12/06	0.51 ± 0.21	0.65 ± 0.27	0.83 ± 0.60	0.59 ± 0.36	-1.4 ± 3.0	-0.2 ± 1.1	0.07 ± 0.34	0.11 ± 0.32
Central Furnace Stocking System	0858 - 1145 12/07	0.90 ± 0.27	0.80 ± 0.31	1.9 ± 0.7	0.78 ± 0.43	0.5 ± 3.3	0.3 ± 1.3	0.11 ± 0.36	0.15 ± 0.35
Central Furnace Stocking System	1208 - 1546 12/07	0.93 ± 0.29	0.73 ± 0.30	0.47 ± 0.56	0.47 ± 0.35	-0.3 ± 3.0	0.2 ± 1.2	0.06 ± 0.34	0.10 ± 0.33
Furnace Tapehole Fume Collector	1355 - 1545 12.04	-0.24 ± 0.32	-0.10 ± 0.42	0.03 ± 0.93	-0.04 ± 0.54	-3.0 ± 5.8	0.6 ± 2.2	-0.14 ± 0.57	-0.08 ± 0.55
Furnace Tapehole Fume Collector	1356 -1546 12/04	-0.15 ± 0.36	0.05 ± 0.42	-0 ± 1.0	-0.10 ± 0.58	2.4 ± 6.9	1.5 ± 2.5	0.66 ± 0.69	-1.2 ± 0.8
Furnace Tapehole Fume Collector	0844 - 1508	-0.001 ± 0.17	-0.06 ± 0.20	-0.10 ± 0.44	-0.09 ± 0.26	15 ± 3	0.001 ± 0.38	-0.04 ± 0.29	0.04 ± 0.26
Furnace Tapehole Fume Collector	1525 - 1808 12/05	-0.06 ± 0.16	-0.06 ± 0.20	0.10 ± 0.47	-0.04 ± 0.26	3.2 ± 3.1	1.1 ± 1.2	-0.07 ± 0.28	-0.53 ± 0.38

a) Picocuries (10^{-12} curies) per actual cubic meter (volume measured at ambient or stack conditions) plus or minus twice the standard deviation based on counting results only.

b) Picocuries per dry standard cubic meter (volume of dry air sampled at 21 C, 760 mm Hg).

c) Duplicate sample.

TABLE 4. RADON-222 STACK EMISSIONS

Source	Time	Date	Concentration (nCi/m^3)[a] Gross	Net	Annual Emissions (Ci/y)[b]
No. 3 Kiln Demister	1500-1600	12/6	2.0 ± 0.1	1.8 ± 0.1	
	1335-1615	12/12	1.7 ± 0.1	1.4 ± 0.1	
	0825-1045	12/13	2.1 ± 0.1	2.0 ± 0.1	
	1050-1500	12/13	2.1 ± 0.1	1.8 ± 0.1	
Source Average				1.8 ± 0.3	9.6[d]
No. 3 Kiln Nodule Cooler	1330-1630	12/11	0.13 ± 0.06	0.05 ± 0.07	
	0820-0930	12/12	0.09 ± 0.05	-0.51 ± 0.09	
	0815-1100	12/13	0.32 ± 0.07	0.06 ± 0.09	
	1050-1330	12/13	0.16 ± 0.04	-0.11 ± 0.08	
	1325-1452	12/13	0.26 ± 0.06	-0.01 ± 0.09	
Source Average				-0.10 ± 0.24	-0.19[d]
Screening Plant Dust	0910-1200	12/10[c]	0.30 ± 0.04	0.15 ± 0.07	
	1330-1530	12/10	0.24 ± 0.05	0.08 ± 0.07	
	0940-1400	12/11	0.08 ± 0.04	-0.01 ± 0.06	
	1100-1320	12/12[c]	0.15 ± 0.03	-0.44 ± 0.08	
	0830-1055	12/13	0.30 ± 0.06	0.04 ± 0.08	
Source Average				-0.04 ± 0.23	-0.04[e]
No. 3 Furnace Taphole Fume Collector	0830-1500	12/15[c]	0.16 ± 0.03	0.02 ± 0.07	0.05[f]

a) Nanocuries (10^{-9} curies) per cubic meter plus or minus twice the standard deviation based on counting statistics. Source average uncertainties are calculated from the variance about the mean of the samples.

