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An Integrated Assessment of the Phosphate Industry

M. T. Ryan
S. J. Cotter

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AN INTEGRATED ASSESSMENT OF THE PHOSPHATE INDUSTRY

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Health and Safety Research Division

Prepared for:

U. S. DEPARTMENT OF ENERGY
Assistant Secretary for Environment
Office of Technology Impacts

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Summary

The continued growth in domestic phosphate fertilizer production combined with price increases in uranium makes the extraction of uranium from wet process phosphoric acid economical. A review of actual and proposed phosphate mining activities in the United States was provided and contemporary phosphate industry production capacities were analyzed for growth trends. It was determined that the average annual capacity growth rate since 1955 is 7%. Uranium production capacity estimates were made and were compared to actual uranium production of recent years. It was optimistically estimated that with the continued development of (improved) recovery from wet process phosphoric acid and the development of technologies for the recovery of uranium from marketable phosphate rock that uranium production may increase to 9600 short tons per year. This value is approximately 3 times the average level of uranium production predicted assuming realistic expansion of existing uranium coproduction from the phosphatic resources. Production from conventional uranium mining and milling averaged 11,000 short-tons per year during the period 1965 to 1971.

There are three significant areas of radiological impacts, namely occupational impacts associated with phosphate facility operations, nonoccupational impacts associated with facility operations, and nonoccupational impacts associated with the use of reclaimed lands. Average occupational gamma radiation doses were estimated to be 1.5 times above

the background gamma radiation dose (∿ 70 millirad/year) and average lung doses were estimated to be 2 to 4 times above the background lung dose (19 to 64 millirad/year). A review of work by the National Institute for Occupational Safety and Health (NIOSH) shows that occupational impacts must be considered for such pollutants as chromium, vanadium, arsenic, uranium, flourine, cadmium, respirable silicon, and respirable dust. Concentrations of these pollutants were measured at four Florida phosphate facilities and these measurements are compared with Occupational Safety and Health Administration limits and NIOSH proposed standards.[20-24]

Calculations for the population dose commitments associated with the routine releases of radionuclides from phosphate facilities were estimated by normalizing the population dose to individual body compartments via individual exposure pathways to the total body dose via all exposure pathways. The radionuclides which contributed the largest portion of the dose to a given compartment via a specific exposure pathway were also identified. This method of dose estimation allows identification of radionuclides and exposure pathways of highest importance and provides a tool to aid in the design of in-plant and environmental airborne effluent sampling programs. The impacts of waterborne effluent releases to ground water are reviewed and it is estimated that effluent discharges from slime ponds associated with beneficiation and with wet process phosphoric acid processing are not as significant as other impacts.

The application of uranium-bearing phosphate fertilizer to crop lands and the subsequent ingestion of crops grown on these lands was also assessed. It was determined that any ingestion of uranium via this pathway is very small when compared to the average dietary intake of uranium.

Hundreds of measurements of radon daughter product air concentrations made in structures built on reclaimed phosphate lands have been reviewed. On these lands, 2% of measured values were greater than 0.05 working levels (WL), 65% were between 0.01 and 0.05 WL and 33% were below 0.01 WL. Comparisons to working level value distributions for structures on undisturbed lands, have been given elsewhere and indicate that an enhancement of the natural radiation environment has occurred resulting in higher dose to populations living on reclaimed lands. Differences in the results of various studies are discussed.

Impacts associated with the phosphate industry are compared with similar impacts resulting from uranium mining and milling. These comparisons indicate that while impacts (both detrimental and beneficial) are associated with corecovery of uranium from wet process phosphoric acid significant impacts are independent of uranium corecovery.

1. INTRODUCTION

The phosphate industry in the United States includes three major activities, namely, mining and milling of phosphate rock, phosphate product manufacture, and phosphate product use. It has been known for more than 20 years that phosphatic materials contain uranium, thorium, and their decay products in greater than background amounts.[1] This assessment of the radiological impacts associated with the redistribution of radioactive components of phosphate materials may provide insight into the effects of uranium extraction from phosphate materials for use in the nuclear fuel cycle. The United States Department of Energy (DOE) has the responsibility to ensure that environmental, health, and safety aspects associated with the development of energy resources are addressed as part of the development process. Thus, as DOE is exploring and assessing the uranium resources under the National Uranium Resource Evaluation Program (NURE), the Office of Technology Impacts is sponsoring this assessment to address the environmental health and safety impacts associated with the uranium potential of the phosphate industry.

Phosphate mining activities have been steadily increasing for the last two decades. Values for production of P_2O_5 (short tons/year) and marketable rock (short tons/year) since 1955 are given in Table 1.[2] (Marketable rock is used to denote ore that has been beneficiated.* If P_2O_5 content is high enough, beneficiation may not be required.) This growth in production can be described by a linear equation. For P_2O_5, the equation

---

*Beneficiation is a physical separation process using screen flotation to remove nonphosphate components.

Table 1. Annual P_2O_5 and marketable rock production figures[a]

Year	P_2O_5 x 10^3 tons	Marketable rock x 10^3 short tons	
		Fla. & N.C.	U.S. total
1955	2284	9797	13737
1960	2572	13800	19618
1961	2645	15444	20786
1962	2807	15623	21708
1963	3073	16343	22238
1964	3378	19161	25715
1965	3512	21563	29482
1966	3897	29827	39044
1967	4305	31910	39700
1968	4453	33032	41251
1969	4666	29930	37725
1970	4574	31278	38739
1971	4803	32151	38886
1972	4864	34121	40831
1973	5085	34427	42137
1974	5099	36980	45686
1975	4511	40699	48816
1976	5215		
1980	6040[b]	48014[b]	57465[b]
1985	6848[b]	56239[b]	66595[b]

[a]Harre, E. A., Goodson, M. N., and Bridge, J. D. *Fertilizer Trends*. 1976.

[b]Predicted values.

$$\text{short tons} \times 10^3 \text{ of } P_2O_5 \text{ produced per year} = 0.16\,(\text{year} - 1955) + 2.00 \qquad (1)$$

describes P_2O_5 production with a correlation coefficient of $r = 0.95$. For total U.S. production of marketable rock, the equation

$$\text{short tons} \times 10^3 \text{ of marketable rock produced per year} = 1826\,(\text{year} - 1955) + 11815 \qquad (2)$$

describes marketable rock production with a correlation coefficient of $r = 0.95$. The average annual growth in marketable rock production has been 7% since 1955. Assuming the characteristics of phosphate supply and demand do not change, these equations could be used to predict phosphate production for the near future. Values of P_2O_5 and marketable rock production have been estimated for 1980 and 1985. These values are also given in Table 1.

The data used in these regression analyses are from a summary of fertilizer production figures from the U.S. Department of Agriculture (USDA) and the U.S. Bureau of Mines (USBM) as compiled by the Tennessee Valley Authority (TVA).[2]

Current phosphate mining activities can be assessed using industrial annual production capacity data for 1978. Actual annual production figures vary from year to year, and use of the phosphate industry's production capacity may provide a better basis for comparison. Production capacity estimates for 1978 are given below.

State	Percentage of total	Short tons of phosphate rock
Florida	77	51×10^6
Western states (Idaho, Utah, California, Montana)	11	7×10^6
North Carolina (7.5%)	12	8×10^6
Tennessee (4.1%)		
Total U.S. production capacity 1978	100	66×10^6

Major new phosphate mining activities are planned in Idaho, Utah, and California* with production estimates of 15×10^6 short tons of product rock per year, 8×10^5 short tons of product rock per year, and 6×10^5 short tons of product rock per year, respectively.[3-5]

These proposed activities represent approximately a 25% increase over present production levels and could serve as replacement for current reserves as they are depleted or as added production capacity if demand increases significantly.[6]

The uranium concentration in these phosphate resources varies from 50-300 ppm with an average of approximately 100 ppm. In the Florida land pebble deposits in the northern and central counties, the uranium concentration (U_3O_8) is about 100-200 ppm, while in Florida hard rock deposits, the U_3O_8 concentration is approximately 100-300 ppm. In Tennessee and North Carolina brown rock, the U_3O_8 content is about

---

*It is assumed that western ores are not beneficiated even though some beneficiation may be performed.

5-20 ppm, while western states rock contains between 50-100 ppm of U_3O_8.[7] Expressing all values as pounds of U_3O_8 per short ton of rock yields the following results.

State	Pounds U_3O_8/short ton of rock
Florida	
land pebble deposits	0.2 to 0.4
hard rock deposits	0.2 to 0.6
Tennessee and North Carolina	0.01 to 0.04
Western states	0.1 to 0.2

The total uranium resource based on phosphate rock production capacity can be estimated using regional phosphate rock production capacity percentages and average regional U_3O_8 content data.

To achieve this estimate, production capacities of phosphate rock must be realized and uranium extraction must be near 100%. This latter assumption is optimistic but provides an upper bound for estimates of uranium production and as such it provides a bound for use in radiological and nonradiological impact assessments. On this basis the regional and total U_3O_8 production based of phosphate industrial capacities are estimated below.

A recent report DeVoto and Stevens (1979) gives phosphoric acid production capacities and contained uranium for sources in the United States and the free world.[8] Their estimate of annual uranium maximum coproduction from phosphatic resources is approximately 4100 short tons. This estimate parallels our estimate of 9600 short tons per year. This difference of a factor of about two arises from differences in estimated average uranium content in phosphatic resources. Our estimates of contained uranium were based on simple arithmetic averages of reported

uranium concentration ranges while the estimates of contained uranium by DeVoto and Stevens are based on a more comprehensive evaluation of report uranium concentration in phosphatic resources. This difference does not affect subsequent calculations in this report.

State	Average U_3O_8 concentration (lbs U_3O_8/ton of rock)	1978 U_3O_8 production capacity (short tons/year)
Florida	0.35	8900
Tennessee and N.C.	0.03	100
Western states	0.15	600
National total U_3O_8 based on phosphate rock production capacity estimate		9600

Estimates of the recoverable amount of uranium in phosphate resources depends on the level of optimism used in the recovery senario. A clear distinction should be made between the amount of uranium contained in phosphate ore that comes out of the mine, in marketable rock after beneficiation and the fraction of the uranium that is likely to be recovered during phosphoric acid manufacture. The value of 9600 short tons of U_3O_8 per year given above is an estimate of the uranium contained in marketable rock. As stated earlier actual production of U_3O_8 at this level would require 100% recovery of uranium from marketable rock and production at full industrial capacity. Higher and still more optimistic estimates of uranium production may be made if it is recognized that only 42% of the uranium contained in the mined material remains in the

marketable rock.[9,10] The remainder stays with the waste products of beneficiation namely slimes and sand tailings. Recovery from these wastes has not been technologically and economically demonstrated. If these factors are overlooked the higher optimistic estimate of U_3O_8 production would be 22,000 short tons of U_3O_8 per year. This value should be viewed as an optimistic upper bound for uranium production from phosphate resoruces including recovery from wastes with as yet unestablished technology. It must be emphasized that actual uranium co-production from phosphate sources is much lower at the present time. A recent review of uranium recovery processes cited that production of 20,000 short tons of U_3O_8 by extraction from wet-process phosphoric acid could be expected through 1986 and that production of 70,000 short tons of U_3O_8 could be expected through 2000 (at $10 per pound).[11] This latter figure represents an average annual production of approximately 3000 short tons of U_3O_8. For perspective, this can be compared to the average uranium industry production from 1965 to 1971 of about 11,000 short tons of U_3O_8 per year.[12] In 1978, 18,500 short tons of U_3O_8 were produced, a significant increase in uranium production over recent years. Of this amount, 1700 tons of U_3O_8 were produced from mine waters, phosphates, and solution mining.[13] If uranium resources can be extracted more economically from phosphate materials, domestic uranium production may be significantly increased. Any estimates of the magnitude of increased uranium production must include appropriate assumptions. The values above must be used in context with the assumptions given.

Several government agencies and contractors have been involved in the assessment of the impacts of the phosphate industry. Table 2 lists these agencies, the sites of phosphate industrial activity, and the types of assessments that have been performed. The date of report publication is also shown.

Table 2. Summary of federal and state phosphate impact assessments

Agency	Site	Assessment activity
Department of Energy (DOE) Oak Ridge National Laboratory (ORNL)	United States	Integrated Assessment of Phosphate Industry (in progress)
Department of the Interior (DOI) Geological Survey (USGS)	Wyoming	Land Reclamation (in progress)
DOE-ORNL	Tampa, Florida Nichols, Florida Mulberry, Florida Bartow, Florida Texas City, Texas Pasadena, Texas	Radiological Assessment of Formerly Utilized Sites (Phosphate) (in press)
Environmental Protection Agency (EPA)-ORNL	Central Florida	Potential Radiological Impact Assessment (1979)
University of Florida Florida Phosphate Council	Florida	Natural Radiation Exposure Assessment (1978)
EPA	Central Florida	Area Wide Environmental Impact Statement (1978)
EPA	Southeastern Idaho	Final Environmental Impact Statement (1978)
EPA-Office of Radiation Programs	Pocatello, Idaho	Radiological Surveys The Wet Process Plant The Thermal Process Plant (1978)
State of Florida, Land Reclamation Study Commission	Florida	Land Reclamation (1978)
DOI-Bureau of Land Management (BLM)	Vernal City, Unitah County, Utah	Preference Right Lease Technical Examination (1976)
DOI-BLM	Los Padres National Forest, California	Final Environmental Impact Statement (1973)

2. RADIOACTIVE MATERIALS IN THE PHOSPHATE INDUSTRY

Phosphate industrial activities can be conveniently divided into mining and milling of phosphate ores, phosphate product manufacture by the wet process, and phosphate product manufacture by the thermal process.

Phosphate mining and milling is distributed by region as cited earlier, and the techniques used to mine phosphate ores differ from area to area depending upon the type of deposit. In Florida and North Carolina, ore is mined by strip mining techniques that have been described elsewhere.[2,9] The basic technique involves stripping the overburden and mining the matrix with draglines creating mining cuts from 50 to 70 ft in depth and from hundreds to thousands of feet in length. Overburden is stored near mined cuts and used as backfill as subsequent cuts are made. The ore is slurried to 40% solids and is pumped via pipe to beneficiation facilities. Marketable rock is then obtained by screen flotation, separating the sand fraction, clay slimes fraction, and the marketable rock. As a rough "rule of thumb," about one-third of the mined rock goes to each of these beneficiation products.[9] Though this "rule" may not apply to individual facilities or specific processing schemes, it may provide an adequate estimate of industry-wide product and waste yields.

