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Idaho Radionuclide Exposure Study—Literature Review

E. G. Baker
H. D. Freeman
J. N. Hartley

October 1987

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IDAHO RADIONUCLIDE EXPOSURE STUDY--
LITERATURE REVIEW

E. G. Baker
H. O. Freeman
J. N. Hartley

October 1987

Prepared for
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Office of Radiation Programs
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SUMMARY

Phosphate ores contain elevated levels of natural radioactivity, some of which is released to the environment during processing or use of solid byproducts. In the Western states, most of the phosphate rock is produced in the State of Idaho. Four phosphate processing plants are located in southeastern Idaho. Two thermal process plants, one located near Soda Springs (Monsanto Co.) and the other near Pocatello (FMC Corporation), produce elemental phosphorus. A wet process plant (J. R. Simplot Co.) producing phosphoric acid and a variety of fertilizers is in operation near Pocatello. A second wet process plant (Beker Industries) located in Conda near Soda Springs is currently shut down.

The effect of radionuclides from Idaho phosphate processing operations on the local communities has been the subject of much research and study. The literature is reviewed in this report. Two primary radionuclide pathways to the environment have been studied in detail:

- airborne release of volatile radionuclides, primarily ^{210}Po, from calciner stacks at the two elemental phosphorus plants
- use of byproduct slag as an aggregate for construction in Soda Springs and Pocatello.

The health impact of both pathways has been estimated by the U.S. Environmental Protection Agency (EPA) (^{210}Po) and the Idaho Department of Health and Welfare (slag). The EPA has issued a standard of 21 Ci/year of ^{210}Po emissions from calciner stacks. Since 1977, the State of Idaho has forbidden use of slag as a construction material in habitable structures; it is still used for other construction activities, mainly in road construction and for railroad ballast.

Despite the research, there is still no clear understanding of the population dose from radionuclide emissions, effluents, and solid wastes from phosphate processing plants. Two other potential radionuclide pathways to the environment have been identified: radon exhalation from phosphogypsum and ore piles and contamination of surface and ground waters. Recommendations on further study needed to develop a data base for a complete risk assessment are given in the report.

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CONTENTS

FIGURES

TABLES

1.0 INTRODUCTION

Phosphate ores contain approximately 60 times the levels of natural radioactivity normally found in the earth's crust. Some of the radioactivity is released to air and water during processing of the ores, and some is distributed in the environment through the use of solid byproduct wastes. In 1979, the U.S. Environmental Protection Agency (EPA) listed radionuclides as hazardous air pollutants and was further required by the Clean Air Act to issue emission standards. In February 1985, the EPA issued a radionuclide standard of 21 Ci/year of ^{210}Po air emission per elemental phosphorus plant under orders of the North District of California, U.S. District Court. This standard was based on partial EPA studies satisfying the court orders for regulation under Section 112 of the Clean Air Act.

The communities of Soda Springs and Pocatello, Idaho, are immediately adjacent to elemental phosphorus and phosphate fertilizer plants containing stockpiles of slag, phosphogypsum,(a) and phosphate ore. Furthermore, slag containing elevated concentrations of naturally occurring radionuclides has been used as an aggregate material for paving and building throughout these communities.

This report is the first phase of a program to evaluate the impact of radioactive air emissions, water effluent, and solid wastes from the phosphorus processing plants in southeastern Idaho. The objective of this literature review is to identify radionuclide source terms and pathways to the environment from phosphate processing plants in Idaho and to determine additional information needed to assess the total radionuclide exposure from these plants and from natural sources in the area.

Radionuclides present in phosphate ore are in the ^{238}U and ^{232}Th decay series. Members of these series are shown in Appendix A. Emphasis in the research to date has been on ^{210}Po, a volatile radionuclide in the ^{238}U series, which is known to be emitted from elemental phosphorus plants. However, other members are also of concern. A variety of terms dealing with radioactivity,

(a) Solid byproduct of the wet process; primarily calcium sulfate.

radionuclides, and phosphate processing are used throughout this report. A glossary of terms and abbreviations is included as Appendix B. Appendix C is a bibliography of literature reviewed during the preparation of this report.

2.0 IDAHO PHOSPHATE INOUSTRY

The phosphate industry in Idaho is an integrated operation from mining to production of phosphorus, phosphoric acid, and fertilizers. The locations of phosphate operations in Idaho are shown in Figure 2.1.

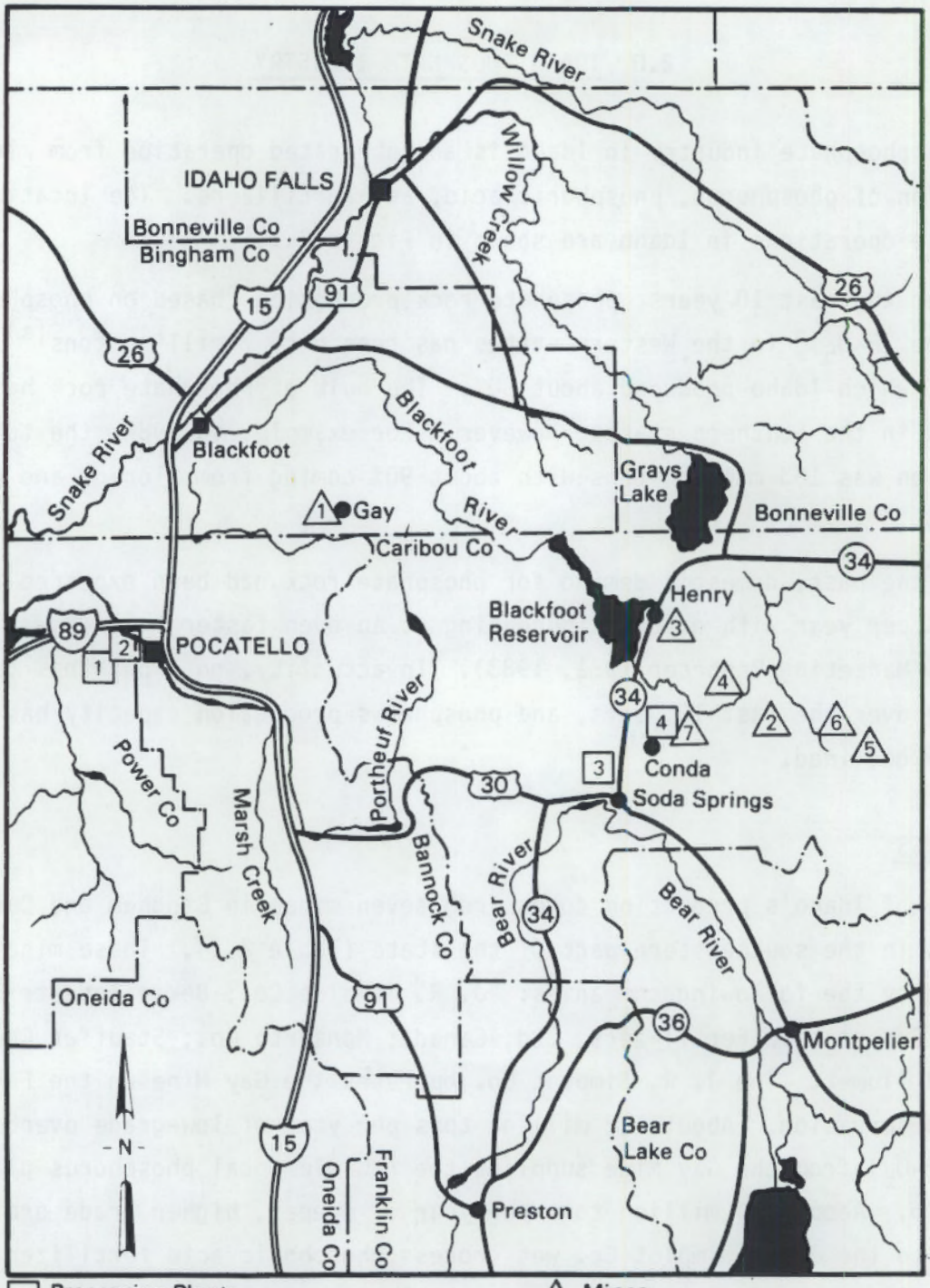
Over the past 10 years, phosphate rock production [based on phosphorus pentoxide (P_2O_5)] in the Western states has been 6 to 7 million tons[(a)] per year, of which Idaho produced about 80%. The bulk of phosphate rock has been produced in the Southern states, however. For example, in 1984, the total U.S. production was 163 million tons with about 90% coming from Florida and North Carolina.

In the past, domestic demand for phosphate rock had been expected to grow 1% to 3% per year with exports increasing at an even faster rate (Powell 1974; Chemical Marketing Reporter 1981, 1983). In actuality, no growth has been observed over the last 10 years, and phosphorus production capacity has actually declined.

2.1 MINES

All of Idaho's production comes from seven mines in Bingham and Caribou counties in the southeastern part of the state (Table 2.1). These mines are operated by the following companies: J. R. Simplot Co.; Beker Industries; Western Cooperative Fertilizers, Ltd, Canada; Monsanto Co.; Stauffer Chemical Co.; and Alumet. The J. R. Simplot Co. operates the Gay Mine on the Fort Hall Indian Reservation. About 1.2 million tons per year of low-grade overburden (24.5% P_2O_5) from the Gay Mine supplies the FMC elemental phosphorus plant at Pocatello. About 0.4 million tons per year of deeper, higher grade ore are shipped to the J. R. Simplot Co. wet process phosphoric acid fertilizer complex in Pocatello. The J. R. Simplot Co. recently began production at the Smoky Canyon Mine, which replaced the nearly exhausted Woodall Peak Mine. The ore goes to the wet process plants at Pocatello and California.

(a) Unless indicated otherwise, "tons" refers to metric tons.



Processing Plants

1 FMC Elemental Phosphorus Plant
2 JR Simplot Wet Process Fertilizer Complex
3 Monsanto Elemental Phosphorus Plant
4 Beker Wet Process Fertilizer Complex

Mines

1 Gay (Simplot)
2 Dry Valley (Beker)
3 Henry (Monsanto)
4 Wooley Valley (Stauffer)
5 Smoky Canyon (Simplot)
6 Diamond Creek (Alumet)
7 Conda Woodall Peak (Simplot)

FIGURE 2.1. Idaho Phosphate Operations

TABLE 2.1. Phosphate Rock Mines in Idaho

Mine	Location	Operator	Production Capacity 10^6 TPY(a)	Destination
Gay	Fort Hall	J. R. Simplot Co.	1.6	FMC, Pocatello; J. R. Simplot Co., Pocatello
Dry Valley	NE of Conda	Beker Industries Western Cooperative	1.8	Beker Industries, Conda; Alberta, Canada
Henry	N of Soda Springs	Monsanto	0.8	Monsanto, Soda Springs
Wooley Valley	NE of Soda Springs	Stauffer	0.7	Stauffer, Silver Bow, Montana
Smoky Canyon	30 miles east of Conda	J. R. Simplot Co.	--	J. R. Simplot Co., Pocatello California
Diamond Creek	NE of Soda Springs	Alumet	--	--
Woodall Peak	Conda	J. R. Simplot Co.	--	--

(a) Tons per year.

The Dry Valley Mine northeast of Conda is operated by the Conda Partnership, a 50/50 association between Beker Industries and Western Cooperative Fertilizers, LTD, Canada. Beker Industries' share of the mined rock was used in its adjacent wet process fertilizer complex at Conda; this plant is currently shut down. Western Cooperative's share is exported to Alberta.

Monsanto obtains phosphate rock for its elemental phosphorus plant in Soda Springs from the Henry Mine near the village of Henry north of Soda Springs. Stauffer operates the Wooley Valley Mine northeast of Soda Springs to produce rock for its elemental phosphorus plant in Silver Bow, Montana.

2.2 PROCESSING PLANTS

The four phosphate processing plants in Idaho and their capacities are listed in Table 2.2. Two processing plants are located near Pocatello (Figure 2.2). The city is triangular-shaped and nestled in a large draw in the Pocatello Range at an altitude of 4500 ft. It is bounded on the west by adjacent mountains and on the east by adjacent mountains with a more gentle rise. The town of Chubbock, a suburb of Pocatello, lies to the northwest on flatlands. The FMC elemental phosphorus plant and the J. R. Simplot Co. fertilizer plants are located about 3 miles northwest of Pocatello at an elevation of 4500 ft at the base of the mountain that bounds the west side of the city. Phosphogypsum waste and slag are deposited on the mountain side about 150 ft above the plants.

The Monsanto elemental phosphorus plant (Figure 2.3) and the Beker Industries wet process fertilizer complex are located near Soda Springs. The city of Soda Springs is adjacent to the Bear River at an altitude of 6000 ft. It is bounded by Chester Hill (6500 ft) to the north-northwest and Rabbit Mountain (6700 ft) 1.5 miles to the east-northeast. The elemental phosphorus plant is located 1.5 miles north-northwest of town at an altitude of 6000 ft. The fertilizer plant at Conda (currently shut down) is located 6 miles northeast of Soda Springs in the foothills of the Aspen Range at an altitude of 6200 ft.

TABLE 2.2. Phosphate Processing Plants in Idaho

Company	Location	Type of Process	Production Capacity, TPY[a]	
Beker Industries[b]	Conda	Wet Process	270,000	Phosphoric Acid
			64,000	Superphosphoric Acid
J. R. Simplot Co.	Pocatello	Wet Process	326,000	Phosphoric Acid
			75,000	Superphosphoric Acid
FMC	Pocatello	Elemental	137,000	
Monsanto	Soda Springs	Elemental	95,000	

(a) (Metric) tons per year; based on P_2O_5 for wet process plants and P_4 for elemental phosphorus plants.
(b) Currently shut down.