b) Calculated assuming 24 hour continuous operation for 50 weeks per year.

c) The results derived from duplicate Samples.

d) Annual emissions are for sum of three kilns.

e) Annual emission is for sum of two stacks.

f) Annual emission is for sum of six furnaces.

are shown in Table 6. Mass emission rates measured from the three samples were quite uniform. This uniformity is also evident in the radioactivity emission rates of the non-volatile nuclides. Lead-210 and polonium-210 emission rates varied by factors of 3 to 4. The reason for this is unknown, but it is probably due to the fact that lead and polonium, as vapors or fumes, are not removed from the exhaust stream as consistently as the particulates. The annual particulate radioactivity emission rates, calculated for the total number of stacks from each type of source are shown in Table 7. Calculated emissions of uranium, thorium-230, and radium-226 from the three kilns averaged 2.2 mCi/y. The annual emission of lead-210 was 480 mCi and that for polonium-210 was 750 mCi.

Nodule Cooler Emissions

Radioactivity concentrations in the nodule cooler exhaust were essentially the same as for the non-volatile nuclides in the kiln demister. The overlapping confidence intervals for all nuclides and the mechanical nature of the particulate generation suggest that the concentrations and emission rates for all nuclides released from the coolers are approximately equal. The annual emission rates of about 1 mCi/y for uranium-234 and -238 and thorium-230 probably hold for the other nuclides as well. Polonium-210 may actually be close to the 0.6 mCi/y emission rate due to some depletion in the kiln. Simultaneous sampling of the inlet to and outlet from the nodule cooler scrubber showed an average removal of uranium chain nuclides of 99.5 percent. Mass analysis of the filters showed an average mass removal efficiency of 98.5 percent.

Nodule Transfer Point Emissions

Concentrations of non-volatile nuclides averaged from about 1.0 to 5.6 pCi/m^3 during the three sampling periods (Table 3). Visible emissions varied noticeably, with the controlling factor seeming to be the temperature of nodules on the conveyor. At times the nodules were still glowing and generated larger quantities of emissions at the transfer points. Non-volatile radionuclide emissions averaged 1.5 mCi/y (Table 7). Polonium-210 was lower most likely due to depletion in the kilns.

TABLE 5. STACK FLOW AND PARTICULATE EMISSION RATES

Source	Collected Time	Collected Date	Exhaust Gas Flow Rate (m^3/min)	Particulate Concentration (mg/m^3)[a]
No. 3 Kiln Demister	1340-1540	12/6	3,399	137.4[b]
	1720-1920	12/6	3,554	114.4[b]
	0952-1152	12/7	3,554	114.4[b]
No. 3 Nodule Cooler	1338-1643	12/11	1.657	68.3
	1012-1242	12/14	1,571	64.6
Nodule Transfer Point Exhaust	0922-1034	12/6	1,223	215.6
	1208-1320	12/6	1,118	437.8
	1505-1617	12/6	1,224	247.4
Screening Plant Dust Collector	1057-1240	12/7	1,105	706.3
	1408-1520	12/7	1,091	1,664
	0933-1045	12/10	1,104	176.1
South Scale Room Dust Collector	1913-1133	12/4	605.1	189.1
	1522-1820[c]	12/4	606.3	207.1
	1524-1822[c]	12/4	622.7	225.8
	0928-1202	12/5	610.1	198.3
Stocking System Dust Collector	1040-1309	12/4	394.8	135.6
	1453-1705	12/4	389.1	108.9
	0905-1117	12/5	386.8	180.4
Central Furnace Stocking System	1351-1609	12/6	578.1	179.1
	0858-1145	12/7	536.1	248.6
	1208-1546	12/7	573.2	168.5
Furnace Taphole Fume Collector	1355-1545[c]	12/4	823.9	17.9
	1356-1546[c]	12/4	787.1	21.6
	0844-1508	12/5	887.3	35.9
	1525-1808	12/15	840.3	15.6

a) Flow rate and concentration volumes are actual cubic meters at stack conditions.

b) No. 3 kiln volumes are dry, standard cubic meters (at 20°C, 760 mm mercury pressure.)

c) Duplicate samples.