The sand fraction or sand tailings rapidly dewater and are used for backfill, occasionally combined with overburden and clays. Sands are also combined with overburden to construct slime retention dams. Slimes dewater very slowly and must currently be stored in holding ponds for

tens of years. Approximately 10,000 gallons of water are consumed per ton of marketable rock, with process water recycled.

In Tennessee and the western states, mined ore contains sufficient P_2O_5 for direct use. Western states and Tennessee ores are mined and shipped directly to processing facilities by truck or rail car.

Analyses of Florida mining and beneficiation products have been performed, and results indicate that the decay products of the uranium and actinium series are in equilibrium with the parent radionuclides and that the decay products of the thorium series are nearly in equilibrium with the parent, ^{232}Th.

Table 3 gives results of the analyses of the concentrations of ^{238}U, ^{226}Ra, ^{230}Th, and ^{232}Th in the products of beneficiation (i.e., marketable rock, clay slimes, and sand tailings).[9] These results indicate that there is a physical separation of portions of the radioactive component of the ore, but beneficiation does not result in a preferential concentration of specific radionuclides. This observation is important when assessing radiological impacts because uranium and radium impacts have different magnitudes for equal levels of radioactivity. Where it can be shown that levels of uranium and radium radioactivity are consistently at or very near to secular equilibrium, radiological impacts assessments are generally simplified.

The concentrations of ^{238}U, ^{226}Ra, and ^{230}Th are nearly equal in the marketable rock and clay slimes. The concentrations of these radionuclides in sand tailings are approximately six times lower than the concentrations in marketable rock or clay slimes. Most of the ^{232}Th

Table 3. Radionuclide concentrations in phosphatic materials[a]

Material	^{226}Ra (pCi/g)	^{238}U (pCi/g)	^{230}Th (pCi/g)	^{232}Th (pCi/g)
Marketable rock	42	41	42	0.44
Clay slimes	45	44	48	1.4
Sand tailings	8	5	4	0.9

[a] Guimond, R. J., and Windham, S. J., *Radioactivity Distribution in Phosphate Products, By-Products, Effluents and Wastes*, ORP/CSD-75-3 (August 1975).

remains with the clay slimes with about 25% contained in the marketable rock. This partitioning results from the physical separation of ore components with which the radioactive material is associated.

Figure 1 shows a flow chart of phosphate mining and beneficiation, and using the "one-third rule of thumb" bulk material flow estimates are provided.[2] The total quantity of mine rock produced is based on the 1978 marketable rock production capacity estimate of 66×10^6 short tons of marketable rock per year.

For specific assumptions of ore assay, production capacity, and partitioning of bulk material to the three products of beneficiation, total radioactive material flow in mining and beneficiation can be estimated. Regional production capacity estimates (1978); radium, uranium and thorium assay data given in Table 4 and a bulk mass partitioning of one-third of mine rock mass to each beneficiation product were used to estimate the total mined amounts of radioactive materials.[14,15] Total mined radioactivity for 1978 is estimated at approximately 4500 Ci of ^{226}Ra, 4300 Ci of ^{238}U, 4100 Ci of ^{230}Th, and 90 Ci of ^{232}Th. Again, within the limits of experimental measurement, the uranium series is approximately in secular equilibrium.

Wet processing of marketable rock yields phosphoric acid as a primary product, which is then used to manufacture fertilizer materials. Normal and triple superphosphate are manufactured in different ways. Normal superphosphate is produced by partial digestion of phosphate rock with sulfuric acid. This procedure converts the phosphate to a soluble form. Triple superphosphate is produced by redigestion of phosphate

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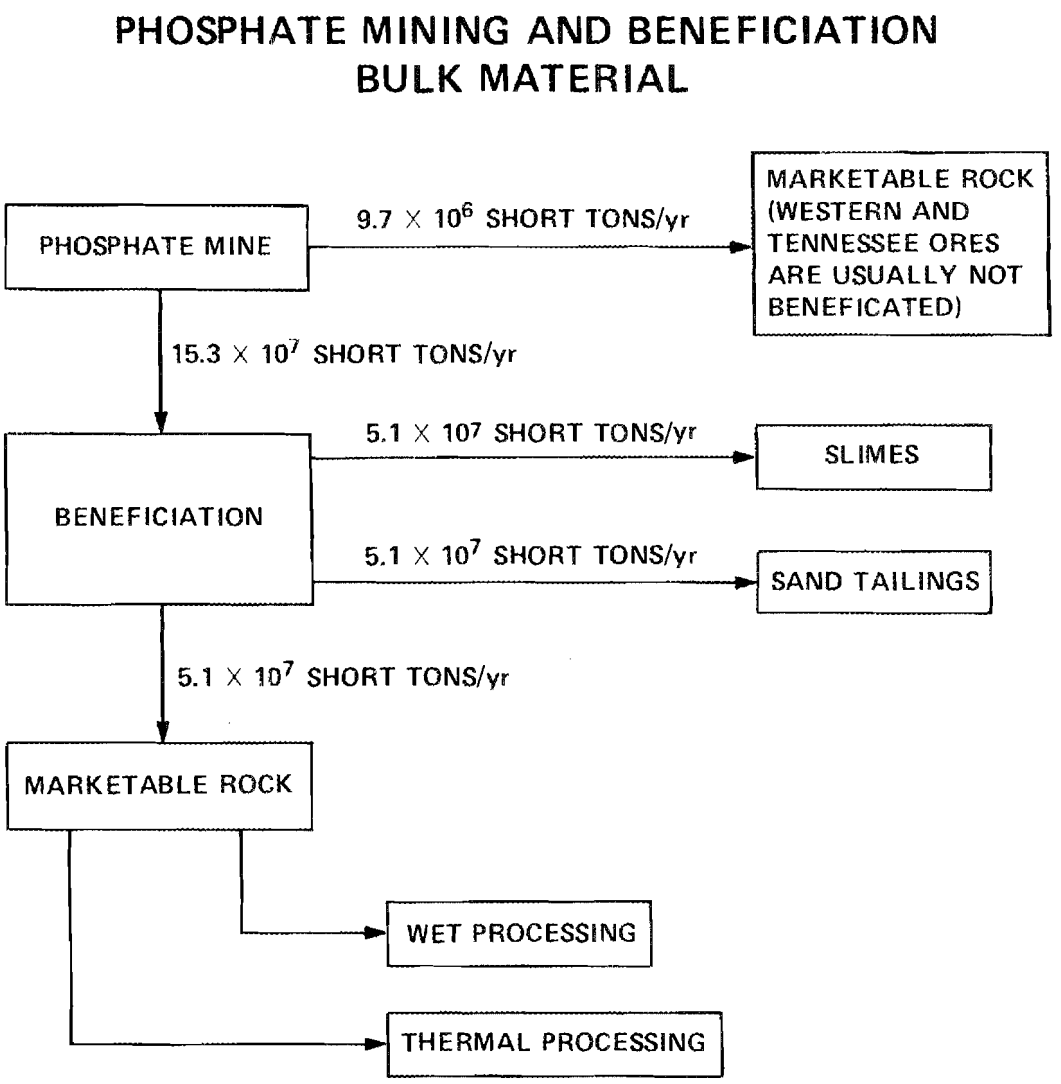

Fig. 1. Bulk material flow in phosphate mining and milling.

Table 4. Regional production capacity estimates for radionuclides in mined phosphate rock

Material	1978 production capacity estimates (short tons/year)	^{226}Ra (Ci/year)	^{238}U (Ci/year)	^{230}Th (Ci/year)	^{232}Th (Ci/year)
Marketable rock					
Western states[a]	7.0×10^6	150	150	150	
Tennessee[a]	2.7×10^6	10	10	10	
Florida	51×10^6	2000	1900	1900	20
Clay slimes[b]	51×10^6	2100	2000	2000	70
Sand tailings[b]	51×10^6	350	250		

[a]Assumes a uranium assay of: Western states - 0.15 lbs U_3O_8/short ton of rock
Tennessee - 0.02 lbs U_3O_8/short ton of rock
and secular equilibrium for ^{226}Ra and ^{230}Th.

[b]Assumes one-third of mine rock to each beneficiation product.

rock with product phosphoric acid to produce a fertilizer with a high phosphate content.

A flow chart showing the percentages of marketable rock required for each wet process product is given in Fig. 2.[14] Estimates of the concentrations of radionuclides in wet process products relative to their concentration in marketable rock are given in Fig. 3. These percentages are calculated,

$$100 \times \frac{\text{measured concentration in fertilizer product}}{\text{measured concentration in marketable rock}} ,$$

and provide an indication of the overall assessment of the enhancement of radionuclide concentrations by fertilizer production. These data indicate that there is a selective separation and concentration of various radionuclides in wet process products and wastes. Initial digestion of phosphate rock for the production of phosphoric acid results in the selective separation of ^{238}U, ^{230}Th, and ^{226}Ra. Most of the ^{226}Ra (80%) goes to gypsum waste, while most of the ^{238}U (86%) and ^{230}Th (70%) goes to phosphoric acid. Further processing results in ^{238}U, ^{230}Th, and ^{226}Ra concentrations in fertilizer products relative to marketable rock also shown in Fig. 3. Though these data do not readily provide an estimate of the total amount of radioactive materials in each product, they do provide a perspective on the technological enhancement of natural radioactive materials in phosphate fertilizers. The analyses from which these percentages were obtained were from several mines, wet process and thermal process facilities, and are assumed to be typical of average radionuclide content.[9,10,16,17]

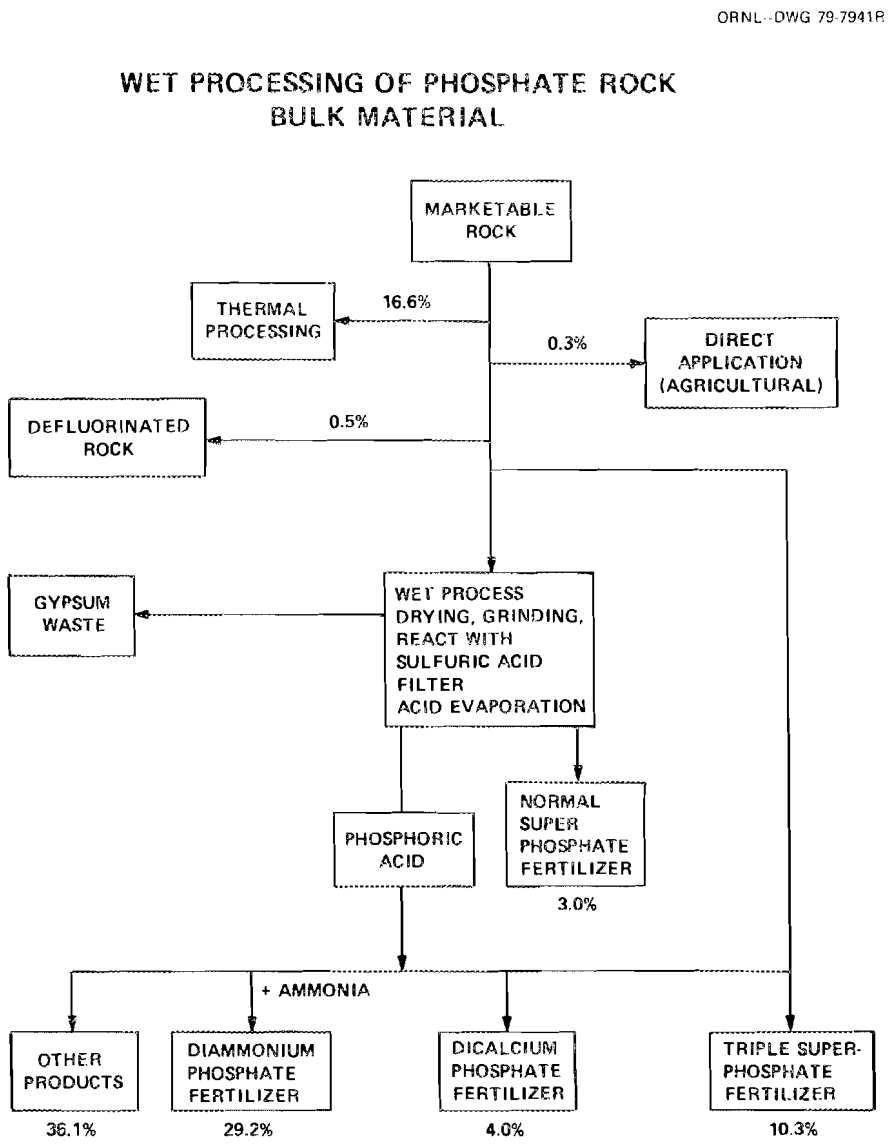

Fig. 2. Marketable rock partitioning to wet process products.