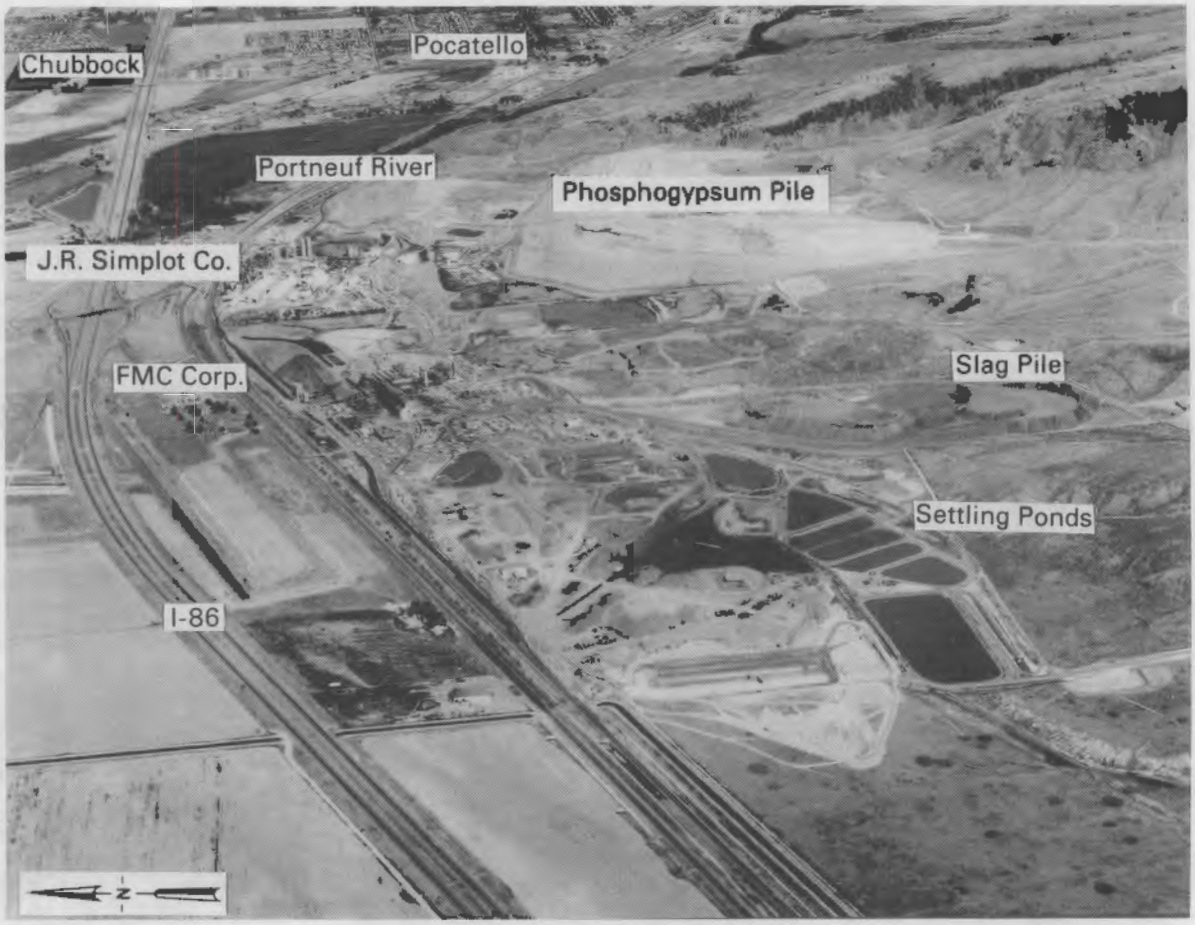


FIGURE 2.2. Aerial View of the Pocatello Area Showing the J. R. Simplot Co. Fertilizer Complex and the FMC Elemental Phosphorus Plant

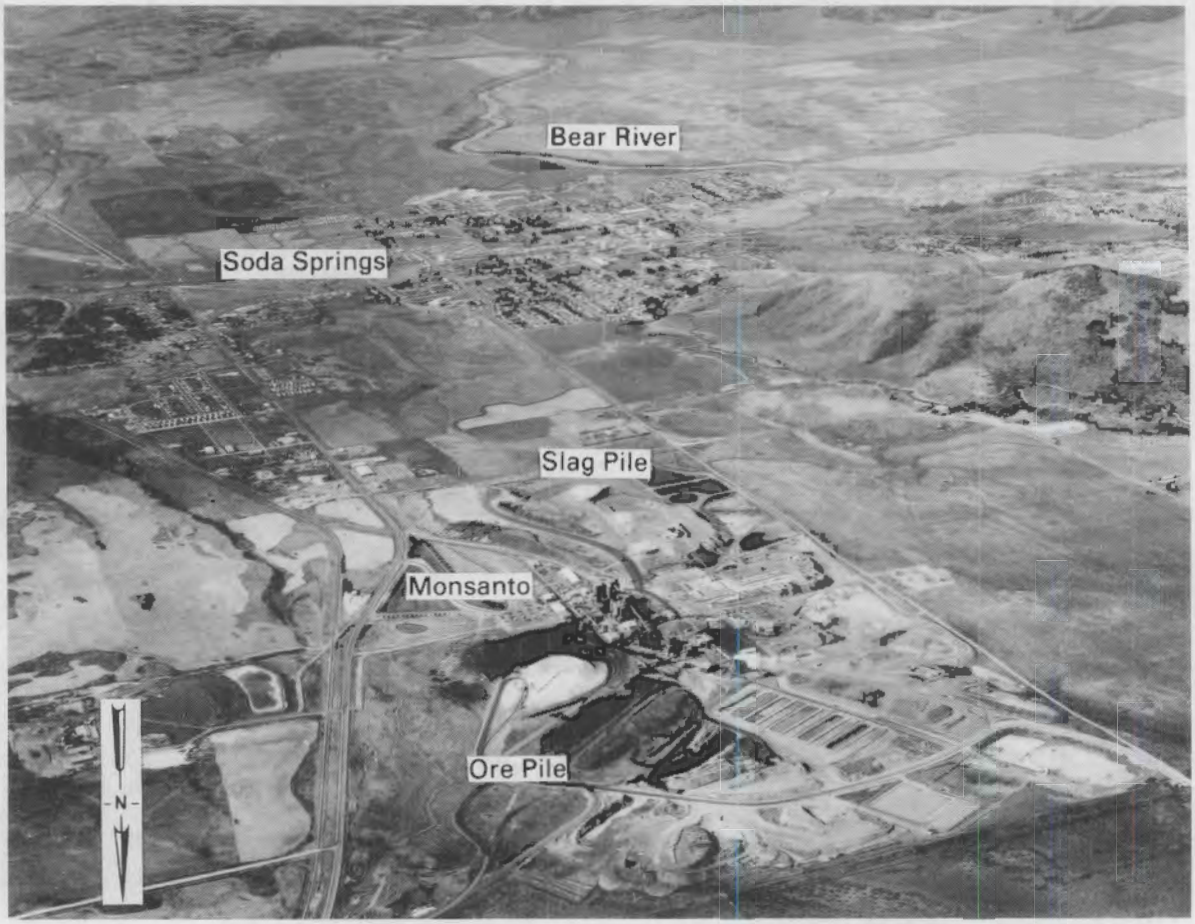


FIGURE 2.3. Aerial View of the Monsanto Elemental Phosphorus Plant and Soda Springs

3.0 RADIONUCLIDE EMISSIONS FROM PHOSPHATE PROCESSING PLANTS

The four phosphate processing plants in Idaho (listed in Table 2.2) are of two types: wet process and thermal process. The Beker Industries plant at Conda and the J. R. Simplot Co. plant at Pocatello are wet process plants that use high-grade ore (~31% P_2O_5) to produce phosphoric acid, which in turn is used to make a wide variety of fertilizers. Low-quality ore (~24% P_2O_5) is converted to elemental phosphorous at FMC's plant in Pocatello and Monsanto's plant in Soda Springs, both thermal process plants.

Table 3.1 shows the radionuclide content of some of the phosphate ores used in the processing plants. The ^{238}U concentration is in the range of 20 to 40 pCi/g and, except for the blended Monsanto feedstock, is in equilibrium with its progeny (^{234}U, ^{230}Th, ^{226}Ra, ^{210}Pb, and ^{210}Po). Concentrations of other radionuclides in the ^{235}U and ^{232}Th series are low.

Radionuclides from phosphate processing can be released to the environment through several different pathways, which vary depending on the type of processing plant. A brief description of each type of plant is given here, and radionuclide pathways to the environment are identified.

TABLE 3.1. Radionuclide Content of Phosphate Rock Used in Idaho Processing Plants, pCi/g (Eadie and Bernhardt 1977; Eadie, Bernhardt, and Boyson 1978; EPA 1984b,c)

Radionuclide	Conda, J. R. Simplot Co.	Gay, J. R. Simplot Co.	Gay, FMC	Gay, FMC	Blended, Monsanto
^{238}U	<20	26	22	21	32
^{234}U	39	28	22	--	--
^{230}Th	45	44	22	--	--
^{226}Ra	28	28	26	--	--
^{210}Pb	12	7.9	27	26	151
^{210}Po	32	35	22	21	91
^{235}U	<9	<2	1	--	--
^{232}Th	<1	3	0.4	--	--
^{228}Ra	<1	1.1	1	--	--

3.1 WET PROCESS PHOSPHORIC ACID AND FERTILIZER PRODUCTION

The wet process plants in Idaho typically have handled 2 to 2.5 million tons per year of high-grade ore. With the shutdown of the Beker Industries plant, about 1 to 1.5 million tons per year are now being processed. Primary products and byproducts from the wet process plants are phosphoric acid, various fertilizers made from phosphoric acid, and phosphogypsum.

3.1.1 Process Description

The following description is based on the J. R. Simplot Co. fertilizer complex near Pocatello (Figure 3.1) and comes primarily from a 1975 radiological survey conducted by the EPA (Eadie, Bernhardt, and Boysen 1978). Additional information on wet process plants in general and the J. R. Simplot Co. complex in particular was obtained from other sources (Smith 1982; PEDCo 1983; EPA 1973; Guimond and Windham 1975a,b).

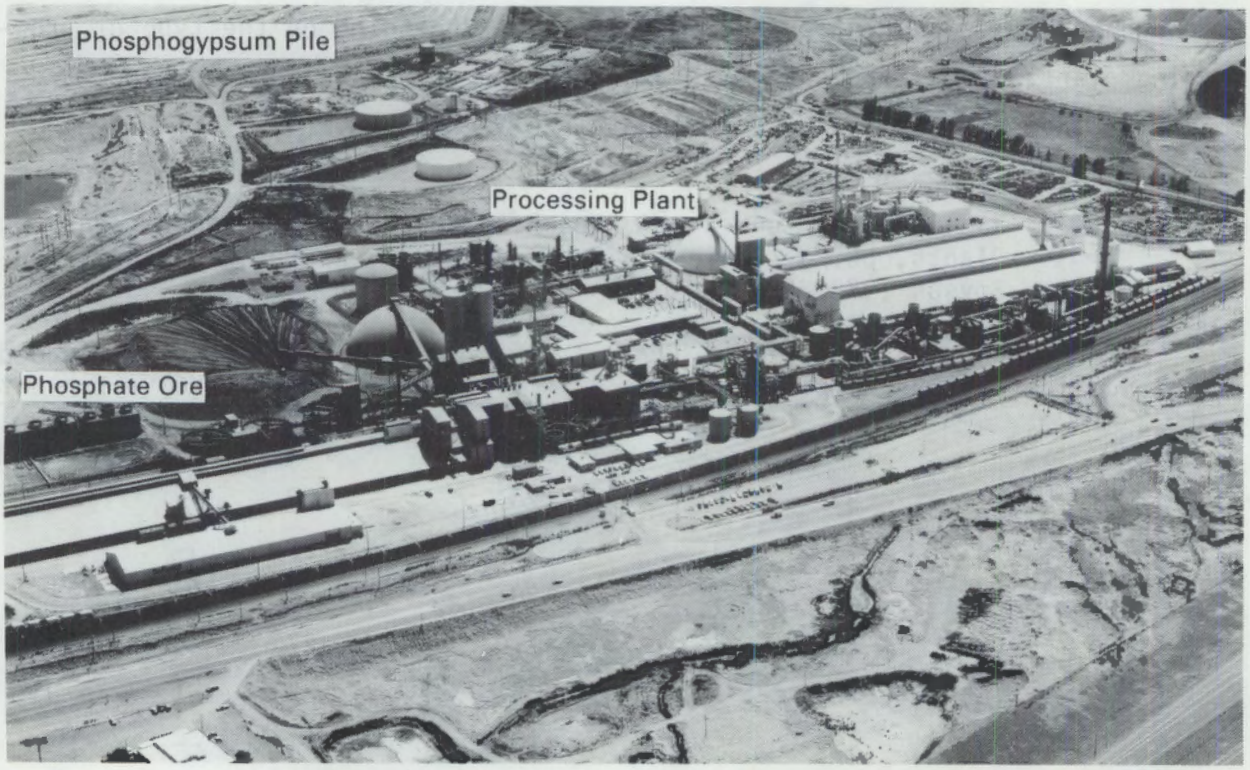


FIGURE 3.1. Aerial View of the J. R. Simplot Co. Wet Process Fertilizer Complex Near Pocatello

At the J. R. Simplot Co. complex, phosphate rock is stockpiled onsite and then fed to a fluid bed calciner where it is heated to about 1400°F (760°C) to burn off the organic material. Gas from the calciner is scrubbed before being vented through a stack to the atmosphere. The calcining also upgrades the ore to approximately 32% to 34% P_2O_5. The calcined ore is ground to a fine powder, which is fed to the phosphoric acid plant. Air from the grinding mill is vented to the atmosphere through a dust collector and a stack.