TABLE 6. PARTICULATE RADIOACTIVITY ANNUAL EMISSION RATES

Source	Time-Date Collected	Radioactivity Emission Rates (pCi/s)[a] U-238	U-234	Th-230	Ra-226	Pb-210	Po-210	Th-232	Th-228
No. 3 Kiln Demister	1340 - 1540 12/06	22 ± 10	24 ± 12	21 ± 29	23 ± 16	6300 ± 400	16000 ± 900	-20 ± 18	18 ± 18
	1720 - 1920 12/06	26 ± 10	54 ± 14	12 ± 24	28 ± 16	1900 ± 200	3800 ± 500	1 ± 15	2 ± 14
	0951 - 1152 12/07	20 ± 9	24 ± 12	13 ± 23	16 ± 13	7000 ± 300	4000 ± 500	3 ± 14	4 ± 14
Source Average		23 ± 3	34 ± 17	15 ± 5	22 ± 6	5100 ± 2800	7900 ± 7000	-5 ± 13	8 ± 9
No. 3 Nodule Cooler	1338 - 1643 12/11	10 ± 9	5 ± 11	2 ± 24	-3 ± 14	6 ± 140	20 ± 580	-5 ± 15	3 ± 14
	1012 - 1242 12/14	11 ± 10	9 ± 12	16 ± 29	-1 ± 14	-40 ± 140	-9 ± 52	-4 ± 15	-2 ± 15
Source Average		11 ± 1	7 ± 3	9 ± 10	-2 ± 1	-17 ± 33	6 ± 21	-5 ± 1	1 ± 4
Nodule Transfer Point Exhaust	0922 - 1034 12/06	39 ± 9	41 ± 11	56 ± 26	36 ± 15	-60 ± 100	13 ± 34	2 ± 11	0 ± 10
	1208 - 1320 12/06	59 ± 11	65 ± 15	100 ± 50	80 ± 29	20 ± 160	39 ± 60	-1 ± 16	-22 ± 19
	1505 - 1617 12/06	19 ± 7	16 ± 8	10 ± 18	41 ± 63	-20 ± 98	0 ± 35	0 ± 11	-36 ± 18
Source Average		39 ± 20	41 ± 25	55 ± 45	52 ± 24	-20 ± 40	17 ± 20	0 ± 2	-19 ± 18
Screening Plant Dust Collector	1057 - 1240 12/07	63 ± 7	75 ± 13	68 ± 29	73 ± 25	-61 ± 97	26 ± 40	-1 ± 10	-20 ± 14
	1408 - 1520 12/07	230 ± 20	230 ± 20	280 ± 90	240 ± 80	160 ± 100	28 ± 92	28 ± 27	34 ± 28
	0933 - 1045 12/10	25 ± 9	29 ± 10	35 ± 23	18 ± 11	20 ± 100	10 ± 38	2 ± 12	2 ± 11
Source Average		110 ± 110	110 ± 110	130 ± 130	110 ± 120	40 ± 110	21 ± 10	10 ± 16	5 ± 27
South Scale Room Dust Collector	0913 - 1133 12/04	11 ± 4	16 ± 5	17 ± 10	11 ± 6	-5 ± 32	-3 ± 17	-2 ± 5	-1 ± 5
	1522 - 1820[b] 12/04	18 ± 3	22 ± 4	21 ± 8	18 ± 6	-14 ± 39	-3 ± 14	0 ± 4	1 ± 4
	0928 - 1202 12/05	20 ± 5	18 ± 6	10 ± 12	20 ± 9	-13 ± 57	-4 ± 23	-2 ± 6	-1 ± 6
Source Average		16 ± 5	18 ± 3	16 ± 6	16 ± 5	-11 ± 5	-3 ± 1	-1 ± 1	0 ± 1
Stocking System Dust Collector	1040 - 1309 12/04	5.3 ± 1.9	6.4 ± 2.2	9.2 ± 9.9	3.8 ± 2.8	-6 ± 23	-1.6 ± 9.2	0.1 ± 2.6	0.3 ± 2.4
	1453 - 1705 12/04	6.2 ± 2.0	5.6 ± 2.3	6.3 ± 4.6	5.3 ± 3.0	1 ± 17	-4.9 ± 8.4	-0.1 ± 2.5	0.2 ± 2.5
	1905 - 1117 12/05	4.3 ± 1.8	6.3 ± 2.1	3.5 ± 4.3	4.3 ± 2.8	-5 ± 28	-2.3 ± 8.4	0.4 ± 2.6	0.7 ± 2.5
Source Average		5.3 ± 1.0	6.1 ± 0.4	6.3 ± 2.9	4.5 ± 0.8	-3.3 ± 3.8	-2.9 ± 1.7	0.1 ± 0.3	0.4 ± 0.3
Central Furnace Stocking System	1351 - 1609 12/06	4.9 ± 2.0	6.3 ± 2.6	8.0 ± 5.8	5.7 ± 3.5	-13 ± 29	-2.0 ± 11	0.7 ± 3.3	1.1 ± 3.1
	0858 - 1145 12/07	8.0 ± 2.4	7.1 ± 2.8	17 ± 6	7.0 ± 3.8	5 ± 29	3.0 ± 12	1.0 ± 3.2	1.3 ± 3.1
	1208 - 1546 12/07	8.9 ± 2.8	7.0 ± 2.9	4.5 ± 5.3	4.5 ± 3.3	-3 ± 29	2.0 ± 11	0.6 ± 3.2	1.0 ± 3.2
Source Average		7.3 ± 2.1	6.8 ± 0.4	10 ± 6	5.7 ± 1.3	-3.7 ± 9.0	1.0 ± 2.7	0.8 ± 0.2	1.1 ± 0.2