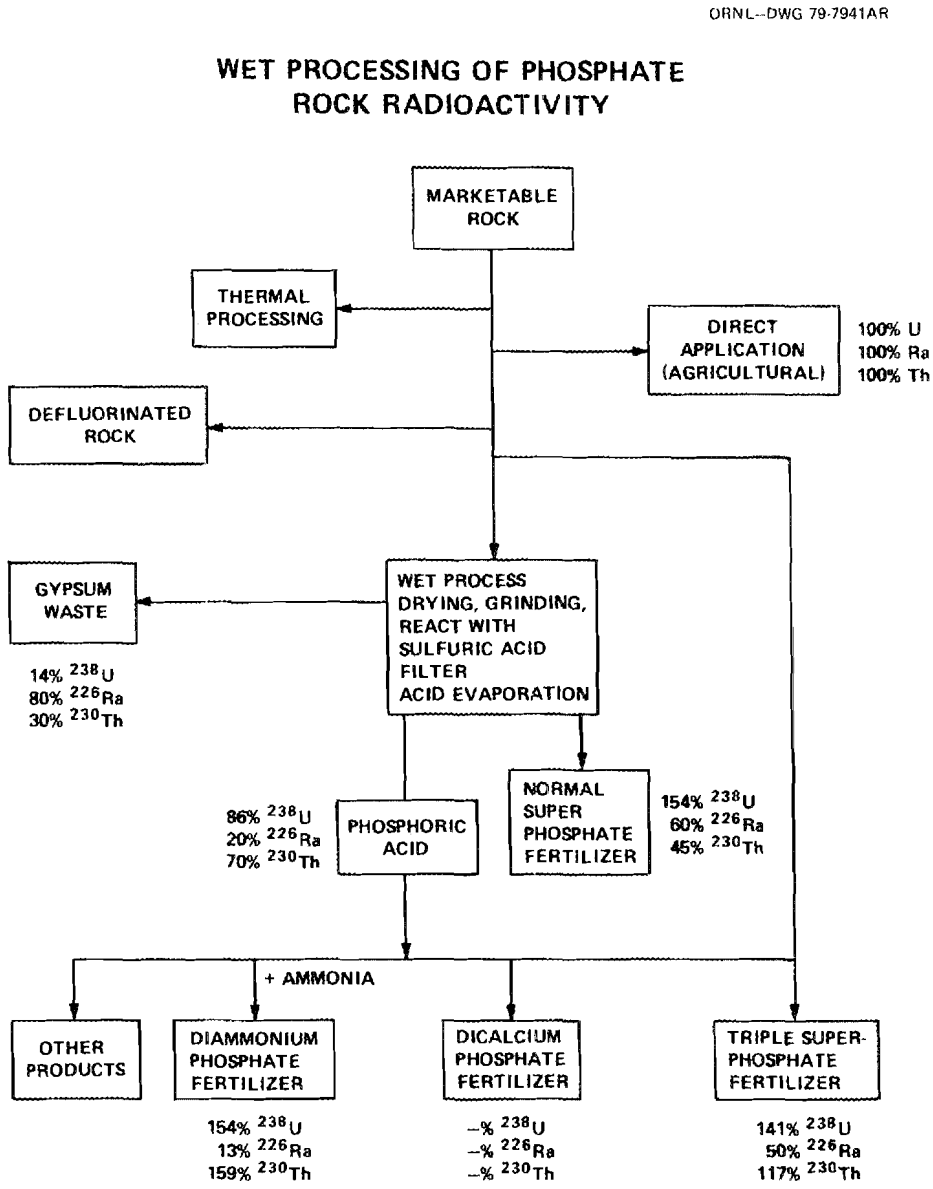

Fig. 3. Radioactivity and technological enhancement in wet process products.

Thermal processing of marketable rock yields elemental phosphorus, which is used to produce reagent elemental phosphorus, furnace phosphoric acid, and anhydrous phosphorus derivatives. Ferrophosphorus and slag are waste products. Marketable rock, silica, and coke are burned in electric arc furnaces producing phosphorus gas and carbon monoxide. The phosphorus is obtained by condensation from a gas stream. Figure 4 gives the percentages of marketable rock that are required for thermal process products.[14] Tables 5 and 6 give the concentrations of ^{238}U, ^{226}Ra, ^{230}Th, ^{232}Th, ^{210}Po, and ^{210}Pb in the furnace input materials, products, and wastes.[16] Figure 5 gives a flow chart of this process and shows that most of the ^{238}U, ^{226}Ra, and ^{230}Th remains in the ferrophosphorus and slag wastes. In thermal and wet processing or any other processing using high temperatures, an important concern is airborne effluents from electric furnace operations as ^{210}Po and ^{210}Pb are volatile at furnace operating temperatures. These concerns will be addressed in detail in the discussion of airborne effluent radiological impacts. It has been shown that these impacts are not as significant for wet process facilities.[17]

The slag and ferrophosphorus wastes have uses as additives in other industries. The slag is used for road ballast, railroad track ballast, aggregate in highway surfaces and concrete block, and in insulation. The Environmental Protection Agency (EPA) is investigating the use of these materials and the radiological impacts that may result from these uses. The ferrophosphorus is used as a metal strengthening additive in the production of steel, vanadium, carbon steel, and cast iron.[18-20]

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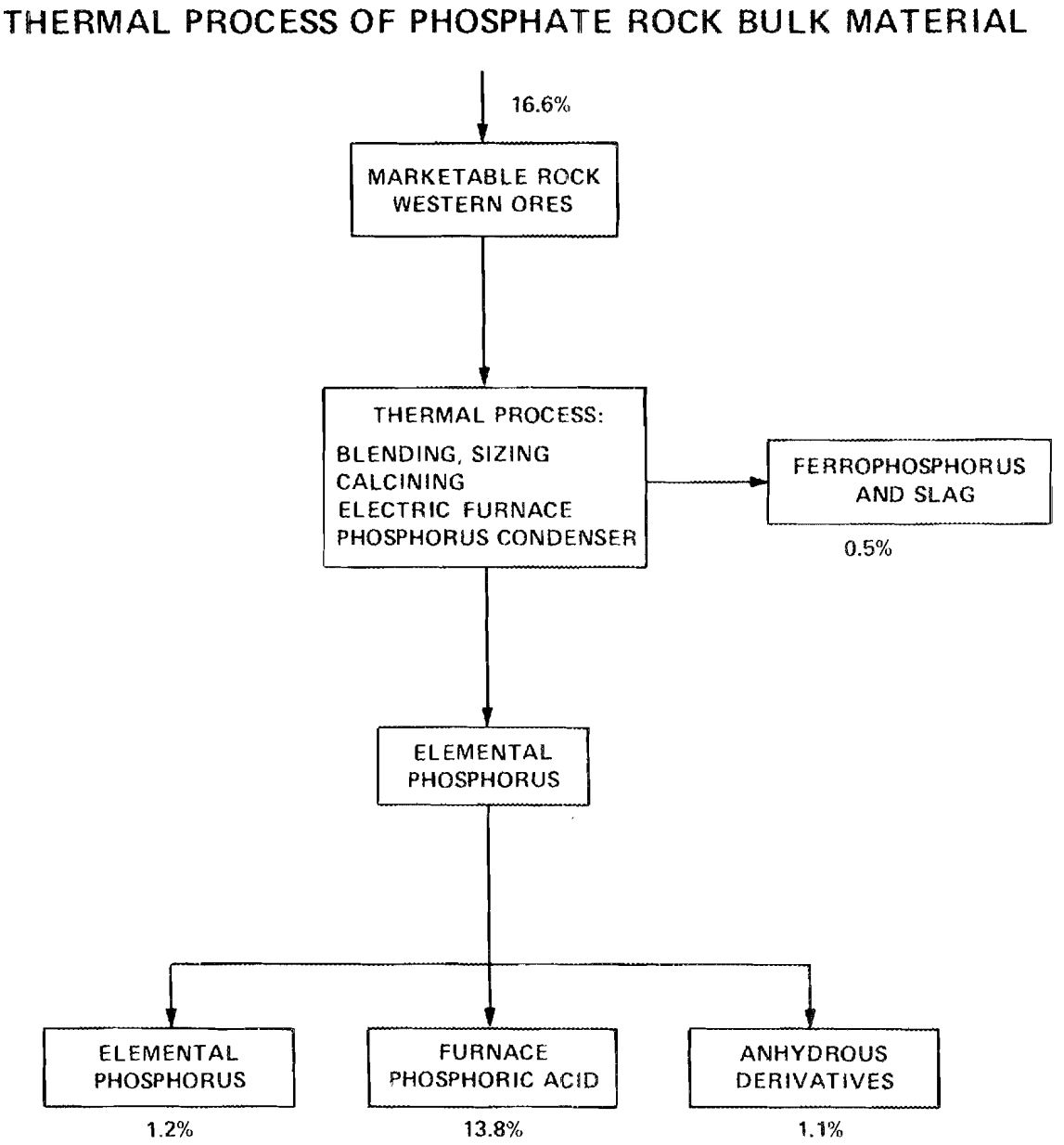

Fig. 4. Marketable rock partitioning to thermal process products.

Table 5. Radionuclides in input materials to thermal processing[a]

Radionuclide	Furnace input products (thermal process) phosphate rock (pCi/g)	Calcined rock (pCi/g)	Silica (pCi/g)	Coke (pCi/g)	Coke supplement (pCi/g)
^{226}Ra	26 ± 19	25 ± 8.6	1.74 ± 0.24	0.78 ± 0.17	0.70 ± 0.16
^{238}U	22 ± 3.2	24 ± 1.8	1.5 ± 1.4		
^{230}Th	22 ± 4.1	22 ± 4.1	1.6 ± 0.53		
^{232}Th	0.43 ± 0.12	0.47 ± 0.10	0.69 ± 0.37		
^{210}Pb	27 ± 12	18 ± 8.6	0.67 ± 0.55	2.4 ± 0.62	0.61 ± 0.54
^{210}Po	22 ± 3.0		2.6 ± 0.90		9.98 ± 0.11

[a]Eadie, G. G., and Bernhardt, D. E., *Radiological Surveys of the Idaho Phosphate Ore Processing-The Thermal Process*, ORP/LV-77-3 (November 1979).

Table 6. Radionuclides in end products of thermal processing (pCi/g)[a]

Radionuclide	Phosphorus product (solid)	Ferrophosphorus	Slag
^{226}Ra	0.021 ± 0.020	13 ± 0.65	32 ± 13
^{238}U		<71	25
^{230}Th		<24	26 ± 11
^{232}Th		<17	0.59 ± 0.29
^{210}Pb	0.21 ± 0.17	42 ± 1.8	11 ± 7.9
^{210}Po		440 ± 27	<16

[a]Eadie, G. G., and Bernhardt, D. E., *Radiological Surveys of Idaho Phosphate Ore Processing-The Thermal Process*, ORP/LV-77-3 (November 1977).

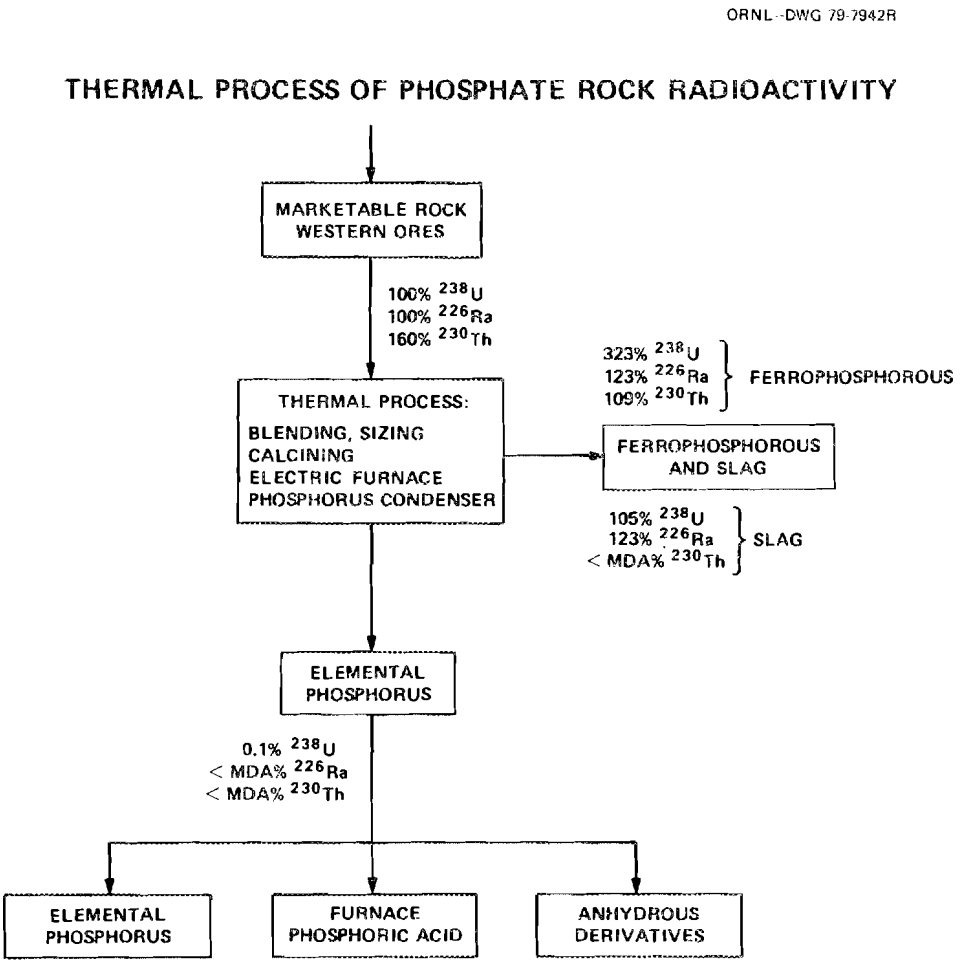

Fig. 5. Radioactivity and technological enhancement in thermal process products.

In 1972, 1.18×10^3 short tons of ferrophosphorus were shipped for use at steel facilities;[18] and based on the average uranium and radium concentrations given in Table 6, it is estimated that 0.5 Ci of ^{210}Po, 0.08 Ci of ^{238}U, 0.06 Ci of ^{210}Pb, 0.03 Ci of ^{230}Th, 0.02 Ci of ^{232}Th, and 0.01 Ci of ^{226}Ra were processed in steel production. More detailed information regarding the fate of these radionuclides in steel production is not available making radiological impact assessment rather speculative.

3. DOSE ESTIMATES

3.1 Dose Estimates Associated with Occupational Exposure in the Phosphate Industry

Radiation dose to persons occupationally exposed in phosphate industries can be considered for mining and milling, marketable rock processing, and product use. Most of the available assessments of radiation levels, airborne radioactivity levels, and occupational exposure are associated with marketable rock processing.[6,16,17,21-25] The EPA has performed studies at Florida and Idaho facilities to estimate in-plant radiation exposures.[16,17] These studies include evaluation of external and inhalation doses.

Other studies have addressed the occupational doses associated with phosphate product use. Some internal dose estimates associated with fertilizer application have been made, but these estimates are very limited. Population dose estimates are addressed in more detail.

A study specifically addressing radiation dose estimates to phosphate industry personnel was performed by the EPA in 1976, and data were obtained at several phosphate processing facilities in Florida.[6] Measurements of external gamma radiation levels were made with integrating thermoluminescent dosimeters (TLD) at locations where no phosphate industry existed. The values reported reflect time averaged conditions; the range of values reported were 4 to 6 μR/hr and provide a basis for comparison of occupational exposure levels discussed below. Other gamma radiation level measurements were made with TLDs at specific

locations inside the various phosphate processing facilities. The average exposure levels ranged from 6 μR/hr in parking lots and general office areas to 179 μR/hr at a large reaction tank at one facility. All other values were less than or equal to 65 μR/hr. Others have reported mean background gamma radiation of 5 μR/hr on areas where no undisturbed radioactive ores were known to exist.[26] A detailed list of gamma exposure levels is given in Table 7.