Ground phosphate rock is mixed with sulfuric acid in a reactor vessel to produce phosphoric acid and calcium sulfate (phosphogypsum). The reaction is approximated by the following equation:

$$\underset{\text{water}}{2H_2O} + \underset{\text{phosphate ore}}{Ca_3(PO_4)_2} + \underset{\substack{\text{sulfuric}\\\text{acid}}}{3H_2SO_4} \rightarrow \underset{\substack{\text{phosphoric}\\\text{acid}}}{2H_3PO_4} + \underset{\text{phosphogypsum}}{3CaSO_4 \cdot 2H_2O}$$

Phosphoric acid is separated from the phosphogypsum in a pan filter. Fumes from the exothermic reaction are collected and treated with water scrubbers. Phosphogypsum in the pan filters is rinsed and eventually pumped to a phosphogypsum pile. Water from the phosphogypsum pile is returned to the phosphoric acid circuit as process makeup water.

The phosphoric acid is used as the basis for several fertilizer formulations. The J. R. Simplot Co. uses it to produce three types of fertilizers: three grades of solid ammonium phosphate, two grades of liquid ammonium phosphate, and triple superphosphate (TSP). Ammonium sulfate is also made at the plant, but phosphoric acid is not used. To make solid ammonium phosphate, ammonia, phosphoric acid, and sulfuric acid are mixed in a reactor, granulated, and dried. Liquid fertilizer is made by reacting ammonia, water, and "super acid" (70% P_2O_5) to form the liquid products. The TSP is made by reacting phosphate rock with phosphoric acid. The resulting material is granulated and dried. Off-gas from the dryer is scrubbed and vented through a stack.

3.1.2 Radionuclide Pathways to the Environment

Radionuclides contained in phosphate ore and wet process products can enter the environment through a number of pathways. Figure 3.2 shows potential radionuclide pathways to the environment for a wet process plant.

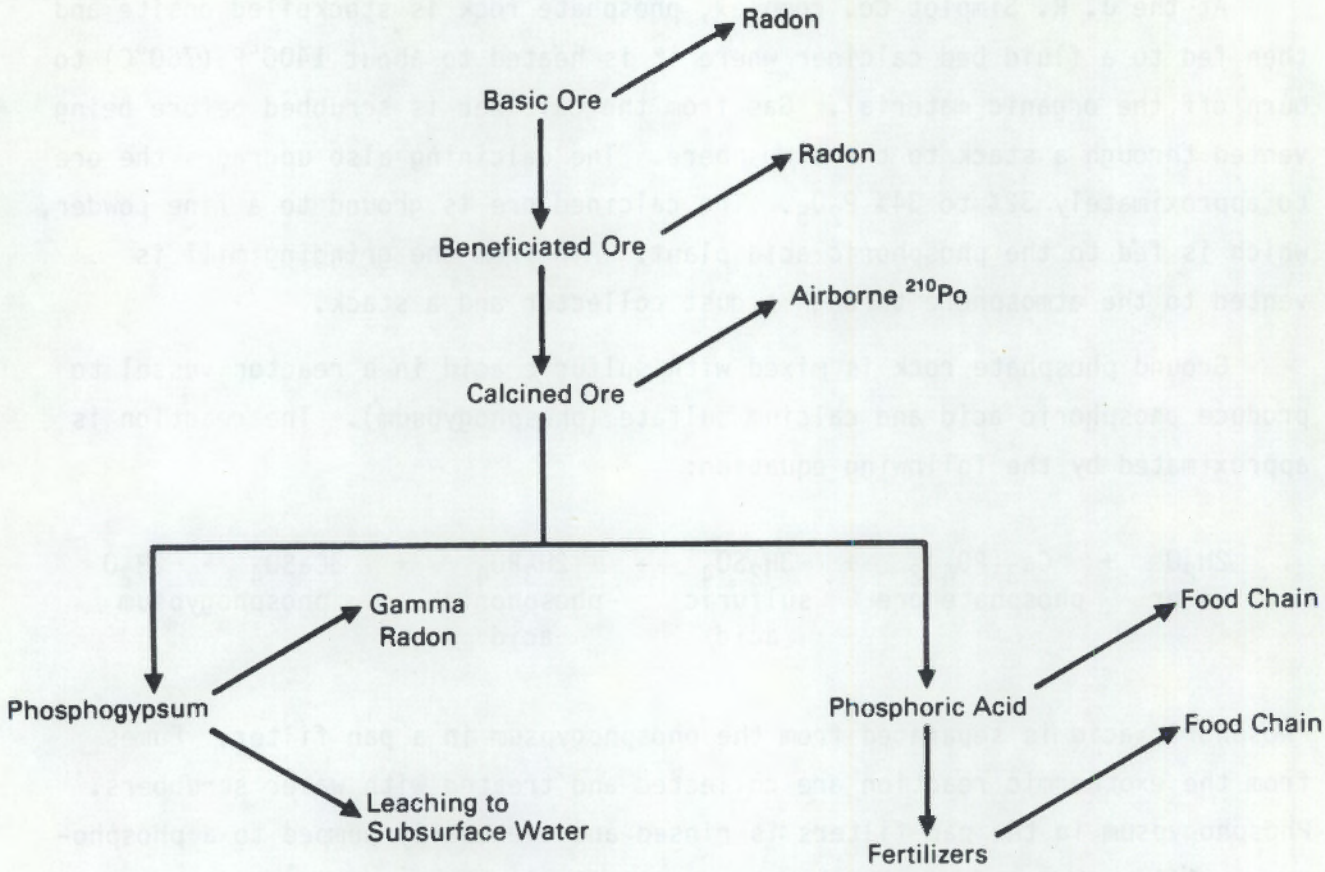


FIGURE 3.2. Radionuclide Pathways to the Environment for a Wet Process Plant (Boothe 1977)

Both the ore and the phosphogypsum stored onsite may release ^{222}Rn, a gaseous progeny of ^{238}U. Furthermore, during production of phosphoric acid or fertilizer, volatile radionuclides such as ^{210}Po, ^{210}Pb, and ^{222}Rn may be emitted in the calciner off-gas. Also, the fertilizer products themselves contain trace quantities of radioactive materials, primarily uranium, which can enter the environment. The main byproduct is phosphogypsum, which contains radium, a source of radon and gamma radiation and a potentially leachable contaminant to subsurface waters (Boothe 1977). Similarly, ore stockpiles are a source of ^{222}Rn and fugitive dust containing radionuclides. The radiological survey of the J. R. Simplot Co. complex identified another significant source of airborne radionuclides to be the TSP dryer stack.

Airborne Radionuclides

Sources of airborne radionuclides are the TSP dryer, the calciners and other stack emissions, and fugitive emissions from ore stockpiles and phosphogypsum piles. There are three fluid bed calciners at the J. R. Simplot Co. complex and two at the Beker Industries plant. The sizes of the calciners and particulate control and emission data from these plants are given in Table 3.2.

In elemental phosphorus plants, the particulate emissions from the calciners contain significant quantities of ^{210}Po and ^{210}Pb, which are volatilized in the calciner and condensed on particulate matter (see Table 3.9). However, calciners at wet process plants operate at lower temperatures, and recent EPA tests at the J. R. Simplot Co. plant showed that relatively little volatilization of these radionuclides occurred during calcining (EPA 1987). Particulates in the calciner scrubber outlet were only slightly enriched in ^{210}Po. Polonium-210 concentrations in the particulates were 65 to 103 pCi/g compared with 11,000 to 37,000 pCi/g in particulates emitted from elemental phosphorus plant calciners (see Table 3.9).

The measured radionuclide emission rates were 0.06 μCi/h for ^{210}Pb and 0.15 μCi/h for ^{210}Po. The estimated annual emission rates for the calciner tested were 0.5 mCi/year for ^{210}Pb and 1 mCi/year for ^{210}Po. These radionuclide emission rates are based on a particulate emission rate of 1.6 kg/h measured during the test. This measurement corresponds to a particulate removal efficiency of 99.7% and a ^{210}Po removal efficiency of 99.6%. Particulate emission rates that were an order of magnitude higher have been reported for the J. R. Simplot Co. plant as shown in Table 3.2 (Smith 1982). Even with an order of magnitude increase, ^{210}Po emissions from the wet process would still be several orders of magnitude lower than the proposed standard of 21 Ci/year.

TABLE 3.2. Particulate Emissions from Phosphate Rock Calciners at Wet Process Plants (Smith 1982)

Company	Location	Production Rate, tons/h	Type of Calciner	Control Device	Stack Gas Flow, 10^3 scfm	Particulate Emission Rate gr/scf[a]	lb/h	lb/ton
J. R. Simplot Co.	Pocatello	35	Fluid Bed	Entoleter Scrubber[b]	25	0.03	7.4	0.21
		41	Fluid Bed	Entoleter Scrubber[b]	28	0.1	24	0.58
		55	Fluid Bed	Electrostatic	58	0.06	30	0.54
Beker Industries	Conda	65	Fluid Bed	Venturi Scrubber[c]	33	0.032	8.8	0.136
		70	Fluid Bed	Venturi Scrubber[d]	40	0.10	34	0.49

(a) To convert grains/scf to grams/m^3, multiply by 1.83.
(b) Equivalent to a high-energy venturi scrubber.
(c) 8-in. water gauge pressure drop.
(d) 12-in. water gauge pressure drop.

The 1975 EPA survey (Eadie, Bernhardt, and Boysen 1978) found that the TSP dryer stack was the largest source of airborne radionuclide emissions. The concentration of radionuclides (^{238}U chain) was about 10 pCi/m^3. Other stacks sampled during the EPA survey were the phosphoric acid scrubber, the grinding mill stack, and the ammonium phosphate reactor stack. Radionuclide concentrations were at least an order of magnitude lower in these locations than those measured in the TSP stack. Eadie, Bernhardt, and Boysen (1978) estimated the total radionuclide emissions from the J. R. Simplot Co. plant to be about 5 to 10 mCi/year of ^{210}Pb, ^{226}Ra, ^{234}U, and ^{238}U with lower values for ^{230}Th and ^{210}Po.

The TSP process has been extensively modified since these data were taken, and emission of airborne radionuclides has been greatly reduced. Because this process was the major source of radionuclide emissions, the current total radionuclide emissions from the J. R. Simplot Co. plant should be much lower than those reported by Eadie, Bernhardt, and Boysen (1978).

Phosphogypsum

Phosphogypsum, a byproduct produced by the reaction of phosphate rock and sulfuric acid, is another source of radionuclide emissions to the environment. Most of the ^{226}Ra in the rock is not dissolved by acidulation with sulfuric acid and remains with the phosphogypsum. Some of the uranium and the thorium does enter solution and is transferred to the phosphoric acid product (Guimond and Windham 1975a). Radionuclide concentrations for three phosphogypsum solid samples from Idaho phosphoric acid plants are shown in Table 3.3, along with a feed ore for comparison.

About 3 to 4 tons of phosphogypsum are produced per ton of P_2O_5 processed, or about 1 ton of phosphogypsum per ton of rock fed. Annual production of phosphogypsum in Idaho is estimated at 2 million tons per year with both plants operating. With the shutdown of the Beker Industries plant, production of phosphogypsum will drop to about 1 million tons per year.

Phosphogypsum slurry from the phosphoric acid plant is pumped to piles for storage. Characteristics of the phosphogypsum piles at the J. R. Simplot Co. and Beker Industries facilities are shown in Table 3.4.

TABLE 3.3. Radionuclide Concentration in Phosphogypsum Solids, pCi/g

Radionuclide	Calcined Feed Ore (a)	Phosphogypsum (a)	(b)	(c)
^{238}U	38	<14	--	--
^{234}U	64	24	--	--
^{230}Th	65	26	--	--
^{226}Ra	38	23	7.9	16
^{210}Pb	1	1.1	--	--
^{210}Po	42	26	--	--
^{235}U	1	<6.8	--	--
^{232}Th	0.4	<1.3	--	--
^{228}Ra	<1	<1.0	--	--

(a) Eadie, Bernhardt, and Boysen (1978).
(b) PEDCo (1983).
(c) EPA (1973).

TABLE 3.4. Phosphogypsum Piles in Idaho (PEI 1985)

Company	Location	Number of Piles	Size, ha (acres)	Height, m (ft)	Quantity, metric tons (short tons)
Beker Industries	Conda	1	36 (90)	24 (80)	7.9×10^6 (8.7×10^6)
J. R. Simplot Co.	Pocatello	2	24 (60)	15 (50)	3.3×10^6 (3.6×10^6)
			81 (200)	18 (60)	15.0×10^6 (17.0×10^6)

During the 1975 survey, EPA measured a net gamma exposure rate (gross gamma exposure minus background) of 31 μR/h or 52 mrem/year on the phosphogypsum pile at the J. R. Simplot Co. facility. This value was typical for the general work areas tested in the plant. A radon concentration of 0.31 pCi/l was measured on the phosphogypsum pile (Eadie, Bernhardt, and Boysen 1978).