(continued)

TABLE 6. (Continued)

Source	Time-Date Collected	Radioactivity Emission Rates (pCi/s)[a]							
		U-238	U-234	Th-230	Ra-226	Pb-210	Po-210	Th-232	Th-228
Furnace Taphole	1355 - 1545 12/04	-2.7 ± 3.2	1.1 ± 4.0	-2.5 ± 9.2	-0.5 ± 5.1	-40 ± 55	8 ± 21	-0.1 ± 5.4	-0.1 ± 5.2
Fume Collector	0844 - 1508 12/05	0.0 ± 2.5	-0.5 ± 3.0	-1.5 ± 6.5	-1.4 ± 3.9	230 ± 50	0.0 ± 5.7	-0.5 ± 4.2	0.6 ± 3.9
	1525 - 1808 12/05	-0.9 ± 2.3	-0.9 ± 2.8	1.4 ± 6.5	0.5 ± 3.7	44 ± 43	5 ± 17	-1.0 ± 3.9	-7.4 ± 5.3
Source Average		-1.2 ± 1.4	-0.1 ± 1.1	-0.9 ± 2.0	-0.5 ± 1.0	78 ± 140	4.3 ± 4.0	-0.5 ± 0.6	-2.3 ± 4.4

a) Picocuries (10^{-12} curies) per second plus or minus the standard deviation based upon counting statistics. Source average uncertainties are calculated from the variance about the mean of the samples.

b) Derived from duplicate samples.

c) Exhaust fans were shut down several times.