Other measurements at phosphate facilities were made with pressurized ionization chambers and gamma scintillation instruments.[6] These measurements reflect exposure conditions at a particular time rather than average exposure conditions. The pressurized ionization chamber readings varied from 12 μR/hr in dry fertilizer product storage areas to 132 μR/hr in a general area around chemical reaction tanks. Scintillator readings ranged from 15 μR/hr in dry products storage areas to 500 μR/hr inside a chemical reaction tank. (This reading was obtained during a routine cleanout operation.) The difference in the readings obtained with the two measuring systems (where comparable data were available) was not greater than 36 μR/hr, however, reliance upon scintillator measurements to calculate exposure may not be justified in all cases. Other investigators have shown that variations of up to 100% are obtained between scintillation measurements and a shielded, nearly energy-independent Geiger-Mueller counter.[27]

In the EPA study sited above lung doses were estimated from air samples analyzed for ^{226}Ra and the isotopes of uranium and thorium. Using lung dosimetry models by the International Commission on Radiological

Table 7. External gamma radiation levels at Florida phosphate processing facilities[a]

	Location	Average exposure (μR/hr)
(1)	Dundee, Florida—background	4
(2)	Lake Wales, Florida—background	6
(3)	Polk City, Florida—background	4
(4)	Grounds—general office	14
(5)	Parking lot—general office	6
(6)	Ore unloading	19
(7)	Ore unloading	38
(8)	Ore unloading	54
(9)	Ore unloading	39
(10)	Ball mill area	37
(11)	Phosphoric acid plant	179
(12)	Fertilizer plant	7
(13)	Fertilizer product storage	16
(14)	Fertilizer product storage	16
(15)	Furnace Area—thermal process plant	65
(16)	Ore drying and storage	57

[a]Windham, S. T., Partridge, J., and Horton, T. *Radiation Dose Estimates to Phosphate Industry Personnel.* EPA-52015-76-014, December 1976.

Protection (ICRP) and the EPA, lung dose estimates were made.[6] The radionuclides included in the sample analyses were ^{238}U, ^{235}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th, ^{227}Th, ^{226}Ra, ^{222}Rn, and ^{210}Po. Working level* measurements were not made. The range of average lung doses, using the ICRP model, was 0.05 rem/year in a product storage and shipment area to 3.0 rem/year atop a waste gypsum pile (the dose assessment atop the gypsum pile considers radon plus daughters only). Using the EPA dose conversion factors, calculated doses ranged from 0.15 rem/year in the product storage and shipment area to 3.6 rem/year in a ball-milling area. The averages of all values were 1.0 rem $\pm$ 98% and 2.3 rem $\pm$ 92% using the ICRP and EPA models, respectively.[6]

This report concluded that

1. external gamma radiation doses are below the nonoccupational dose limit of 0.5 rem/year if continuous occupancy is assumed, and
2. all lung dose estimates were below the occupational guideline of 15 rem/year and in most cases were below 1.5 rem/year, the nonoccupational limit.[28]

Studies were performed by the EPA at two Idaho phosphate facilities: a thermal process facility and a wet process facility.[16,17] External gamma radiation dose rates at the wet process facility ranged from background in control and storage areas to 222 millirem/year near a condensate pipe. At the thermal process facility, external gamma radiation

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*A working level is defined as any combination of short-lived radon daughters in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha particle energy.

dose rates ranged from background in control areas to 182 millirem/year in an outdoor slag storage area. Dose rates were calculated on the basis of 40 hr/week and 50 week/year occupancy. Because the gamma scintillation meter was calibrated against a ^{226}Ra standard source (atypical of field conditions), large errors may be associated with these dose rate estimates as discussed above.

Measured ^{222}Rn concentrations in the thermal process facility ranged from 170 pCi/liter to 11,000 pCi/liter.[6] Corresponding lung dose rates were calculated to be 0.2 rem/year to 10 rem/year. In the wet process facility, ^{222}Rn concentrations varied from 180 pCi/liter to 1900 pCi/liter. The calculated dose rates were 0.2 to 1.7 rem/year.[6]

Aerosol analyses were performed by EPA at both facilities to determine the size distributions of airborne particles and the particles with which the radioactivity was associated.[16,17] In the wet process facility, data for various sampling locations indicated 45-85% of the ^{226}Ra, 5-75% of the uranium, and 7-75% of the ^{230}Th were associated with particles of equivalent aerodynamic diameter (EAD) of 0.5 μ or less.[16] The lowest percentages were obtained in an Ammophos plant where dusts were of much larger particle size.[16] In the thermal process plant approximately 50% of ^{226}Ra, uranium and ^{230}Th were associated with particles of equivalent aerodynamic diameter of 0.5 μ or less.[17] These results are significant when it is realized that dust particles with these smaller sizes (EAD approximately equal to tenths of microns) penetrate into the pulmonary region of the respiratory tract from which clearance is relatively slow.

Other limited studies have been conducted at four Florida facilities and one Utah facility by the National Institute for Occupational Safety and Health (NIOSH).[21-25] These industrial hygiene surveys included reviews of the medical, safety, and industrial hygiene programs.

In the NIOSH study, air concentration sampling was performed for heavy metals (Cd, Cr, and V), free silica, fluoride, and phosphoric acid mists. Not all pollutants were sampled at each facility; and for some pollutants, there is a reported value for a small percentage of the sampled locations. Table 8 gives a summary of measurements made at each facility. Ranges of concentrations of each pollutant, Occupational Safety and Health Administration (OSHA) legal limit concentrations, the NIOSH proposed standards, and threshold limit values are given for each pollutant in Table 9. The NIOSH proposed standards for Cr, V, As, U, F, respirable silicon, and respirable dust were exceeded at one or more of the four plants where sampling was performed. These NIOSH proposed standards for chromium were exceeded at all four facilities. The OSHA legal limits for uranium and fluorine were exceeded at one and two of the four plants, respectively. Also, a question must be raised about the validity of reporting 0.0 working levels (WL)* for radon daughter product concentration for plants 3 and 4. Natural background ^{222}Rn progeny concentrations have been measured in outdoor areas with no high activity natural deposits, and values as low as 0.002 WL $\pm$ 100% have been reported.[29] It is true that these values are low, but it is usually

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*A working level is defined as any combination of short-lived radon daughters in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha particle energy.

Table 8. Measurements at phosphate facilities reported by National Institute for Occupational Health and Safety (NIOSH)

Plant No.	Heavy metals Cd, Cr, V	Respirable silica	Respirable dust	Fluoride	Uranium	Arsenic	Rn or Rn daughters	H_2SO_4	H_3PO_4
1	*b*	*b*	*b*	*b*	*b*	*b*	*b*	*b*	*b*
2	*a*	*c*	*b*	*a*	*d*	*d*	*a*	*a*	*a*
3	*a*	*d*	*d*	*a*	*d*	*d*	*a*	*b*	*b*
4	*a*	*d*	*b*	*a*	*c*	*a*	*a*	*a*	*a*
5[e]	*a*	*b*	*b*	*a*	*c*	*b*	*a*	*a*	*a*

[a]Sampled, values reported.

[b]Not sampled.

[c]Sampling method given, no values reported.

[d]Values reported for less than 20% of sampling locations.

[e]Only ranges of values given.

Table 9. Results of National Institute for Occupational Safety and Health (NIOSH) industrial hygiene surveys at phosphate facilities[a]

Pollutant	Plant 2	Plant 3	Plant 4	Plant 5	OSHA standard	NIOSH proposed standard	AGGIH TLV
Cd	<0.001-<0.002	0.001-0.007	0.002-0.004	<MDL-0.003	0.2 (dust) 0.2 (fume)	0.040	0.05
Cr[b]	0.01-0.05	<0.001-0.02	0.002-0.004	0.0007-0.002	0.5	0.001	0.5
V[b]	<0.001	0.008-0.05	0.03-0.39	0.006-0.01	0.5	0.5	0.5
As[b]	<0.001-<0.01	0.001-0.005	0.03-0.05		0.5	0.002	0.5
U[b,c]	0.005-0.014	0.022-0.514			0.25 (insoluble)[d] 0.05 (soluble)		0.02
H_2SO_4	0.013-0.22		0.16-0.67	0.08-0.13	1.0	1.0	1.0
H_3PO_4	0.03-0.52		0.018-0.129	0.02-0.08	1.0		1.0
F[b,c]	0.02-13.24	0.21-0.88	0.013 + 0.84 (particulate) 0.04 + 08.02 (impinger)	0.03-0.07	2.5	2.5	2.0
Respirable Si[b]		0.04-0.09	0.015-0.06			0.05	0.2
Respirable dust[b]		0.45-3.63			3.38		3.38
Radon daughters	0.0-0.02 WL[e]	0.0 WL	0.0 WL	0.27 pCi/liter + 0.56 pCi/liter of ^{222}Rn	0.33 WL for 40 hr/week 50 week/year (10 CFR 20 App. B)		

[a]Values are in mg/m^3 unless otherwise indicated.

[b]Indicates that readings at one or more plants exceeded NIOSH proposed standards.

[c]Indicates that readings at one or more plants exceeded OSHA legal limits.

[d]0.25 mg $U_{NAT}/m^3 = 9.57 \times 10^{-11}$ μCi/cc of U_{NAT} (insoluble).
0.05 mg $U_{NAT}/m^3 = 1.91 \times 10^{-11}$ μCi/cc of U_{NAT} (soluble).
NRC occupational MPC_a for U_{NAT}
1×10^{-10} μCi/ml (insoluble)
1×10^{-10} μCi/ml (soluble)

[e]A working level (WL) is defined as any combination of short-lived radon daughters in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of alpha particle energy.

more informative to report values as less than a given detectable limit rather than zero. It is quite unlikely that 0.0 WL was the real value of ^{222}Rn progeny concentration. It is more likely that the concentrations of ^{222}Rn daughters reported as 0.0 WL were simply below detectable limits of the sampling and detection techniques employed.

Statistical data on the number of people employed in the phosphate industry are compiled by the U.S. Department of the Labor's Mine Safety and Health Administration (formerly the U.S. Department of the Interior Mine Enforcement and Safety Administration).[30-34] Table 10 was compiled from these data and presents the average number of full-time workers in phosphate mines and mills for the years 1973 through 1977. The number and frequency of fatal accidents are also given.

The occupational work force external gamma radiation dose can be estimated from the data in Table 10 and the gamma radiation exposure level data in Table 7. The average annual occupational dose estimate is calculated by multiplying the average external gamma radiation level (μR/hr) by 2000 work hours per year and the average number of persons working daily. This value may be converted to person-rem per year assuming that 1 Roentgen is approximately equal to 1 rad and that the quality factor for gamma radiation is 1 rem per rad. The average external gamma radiation level in Florida phosphate facilities is estimated by averaging readings 4 through 10 and 12 through 16 from Table 7. The background values and value 11 were excluded from the average. Value 11 is an exposure level associated with a special tank

Table 10. Work force and accident statistics for the phosphate industry, 1973 through 1977[a]

Year	Average number of men working daily			Fatalities	Rate of fatal accidents (fatalities/10^6 man-hours)
	Mill	Mine	Total		
1973	2019	2737	4756	2	0.19
1974	2017	3635	5652	4	0.36
1975	2549	3730	6279	3	0.22
1976	3200	3661	6861	3	0.22
1977	3159	3391	6550	4	0.30

[a]Compiled from U.S. Department of Labor and the U.S. Department of Interior Reports.[30-34]

cleanout operation conducted only a few times per year by a limited number of people. The average gamma exposure level is then 30 μR/hr ± 70%. The associated work force dose commitment for the 1977 work force data is estimated to be 400 person-rem with an error of ≥70%. This error arises from uncertainties in the average gamma radiation exposure level and the average number of full-time employees. Also, the simplified method of multiplying average exposure or dose by the average number of employees may be inaccurate if a large fraction of new employees is exposed at a level that deviates significantly from the calculated average exposure. A description of the number of persons working at specific plant locations and the exposure levels at those locations would be required for more detailed estimates of work force dose. Work force gamma radiation dose commitment estimates were made for the years 1973 through 1977 and are presented in Table 11.

Occupational external gamma radiation doses at phosphate facilities in Germany have been estimated and these values may be compared to values obtained in the EPA work cited earlier.[6,35,36] Annual occupational external radiation doses from fertilizer application were estimated to be on the order of a few millirads per year in the German study. Also, occupational radiation dose in phosphate industries were highest for fertilizer production and storehouse personnel, and dose levels in some cases may be approximately 2.0 times natural background external gamma radiation levels (50 to 55 millirad per year is the value cited for natural background gamma radiation dose in Germany).[35,36]

Table 11. Dose estimates (person-rem) for the phosphate industry personnel

Year	External gamma radiation[a]	Background work force gamma dose	Lung dose[b]		Background work force lung dose
			ICRP Group II	EPA	
1973	300	200	4800	11000	520-5200
1974	400	300	5700	13000	620-6200
1975	400	300	6300	14000	690-6900
1976	400	300	6900	16000	750-7500
1977	400	300	6600	15000	720-7200

[a]It is assumed that the average gamma radiation exposure includes background exposure and that workers are exposed 40 hr per week for 50 weeks per year to occupational exposure levels. The average individual annual gamma radiation background dose is approximately 47 millirad.

[b] The average individual lung dose from all sources of exposure may vary up to an order of magnitude due to variation in radioactivity content of different building materials used to construct residences.

A gamma exposure of 2.0 times this background value is calculated to be 100 to 110 millirad per year. If it is assumed that the excess exposure occurs during an occupational period of 2000 hrs per year the average gamma radiation levels is calculated to be approximately 30 μR/hr in the working environment. This value is in excellent agreement with the average value calculated for Florida facilities given above.

Work force lung dose estimates were also made by averaging values for annual lung dose at specific locations in phosphate facilities presented earlier and multiplying by the number of people in the work force for a given year. The figures given for average annual lung doses are for processing operations but not necessarily open pit or mining operations. Average annual individual lung doses of 1.0 rem ± 98% and 2.3 rem ± 92% were calculated using the International Commission on Radiological Protection (ICRP) Task Group II lung dosimetry model and the EPA lung dosimetry model, respectively. Lung dose estimates ranged from 4800 person-rem per year in 1973 to 6900 person-rem per year in 1976 using the ICRP model and from 11,000 person-rem per year in 1973 to 16,000 person-rem per year in 1976 using the EPA model. Values using the ICRP and EPA models are presented for the years 1973 through 1977 in Table 11.