More recent radon flux measurements and radon-in-air measurements on phosphogypsum piles have been made in Florida (Freeman and Hartley 1985; PEI

1986). Radon was also measured in the air short distances from the piles (PEI 1986). Evaluation of these data may indicate the potential significance of radon exhalation from Idaho phosphogypsum piles.

Wastewater

Plant effluents are another potential source of radionuclide releases to the environment. At a wet process phosphoric acid plant, wastewater is generated from two main sources: scrubber liquids and water associated with the phosphogypsum byproduct. Table 3.5 shows the radionuclide concentrations for some water streams at the J. R. Simplot Co. plant. Suspended solids were allowed to settle out in some of the samples, and both the solid and liquid were analyzed.

Scrubber liquids are treated prior to discharge or recycled back to the process. The radionuclide content of the outfall from the J. R. Simplot Co. plant measured in 1973 and 1975 is shown in Table 3.6. The incoming well water for the plant is shown for comparison. There has been no outfall from the plant since 1980.

Fertilizer Products

The radionuclide concentration in some phosphate fertilizers from the J. R. Simplot Co. complex is shown in Table 3.7, along with an analysis of ammonium sulfate for comparison. The ammonium phosphates made from phosphoric acid show elevated levels of ^{238}U and ^{230}Th (^{226}Ra remains primarily in the phosphogypsum). The TSP made from both phosphoric acid and phosphate rock shows elevated levels of ^{226}Ra, ^{210}Po, and ^{234}U, as well as ^{238}U and ^{230}Th.

3.1.3 Radionuclide Material Balance

Sufficient data are not available to determine a radionuclide material balance for a wet process plant, although some generalizations can be made. Typical feed ore for the plant has a radionuclide activity of 30 to 40 pCi/g for ^{238}U, with progeny usually close to equilibrium. This activity corresponds to 30 to 40 μCi/ton of feed ore. Most of the radium goes to the phosphogypsum byproduct, but some of the uranium and thorium end up in the phosphoric acid products. In a study of Florida ores, Guimond and Windham (1975a,b) indicate that ^{238}U, ^{234}U, and ^{230}Th are significantly dissolved by the sulfuric acid in

TABLE 3.5. Radionuclide Content of Process Waters at Wet Process Phosphoric Acid Plant (Eadie, Bernhardt, and Boysen 1978)

Radionuclide	Phosphoric Acid Scrubber		Calciner Scrubber		TSP Scrubber		Phosphogypsum Pile Water Underflow, pCi/l	Phosphogypsum Pond Water, pCi/l	Phosphogypsum Pond Water Overflow, pCi/l
	Suspended, pCi/g	Liquid, pCi/l	Suspended, pCi/g	Liquid, pCi/l	Suspended, pCi/g	Liquid, pCi/l			
^{238}U	--	32	5.0	5.0	1.0-3.5	41-55	560	900	270
^{234}U	--	37	4.7	6.8	0.8-3.2	43-56	550	910	270
^{230}Th	--	0.7	17	0.20	18-40	16-32	78	2.6	77
^{226}Ra	2.5	13	33	3.6	10-22	27-32	36	61	40
^{210}Pb	--	2.2	3.1	2.8	3.1-6.7	24-40	100	68	50
^{210}Po	--	45	76	0.67	32-53	--	37	<5.4	--
^{235}U	--	1.1	<0.18	0.32	<0.10	1.6-1.8	28	35	9.1
^{232}Th	--	<0.03	0.34	<0.04	0.40	0.24-0.32	0.64	<0.2	0.8
^{228}Ra	<10	5.9	1.0	<3.4	<11	<3.5	<3.7	<3.7	<3.7

TABLE 3.6. Radionuclide Content of Outfall and Incoming Water at J. R. Simplot Co. Plant, pCi/l

Radionuclide	Plant Outfall (a)	Plant Outfall (b)	Incoming Well Water (a)
^{238}U	6.2	--	1.2
^{234}U	9.6	--	3.2
^{230}Th	1.6	--	<0.04
^{226}Ra	0.63	0.14-0.58	0.7
^{210}Pb	3.4	--	1.2
^{210}Po	1.4	--	<0.8
^{235}U	0.35	--	0.12
^{232}Th	<0.1	--	<0.04
^{228}Ra	4.0	--	<3.7

(a) Eadie, Bernhardt, and Boysen (1978).
(b) EPA (1973).

TABLE 3.7. Radionuclide Content of Fertilizer Products, pCi/g (Eadie, Bernhardt, and Boysen 1978)

Radionuclide	Ammonium Phosphate (various grades)	Triple Super-phosphate	Ammonium Sulfate
^{238}U	21-27	44	<1.2
^{234}U	<20	45	<1.3
^{230}Th	6.2-6.3	54	<0.16
^{226}Ra	0.65-1.6	14	0.1
^{210}Pb	0.62-1.8	3.6	1.3
^{210}Po	2.1-4.6	18	<0.47
^{235}U	<13	<3.3	<0.84
^{232}Th	0.08-2.0	<1.6	<0.11
^{228}Ra	<0.92-1.8	<0.88	<0.89

the phosphoric acid plant. About 50% to 80% of the thorium and 80% of the uranium is dissolved and ends up in the phosphoric acid products. A comparison of the radionuclide concentrations in the phosphogypsum and the calcined feed ore in Table 3.3 indicates that about 50% of the ^{238}U, ^{234}U, and ^{230}Th is solubilized.

Some of the soluble radionuclides also end up in the phosphogypsum water, particularly ^{234}U and ^{238}U. No information is available to determine what fraction of the total radioactivity is accounted for in the water stream. Very small quantities of radionuclides (about 5 to 10 x 10^{-3} μCi/ton of feed ore) have been detected in the particulates in stack gases.

3.2 THERMAL PROCESS FOR PRODUCTION OF ELEMENTAL PHOSPHORUS

About 2 to 2.5 million tons per year of low-grade ore are processed in the two elemental phosphorus plants in Idaho. The primary product is elemental phosphorus. Phosphorus slag, an amorphous calcium silicate, is also produced and has been sold commercially as an aggregate.

3.2.1 Process Description

The Monsanto and FMC thermal process plants (Figures 3.3 and 3.4) are similar in design and operation (EPA 1984b,c; Gallaghar 1976; Eadie and Bernhardt 1977). At the FMC plant, phosphate rock is crushed, screened, and formed into briquettes before being fed to two moving-grate calciners. At the Monsanto plant, phosphate rock is fed directly to a rotary kiln calciner.

In both plants, rock (or briquettes) is heated to 1200°C to 1300°C in the calciners to remove organic materials and to form heat-hardened nodules that will withstand further processing without disintegrating. The nodules are cooled and passed through a proportioning building where they are blended with sized coke and silica before being fed into an electric arc reducing furnace. High-temperature reactions in the furnace (2500°C) drive off gaseous phosphorus and carbon monoxide and leave molten residues of slag and ferrophosphorus. Furnace off-gases pass through electrostatic precipitators to remove dust before entering a condenser, where phosphorus is condensed, collected in a sump, and pumped to storage.

FIGURE 3.3. Monsanto Elemental Phosphorus Plant Near Soda Springs

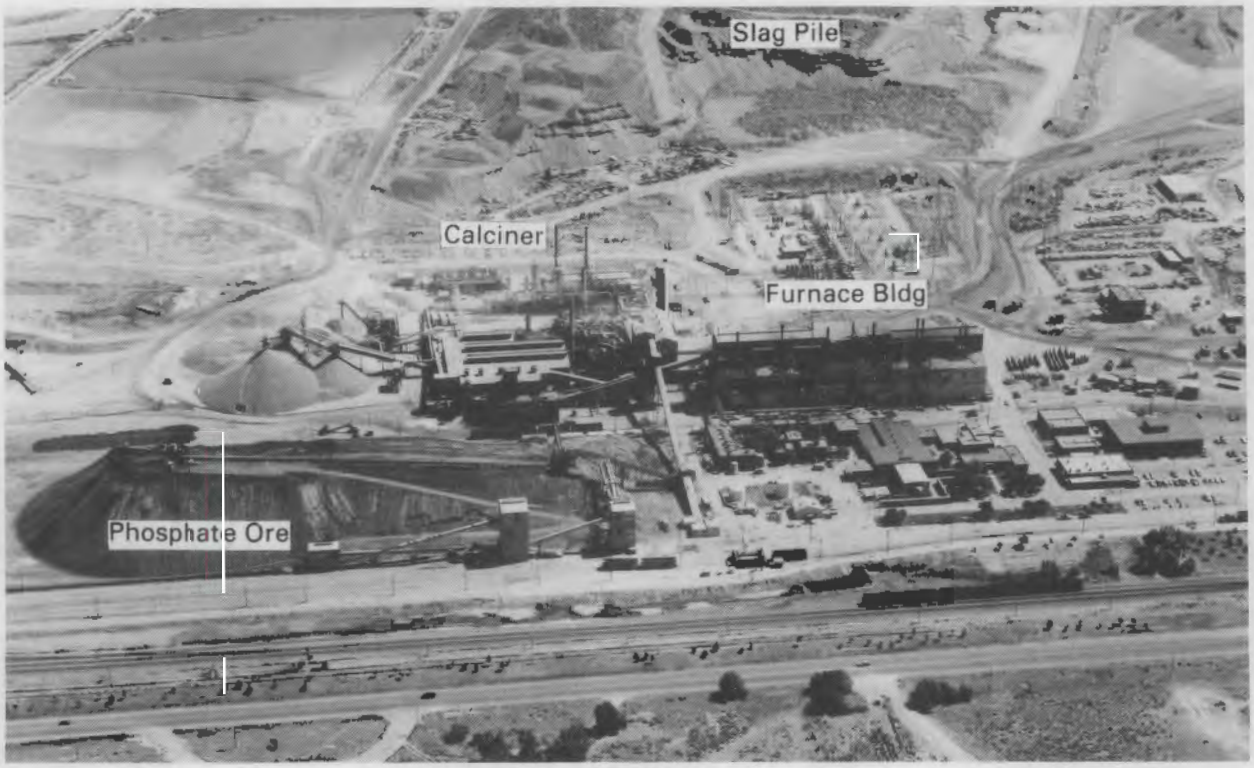


FIGURE 3.4. FMC Elemental Phosphorus Plant Near Pocatello

Cleanup of the calciner off-gases is different for each plant. At the FMC plant, exhaust gases pass sequentially through a fan, a spray quench chamber, a horizontal scrubber, a demister, a second fan, and out the stack. Two parallel gas-cleanup trains are used; one with chevron demisting pads, the other with a cyclonic demisting system. Water from the scrubber system is transported to a settling pond.

Off-gases from the Monsanto kiln pass through a settling chamber, a spray tower, and a demister prior to discharge to the air. Spray tower effluent goes to a settling pond, and the water is recycled to the spray tower.

3.2.2 Radionuclide Pathways to the Environment

Figure 3.5 shows the potential radionuclide pathways to the environment for an elemental phosphorus plant. The ore pile at the plant may emit ^{222}Rn and fugitive dust. Because of higher calciner temperatures, radionuclide emissions from thermal process plant calciners are higher than from wet process plants. Volatile radionuclides, such as ^{210}Po, ^{210}Pb, ^{226}Ra and ^{230}Th, may be emitted in the calciner off-gas, along with ^{222}Rn. Radionuclides also may be volatilized in the furnace.

Another potential pathway is the slag, which contains significant concentrations of radium. These concentrations may lead to gamma radiation exposures associated with use of slag, and potential radon/progeny exposures associated with its use as an aggregate in concrete for building construction.