TABLE 7. PARTICULATE RADIOACTIVITY ANNUAL EMISSION RATES

Source	Radioactivity Emission Rates (mCi/y)[a]							
	U-238	U-234	Th-230	Ra-226	Pb-210	Po-210	Th-232	Th-228
Kiln Demisters	2.2	3.2	1.4	2.1	480	750	-0.5	0.
Nodule Coolers	1.0	1.0	0.9	-0.2	-1.6	0.6	-0.4	0.1
Nodule Transfer Point Exhaust	1.2	1.3	1.7	1.6	-0.6	0.5	0	0.6
Screening Plant Dust Collector	6.9	6.9	8.2	6.9	1.9	1.3	0.6	0.3
South Scale Room Dust Collector	0.50	0.56	0.50	-0.50	-0.35	-0.09	-0.03	0.00
Stocking System Dust Collector	0.33	0.38	0.40	0.28	-0.21	-0.18	0.006	0.025
Central Furnace Stocking System	0.23	0.21	0.32	0.18	-0.12	0.032	0.025	0.035
Furnace Taphole Fume Collector	-0.32	0.02	-0.13	-0.15	15	1.4	-0.095	-0.44
Plant Total	12	14	13	11	490	750	-0.39	1.4

a) Millicuries (10^{-3} curies) per year. Calculate using concentrations found in Table 3 and emission rates from Table 5.

Screening Plant Dust Collector Emissions

As shown in Table 5, the TSP concentrations in emissions from the screening plant dust collector were the highest of any source, and also the most variable. TSP and radioactivity concentrations varied by an order of magnitude over the three sampling periods. This was the most significant source of non-volatile particulate radioactivity emissions from the controlled sources. Emissions from the two stacks accounted for more than half of the plant total.

Other Emission Sources

Emissions from each of the other sources, except the furnace taphole fume collector, had concentrations similar to the preceding sources. Stack flow rates, however, were lower and emissions of non-volatile radionuclides were less than 1 mCi/y for all these sources.

Lead-210 emission rates from the furnace taphole fume scrubber were highly variable, ranging from -40 ± 55 pCi/s to 230 ± 50 pCi/s in the four samples collected (Table 6). It had been shown in the earlier report that the primary source of lead-210 from furnace tapping is FeP tapping. It is probable that the highest lead-210 sample included an FeP tap. The polonium-210 annual average emission rate from furnace tapping was 1.4 mCi.

Particle Size Analysis

Size-fractionated samples were collected from five stacks using an in-stack cascade impactor sampler. Two samples were collected from each source. Sources sampled were the screening plant dust collector, south scale room dust collector, stocking system dust collector, central furnace stocking system, and furnace taphole fume collector. Most of the impactor stages had such a small mass of collected particulates that radiological analysis was not attempted. Considering the sources of particulates collected it is reasonable to assume that the size distribution of non-volatile radionuclides is similar to the mass distribution.

The majority of mass from each source was found on the first impactor stage. Cumulative mass analyses showed the following results for aerodynamic particulate sizes.

TABLE 8. AERODYNAMIC PARTICLE SIZE DISTRIBUTIONS

Source	Run 1 Size(μm)	Run 1 less than	Run 2 Size(μm)	Run 2 less than
Screening plant	14.1	8.7	16.2	8.9
South scale room	17.7	34	17.7	27
Stocking system	14.6	6.8	14.2	48
Central furnace stocking system	Invalid Sample		12.4	6.9
Taphole fume scrubber	12.3	18	12.1	28

High Volume Particulate Samples

Problems were encountered in collecting the high volume particulate samples at the slag dump and Florida ore storage areas. Samplers were unplugged by workers needing electrical outlets or power was accidentally cut to the circuits on several occasions. As a result, sample volumes were smaller than desired and analytical uncertainties were higher. The results are shown in Table 3. When compared to the ambient station high volume samples, the plant site samples show generally higher concentrations with the highest being measured at the rail car dump site adjacent to the Florida ore storage pile. While it is logical to ascribe these higher activities to the slag dumping and ore handling activities no importance can be laid on them because of the relatively uncontrolled sampling performed.