Background dose may provide a basis for comparison of these occupational exposures. The average background external gamma dose associated with terrestrial radionuclides is about 47 millirad/year, while the average background external dose associated with cosmic radiation is approximately 23 millirad/year to the gonads, bone, and lungs. The

average lung dose associated with internally deposited ^{226}Ra is approximately 0.5 millirad/year, while the lung dose due to naturally occurring radon and its daughter products ranges from 19 to 64 millirad/year depending upon the type of structure.[37]

Occupational gamma radiation doses may be 1.5 times background gamma radiation dose lung doses may be 2 times background lung dose if compared to the work force lung dose estimated with the ICRP Task Group II lung model, or 4 times background lung dose if compared with the lung dose estimated with the EPA lung model. While these estimates of background exposure provide some measure for comparison to occupational exposure, it must be realized that background radiation exposure may vary by an order of magnitude from region to region, though the quoted values of background external gamma and lung dose are reasonable national averages. A more detailed estimate of increase in dose commitment of phosphate industry personnel would require detailed study of those communities in Idaho and Florida where most phosphate workers live.

3.2 Population Dose Commitments

3.2.1 Dose commitments from effluent releases

Effluent releases from phosphate facilities have been assumed and subsequent normalized population dose commitment estimates, via different exposure pathways, have been made for the two regions of the United States where phosphate mining and processing activities are highest, namely southeastern Idaho and central Florida. These assessments assumed release rates of 1.0 Ci/year for each of the radionuclides listed in Table 12. The points of release were chosen to be Mulberry, Florida,

Table 12. Radionuclides considered in assessing radiological impact on man from phosphate production

Nuclide (Daughter)	Half-life[a]	Origin
^{238}U	4.4×10^{9} y	Release
(^{234}Th)	24.1 d	Build-up on ground
(^{234}Pa)	6.7 h	Build-up on ground
^{235}U	7.1×10^{8} y	Release
(^{231}Th)	25.5 h	Build-up on ground
^{234}U	2.4×10^{5} y	Release
^{232}Th	1.4×10^{10} y	Release
(^{228}Ac)	6.13 h	Build-up on ground
^{228}Th	1.91 y	Release and build-up
(^{224}Ra)	3.66 d	Build-up on ground
(^{212}Pb)	10.64 h	Build-up on ground
(^{212}Bi)	60.55 m	Build-up on ground
(^{208}Tl)	3.07 m	Build-up on ground
^{230}Th	7.7×10^{4} y	Release
^{227}Th	18.7 d	Release
^{226}Ra	1.60×10^{3} y	Release
(^{210}Pb)	22.3 y	Build-up on ground
(^{210}Po)	138.4 d	Build-up on ground
(^{214}Pb)	26.8 m	Build-up on ground
(^{214}Bi)	19.9 m	Build-up on ground
(^{218}Po)	3.05 m	Build-up on ground
^{222}Rn	3.82 d	Release

[a]Data from Kocher, D.C., *Nuclear Decay Data for Radionuclides Occurring in Routine Releases from Nuclear Fuel Cycle Facilities*, ORNL/NUREG/TM-102.

and Pocatello, Idaho. A large portion all phosphate activities are centered at these two locations with distributions indicated previously. Because of variability in operating procedures among different facilities and uncertainty in industrial average radionuclide release data, accurate calculations of population doses are difficult. In order to make a first estimate about the characteristics of population doses, single release points were assumed for each region and population doses were calculated per curie per year of released radionuclides. It is recognized that the location of this single source will influence population dose calculations and that source terms are not accurately known. For the Florida case, approximately 98% of the population living within a 55-mile radius of Mulberry, Florida lives beyond a ten-mile radius from the chosen release point, and for the Idaho case, 68% live beyond a ten-mile radius. Tables 13 and 14 give the percentage of the population within a 55-mile radius as a function of radial distance from source release points. The sensitivity of estimated population dose on source location is an area needing significant investigation, and consideration should be extended to different modes of release such as arrays of point sources and area sources. It is felt that for relative population dosimetry, the calculations presented below are instructive.

The AIRDOS-EPA computer code was used in these assessments.[38] This code, which is based on the Gaussian plume model, estimates concentrations of radionuclides in air, their rates of deposition on ground surfaces through both dry and wet deposition processes, and concentrations of radionuclides on ground surfaces as a function of direction and

Table 13. Florida site cumulative population distribution

Radial distance from release point (miles)	Cumulative % of total population
0-1	0
1-2	0.09
2-3	0.09
3-4	0.25
4-5	0.29
5-10	1.84
10-15	6.79
15-25	17.47
25-35	41.06
35-45	61.27
45-55	100

Table 14. Idaho site cumulative population distribution

Radial distance from release point (miles)	Cumulative % of total population
0-1	0
1-2	3.27
2-3	11.09
3-4	18.36
4-5	26.18
5-10	31.36
10-15	31.99
15-25	44.16
25-35	53.91
35-45	65.67
45-55	100

distance from a specified point of release. A period of 25 years was assumed for calculating the buildup of radionuclides on ground surfaces. The calculations include the buildup of daughters on ground surfaces resulting from decay of parent radionuclides after deposition.

Dose conversion factors are put into the code; and annual doses to man, for each specified distance and direction, are estimated for total body, lower large intestine, red marrow, endosteal cells, thyroid, lungs, kidneys, liver stomach wall, ovaries and testes through the following exposure modes: (1) immersion in air containing radionuclides; (2) exposure to ground surface contaminated by deposited radionuclides; (3) immersion in contaminated water; (4) inhalation of radionuclides in air; and (5) ingestion of food produced in the area. External dose conversion factors used are from Kocher.[39] Internal dose conversion factors by Dunning[40] (using ICRP metabolic models) are based on the ICRP Task Group II lung model and the gastrointestinal tract model described by Bernard[41] with parameter values given by Eve.[42] These factors are for 50-year dose commitments resulting from one year of intake. The internal dose conversion factors which have been used in this assessment assume a quality factor (QF) of 10 for alpha particles. However QF of 20 has been recommended by ICRP but has not been officially adopted.[43]

Annual average meteorological data are put into the code and finally the code will estimate maximum annual individual dose (rem/year) or annual population dose person-rem per year of effluent release within a specified area. In this case, a radius of 55 miles was used. Many of

the basic assumptions used in the code are conservative, thus maximizing the estimated dose to man. Factors that would reduce the dose such as shielding by dwellings and time spent away from the point of dose calculation are not considered. For instance, the code assumes that a person lives outdoors at the reference location 100% of the time.

In the absence of site-specific detailed data, conservative approaches are often taken when supplying parameter values to the code. In this assessment, ingestion dose calculations assume that all beef, milk, and vegetables consumed by an individual are produced at his or her reference location. These assumptions lead to a higher dose estimate than would likely be accrued.

Meteorological information used in these assessments was originally obtained through the U.S. Department of Commerce, National Oceanic and Atmospheric Administration's STAR Program for Tampa, Florida, and Pocatello, Idaho. Population distributions were generated using the PANS computer code for locations near Mulberry, Florida, and Pocatello, Idaho, the locations chosen to represent points of release. Both the meteorological data and the population data for the Florida site were prepared for use in similar calculations (using AIRDOS-II) and were used without modification in this assessment.[44] Pocatello population and meteorological data were specifically prepared for this assessment.

Tables 15 through 20 present normalized population dose commitments via the surface exposure, inhalation, and ingestion pathways for all target organs, and for particles with aerodynamic diameters (AMAD) of 0.3, 1.0, and 5.0 μ and for release heights of 10, 25, and 50 m for the Florida and Idaho cases, respectively.

Table 15. Percent of total body dose[a] for each organ of interest by its major exposure pathways resulting from releases during Florida phosphate production — 0.3-μm-particle size

Major exposure pathway	Percent of total body dose (%)									
	Red marrow	Lungs	Endosteal cells	Stomach wall	Lower large intestine wall	Thyroid	Liver	Kidneys	Testes	Ovaries
					10-m release height					
Surface exposure	17	11	20	9	7	13	9	9	11	7
Inhalation	12	37	98	<1	<1	<1	15	2	<1	<1
Ingestion	199	<1	1088	<1	4	4	46	20	4	4
					25-m release height					
Surface exposure	15	9	17	8	6	11	8	8	10	6
Inhalation	11	32	84	<1	<1	<1	12	2	<1	<1
Ingestion	205	<1	1117	<1	4	4	47	21	5	4
					50-m release height					
Surface exposure	15	9	17	8	6	11	8	8	10	6
Inhalation	10	30	81	<1	<1	<1	12	2	<1	<1
Ingestion	205	<1	1119	<1	4	4	47	21	5	4

[a]The total body dose refers to the dose received by the total body from all radionuclides through all exposure pathways.

Table 16. Percent of total body dose[a] for each organ of interest by its major exposure pathways resulting from releases during Florida phosphate production — 1.0-μm-particle size

Major exposure pathway	Percent of total body dose (%)									
	Red marrow	Lungs	Endosteal cells	Stomach wall	Lower large intestine wall	Thyroid	Liver	Kidneys	Testes	Ovaries
					10-m release height					
Surface exposure	17	11	20	9	7	13	10	9	11	7
Inhalation	8	22	62	<1	<1	<1	9	1	<1	<1
Ingestion	203	<1	1106	<1	4	4	47	21	5	4
					25-m release height					
Surface exposure	15	9	17	8	6	11	8	8	10	6
Inhalation	7	19	53	<1	<1	<1	8	1	<1	<1
Ingestion	208	<1	1132	<1	4	4	48	21	5	4
					50-m release height					
Surface exposure	15	9	17	8	6	11	8	8	10	6
Inhalation	6	18	51	<1	<1	<1	7	1	<1	<1
Ingestion	208	<1	1134	<1	4	4	48	21	5	4

[a]The total body dose refers to the dose received by the total body from all radionuclides through all environmental exposure pathways.

Table 17. Percent of total body dose[a] for each organ of interest by its major exposure pathways resulting from releases during Florida phosphate production — 5.0-μm particle size

Major Exposure Pathway	Percent of total body dose (%)									
	Red marrow	Lungs	Endosteal cells	Stomach wall	Lower large intestine wall	Thyroid	Liver	Kidneys	Testes	Ovaries
					10-m release height					
Surface exposure	17	11	20	9	8	13	10	9	11	7
Inhalation	4	8	34	<1	<1	<1	5	1	<1	<1
Ingestion	205	<1	1120	<1	4	4	47	21	5	4
					25-m release height					
Surface exposure	15	9	18	8	7	11	8	8	10	6
Inhalation	4	7	29	<1	<1	<1	4	1	<1	<1
Ingestion	210	<1	1145	<1	4	4	48	21	5	4
					50-m release height					
Surface exposure	15	9	18	8	7	11	8	8	10	6
Inhalation	4	6	27	<1	<1	<1	4	1	<1	<1
Ingestion	210	<1	1146	<1	4	4	48	21	5	4

[a]The total body dose refers to the dose received by the total body from all radionuclides through all environmental exposure pathways.

Table 18. Percent of total body dose[a] for each organ of interest by its major exposure pathways resulting from releases during Idaho phosphate production — 0.3-μm-particle size

Major exposure pathway	Percent of total body dose (%)									
	Red marrow	Lungs	Endosteal cells	Stomach wall	Lower large intestine wall	Thyroid	Liver	Kidneys	Testes	Ovaries
					10-m release height					
Surface exposure	9	6	10	5	4	7	5	5	6	4
Inhalation	7	22	58	<1	<1	<1	9	1	<1	<1
Ingestion	217	<1	1182	<1	4	5	49	22	5	4
					25-m release height					
Surface exposure	12	8	15	7	5	10	7	7	8	5
Inhalation	10	31	81	<1	<1	<1	12	2	<1	<1
Ingestion	209	<1	1134	<1	4	4	47	22	5	4
					50-m release height					
Surface exposure	16	10	19	9	7	12	9	9	10	7
Inhalation	12	36	96	<1	<1	<1	14	2	<1	<1
Ingestion	201	<1	1095	<1	4	4	45	21	5	4

[a]The total body dose refers to the dose received by the total body from all radionuclides through all environmental exposure pathways.

Table 19. Percent of total body dose[a] for each organ of interest by its major exposure pathways resulting from releases during Idaho phosphate production — 1.0-μm-particle size

Major exposure pathway	Percent of total body dose (%)									
	Red marrow	Lungs	Endosteal cells	Stomach wall	Lower large intestine wall	Thyroid	Liver	Kidneys	Testes	Ovaries
					10-m release height					
Surface exposure	9	6	11	5	4	7	5	5	6	4
Inhalation	5	13	37	<1	<1	<1	5	1	<1	<1
Ingestion	219	<1	1194	<1	4	5	49	22	5	4
					25-m release height					
Surface exposure	13	8	15	7	6	10	7	7	8	5
Inhalation	6	18	51	<1	<1	<1	8	1	<1	<1
Ingestion	211	<1	1150	<1	4	4	47	22	5	4
					50-m release height					
Surface exposure	16	10	19	9	7	12	9	9	11	7
Inhalation	8	21	61	<1	<1	<1	9	1	<1	<1
Ingestion	204	<1	1113	<1	4	4	46	21	5	4

[a]The total body dose refers to the dose received by the total body from all radionuclides through all environmental exposure pathways.

Table 20. Percent of total body dose[a] for each organ of interest by its major exposure pathways resulting from releases during Idaho phosphate production — 5.0-μm-particle size

Major Exposure Pathway	Percent of total body dose (%)									
	Red marrow	Lungs	Endosteal cells	Stomach wall	Lower large intestine wall	Thyroid	Liver	Kidneys	Testes	Ovaries
					10-m release height					
Surface exposure	9	6	11	5	4	7	5	5	6	4
Inhalation	3	5	20	<1	<1	<1	3	<1	<1	<1
Ingestion	221	<1	1203	<1	4	5	50	23	5	5
					25-m release height					
Surface exposure	13	8	15	7	6	10	7	7	8	5
Inhalation	4	6	28	<1	<1	<1	4	<1	<1	<1
Ingestion	214	<1	1163	<1	4	4	48	22	5	4
					50-m release height					
Surface exposure	16	10	19	9	7	12	9	9	11	7
Inhalation	4	8	33	<1	<1	<1	5	1	<1	<1
Ingestion	207	<1	1127	<1	4	43	46	21	5	4

[a]The total body dose refers to the dose received by the total body from all radionuclides through all environmental exposure pathways.