Calciner Off-Gases (and other airborne emissions)

Emission tests of the calciner off-gases at the two Idaho elemental phosphorus plants have recently been completed (Jongleux 1984a,b; EPA 1984b,c). Comparison of the radionuclide content of the phosphate rock feedstock and the calcined product shown in Table 3.8 indicates that about 98% of the ^{210}Po and about 95% of ^{210}Pb are volatilized in the calciner at the Monsanto plant near Soda Springs. At the FMC plant near Pocatello, about 60% of the ^{210}Po is volatilized, while almost no ^{210}Pb is volatilized. At both plants, very little or no ^{238}U is volatilized. Both calciners operate at similar temperatures (1200°C to 1300°C). The reduced volatilization at the FMC plant is apparently

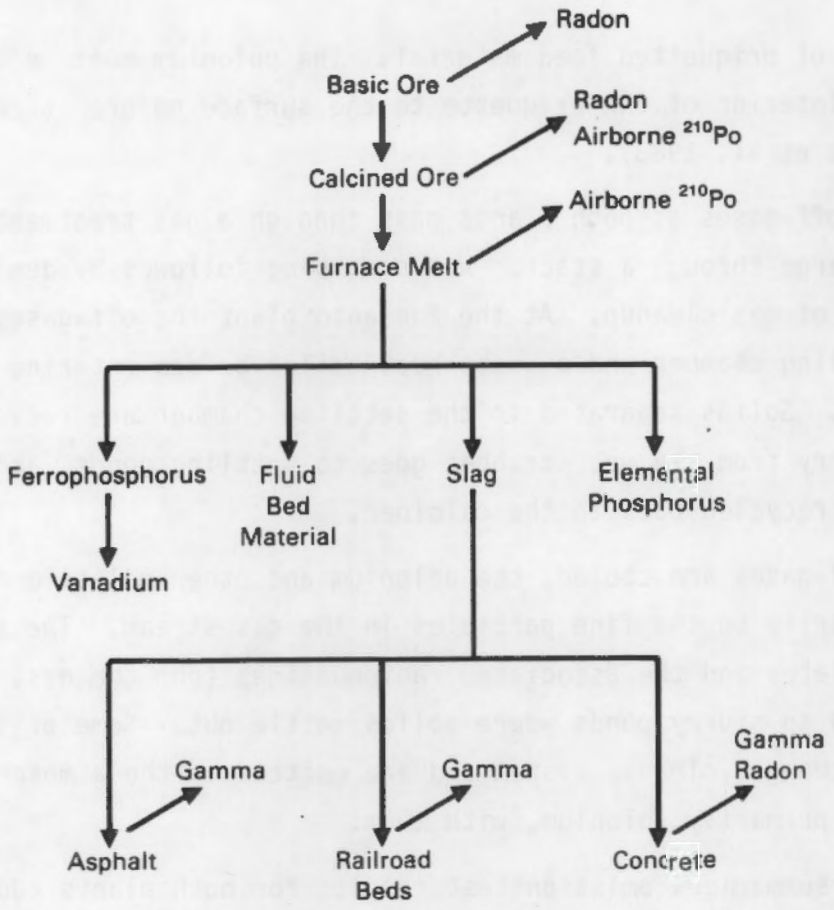


FIGURE 3.5. Radionuclide Pathways to the Environment for a Thermal Process Plant

TABLE 3.8. Radionuclide Content of Phosphate Rock Feedstock and Calcined Product at Idaho Elemental Phosphorus Plants (EPA 1984b,c)

Radionuclide		Feedstock, pCi/g	Calcined Product, pCi/g
Monsanto	^{238}U	32	37
	^{210}Pb	150	6
	^{210}Po	91	2
FMC	^{238}U	21	22
	^{210}Pb	26	27
	^{210}Po	21	8

due to the use of briquetted feed material. The polonium must be transported from the bulk interior of the briquette to the surface before it can be released (Stula et al. 1983).

Calciner off-gases at both plants pass through a gas treatment system prior to discharge through a stack. Wet scrubbing followed by demisting is the primary method of gas cleanup. At the Monsanto plant the off-gases pass through a settling chamber and a waste heat boiler before entering the gas-cleanup system. Solids separated in the settling chamber are recycled to the calciner. Slurry from the wet scrubber goes to settling ponds, and the solids are eventually recycled back to the calciner.

As the off-gases are cooled, the polonium and other volatile radionuclides condense, primarily on the fine particles in the gas stream. The scrubbers remove particulates and the associated radionuclides from the gas. Scrubber water is pumped to slurry ponds where solids settle out. Some of the fine particles escape the gas-cleanup system and are emitted to the atmosphere carrying radionuclides, primarily polonium, with them.

Table 3.9 summarizes emission test results for both plants (Jongleux 1984a,b; EPA 1984b,c). Estimated yearly radionuclide emissions from the calciner stack for the Monsanto plant are 21 Ci of ^{210}Po and 5.6 Ci of ^{210}Pb. For the FMC plant, estimated yearly radionuclide emissions are 9 Ci of ^{210}Po and 0.1 Ci of ^{210}Pb. Also included in Table 3.9 are recent results obtained by Monsanto and FMC, which show totals of 20.8 and 6.7 Ci/year of ^{210}Po, respectively. The FMC results were obtained by using EPA Method 5 and EPA Method III calculation procedure. The stack sampling procedure is described in detail by Buttelman (1986). Details on the Monsanto testing are not available.

As part of the tests by Jongleux (1984a,b) and EPA (1984b,c), samples taken of the inlet/outlet gas of one scrubber at the FMC plant indicated that 77% of the ^{210}Po and 85% of the ^{210}Pb were removed from the calciner off-gas. About 70% of the polonium discharged from the stack was associated with particles smaller than 0.5 μm. At the Monsanto plant, about 95% each of the ^{210}Po and the ^{210}Pb was associated with particles smaller than 1 μm.

TABLE 3.9. Airborne Emissions from Phosphate Rock Calciners at Elemental Phosphorus Plants (Jongleux 1984a,b; EPA 1984b,c)

Company	Location	Type of Calciner	Control Device	Stack Gas Flow, 10^3 dscfm	Particulate Emission Rate[a] gr/dscf[c]	Particulate Emission Rate[a] lb/h[d]	Particulate Radionuclide Concentration, pCi/g ^{210}Po	Particulate Radionuclide Concentration, pCi/g ^{210}Pb	Radionuclide Emission Rate µCi/h[b] ^{210}Po	Radionuclide Emission Rate µCi/h[b] ^{210}Pb	Radionuclide Emission Rate Ci/yr[b] ^{210}Po	Radionuclide Emission Rate Ci/yr[b] ^{210}Pb
Monsanto	Soda Springs	Rotary kiln	Settling chamber, spray tower, demister	131	0.153	171	37,000	9,700	2,900	760	21	5.6
Monsanto[e]	Soda Springs	Rotary Kiln	(Same as above)	--	--	--	--	--	--	--	20.8	--
FMC	Pocatello	#1 Moving Grate	Spray chamber, slinger scrubber, demister	67[f] 56[f]	0.122 0.063	70 30	11,000 26,000	70 370	350 360	2 5	5.2	0.06
		#2 Moving Grate	Spray chamber	69	0.039	26	16,000	330	190	4	3.4[g]	0.06
									FMC Total		8.6	0.12
FMC[h]	Pocatello	#1 Moving Grate	(Same as above)	91 75	0.037 0.061	29 39	--	--	--	--	3.7	--
		#2 Moving Grate	(Same as above)	85 83	0.025 0.028	19 20	--	--	--	--	3.0	--
									FMC Total		6.7	

(a) EPA Method 5.
(b) Based on 7,400 h of calciner operation per year (0.85 operating factor).
(c) To convert to g/m^3 multiply by 1.83.
(d) To convert to g/h multiply by 454.
(e) Data provided by Mark Hooper, EPA Region X, April 8, 1987.
(f) Two stacks on the #1 calciner.
(g) Test data are for only one stack (of two total) with feed rate 85% of normal; this number is obtained by multiplying test data by 2 and then by 1.2 (capacity factor).
(h) Data provided by Marc Bowman, FMC.

The 1975 EPA study, which also covered the FMC plant, estimated the total airborne ^{210}Po emissions from the plant to be 7.4 Ci/year (Eadie, Bernhardt, and Boysen 1978), in good agreement with another study (Eadie and Bernhardt 1977). Stula et al. (1983) estimated ^{210}Po emissions from the FMC and Monsanto plants to be 8 Ci/year and 3 Ci/year, respectively. The FMC estimate is based on information provided by FMC and is in good agreement with the other tests. The estimate for the Monsanto plant is much lower than the 21 Ci/year measured in 1984. One reason for the high ^{210}Po emissions measured in 1984 was the high ^{210}Po concentration in the feedstock, 91 pCi/g.

Potential fugitive airborne emissions include dust from the ore and slag piles and radon exhalation from the slag and ore piles. Radon exhalation rates measured by the Montana Department of Health and Environmental Sciences were 3400 to 3800 $pCi/m^2/min$ for phosphate ore and only 18 to $30/m^2/min$ for slag even though they had essentially the same ^{226}Ra content (Lloyd 1983). These measurements indicate that radon exhalation from slag piles is not likely to be a problem.

Slag and Other Solid Byproducts

Slag has been sold as an aggregate for concrete in foundations of buildings, primarily in the Soda Springs area, and is also used as an aggregate for roads and railroad bed ballast. However, since 1977, the State of Idaho has prohibited the use of slag material in the construction of habitable structures (Peterson 1979).

About 85% by weight of the phosphorus ore ends up as slag. The slag, which is primarily calcium silicate, also contains most of the nonvolatile radionuclides originally present in the ore. Table 3.10 shows the radionuclide content of various slags from Idaho elemental phosphorus plants. Except for ^{210}Po and ^{210}Pb, the radionuclides in the slag are in equilibrium at concentrations about 15% to 20% higher than the phosphate rock fed to the plant. The ^{210}Po and ^{210}Pb are partially volatilized in the calciner, which reduces their concentration in the slag.

TABLE 3.10. Concentration of Radionuclides in Slags from Idaho

Radionuclide	Concentration, pCi/g (a)	(b)	(c)	(c)	(d)
^{238}U	25	--			29
^{234}U	25	--			38
^{230}Th	26	--			42
^{226}Ra	32	15.2	39	41	50
^{210}Pb	11	--			--
^{210}Po	<16	--			--
^{232}Th	0.6	--			0.5
^{228}Ra	0.96	--			--

(a) Eadie and Bernhardt (1977).
(b) PEDCo (1983).
(c) Boothe (1977).
(d) Hans et al. (1978).

About 1.8 to 2.3 million tons of slag are produced by the two plants each year. The slag is stockpiled on the plant sites in Pocatello and Soda Springs (Boothe 1977; Melville 1980; Peterson 1979; Gallaghar 1976). Because of the low moisture conditions associated with the slag, no seepage collection systems are utilized.

Another solid byproduct from the elemental phosphorus plant is the dust removed from the electrostatic precipitators. This material is dried in a fluidized bed dryer, and the dried solids are referred to as "fluid bed prills." The fluid bed prills contain a high concentration of radionuclides that were volatilized in the furnace. Reported concentrations of radionuclides are 440 to 1550 pCi/g ^{210}Po, 52 pCi/g ^{210}Pb, and 10 to 13 pCi/g ^{226}Ra (Eadie and Bernhardt 1977; Gallaghar 1976). Using a production rate of 0.13 tons per ton of product, PEDCo (1983) estimated that 30,000 tons of fluid bed prills are produced in Idaho each year and stored on the plant sites.

A third solid byproduct is ferrophosphorus, the dense metallic material tapped from the bottom of the furnace. Ferrophosphorus from Monsanto is sold to the Kerr-McGee Corporation, which extracts the vanadium from it at a plant

in Soda Springs. The FMC plant is currently stockpiling its ferrophosphorus. The radioactivity in the ferrophosphorus, about 30 to 50 pCi/g, is primarily from ^{234}U and ^{238}U (Eadie and Bernhardt 1977; Gallaghar 1976).

Wastewater

Only limited information on wastewater streams from elemental phosphorus plants is available (Eadie and Bernhardt 1977; Gallaghar 1976; PEDCo 1983). The calciner scrubber liquid, the phosphorus condenser wastewater, and the fluid bed scrubber liquor are pumped to waste ponds onsite to separate the solids from the stream. These streams show some elevated radioactivity, but no contamination of the local environment by water from the elemental phosphorus plants has been observed.

3.2.3 Radionuclide Material Balance

Figure 3.6 shows an estimate of a radionuclide material balance for a thermal process plant, assuming a feed ore with a ^{238}U concentration of 30 pCi/g and progeny nuclides in equilibrium. The results are based on data from the FMC plant (Eadie and Bernhardt 1977; Gallaghar 1976; Stula et al. 1983). The majority of the radionuclides end up in the slag. A significant fraction of the ^{210}Po and some of the ^{210}Pb are volatilized. The volatilized radionuclides ultimately end up as airborne emissions released out the calciner stacks or in a slurry from the calciner gas treatment system. The fluid bed prills contain the majority of radionuclides volatilized in the furnace. Little or no radioactivity has been reported in the elemental phosphorus.

Based on an estimated feed rate of 1.2 million tons per year of ore at the FMC plant, about 35 Ci/year of ^{210}Po are charged to the plant. About 6 to 10 Ci/year are discharged to the atmosphere from the calciner stack. The rest is distributed as indicated in Figure 3.6.