SECTION 7

DISCUSSION OF RESULTS

As was expected, several sources of enhanced radioactivity emissions were found at this elemental phosphorus plant. The gaseous and volatile radionuclides - radon, polonium-210, and lead-210 - were driven off the process materials in either the kilns or furnaces and ultimately exhausted to the atmosphere via the kiln exhausts. These radionuclides were also those released in greatest quantity. Annual radon emissions from the kilns were measured at 9.6 Ci/y. Although the kiln emission control system was not designed to remove lead and polonium fumes most of them were removed and annual emission rates were 0.48 Ci for lead-210 and 0.75 Ci for polonium-210, about an order of magnitude below the radon.

The screening plant dust collector emissions were found to account for more than half of the non-volatile radioactive nuclides emitted from the plant's controlled sources. The mass emission rates varied from 11.7 to 109 kg/h, compared to a rate of 2.9 kg/h reported by Monsanto. Mass emission rates measured at the other sources were generally about twice as much as reported by Monsanto 2 years earlier. The large amount of particulates from the screening plant dust collector, and their greatly varying quantity suggest that the emission control system was not operating properly during the survey. If the average observed mass emission rate of 56 kg/h was reduced to 2.9 kg/h the average non volatile radionuclide emission rates would be reduced from 7.2 mCi/y to 0.36 mCi/y. That would put it in line with the other material handling processes following the nodule transfer point control system and would reduce the plant emissions of the non-volatile nuclides by about one half.

If the screening plant dust collector was not operating at design efficiency, correcting the problem would reduce mass emissions, but would probably not reduce the respirable fraction by the same proportion. As shown before only 8.8 percent, on the average, of the mass emissions were below 14.1 to 16.2 μm aerodynamic diameter. Large particles would normally be removed with more efficiency than small particles. This is shown to some degree by the size distributions found for the other sources measured.

The two major sources of uncontrolled emissions, materials handling in the kiln-nodule cooler area and open nodule craneway storage area, produce primarily large particles that are deposited in the immediate area. Very little airborne material was visibly evident at any appreciable distance from these sources.

REFERENCES

1. National Council on Radiation Protection and Measurements. Natural Background Radiation in the United States, NCRP Report No. 45, Washington, D.C., 1975.

2. Habashi, Fathi. Uranium in phosphate rock. Special Publication 52, Montana Bureau of Mines and Geology, Montana College of Mineral Science and Technology, Butte, Montana, December 1970.

3. Menzel, F.G. Uranium, radium, and thorium content in phosphate rocks and their possible radiation hazards. Journal of Agriculture and Food Chemistry, Vol. 16, No. 2, pp. 231-234, 1968.

4. Eadie, Greogry G., and David E. Bernhardt. Radiological surveys of Idaho phosphate ore processing - the thermal process plant. U.S. Environmental Protection Agency, Technical Note OPR/LV-77-3, Las Vegas, Nevada, November 1977.

5. Engineering-Science, Inc. Collection of airborne radon and radioactive particulates at the Monsanto Chemical Intermediates Columbia Elemental Phosphorus Plant Columbia, Tennessee. McLean, Virginia, April 1980.

6. Code of Federal Regulations, Title 40, Chapter 1, Part 60, Appendix A.

7. Code of Federal Regulations, Title 40, Chapter 1, Part 50, Appendix B.

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16. ABSTRACT
Naturally occurring radioactivity was measured in the atmospheric emissions and process materials of a thermal phosphate (elemental phosphorus) plant. Representative exhaust stack samples were collected from each process in the plant. The phosphate ore contained 12 to 20 parts per million uranium. Processes, emission points, and emission controls are described. Radioactivity concentrations and emission rates from the sources sampled are given.

17. KEY WORDS AND DOCUMENT ANALYSIS

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