The method of data presentation provides an insight into the dosimetric importance of exposure pathways and body compartments. The numbers provided in Tables 15 through 20 are ratios of the dose to a specific body compartment delivered via a given pathway from all radionuclides divided by the dose to the total body delivered via all pathways from all radionuclides. By comparing these ratios, important compartments (those that receive the highest doses) and important pathways can be identified. Table 21 gives the radionuclides that contribute the largest fraction of dose to a given compartment and the percentage contribution. In cases where the contributions of two radionuclides were significant, both radionuclides are listed. This method of presentation allows the identification of those radionuclides that may contribute to doses in various body compartments via individual pathways.

The estimated relative population doses to any individual organ did not differ by more than a factor of 3 with AMAD and not more than a factor of 1.3 with release height. The largest relative population organ doses were to endosteal cells and red bone marrow with ^{226}Ra and thorium radionuclides contributing greatly to the doses.

Apart from identifying those pathways, body compartments, and radionuclides which are dosimetrically important, these calculations may aid in the design of environmental monitoring and radiation surveillance programs. For example, in both the Florida and Idaho cases the ingestion pathway results in the highest relative doses to nearly all organs. The thorium radionuclides and ^{226}Ra are major contributors to the dose via this pathway. Therefore, a sampling program designed to evaluate population

Table 21. Primary radionuclides contributing to each organ dose through the major exposure pathways

Exposure pathway	Total body	Red marrow	Lungs	Endosteal cells	Stomach wall
Surface exposure	^{234}Th (47%) ^{214}Bi (18%)	^{234}Th (57%)	^{234}Th (44%) ^{214}Bi (19%)	^{234}Th (58%)	^{234}Th (40%) ^{214}Bi (21%)
Inhalation	^{232}Th (36%) ^{230}Th (35%)	^{232}Th (46%) ^{230}Th (44%)	^{228}Th (21%) ^{234}U (16%) ^{230}Th (16%)	^{232}Th (50%) ^{230}Th (44%)	^{228}Th (50%) ^{235}U (21%)
Ingestion	^{226}Ra (76%)	^{226}Ra (82%)	^{226}Ra (75%)	^{226}Ra (75%)	^{226}Ra (38%)

	Lower large intestine wall	Thyroid	Liver	Kidneys	Testes	Ovaries
Surface exposure	^{234}Th ^{214}Bi (23%)	^{234}Th (52%)	^{234}Th (43%) ^{214}Bi (20%)	^{234}Th (44%) ^{214}Bi (19%)	^{234}Th (47%) ^{214}Bi (15%)	^{234}Th (37%) ^{214}Bi (24%)
Inhalation	^{228}Th (60%)	^{230}Th (28%) ^{232}Th (31%)	^{230}Th (45%) ^{232}Th (51%)	^{230}Th (39%) ^{232}Th (44%)	^{230}Th (34%) ^{232}Th (39%)	^{230}Th (23%) ^{232}Th (25%)
Ingestion	^{226}Ra (39%)	^{226}Ra (57%)	^{230}Th (43%) ^{232}Th (37%)	^{210}Po (55%)	^{226}Ra (54%)	^{226}Ra (59%)

doses must include careful evaluation of the vegetables, beef, and milk in which all of these radionuclides may concentrate and subsequently be eaten by the local population. Further, it should be noted that estimates of actual doses to various organs via given pathways from radionuclides may be obtained. By using a ratio of actual released amounts of radionuclides to the unit releases assumed in these calculations (presented in Tables 15 through 20) and by accounting for the percent contribution to dose from given radionuclides the actual relative compartment dose is obtained. Estimates of absolute population doses to body compartments can be obtained by multiplying by the normalization factor defined previously (i.e., the whole body dose from all radionuclides released via all pathways).

A limited field study has been conducted by the EPA to evaluate the doses to populations in the vicinity of selected phosphate facilities.[45] This study was limited to wet process operations. Activity release rates from specific points in the process were obtained from particulates, emissions, and specific activity data. Some stack sampling was performed, and EPA measured values of released activity were generally lower than estimated facility release data. Lung dose calculations made with the AIREM computer code led to the conclusions that lung dose commitments of a few millirem per year would be received by persons living in the vicinity of phosphate facilities.[45]

3.2.2 Population doses from water borne effluent releases

The impacts associated with water effluent pathways have also been documented.[9,10,13,46] The major sources of water borne effluents include

slime pond discharge associated with beneficiation and runoff from gypsom waste piles associated with wet process phosphoric acid production. As summarized by Guimond

> "Several slime ponds have discharges to the environment. The discharge quantities depend upon the facility's degree of recycle, overall water consumption, and local precipitation. Since most of the radioactivity in the waste products of beneficiation is present in the slimes, this could pose potential problems to receiving streams if the radioactivity was not removed prior to discharge. To examine this aspect, the concentration of radium-226 was determined for slime discharges and effluent discharges from seven mine and beneficiation plants. The concentration of dissolved radium-226 in slime discharges was less than 5.0 picocuries per liter at all seven facilities. The concentration of radium-226 in the undissolved fraction varied from 10 to 2050 pCi per liter. This great variation was a function of the total suspended solids in the slime discharge. The radium-226 concentrations in undissolved fraction ranged from 10 to 80 pCi per gram, thus emphasizing the importance of the total suspended solids concentration in determining the total concentration of radium-226 in picocuries per liter in the slime discharge. Although no chemical process is used to treat the discharge from the slime ponds, dissolved radium-226 generally less than 1 pCi per liter were observed in the effluents. The total concentration of radium-226 in every effluent discharge sample analyzed was less than 3.0 pCi per liter."[13]

A summary of an EPA assessment of the water borne releases from wet process phosphoric acid facilities has been given.[46,47] This was an assessment of individual and population doses associated with the intake of drinking water from the Alafia River in Florida, a river into which five phosphate facilities discharge liquid effluents. The study concluded

> The Agency's proposed Federal Water quality criteria for input to drinking water supplies is a concentration of 5 pCi/liter for the naturally occurring radionuclides radium-226 and 228; and a maximum acceptable aggregate dose to the population served by the water supply of 3,000 man-rem, unless the radium-226 activity is less than 0.5 pCi/liter. The impact of the effluents of the five facilities would be to increase the radium-226 concentration by 0.34 pCi/liter for two months with a resultant population dose of 5.2 man-rems. Clearly, at an effluent radium-226 concentration of 9. pCi/liter, the impact of the releases are well within proposed water quality criteria for municipal water intake.[47]

These studies indicate that the impacts associated with discharges to surface waters may not be as significant as some of the impacts previously discussed. Groundwater, an important source of potable water in Florida, contains radionuclides which contribute to background dose. Values of radium concentration of 0 to 76 pCi per liter with 90% being less than 4.6 pCi per liter. Though detailed assessments of the impacts of phosphate facility radionuclide releases on groundwater concentration are not available, consideration of this pathway should not be eliminated.

In Idaho, grab water samples were taken in the Portweuf River, from rainwater runoff and at various water discharge points in both the wet process and thermal process plants. Generally, the liquid fraction was separated from the suspended solids, and these fractions were analyzed separately. Because only a few samples were taken, no definite conclusions could be made regarding the analysis of river samples or about average concentration in process effluents. Due to a slightly higher concentration of ^{226}Ra, uranium, and thorium in sediments, it was suggested that future sampling include detailed analysis of sediments.[17]

It is suggested that extensive sampling be considered in aquatic systems in both Florida and Idaho to assess adequately any releases of radionuclides in waterborne effluents into the environs of phosphate facilities.

3.2.3 Dose estimates associated with the application of uranium bearing fertilizers

The annual distribution of radioactive materials on lands enriched with phosphate fertilizers has been estimated by Pfister in Germany.[35]

To estimate potential dosimetric impact of these radioactive materials, calculations of the annual intake rate of uranium by man via vegetable consumption were made. It was assumed that

1. previously unfertilized land was enriched with phosphate fertilizer;
2. the plow layer, in which the radioactive materials were distributed, is 15-cm deep and the soil bulk density is 1.43 g/cm^3;
3. addition of fertilizers adds only radioactivity and not mass (i.e., no dilution of the radioactivity by the fertilizer mass is assumed);
4. an additional area concentration of 3.9 μCi of uranium per hectare of land resulted from the annual application of the fertilizer.

A concentration of 1.8×10^{-6} μCi uranium per kilogram of soil from a one year addition of fertilizer was calculated. It was assumed that the fertilizer contained only uranium. This can be compared to an average background concentration of 1.5×10^{-3} μCi of uranium per kilogram of soil.[48] The terrestrial background concentration of uranium is approximately 10^3 times the concentration resulting from a single fertilizer application. Using a plant:soil concentration ratio for uranium of 2.9×10^{-4} and an average adult intake of 184 kg/year of fruits, vegetables, and grains, an annual total intake rate (from terrestrial uranium and fertilizer-added uranium) of 85.4 pCi/year is calculated.[48] The intake due to the fertilizer application is 0.1 pCi/year of 0.1% of the total intake of uranium.

To arrive at an estimate of 50-year dose commitment for a person eating foods grown in lands undergoing periodic refertilization, an intake rate function must be developed that accounts for refertilization inputs and losses (leaching, mobility, washout, and erosion) of radionuclides from the plow layer. These parameters are not well known, and intake rate estimates would have large uncertainties.

An alternate assessment of the impact may be obtained by comparing the intake of uranium in a standard diet (both food and fluids) with the intake associated with fertilization of crop lands. The annual dietary intake of reference man* is approximately 265 pCi/year.[49]

If we had assumed that all uranium added by fertilization accumulated without any losses over the 50-year period of interest, the intake rate (associated with fertilizer-added uranium) in the 50th year would be 5 pCi/year. This additional maximum intake rate is still 53 times lower than the yearly intake of uranium in food and fluids by reference man. Any dose commitments associated with the uranium added by cropland fertilization would be correspondingly small.

3.2.4 Dose estimates associated with the use of mined, unmined, and reclaimed lands

The nonoccupational radiological impacts of the phosphate industry that have been examined most thoroughly to date are those associated with technologically enhanced sources of ^{222}Rn (hereafter referred to as radon) and inhalation dose commitments associated with radon progeny.

The EPA, the Florida Land Reclamation Study Commission (FLRSC), the Florida Department of Health and Rehabilitative Services (FDHRS), and

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**Reference Man* (ICRP-23) is a compilation of anatomic, physiological, metabolic, and dietary data recommended for use in the assessment of radiation exposure.[49]

the University of Florida have performed studies of the various aspects of these impacts.[26,48,50,51] The studies by the Office of Radiation Programs (ORP) of EPA involved measurement of radon daughter product concentrations in 125 structures in Polk County, Florida. Track-etch film dosimetry and thermoluminescent dosimetry were used in each structure.

The results were separated into two categories according to land use. These categories are reclaimed phosphate lands and non-reclaimed phosphate lands. The percentile range of radon daughter level concentrations was determined for each land category and for the total sample of structures. For reclaimed lands, the values were:

> 0.05 WL 33.3%

$0.05 \geq x \geq 0.01$ WL 33.3%

< 0.01 WL 33.3%

and for non-reclaimed land:

> 0.05 WL 0%

$0.05 \geq x \geq 0.01$ WL 22.2%

< 0.01 WL 78.8%

For all structures grouped together the percentile range of radon daughter levels were:

> 0.05 WL 19%

0.05 WL $\geq x \geq 0.01$ WL 29%

< 0.01 WL 52%

A more comprehensive study was conducted in Polk and Hillsborough counties (Florida) by FDHRS. In this study 997 structures were examined. Measurements of external gamma radiation levels (both indoors and outdoors), indoor WL concentrations and average annual gamma doses were

Table 22. Radiological measurement associated with structures built on Florida phosphate lands[a]

Land type	Total number of structures (%)	Outdoor mean gamma radiation level (μrad/hr)	Indoor mean gamma radiation level (μrad/hr)	^{222}Rn progeny concentration percentile range (integrating radon daughter samples)		^{222}Rn progeny concentration percentile range (track-etch dosimetry)	
Reclaimed lands	56	10 ± 10	8 ± 4	>0.05 WL	2%	9.2% > 0.029 WL[b]	>0.05 WL 2%
				0.05 ≥ x ≥ 0.01 WL	37%		0.05 ≥ x ≥ 0.01 WL 65%
				<0.01 WL	61%		<0.01 WL 33%
Undisturbed lands no phosphate deposits	33	6 ± 1	6 ± 1			0.0% ≥ 0.029 WL[b]	>0.05 WL 0%
							0.05 ≥ x ≥ 0.01 WL 1%
							<0.01 WL 99%
Undisturbed lands with phosphate deposits	9	7 ± 1	7 ± 1	>0.05 WL	0%	18.6% ≥ 0.029 WL[b]	>0.05 WL <1%
				0.05 ≥ x ≥ 0.01 WL	37%		0.05 ≥ x ≥ 0.01 WL >99%
				<0.01 WL	61%		<0.01 WL 0%
Unknown	2	8 ± 3	7 ± 2				>0.05 WL 0%
							0.05 ≥ x ≥ 0.01 WL 100%
							<0.01 WL 0%
Total	100	8 ± 8	7 ± 4	>0.05 WL	2%		>0.05 WL 1%
				0.05 ≥ x ≥ 0.01 WL	29%		0.05 ≥ x ≥ 0.01 WL 49%
				<0.01 WL	69%		<0.01 WL 50%

[a]U.S. Environmental Protection Agency, *Central Florida Phosphate Industry Areawide Impact Assessment Program*. EPA 904/9-78-006h, March 1978; Florida Department of Health and Rehabilitative Services. *Study of Radon Daughter Concentrations in Structures in Polk and Hillsborough Counties*. January 1978.

[b]0.029 WL control level of the State of Florida Department of Health and Rehabilitative Services.

measured. Of these 997, 56% were built on reclaimed lands and 42% on undisturbed lands (33% of the total were on undisturbed lands with phosphate deposits). The nature of the lands associated with 2% of the structures were unknown. Mean external gamma radiation levels for indoor and outdoor locations are given in Table 22. Indoor gamma radiation levels in structures on undisturbed land were approximately 22% lower than indoor gamma radiation levels for reclaimed lands. The indoor and outdoor measurements taken on undisturbed lands with phosphate deposits did not differ significantly.