The limited data available for the Monsanto plant makes a radionuclide material balance impossible at this time. The ^{210}Po and ^{210}Pb activity in the feedstock used during the recent tests is quite high and not in equilibrium (see Table 3.8). This is unusual because most phosphate ores show progeny nuclides to be in equilibrium with ^{238}U. The high ^{210}Pb and ^{210}Po content is

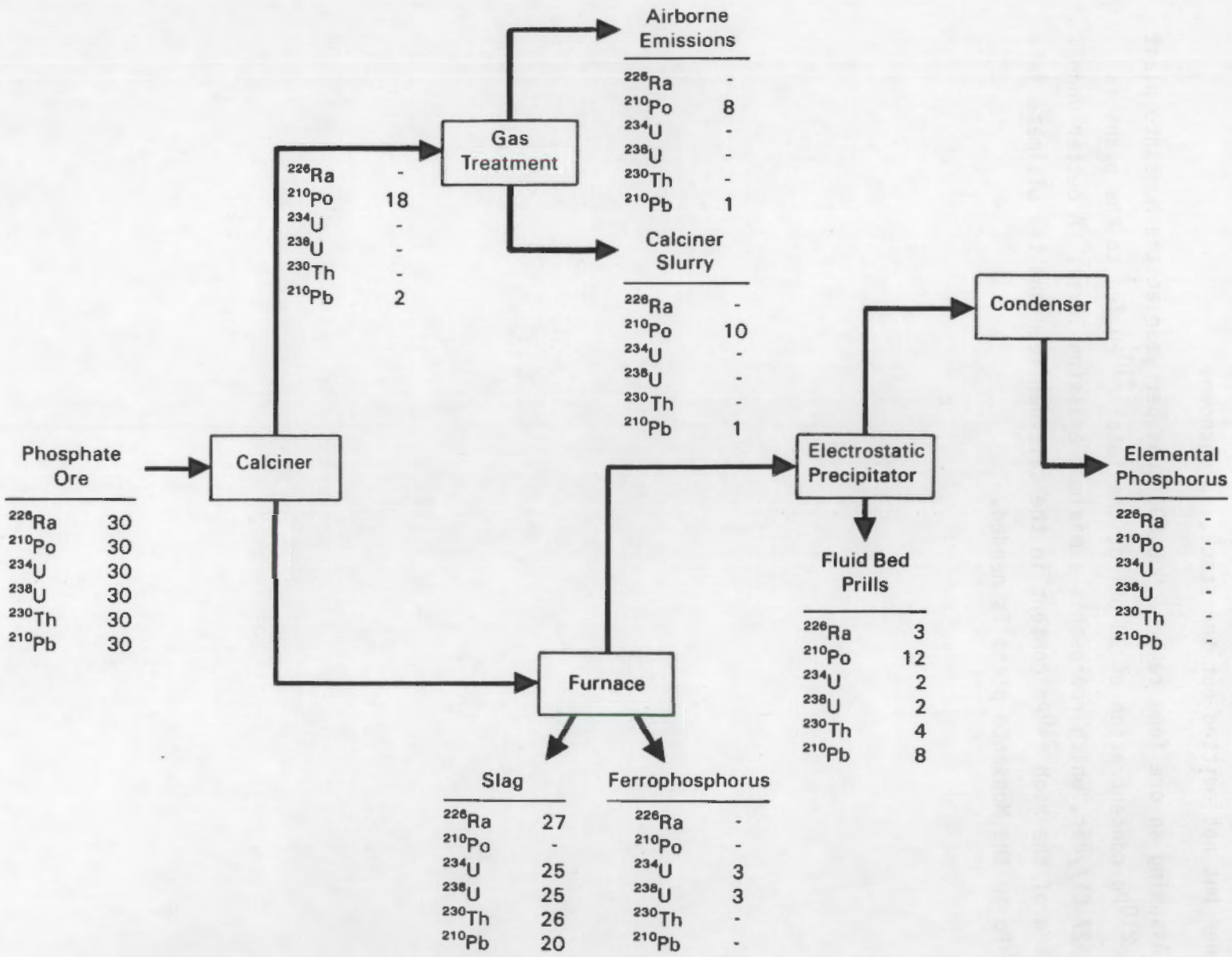


FIGURE 3.6. Radionuclide Material Balance for a Thermal Process Plant, pCi/g

apparently due to recycling the settling chamber dust and the calciner scrubber solids. The ultimate fate of the ^{210}Po and ^{210}Pb, which are volatilized in the calciner but not emitted out the stack, is unknown.

Assuming an ore feed rate of 900,000 tons per year at the Monsanto plant and a ^{210}Po concentration of 30 pCi/g, the total ^{210}Po fed to the plant is about 27 Ci/year, which represents a maximum emission level. A better understanding of the high ^{210}Po content in the calciner feed and the ultimate fate of ^{210}Po in the Monsanto plant is needed.

4.0 ENVIRONMENTAL RADIOACTIVITY FROM PHOSPHATE PROCESSING PLANTS

Two major pathways by which radionuclides from phosphate processing plants are distributed within the environment are 1) radioactive particulates and radon gas emitted from stacks or from fugitive emissions from ore, slag, and phosphogypsum piles or 2) commercial use of byproducts such as slag or phosphogypsum. A third potential pathway has been theorized: contamination of ground or surface waters from plant discharges or leaching of waste ponds.

4.1 AIRBORNE RADIONUCLIDES

Elevated ^{210}Po concentrations and alpha activity have been detected in the Pocatello area as an apparent result of phosphate processing operations. Figure 4.1 compares gross alpha air concentrations at three locations in Idaho measured in 1974 by the Health Services Laboratory, Idaho Operations Office, U.S. Department of Energy. The bottom curve is at a location (Howe) removed from all industrial activity and can be considered background. The top curve is for the Pocatello Sewage Treatment Plant located 0.5 mile downwind from the two phosphate processing plants (location A in Figure 4.2). The middle curve is for a location 15 miles downwind from these plants near Blackfoot (Boothe 1977).

In an earlier study conducted by the U.S. Atomic Energy Commission (AEC 1970), the average gross alpha concentration in the air was 0.034 pCi/m^3 at the Pocatello Sewage Treatment Plant and 0.0087 pCi/m^3 at the Pocatello Fire Station during a 6-month period in 1969-1970. These concentrations compare with a background of 0.0016 pCi/m^3. The study determined that most of the elevated alpha activity at the Pocatello Sewage Treatment Plant was ^{210}Po. The ^{210}Po concentrations averaged 88% of the gross alpha activity.

Another study (Eadie and Lambdin 1980) found that airborne concentrations of ^{234}U, ^{238}U, ^{226}Ra, and ^{230}Th at the Pocatello Sewage Plant were higher than at other sites near Pocatello and were about an order of magnitude greater than levels measured at Howe, the "background location." The other sites (B, C, D, and E) in Figure 4.2 were at least 3 miles from the phosphate plants and not in the predominant downwind direction. Concentrations at these sites are similar

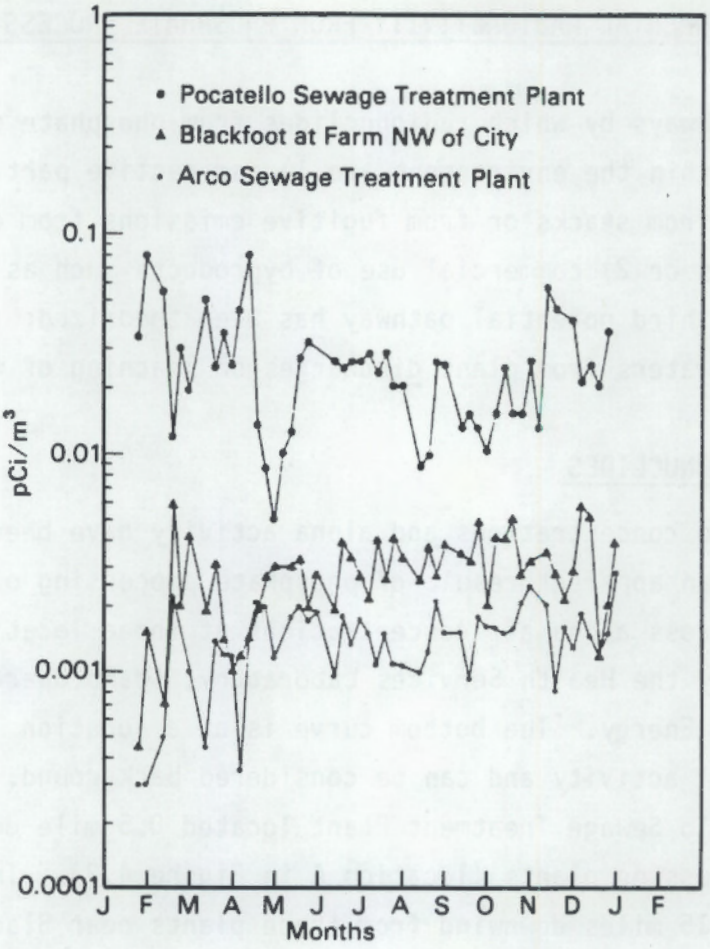


FIGURE 4.1. Gross Alpha Air Concentrations, 1974 (Boothe 1977)

to background levels reported for Chicago and New York but a factor of 3 higher than measured at Howe. Average annual lung dose in mrem/year for these sites is shown in Table 4.1. The ^{210}Po and ^{210}Pb concentrations were not reported because of technical difficulties encountered during the study; however, the authors concluded that these two radionuclides would be statistically above background.

Elevated ^{210}Po levels have also been detected in soil in the Pocatello area (McNabb, Kirk, and Thompson 1979). Soil samples were collected at four locations (F, G, H, and I in Figure 4.2) in the vicinity of the phosphate processing plants where land appeared not to have been disturbed by

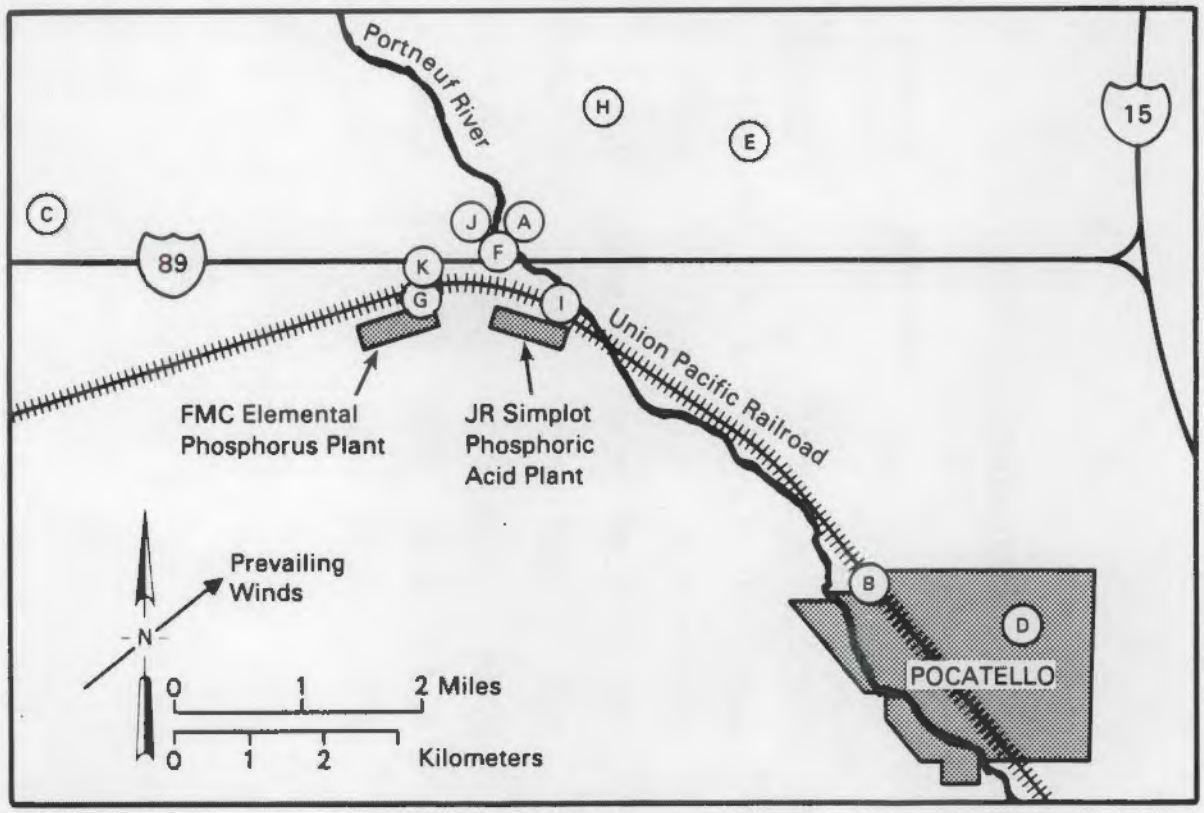


FIGURE 4.2. Sampling Locations in the Pocatello Area

industrial or farming activities. Subsurface samples contained ^{238}U in equilibrium with its progeny, ^{234}U, ^{230}Th, and ^{210}Po, at about 1 pCi/g of sieved soil. Thorium-232 and its progeny, ^{228}Th, were also in equilibrium at about 0.5 to 1.0 pCi/g depending on the site. Surface samples contained ^{238}U and progeny in concentrations significantly higher than the 1 pCi/g found in the subsurface soils, as shown in Table 4.2.

As indicated in Table 4.2, the ^{210}Po values exceeded the equilibrium with ^{238}U at the first three sites, suggesting that the ^{210}Po contamination came from the FMC calciner stack. The difference is most apparent at Site G, the closest site to the FMC elemental phosphorus plant. The high ^{238}U content shown for Location I may have resulted from blowing phosphate ore dust (McNabb, Kirk, and Thompson 1979).