Radon progeny measurements (WL) were attempted using radon air sampling techniques in 176 structures from the study sample. Valid measurements were obtained at only 71 of these locations due to technical difficulties with the sampling system. The percentile radon progeny concentration ranges are also given in Table 22 by land type. Measurements of radon progeny concentrations were also made using track-etch dosimetry techniques. Several significant differences exist between the EPA preliminary study and the FDHRS study. In the FDHRS study, 17% less structures had average WL concentrations above 0.05 WL but approximately 20% more structures had WL concentrations in the range of 0.05 to 0.01 WL. The distributions of structures by land type are nearly equal in both studies. In the EPA study, 66% of the structures were on reclaimed land, and in the FDHRS study, 56% of the structures were on reclaimed land. The characteristics of phosphate deposits in nonreclaimed lands and the similarity of these lands to undisturbed lands cannot be determined from the published data.

The FLRSC has performed a study to characterize phosphate lands. Land classifications, acreages, and potential land uses are given in Table 23.[52] In addressing the radiological impacts the FLRSC concluded that:

> "Preliminary data might well indicate an area of significant concern but the data base was so localized and the results so premature that no positive remedial action could be implemented without considerably more definitive and regionalized collection analysis."

Further the Commission stated that:

> "The level of confidence in the measurement of the Radon-222 emanations, the low level of the gamma radiation and the lack of a technique or model to test lands prior to the building of structures does not justify specific reclamation recommendations at the present time. However, the lack of agreement as to the magnitude of the 'problem' between various investigators is of substantial concern to the Commission. The Commission recommends a dedicated effort by the various State and Federal agencies and industry to provide a reliable data base from which the proper course of action can be charted."

A study of the radioactivity of phosphate lands was conducted by the University of Florida from March 1976 to February 1978 and was supported by the Florida Phosphate Council.[26] The six specific objectives of the work were:

"(1) *Independently cross-check indoor radon progeny levels in the phosphate mining region* - sampling in a limited number of structures also sampled by EPA and HRS;

(2) *Determine the variables affecting the radiological characteristics of lands and structures* particularly the relationship to mining and land reclamation;

Table 23. Florida lands classification and potential use[a]

Reclamation classification	1976 Acreage	% of total	Potential land use
Land filled with overburden	83.3	2	Agricultural, industrial, pasture, residential
Land and lakes	2,205.60	51	Agricultural, industrial, pasture, recreational, residential
Land filled with sand	542.0	13	Industrial, pasture, residential
Land filled with sand and capped w/over-burden	351.3	8	Agricultural, industrial, pasture, residential
Settling ponds filled w/clay	680.0	16	Pasture
Settling ponds filled w/clay and capped w/overburden	168.3	4	Pasture
Settling ponds filled w/clay and capped w/overburden and sand	275	6	Agricultural, pasture
TOTAL	4,305	100	

[a]Data from Florida Land Reclamation Study Commission. *Report on Phosphate Mining and Reclamation*. 1978.

(3) *Evaluate the distribution and scope of altered radiological parameters* using data collected in this study and from the reports of others;

(4) *Study and model radon behavior in soil* an input to developing land criteria, land testing methods and recommendations for modifying land radioactivity;

(5) *Evaluate lands criteria and test methods*; and

(6) *Recommend methods for controlling land radioactivity in mining, reclamation and land development.*"

By an independent measurement of radon progeny concentrations (WL) in structures previously measured by the EPA and FDHRS, it was concluded that the University of Florida results are similar to those of the previous studies and that average long-term radon progeny concentrations ranged from <0.001 to ∿0.1 WL. The relationship between the University of Florida measurements and those of the EPA and FDHRS studies is demonstrated in Fig. 6.

Surface soil (0-0.3 m) radon flux was used to characterize the radiologic conditions by land type. The six land types examined were unaltered, unmoved radioactive fill, tailings, overburden, capped and mixed clays, and debris lands. All four land characterization techniques namely surface soil ^{226}Ra concentration, deep soil ^{226}Ra concentration, above ground gamma radiation level and ^{222}Rn flux showed unaltered lands to have the lowest average values for each measurement. Debris lands had the highest average values except for core soil ^{226}Ra concentration, which averaged higher for capped and mixed clays.

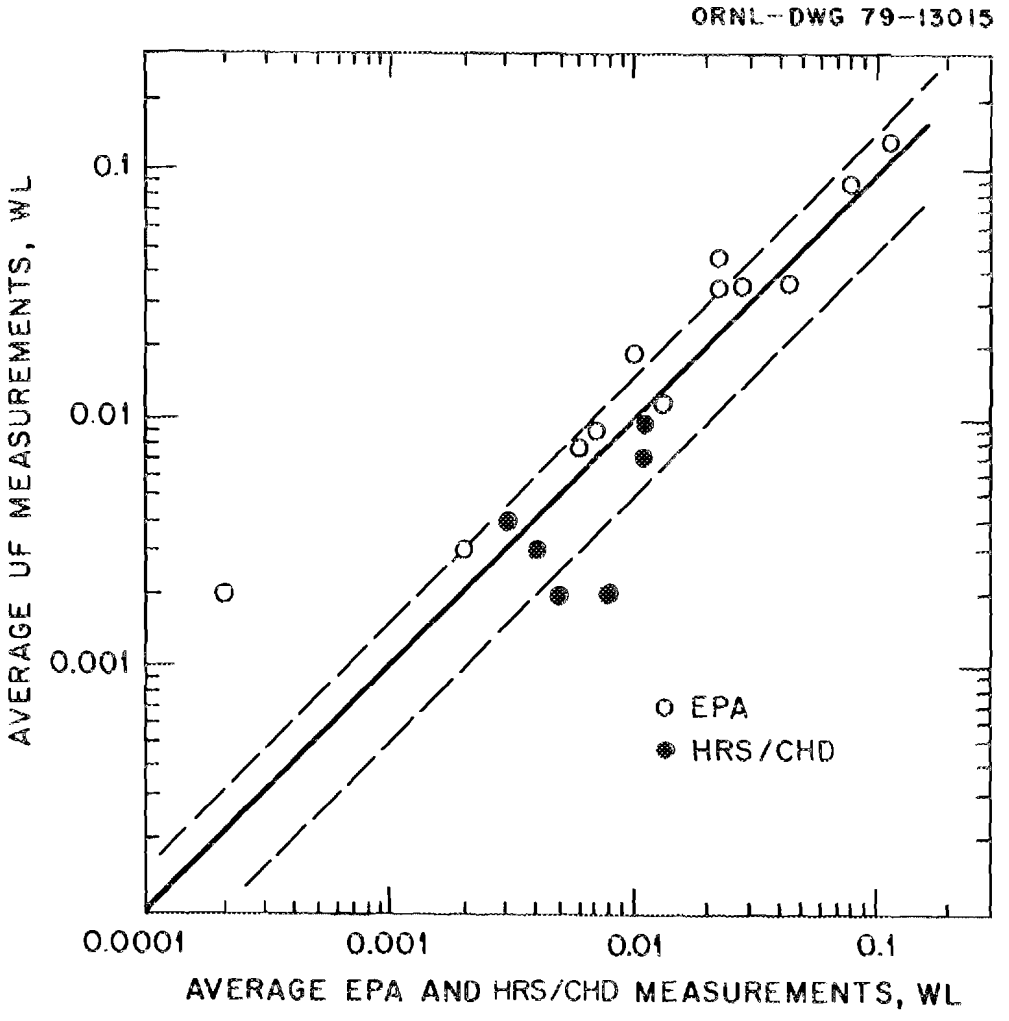

Fig. 6. Correlation of radon progeny concentration measurements made by University of Florida and Florida Department of Health and Rehabilitative Services. Source: Roessler, C. E., Wethington, J. A., and Bolch, W. E. *Natural Radiation Exposure Assessment Radioactivity of Lands and Associated Structures.* Cumulative Summary Report covering March 1976 - February 1978.

For slab-on-grade structures, measurements were made indoors and outdoors. (Slab-on-grade structures are those where floor surfaces are closely coupled with the ground.) Radon progeny concentrations (WL), radon concentrations, and external gamma radiation levels were measured indoors while external gamma radiation levels, radon flux, and surface soil ^{226}Ra concentrations were measured outdoors. The lowest values were obtained for unaltered lands, while highest levels were associated with debris lands.

Through a reexamination of FDHRS along with University of Florida (UF) data and a reconciliation of land classifications, a summary of radon progeny concentrations in structures based on land type and structure type has been made by UF. Included in this summary are estimates of excess population exposure expressed as excess WL-persons above background. These exposures are presented in Table 24 by land type.

Other assessments are being performed in Montana, Idaho, and Alabama by the EPA in order to evaluate gamma radiation and radon progeny exposures in structures where phosphate slag (from the thermal process) has been used as an additive in building and road construction materials.

Screening surveys have been conducted in three areas in Idaho and two areas in Montana. Anomalous gamma radiation levels were noted, and structures suspected of containing slag were identified for radon progeny measurements. These preliminary investigations indicate a need for comprehensive long-term gamma and radon progeny exposure data so that potential health impacts on the public may be identified properly.[53]

Table 24. Estimation of population exposure from elevated radon progeny concentrations[a,b]

Exposure and category	Estimates			Radon progeny concentration, WL		Excess population exposure	
	% of residences	Residences	Persons	Total	Above background	WL-persons	%
A. Reclaimed Land							
1. Slab-on-grade:							
a. High activity overburden and debris lands	0.41	322	1128	0.043	0.040	45.1	37
b. Low activity overburden	1.07	843	2951	0.008	0.005	14.8	12
c. Tailings	1.26	995	3482	0.008	0.005	17.4	14
2. Crawl space and mobile homes: all reclaimed	1.82	1440	5039	0.006	0.003	15.1	12
B. Radioactive deposits and fill							
1. Slab-on-grade	0.60	473	1656	0.019	0.016	26.5	22
2. Crawl space and mobile homes	0.40	316	1105	0.006	0.003	3.3	3
C. Undisturbed lands							
All structures	94.44	74468	260639	0.003			
Total	100.00	78857	276000			122.2	100
		Weighted averages		0.0034	0.00044		

[a]Distribution of percentages, structures and population estimated from (a) total population of the county (1970 census), (b) the HRS estimate that approximately 25% of reclaimed lands structures were included in the HRS study, and (c) land type and structure type distribution of structures sampled by HRS.

[b]Roessler, C. E., Wethington, J. A., and Bolch, W. E. *Natural Radiation Exposure Assessment Radioactivity of Lands and Associated Structures.* Cumulative Summary Report covering March 1976 - February 1978.

4. SUMMARY OF IMPACTS

In this section a summary of 25-year cumulative impacts of the phosphate industry on land use, water use, energy commitments, nonradioactive effluent releases, radioactive effluent releases and radiological dose commitments is given. Some of these impacts have not been discussed in great detail above because they are fully discussed elsewhere.[3,52] These impacts are then put in perspective by comparing them with similar 25-year cumulative impacts of the uranium mining and milling industries as presented in the "Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors" (GESMO).[54] The summary of the land use, water use, and energy use data is given in Table 25 and the summary of effluent data and dose commitment data is given in Table 26.

4.1 Land Use

A total of 201,730 acres of lands have been impacted by the Florida phosphate industry through 1977. Of this total, 127,600 acres are mined out lands, 38,685 acres are disturbed lands, and 35,445 acres are reclaimed lands.[52] In the Idaho phosphate activity there is a proposed commitment of 16,000 acres of new development through the year 2000.[3] A total of 366,300 acres of land have been or will be disturbed during the period 1975 to 2000 for uranium mining and milling activities and an additional 41,500 acres of land will be permanently committed during this period.[54]

4.2 Water Use

The current (1976) water use by the Florida Phosphate Industry is 1.4×10^{15} gal/day and it is projected to be 2.0×10^{15} gal/day in the year 2000.[46] These total water consumption rates include both municipal

Table 25. A summary of some important resource impacts associated with the phosphate industry

	Land use	Water use	Energy use
Uranium Mining[a] 1975 to 2000 (No plutonium recycle)	3.34×10^5 acres disturbed 1.1×10^4 acres permanently committed	3×10^{12} gallons discharged to ground	2.87 GWY electric 2×10^9 gallon fuel oil
Uranium Milling[a] 1975 to 2000 (No plutonium recycle)	3.2×10^4 acres distributed 3.1×10^4 acres permanently committed	1.15×10^{12} gallons discharged to air	6.70 GWY electric 1.3×10^9 therms (gas) (1.06×10^8 J/therm)
Florida Phosphate Industry	through 1977[b] 1.3×10^5 acres of mined out lands 3.5×10^4 acres of reclaimed lands	1.4×10^{15} gallon/day (1976) 2.0×10^{15} gallon/day (2000) Water consumption rates include municipal and mining needs.[d]	for 1978 to 2000 10 GWY electric Includes industrial demand for the period 1978 to 2000.[d]
Idaho Phosphoria Region Proposed Mining Activity	1.6×10^4 acres of new development[c]	9.75×10^9 gallon/day[c] Water consumption rates include municipal and mining needs.	for 1976 to 2000[c] 16 GWY electric[c] 2.73×10^9 cu. ft of gas 3.10×10^5 short-tons coal 2.9×10^8 gallon diesel fuel 1.6×10^{10} gallon gasoline

[a]U.S. Nuclear Regulatory Commission, *Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors*, NUREG-0002 (GESMO) (1976).

[b]Florida Land Reclamation Study Commission. *Report on Phosphate Mining and Reclamation*. 1978.

[c]U.S. Department of the Interior, U.S. Department of Agriculture. *Final Environmental Impact Statement – Development of Phosphate Resources in Southeastern Idaho*. (not dated).

[d]U.S. Environmental Protection Agency, *Central Florida Phosphate Industry Areawide Impact Assessment Program*. EPA 904/9-78-006h, March 1978.