In another study, ^{210}Po concentrations were determined for soil, vegetation, and deer mouse tissues collected at a solid radioactive waste disposal facility near Idaho Falls, near the phosphate plants at Pocatello, and at two

TABLE 4.1. Average Annual Lung Dose (mrem/yr) for Insoluble Radionuclides (Eadie and Lambdin 1980)

Radionuclide	A Sewage Plant	B Hayes Fire Station	C Airport	D Courthouse	E Chubbuck School	Howe, Idaho	Arco, Idaho	New York City (NCRP 1975)	Argonne National Lab, Chicago (NCRP 1975)
^{238}U	1.8	0.32	0.32	0.29	0.58	0.10	0.17	0.08	0.38
^{234}U	2.3	0.37	0.41	0.41	0.63	0.12	0.23	0.93	0.44
^{230}Th	3.0	0.54	0.48	0.48	0.82	0.24	0.31	No data	0.15
^{226}Ra	2.7	1.0	0.56	1.7	1.8	0.28	0.26	0.37	No data
^{235}U	0.13	0.033	0.061	0.11	0.068	0.023	0.06	0.04	0.023
^{232}Th	0.22	0.099	0.11	0.045	0.14	0.14	0.14	No data	0.090
Total Dose (mrem/yr)	10	2.4	1.9	3.0	4.0	0.90	1.1	1.4	1.1

TABLE 4.2. Radionuclide Activity of Surface Soil Samples from the Pocatello Area, pCi/g (McNabb, Kirk, and Thompson 1979)

Location	^{238}U	^{234}U	^{230}Th	^{210}Po	^{232}Th	^{228}Th
F	2.2	2.2	3.1	3.8	0.7	0.8
G	2.9	2.6	1.1	8.0	0.4	0.4
H	1.4	1.5	1.2	2.8	0.7	0.8
I	13	13	9	9	0.5	0.5

rural areas in southeastern Idaho (Arthur and Markham 1984). Polonium concentrations in the media sampled near the radioactive waste disposal facility were equal to or less than the values for the rural areas. Concentrations of ^{210}Po in soils and deer mouse hide and carcass samples at locations C, J, and K (Figure 4.2) were statistically greater than the other locations. The mean ^{210}Po concentration in soils and deer mouse tissues near the phosphate plants were four and three times greater than the control values. No statistical difference was observed for ^{210}Po concentrations in vegetation at the different sampling locations, which suggests that the deer mouse concentration of ^{210}Po is probably due to contact with contaminated soils. The ^{210}Po concentrations were highest at the sample locations K and J (6.2 and 2.6 pCi/g) downwind of the phosphate plants and lowest at the airport (2.3 pCi/g), 3.2 km (2 miles) west of the plants. The ^{210}Po concentrations in the soils at the background sites, Arco and Idaho Falls, were 1.4 to 1.5 pCi/g.

Severson and Gough (1976) found that the processing plants at Pocatello have a direct influence on the elemental content of local soil and vegetation. Samples taken at approximate logarithmic intervals up to 64 km (40 miles) northeast (downwind) and southwest (upwind) of the phosphorus processing plants showed a direct correlation between concentration of certain elements and distance from the processing plants.

Very similar results were obtained by Johnson et al. (1980), who measured the uranium content in soils 10 miles upwind to 10 miles downwind of the phosphorus processing plants in Pocatello. The concentration of uranium in the soils was about 3 ppm (1 pCi/g ^{238}U) in both the surface and subsurface soils upwind and at distances greater than 6 miles downwind. At less than 6 miles

downwind, the uranium concentration increased closer to the plant to a high of 6.9 ppm (~2 pCi/g ^{238}U) at only 0.5 mile from the plants.

4.2 DISTRIBUTION OF RADIONUCLIDES FROM COMMERCIAL USE OF BYPRODUCTS

The byproduct slag from the thermal process plants has been used as a construction material in both Pocatello and Soda Springs. Most of the uranium and radium in the ore are contained in the slag, which then becomes a source of gamma radiation. In addition, ^{222}Rn is produced from the decay of ^{226}Ra. Due to the refractory nature of the slag, the radon-emanating fraction is likely to be low, and radon release may not be significant.

The State of Idaho prohibited use of slag in habitable structures in 1977, but it is still being used for road construction, for railroad bed ballast, and for other general construction uses. In 1974, about 15 Ci of radium were distributed into the environment around Pocatello, as shown in Table 4.3. In the Soda Springs area from 1972-1974, an average of 6 Ci/year of radium was distributed into the environment, as shown in Table 4.4 (Boothe 1977).

During 1977, a gamma survey truck operated by the EPA and the Idaho Radiation Control Office scanned the towns of Soda Springs and Pocatello to detect elevated radiation levels coming from phosphate slag used within structures. A detailed description of the mobile gamma ray logging system is given by Hans et al. (1978). A summary of the results is provided by Peterson (1979).

TABLE 4.3. Distribution of Radium in Environs from the Commercial Use of Slag--Pocatello Area, 1974. Assumes slag contains 35 pCi/g radium (Boothe 1977).

Use	Location	mCi/yr
Roads	Streets of Pocatello	562.9
Roads	Streets of American Falls	251.0
Roads	Bannock County	5,514.2
General construction	Bannock County	886.9
Railroad bedding	Unknown	4,741.0
Farmers	Bannock County	1,885.7
Concrete	Pocatello	91.6
Miscellaneous	Unknown	1,519.0
	Total	15,462.0

TABLE 4.4. Distribution of Radium in Environs from the Commercial Use of Slag--Soda Springs Area, 1972-74. Assumes slag contains 35 pCi/g radium (Boothe 1977).

Use	Location	mCi/yr
Roads	Streets of Soda Springs	46.8
Roads	Streets of Montpelier	37.2
Roads	Caribou County	2,131.2
Concrete	Caribou County	265.2
General Construction	Caribou County	732.9
Railroad bedding	Unknown	2,857.7
	Total	6,071.0

Of the 1,082 structures in Soda Springs, 340 showed elevated gamma levels. In Pocatello, 2,035 structures with elevated gamma levels were detected. The survey was unable to positively identify slag-containing structures because of the added presence of slag in streets, sidewalks, and driveways.

A door-to-door survey in Soda Springs checked 278 structures suspected of being built with slag. Of these, 156 (56%) were found to contain slag. Observed average gamma levels in these structures ranged from 11 to 50 μR/h including background. The mean value was slightly over 28 μR/h. Normal background was approximately 10 μR/h.

During the summer of 1978, radon progeny measurements were made by the EPA and the Idaho Radiation Control Office in 107 of the locations where slag was used in construction. The measurements ranged from 0.0005 to 0.046 working level (WL) with a median value of 0.0053 WL. Estimated normal background is 0.005 WL; the EPA guideline is 0.02 WL. Radon measurements made at the same locations at the same time ranged from 0.05 to 7.42 pCi/l with a median value of 1.2 pCi/l. When comparing the radon and radon progeny data from each location, the median equilibrium was about 43%. The value of these determinations is limited as they are grab samples taken during the summer, and it is well known that radon and radon progeny levels vary greatly with barometric pressure, temperature, ventilation, season of the year, etc.

Year-long, integrated WL samples were made at four locations in Soda Springs where slag was used. The annual average values were 0.0077 WL, 0.0113 WL, 0.0089 WL, and 0.0141 WL. Results from the year-long samples also

indicate a slight increase during the summer, in contrast to the traditional understanding of seasonal variations in radon and radon progeny levels (Peterson 1979).

A study of slag use in homes in Butte, Montana, found the radon exhalation rate from slag to be very low, 18 to 30 $pCi/m^2/min$ compared with 3400 to 3822 $pCi/m^2/min$ for phosphate ore, even though the ^{226}Ra content was essentially the same for the ore and the slag (Lloyd 1983). Radon exhalation was lower from slag than from natural decomposed granite soils in the Butte area. The author concluded that the radon exhalation from slag did not contribute to the elevated radon levels in Butte homes.

An extensive door-to-door survey similar to the Soda Springs survey has not been conducted in Pocatello; however, gamma levels have been reported for a variety of locations in Pocatello (Table 4.5). Table 4.5 gives the background level as 6 μR/h. Others have reported it as high as 11 μR/h (Hans et al. 1978). Readings throughout the city have been as high as 44 μR/h (Hans et al. 1978) and average about 15 μR/h as indicated in Table 4.5.

Tables 4.3 and 4.4 showed that the primary use of slag was for road construction. In addition to elevated gamma levels shown in Table 4.5, the possibility of contamination of soils next to roadways exists. Surface soils next

TABLE 4.5. Gamma Survey, Pocatello Area [gross readings taken 1 m (3.3 ft) above surface (Boothe 1977)]

Location	μR/h
Average background in city	6
Indian Hills School (new asphalt)	9
Cedar Hills Drive	12
Railroad grade throughout city	9
Second Avenue along Ross Park	12
South end of Arthur Avenue	12
Benton Avenue from Arthur to Fifth	23
Yellowstone Avenue	18-35
Pocatello shopping mall parking lot	37
Holiday Inn parking lot	29
Holiday Inn sidewalk	10
Wilcox School area	32
Chubbuck School playground	26
Approximate average	15

to road beds were found to have elevated levels of uranium. Up to 18 ppm of uranium was measured in soils 1 m (3.3 ft) from roads paved using slag. This amount compares with 3.6 ppm background levels. The roads were paved with a 94% slag/6% asphalt mixture, a typical aggregate/asphalt mix for road paving. The study found that contamination of the soils is probably due to abrasion of slag particles in the road rather than from leaching.

Aerosol samples were taken 1.5 m (4.9 ft) above ground next to a roadway paved with slag (Melville 1980; Melville et al. 1981). The dust particles >5 μm that were collected from the aerosol samples were analyzed, and the uranium content calculated to be 0.043 pCi $^{238}U/m^3$. These large particles are not an important radiological health consideration. Only smaller particles 0.01 to 5.0 μm are retained in the respiratory system. Based on the results for the larger particles, the ^{238}U content in the small particle range (0.4 to 5.0 μm) was estimated to be less than 0.03 $\mu g/m^3$ of air. The authors concluded that this level was sufficiently low as to not cause serious concern but recommended that uranium progeny products such as ^{226}Ra, ^{222}Rn, ^{210}Po, and ^{210}Pb also be evaluated.

4.3 RADIONUCLIDES IN WATER

No contamination of surface or ground waters in southeastern Idaho has been linked directly to phosphate processing plants. However, elevated beta activity has been found in the Old Airport well, 1 mile north of the FMC thermal process plant near Pocatello (Boothe 1977). The beta activity has been identified as ^{40}K. If this potassium, which is highly soluble, is entering the ground water from phosphate ponds or discharges, then the possibility of radionuclide contamination exists as well (Boothe 1977).

The ^{40}K concentration and beta activity for several wells near the FMC plant are listed in Table 4.6. The ^{40}K concentration in the Old Airport tap jumped dramatically between 1972 and 1973. Furthermore, the concentration in the FMC well increased between 1973 and 1975. The other wells in the area show no increase in ^{40}K concentration over the same time.

It has been theorized that the elevated ^{40}K levels may be due to leaching from waste ponds at the FMC plant. While the hydrology for the area is not

TABLE 4.6. Beta Activity and ^{40}K Concentration in Wells Near the FMC Pocatello Plant (Baston 1976; Boothe 1977)

Well	^{40}K, pCi/l				Beta Activity, pCi/l	
	1970	1972	1973	1975	1975	(a)
Old Airport Tap	60.6	11.8	131.0	138.5	113.0	198.7
Batiste Spring	7.3	19.8	10.7	18.0	15.0	-
Pocatello Airport	-	-	-	-	-	2.4
Lindley Well	-	-	-	-	-	6.1
FMC Wells #1-5	5.9-11.5	7.8-12.0	5.8-14.8	9.7-20.9	-	-
FMC Well #6	24.2	-	18.9	86.5	-	-

(a) Data taken from Boothe (1977); no date given.

well established, the direction of flow is generally accepted as northerly. The Old Airport tap (close to Location K in Figure 4.2) is less than 1 mile north of the FMC plant. The Lindley well and Batiste Springs (close to Location F in Figure 4.2) are about 1 mile from the plant. The Pocatello Airport well is 2 miles west northwest of the plant. The FMC wells are all located just north of waste ponds at the plant. The fact that other wells do not show contamination suggests that the mechanism of hydrologic transport is not appropriate or is limited to specific isolated coordinates (Baston 1976).

In evaluations of surface water, the Portneuf River has been sampled upstream and downstream of the two phosphate processing plants near Pocatello (Eadie and Bernhardt 1977; Eadie, Bernhardt, and Boysen 1978). Prior to the analysis, the suspended solids were allowed to settle out, and both the liquid and the insoluble material were analyzed. The radionuclide concentrations in the upstream and downstream liquid were basically identical. However, the uranium and thorium contents of the suspended solids in the downstream sample were an order of magnitude higher than in the upstream sample. The radionuclide content of both the liquid and suspended solids is well within the range of naturally occurring radioactivity. Any future efforts to evaluate the radionuclide content in rivers and streams near phosphate plants should include sediment sampling (Eadie and Bernhardt 1977; Eadie, Bernhardt, and Boysen 1978).

5.0 ESTIMATE OF HEALTH IMPACT

The two areas are of primary concern with respect to the health impact of phosphate rock processing and byproduct utilization in southeastern Idaho are as follows:

- inhalation of airborne radionuclides emitted from calciner stacks
- gamma and radon progeny exposure associated with use of byproduct slag in construction.

5.1 AIRBORNE RADIONUCLIDES FROM THERMAL PROCESS PLANTS

The EPA has estimated the radiation exposure and health risks associated with the elemental phosphorus plant operations in Pocatello and Soda Springs (EPA 1984a). Doses were calculated using the AIRDOS-EPA/DARTAB computer model developed by Oak Ridge National Laboratory (EPA 1979; Begovich 1981). Data required as input included the calciner stack emissions (see Table 3.9), population distributions, and meteorological data as shown in Table 5.1. The estimated radiation dose to lung and the estimated fatal cancer risks are shown in Table 5.2. The airborne emissions and the risk of fatal cancer for the thermal process plants are much higher than estimated for the wet process plants. (The risks from wet process plants were made using a generic Florida-based plant.)