Table 26. A summary of some important radiological and nonradiological impacts associated with the phosphate industry

	Effluents		Radiological dose commitments	
	Nonradioactive	Radioactive	Occupational	Nonoccupational
Uranium Mining[a] 1975 to 2000 (No plutonium recycle)	SO_x 8.3 x 10^4 MT NO_x 6.8 x 10^4 MT Particulates 9.9 x 10^3MT	^{222}Rn 23.7 MCi Uranium's daughters	1.2 x10^6 person-rem	2.9 x 10^6 person-rem
Uranium Milling[a] 1975 to 2000 (No plutonium recycle)	SO_x 550 MT NO_x 1.09 x 10^5 MT	^{222}Rn 4.4 MCi Uranium and daughters 5.4 MCi	5.6 x 10^5 person-rem	5.79 x 10^5 person-rem
Florida Phosphate Industry	SO_x little impact on air quality expected[b] NO_x ——— Particulates[b]	^{222}Rn 1975 to 2000 2.6 MCi @ 10.1 x 10^4 Ci/yr[c]	Gamma dose (1975 to 2000) 10^5 person-rem Bkg gamma dose = 7.5 x 10^4 person-rem Total lung dose = 1.5 x 10^5 to 3.5 x 10^5 person-rem Bkg lung dose = 1.65 x 10^4 to 1.65 x 10^5 person-rem	1975 to 2000 ^{222}Rn 5.75 x 10^5 person-rem (0.034% of the total ^{222}Rn population dose[c])
Idaho Phosphoria Region Proposed Mining Activity	SO_x[d] NO_x Particulates[d]	No data available	No data available	No data available

[a]U.S. Nuclear Regulatory Commission, *Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors*, NUREG-0002 (GESMO) (1976).

[b]"The proposed action will have little impact on the air quality in the study area. SO_2 and dust emissions in Polk County because of drying, grinding, and transportation will decrease as mines in that county are depleted and new mines are opened elsewhere. Other nonphosphate industry sources, however, will offset the decreases. The new mines will ship wet rock, thus preventing the migration of an estimated 1140 metric (1250 short) tons per year of dust and 7090 metric (7800 short) tons per year of SO_2 emissions from dryers in Polk County into adjoining areas."[45]

[c]Travis, C. C., Watson, A. P., McDowell-Boyer, L. M., Cotter, S. J., Randolph, M. L., and Fields, D. C., *A Radiological Assessment of ^{222}Rn Released from Uranium Mills and Other Natural and Technologically Enhanced Sources*, NUREG/CR-0573, ORNL/NUREG-55 (February 1979).

[d]"Based upon mining plans as originally submitted and the indicated plant expansions, the primary impact on air quality attributable to the development of phosphate resources in southeastern Idaho would be from the growth of the existing plants. The additional five plants scheduled during the next two decades, with one exception, are expected to exceed any of the primary or secondary ambient air quality standards for SO_2, particulate or fluoride as well as the Class I and II PSD Standards."[3]

and mining needs. A similar total water consumption rate for the Idaho phosphate proposed activity is projected to be 9.8×10^9 gal/day.[3] The total commitment of water resources to uranium mining and milling is projected to be 4.2×10^{12} gallons discharged to the environment during the period 1975 to 2000. This corresponds to a daily discharge rate of approximately 4.6×10^8 gal/day.

4.3 Energy Use

Energy use by the Florida Phosphate Industry is estimated to be 10 GWY (electric) for the period 1978 to 2000 and 16 GWY (electric) for the proposed Idaho phosphate activity.[46] The total electric consumption for the uranium mining and milling industry is estimated to be approximately 10 GWY for the period 1975 to 2000.[44] Where data are available consumption of fossil fuels namely, natural gas, petroleum fuels and coal are also given in Table 25.

4.4 Effluents

Comparisons of effluent releases associated with the phosphate industry can be separated by effluent type. Discussions of major effluent releases are divided into nonradiological effluents namely SO_x, NO_x, and suspended particulates and radiological effluents which include ^{222}Rn and its daughter products and uranium and its immediate daughter products.

4.4.1 Nonradioactive effluents

The impacts of SO_x and particulate releases in the Florida phosphate region are expected to be mitigated by newly applied techniques of shipping of marketable rock. The shipping of wetted rock is projected to mitigiate the release of 141,800 metric tons of SO_2 at drying facilities during the period 1980 to 2000.[46] As a result little impact on air

quality is expected from SO_2 releases. Similarly, the mitigation of 22,800 metric tons of particulates is expected during the same period.[46]

In the case of the Idaho proposed phosphate mining activity some impacts due to SO_2 and particulate releases are expected. As stated in the regional environmental impact statement:

> "Based upon mining plans as originally submitted and the indicated plant expansions, the primary impact on air quality attributable to the development of phosphate resources in southeastern Idaho would be from the growth of the existing plants. The additional five plants scheduled during the next two decades, with one exception, are expected to exceed any of the primary or secondary ambient air quality standards for SO_2, particulate or fluoride as well as the Class I and II PSD Standards."[3]

The estimated releases of SO_2 and particulates from facilities associated with the uranium mining and milling industry are 83,500 metric tons of SO_x and 15,200 metric tons of particulates during the period 1975 to 2000.[54] Further, it is estimated that releases of NO_x will amount to approximately 120,000 metric tons during the same period. No values for NO_x are reported for phosphate industrial activities.

4.4.2 Radioactive effluents

The radiological quality of the environs of phosphate facilities and the radiological impacts of phosphate industrial activities on occupational and nonoccupational populations have been discussed in detail in Sections 3.1 and 3.2. While these impacts cannot be comprehensively summarized by evaluation of a single release or insult, a course comparison may be obtained by doing so. Releases of ^{222}Rn may provide such a value for comparison.

It is expected that 2.6×10^6 Ci of ^{222}Rn will be released as a result of phosphate activities in Florida during the period 1975 to 2000.[55]

No estimates have been given for the Idaho proposed action. Further, it is estimated that 2.8×10^7 Ci of ^{222}Rn will be released during the normal operations of uranium mining and milling facilities during the period 1975 to 2000.[54] It is also estimated that 5.4×10^6 Ci of uranium and its immediate daughter products will be released in airborne effluents during the same period from uranium production facilities.[54]

4.5 Radiological Dose Commitments

It has been estimated that the cumulative occupational gamma dose is 10^5 person-rem and that the cumulative occupational lung dose is between 1.5×10^5 and 3.5×10^5 person-rem to the lung for phosphate industry workers for the period 1975 to 2000. The background gamma and lung doses to this population for this period are 7.5×10^4 person-rem and 1.7×10^4 to 1.7×10^5 person rem, respecitvely. The occupational dose for the same time period for uranium mining and milling workers is estimated to be 1.8×10^6 person-rem.[54]

The nonoccupational dose associated with the phosphate industry is characterized by estimating the total cumulative population dose from ^{222}Rn for the period 1975 to 2000. This dose is estimated to be 5.8×10^5 person-rem.[55] The nonoccupational total cumulative population dose associated with uranium mining and milling for the same period is estimated to be 3.6×10^6 person-rem.[54]

4.6 Discussion

The comparison of radiological and nonradiological impacts associated with the phosphate industry to analogous impacts of the uraniun mining and milling industries indicate that most impacts are within an order of

magnitude. If the magnitude of various impacts are normalized per unit U_3O_8 production, the impacts are comparable. Recall that the realistic estimate for annual uranium production from phosphate resources was 3000 short-tons of U_3O_8 and that the reported average U_3O_8 production during the period 1965 to 1971 from uranium mining and milling was 11,000 short-tons per year. The relative production of U_3O_8 from phosphate sources is approximately a factor 3 lower than from uranium mining and milling sources. The ^{222}Rn releases were a factor of 11 lower for phosphate industrial activities as compared to uranium mining and milling while occupational doses and nonoccupational doses were factors of approximately 7 and 6 lower, respectively. These numbers are not presented as quantitative assessments of the relative impacts of the phosphate industry to those associated with the uranium mining and milling industries. These values do indicate that the impacts of the phosphate industrial activity may be comparable to uranium mining and milling and that these impacts are associated with normal phosphate industrial activities apart from uranium recovery. This results from the fact most significant radiological impacts result from the release and redistribution of ^{226}Ra and ^{222}Rn and its daughter radionuclides during mining and processing of phosphate ore as discussed in Section 3.2.

The radiological impacts of intensified uranium recovery in central Florida from wet process phosphoric acid have been fully studied elsewhere.[44] The study by Davis, et al. concluded that:

> "Releases of radioactive materials from uranium recovery plants result in a negative impact (increased dose commitment) on the populations surrounding the plants. On the other hand, removal of uranium and other radionuclides from phosphoric acid prevents their distribution on farm lands, and urban gardens and grasses via fertilizers; this results in a positive impact (decreased dose commitment) on the associated populations."[44]

Further, it was concluded that assessment of the impacts of increased uranium recovery from wet process phosphoric acid should include assessment of transportation, siting, economic, social, environmental, and health impacts.[44]

5. CONCLUSIONS

The phosphate industry in the United States currently has a realistic potential of delivering approximately 3000 tons of U_3O_8 per year (through the year 2000) as a co-product of wet process phosphoric acid production. If phosphate facilities operate at capacity and if the technology for uranium extraction from beneficiation wastes is developed, levels of uranium production could be increased by about a factor of 5. This estimate must be considered optimistic as detailed in Section 1.0.

Two processes are used to produce various phosphate products. The wet process is used to produce wet process phosphoric acid which in turn is used to produce phosphate fertilizer products. The thermal process is a combustion process used to produce elemental phosphorus, reagent grade phosphoric acid and other products. It is in these processes that enhancement of the radioactive components of phosphatic input materials occurs. The concentration of uranium in normal superphosphate, triple superphosphate, and diammonium phosphate is approximately 1.5 times the concentration in the marketable rock. Radium (about 80% of the amount in the input rock) remains with the gypsum waste. Though it is concentrated in phosphoric acid, a significant amount of thorium goes into gypsum waste because on a mass basis about 5 times more gypsum than phosphoric acid is produced.

In thermal processing, essentially all of the radioactivity is contained in the slag and ferrophosphorus. Slag is used as a lightweight aggregate in building materials and road ballast. Ferrophosphorus is used as an additive in steel production.

Occupational impact assessments are concerned with radiation doses that workers receive as a result of their employment, whereas nonoccupational impact assessments are concerned with radiation doses received by persons in the general population as a result of phosphate industry activities. The annual gamma radiation dose to the phosphate industry worker is approximately 1.5 times the background dose. The annual lung dose may be from 2 to 4 times the annual background lung dose depending upon the model used in calculating average lung dose and the actual background gamma and lung doses associated with workers' location and type of residence.

The most significant nonoccupational impacts are associated with airborne releases of particulate matter from wet process and thermal process phosphate facilities and the use of reclaimed phosphate lands. The red bone marrow and endosteal cells of the bone receive the highest relative doses from atmospheric releases from phosphate facilities via the ingestion pathway. The liver and kidneys also receive significant doses via this pathway. (The ingestion pathway begins with radioactive materials depositing on the food crops, and these crops are subsequently eaten by the local population.) The thorium radionuclides and ^{226}Ra contribute the largest part of the dose with kidneys receiving the largest dose from ^{234}Th and ^{214}Bi. Details of the calculations, which are presented above, may provide a tool with which radiological surveillance programs may be designed.

The impacts associated with waterborne effluents result from water discharges associated with slime waste storage ponds and gypsum pile

runoff to surface waters. No significant impacts are projected for this pathway. Impacts on groundwater quality need to be examined further.

The application of uranium-bearing phosphate fertilizer to crop lands and the subsequent ingestion of crops grown on these lands was also assessed. This pathway of exposure is distinct from the ingestion pathway associated with airborne releases in that the dose results from root uptake of radionuclides contained in the soil medium. It was determined from the calculations presented in Section 3.2.3 that any dose due to ingestion of uranium associated with this pathway is very small when compared to the total dietary intake of uranium by reference man.

The use of reclaimed phosphate lands has the radiological impact that has come under the most extensive examination. In Florida, both state and federal agencies and the University of Florida have examined the exposure of people living on reclaimed phosphate lands. For 42 locations (33 with no phosphate deposits and 9 with phosphate deposits), working levels between 0.01 WL and 0.05 WL were measured in 24% of the structures while in 76% of the structures, working level values of less than 0.01 WL were obtained. On reclaimed lands, 2% of measured values were greater than 0.05 WL, 65% were between 0.01 WL and 0.05 WL, and 33% were less than 0.01 WL. These studies indicate that an enhancement of the natural radiation environment has occurred due to phosphate mining activities. The Florida Department of Health and Rehabilitative Services has established 0.029 WL as a suggested control level. For those structures on phosphate lands with no significant deposits, working level values did not exceed 0.029 WL while in 18.6% of the structures built on undisturbed lands with phosphate deposits, working levels in excess of 0.029

WL were found. On reclaimed lands, 9.2% of the values exceeded 0.029 WL. Efforts aimed at evaluating these same impacts are underway in Montana and Idaho.

As a result of this assessment of the phosphate industry in the United States, it is concluded that though some of the radiological impacts are known and understood, further work is required to detail these impacts. It is recommended that long-term sampling programs be established both in phosphate facilities and in the environs of phosphate facilities in order to measure radiation exposures to phosphate industry workers and the transport of radionuclides via airborne and waterborne effluents, respectively. From environmental transport data, radiation dose commitments to populations living in the environs of phosphate facilities may be estimated. Further, it is recommended that consideration be extended to nonnuclear pollutants such as arsenic, chromium, cadmium, vanadium, fluorine, and respirable dusts because, as indicated by the NIOSH surveys, these pollutants have been found to exist in air in significant concentrations in and around phosphate facilities. Measurements in residential structures built on reclaimed lands indicate that the radiation environment has been enhanced in some structures. Careful attention to reclamation practices and the eventual uses of reclaimed lands may allow for further reduction of population exposures. The information presented above along with new data obtained from more detailed and long-term in-plant and environmental sampling programs, more definitive answers regarding radiological impacts of the phosphate industry can be made.

Even though more detailed sampling information would be useful in assessing the radiological and nonradiological impacts of the phosphate industry certain comparisons can be made between existing data and data for similar impacts associated with uranium mining and milling. These comparisons, discussed in Section 4 indicate that radiological impacts comparable to uranium production impacts are associated with ore mining and beneficiation; thermal processing of marketable rock; wet processing of marketable rock and land reclamation, independent of uranium corecovery. Further nonradiological impacts associated with the phosphate industry also exist independant of uranium corecovery. While the impacts, both detrimental and beneficial, associated with uranium corecovery have been documented and discussed in Section 4.5 the assessment of net impacts should include an evaluation of the net gain in uranium resources as well.

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