Emission limits as low as 1.0 Ci/year of ^{210}Po were considered by the EPA (Stula et al. 1983), but in 1985 the EPA issued a standard of 21 Ci/year of ^{210}Po emissions from calciner stacks at elemental phosphorus plants. This

TABLE 5.1. Input Data used in Dose and Risk Calculations (EPA 1984a)

Plant	Calciner Emissions, Ci/yr		Number of Persons within 80 km	Source of Meteorological Data
	^{210}Po	^{210}Pb		
FMC Pocatello	9	0.1	140,000	Pocatello Airport
Monsanto Soda Springs	21	5.6	80,000	Pocatello Airport

TABLE 5.2. Estimate of Radiation Dose and Fatal Cancer from Airborne Radionuclide Emissions from Calciners at Elemental Phosphorus Plants (EPA 1984a)

Plant	Radiation Dose to Lung		Fatal Cancer Risks	
	Nearby Individuals, mrem/yr	Regional Population, person-rem/yr	Lifetime Risk to Nearby Individuals	Regional Population Fatal Cancers/yr
FMC Pocatello	290	1170	0.0005	0.027
Monsanto Soda Springs	610	750	0.001	0.018

standard would not decrease emissions but would assure that emissions would not increase. The decision to adopt the higher standard was based on the low aggregate risk and the high cost of controls versus the public health benefits (EPA 1984a).

Because of the complex topography of the area and the lack of appropriate meteorological data, in particular for Soda Springs where Pocatello Airport data were used for the analysis, dose and risk estimates using AIRDOS-EPA are probably in error. Further analysis and modeling using appropriate meteorological data are needed.

5.2 GAMMA AND RADON/PROGENY EXPOSURE FROM SLAG

As discussed earlier, a survey of 278 homes in Soda Springs suspected of being built with slag found the mean gamma level to be 28 μR/h compared with a normal background of 10 μR/h. Using a cancer induction risk from gamma radiation of 0.3 per 1000 man-rem, Peterson (1979) estimated about 5 excess cancer deaths/70 years based on the current population in Soda Springs. Based on these results, Peterson concluded that it would be prudent to continue the ban on construction use of slag in structures but that remedial action on existing structures was not warranted.

Grab-sample radon progeny WL measurements were made at 107 slag-contaminated structures in Soda Springs during the summer of 1978 (Peterson 1979). These samples had a mean average of 0.0053 WL compared with normal background of 0.005 WL. Four year-long integrated WL samples were taken with a

radon progeny integrating sampling unit (RPISU) in Soda Springs; the average values were 0.0077, 0.0113, 0.0089, and 0.0141 WL. The year-long samples are generally believed to be more accurate than grab samples, particularly samples taken during the summer as in Soda Springs (Ronca-Battista, Magno, and Windham 1986). If the year-long WL values are used, the mean value is 0.009 WL, and an estimated 23% of the structures would exceed the EPA guideline of 0.02 WL (Peterson 1979). Clearly, additional sampling is needed to determine if radon represents a health impact in Soda Springs. Based on the measurements of radon exhalation from slag by the Montana Department of Health and Environmental Sciences (Lloyd 1983), it is unlikely that slag, because of its low emanating power, is the cause of elevated radon levels in Soda Springs homes.

5.3 RADIONUCLIDE EMISSIONS FROM WET PROCESS PLANTS

The EPA (1984a) also estimated the health impact of radionuclides in particulate emissions from a reference wet process phosphoric acid plant. Emissions of primary concern for the reference plant were from diammonium phosphate (DAP) and TSP dryers. Previous estimates of radionuclide emissions from the J. R. Simplot Co. plant in Pocatello were very similar to those used by the EPA for their reference plant (Table 5.3) (Eadie, Bernhardt, and Boysen 1978). The radiation dose and fatal cancer risk to nearby individuals calculated for the reference plant and shown in Table 5.4 can be used as a preliminary estimate for the J. R. Simplot Co. plant.

The 1975 study of the J. R. Simplot Co. plant (Eadie, Bernhardt, and Boysen 1978) identified the TSP dryer as the primary source of emissions. The TSP plant has been modified since this study was conducted, and is no longer a significant source of airborne radionuclides. The 1975 study did not include calciner emissions, but a recent study has shown these emissions to be low (EPA 1987). Current emissions from the J. R. Simplot Co. facility may be significantly lower than previously reported. As a result, the numbers in Table 5.4, already several orders of magnitude lower than those in Table 5.1 for the thermal process plants in Idaho, probably overestimate the risk for the J. R. Simplot Co. facility.

TABLE 5.3. Airborne Radionuclide Particulate Emissions from Wet Process Phosphoric Acid Plants, mCi/yr

	EPA Reference Plant[a]	J. R. Simplot Co. Plant, Pocatello[b]
^{238}U	6.7	5.10
^{234}U	6.7	5.10
^{230}Th	6.7	<5
^{226}Ra	0.97	5.10
^{210}Pb	3.4	5.10
^{210}Po	3.4	<5

(a) EPA (1984a).
(b) Eadie, Bernhardt, and Boysen (1978).

TABLE 5.4. Health Impact of Airborne Radionuclide Particulate Emissions from a Wet Process Phosphoric Acid Plant (EPA 1984a)

Source	Radiation Dose to Nearby Individuals, mrem/yr		Lifetime Fatal Cancer Risk to Nearby Individuals
DAP and TSP Dryer Emissions	Lung	1.2	2×10^{-6}
	Endosteum	2.2	
	Red Marrow	0.15	
	Kidney	0.07	

6.0 CONCLUSIONS AND RECOMMENDATIONS

Dispersion of radionuclides into the environment by phosphate processing plants has been the subject of much research and study; however, there is still no clear understanding of the subsequent total dose. The following conclusions and recommendations were drawn from the review of pertinent literature.

Two primary radionuclide pathways to the environment have been identified:

- airborne release of volatile radionuclides, primarily ^{210}Po, from calciner stacks at the two elemental phosphorus plants
- byproduct slag use as an aggregate for construction in Soda Springs and Pocatello.

The health impact of both scenarios has been estimated by the EPA (for ^{210}Po) and the Idaho Department of Health and Welfare (for slag).

Two other radionuclide pathways to the environment that require further study were identified:

- radon exhalation from phosphogypsum and ore piles
- contamination of surface and ground waters by wastewater from the processing plants.

Recommendations for further study to provide a basis for a complete dose assessment are as follows:

- Determine gamma exposure caused by use of slag in roads, sidewalks, etc. in the Pocatello and Soda Springs area.
- Measure radon flux from phosphogypsum, ore, and slag piles.
- Measure airborne particulate activity in Pocatello and Soda Springs and correlate results with airborne emissions from phosphate processing plants (calciner off-gas, dust from ore piles, etc.) through air dispersion modeling.
- Determine the background contribution of ^{210}Po, ^{210}Pb, ^{222}Rn, ^{238}U, ^{226}Ru, and gamma radiation to the total radiation dose to the communities of Pocatello and Soda Springs.

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APPENDIX A

^{238}U AND ^{232}Th DECAY SERIES

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^{238}U AND ^{232}Th DECAY SERIES

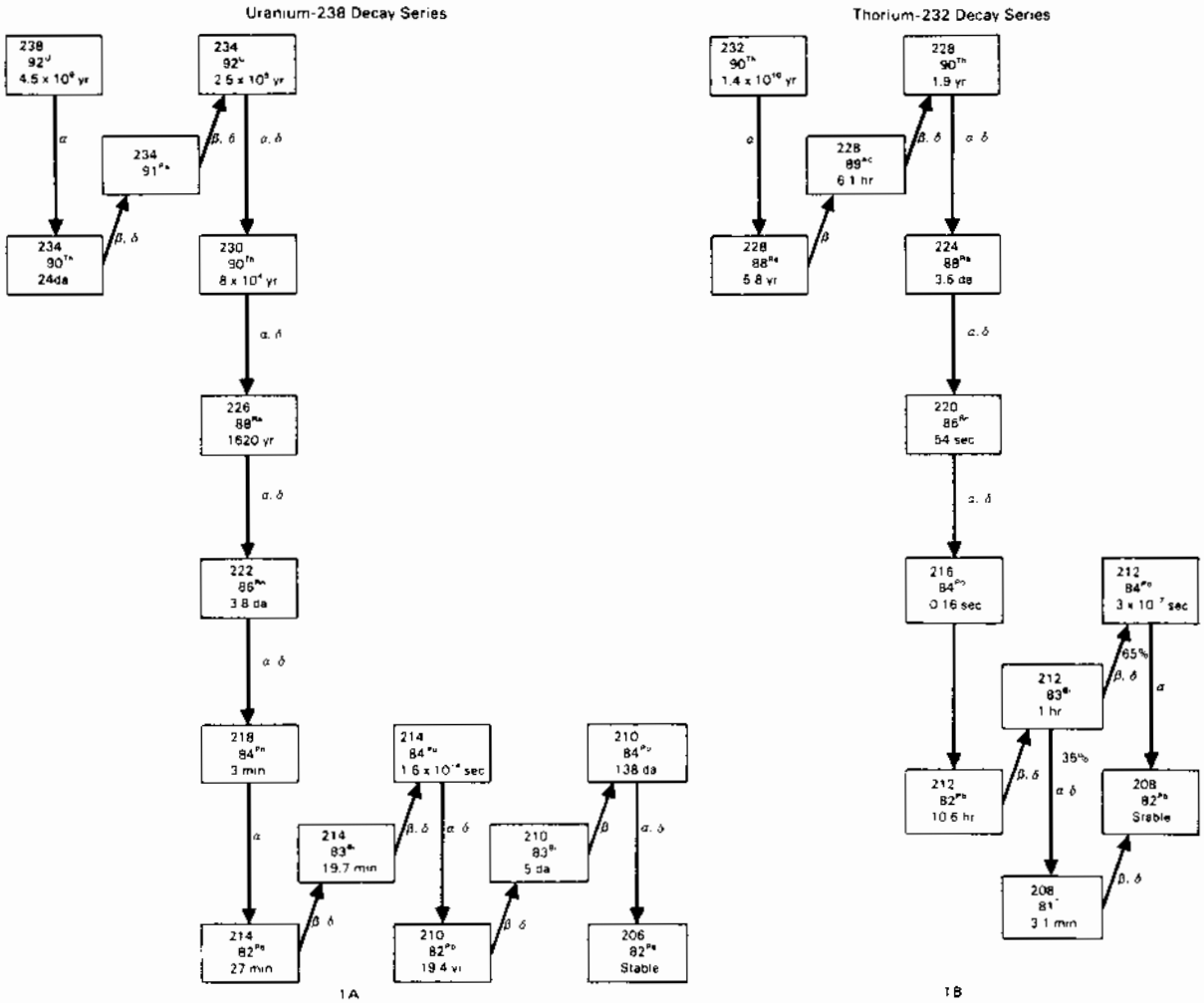

FIGURE A.1. ^{238}U and ^{232}Th Decay Series

APPENDIX B

GLOSSARY OF TERMS AND ABBREVIATIONS

APPENDIX B

GLOSSARY OF TERMS AND ABBREVIATIONS

AIROOS-EPA/DARTAB	computer model used by the EPA to predict offsite concentrations of radionuclides from sources of radioactive materials
alpha	positively charged particles emitted from radioactive elements; particles have very low penetrating power and can be stopped by a sheet of paper
beta	electrons emitted by radioactive elements; beta radiation has a higher penetrating power than alpha radiation
calciner	equipment used to drive off volatile material in the phosphate rock
Ci	(curie) unit of radioactivity equal to 3.7×10^{10} disintegrations per second of any radionuclide
DAP	diammonium phosphate fertilizer
dscf	dry standard cubic feet
dscfm	dry standard cubic feet per minute
EPA	U.S. Environmental Protection Agency
EPA Method 5	standard EPA method for stack particulate sampling
g	gram
gamma	electromagnetic radiation with very high penetrating power
gr	grains (7000 grains = 1 lb)
grab sample	instantaneous sample of air that is analyzed for radon daughter products and converted to the units of working level
h	hour
H_3PO_4	phosphoric acid ($P_2O_5 + 3H_2O \rightarrow 2H_3PO_4$)
K	potassium
kg	kilogram

l	liter
lb/h	pounds per hour
m^2	square meters
m^3	cubic meters
mCi	millicurie, 10^{-3} Ci
mrem	millirem, 10^{-3} rem
P_2O_5	phosphorus pentoxide
P_4	elemental phosphorus
Pb	lead
pCi	picocurie, 10^{-12} Ci
ppm	parts per million
phosphate	rock containing 24% to 34% P_2O_5, the remainder being primarily calcium; also contains small quantities of radionuclides
phospho-gypsum	solid byproduct of the wet process; primarily calcium sulfate
Po	polonium
Ra	radium
radionuclide	radioactive element
radon emanating fraction	the fraction of radon produced from radionuclide decay that escapes to the pore spaces
radon progeny	decay products of radon
rem	a measure of radiation dose devised to take into account the biological effects of different types of ionizing radiation on humans
Rn	radon
RPISU	radon progeny integrating sampling unit
scf	standard cubic feet

scfm	standard cubic feet per minute
slag	solid byproduct of the thermal process, primarily calcium silicate
T	metric tons unless otherwise indicated, 1 T = 2200 lb
Th	thorium
thermal process	processing using high temperatures to convert phosphate rock to elemental phosphorus
TPY	tons per year
TSP	triple superphosphate fertilizer
U	uranium
wet process	process to convert phosphate rock to liquid phosphoric acid
WL	(working level) alpha energy of 1.3×10^5 MeV in 1 liter (l) of air, equivalent to 100 pCi/l of radon in equilibrium with its progeny
µCi	microcurie, 10^{-6} Ci
µm	micrometer, 10^{-6} meter
µR	microroentgen, 10^{-6} R (Roentgen is a unit of exposure of X or gamma radiation based on the ionization that these radiations produce in air.)

APPENDIX C

BIBLIOGRAPHY

APPENDIX C

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