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RADIOISOTOPIC COMPOSITION OF YELLOWCAKE An Estimation of Stack Release Rates

by

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December 1979

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Prepared for
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555
Under Interagency Agreement DOE 40-550-75
NRC FIN No. A2046

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FOREWORD

The Argonne National Laboratory, under contract with the U.S. Nuclear Regulatory Commission (NRC), Division of Safeguards, Fuel Cycle and Environmental Research, has conducted extensive long-term field studies at operating uranium mills to provide input for the Generic Environmental Impact Statement on Uranium Milling being prepared by the NRC.

The overall field study program was intended to:

- (1) Provide measurements that could be used to characterize and to estimate the rates of release of airborne radioactive effluents (source terms) for uranium milling activities. This would include release rates of radioactive particulates (specific nuclides) and radon-222 from (a) mill stacks and vents, (b) ore piles, and (3) tailings piles.
- (2) Provide data that could be used to verify predicted offsite environmental concentrations of radionuclides based on source terms and dispersion calculations.
- (3) Provide data that could be used to evaluate the potential radiological significance of food ingestion pathways for airborne effluents released from uranium mills, focusing primarily on ingestion exposures associated with grazing animals and locally raised food crops or garden produce.
- (4) Serve as a means to test, demonstrate, and evaluate environmental monitoring methods and techniques in order to provide information that could be used in the development of guidance for monitoring programs.

As one part of this broad field study program, measurements were made in order to characterize the particulate emissions and release rates from yellowcake packaging and drying stacks. The results are presented in this report. Uranium mill concentrate (yellowcake) from active operating mills was analyzed for U-238, U-235, U-234, Th-230, Ra-226, and Pb-210 by radiochemical methods. The results are interpreted and correlated with stack discharge measurements made by Fort (1979) using isokinetic sampling procedures. A brief description of the milling processes associated with the mills where data were collected and details of the procedures for radiochemical analysis are also included.

The authors express their appreciation to Drs. L. Roberts, G. Montet, R. Holtzman and Mr. W. Hallett (Argonne National Laboratory), L. Schwendiman (Battelle-Pacific Northwest Laboratory), P. Magno, W. Fort and D. Hendricks (U.S. Environmental Protection Agency) for their critical reviews of this report before publication. The authors are indebted to the management and staff of Kerr-McGee Nuclear Corporation, Anaconda Company (Bluewater NM Operation), Exxon (Highland Uranium Operation), and Union Carbide (Uravan Uranium Operation) for their cooperation and assistance.

The overall study was conducted by the Division of Environmental Impact Studies of Argonne National Laboratory. This activity was monitored by Harry Landon, Paul Magno (now with EPA-Washington), Hubert Miller, and presently Ms. Laura Santos (NRC). Stack release rate measurements were conducted by C. W. Fort (1979) from the U.S. Environmental Protection Agency (EPA), Las Vegas Office of Radiation Programs.

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RADIOISOTOPIC COMPOSITION OF YELLOWCAKE: An Estimation of Stack Release Rate

by

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Abstract

Uranium concentrate (yellowcake) composites from four mills (Anaconda, Kerr-McGee, Highland, and Uravan) were analyzed for U-238, U-235, U-234, Th-230, Ra-226 and Pb-210. The ratio of specific activities of U-238 to U-234 in the composites suggested that secular radioactive equilibrium exists in the ore. The average activity ratios in the yellowcake were determined to be 2.7×10^{-3} (Th-230/U-238), 5×10^{-4} (Ra-226/U-238) and 2×10^{-4} (Pb-210/U-238). Based on earlier EPA measurements of the release rates from the stacks, the amount of yellowcake released was determined to be 0.1% of the amount processed.

INTRODUCTION

Any release of uranium product, i.e., "yellowcake", from mill stacks during the drying process is a source of environmental contamination. The rate of yellowcake escape from the dryer and packaging stacks is dependent on the efficiency of the stack dust-collecting systems. Further, the potential health effects of the yellowcake are dependent on both the particle size and the solubility of the radionuclides in biological tissues. The solubility varies according to the chemical and physical processes used for uranium extraction and drying. Data reported in the literature suggest a thorium-230 activity of 0.9% (ion exchange) to 5.3% (solvent extraction) and radium-226 activity of 0.06 to 0.26% of the U-238 in the yellowcake produced by acid leach processing (Merritt, 1971; HEW, 1952). No estimate for lead-210 activity was found in earlier literature.

The current chemical purity standards are a reflection of industrial purification capabilities rather than concern for the potential effects of environmental releases. The existing specifications limit the maximum thorium content of yellowcake to about 2% by weight (Hardy, 1972; Merritt, 1971; Nelli, 1959; Jodra and Josa, 1962). The elemental lead content of yellowcake has been estimated at 0.1-1% (Galkin, 1963). But, in order to estimate potential environmental and health effects of yellowcake releases, the amount of radioactivity in yellowcake due to the nuclides Th-230, Ra-226, and Pb-210 must be known.

Yellowcake, generally designated as U_3O_8 , is actually a complex mixture of compounds containing uranium in different oxidation states, and often including diuranates, peroxide hydrates, basic uranyl sulfate and other ions (Merritt, 1971). The two common oxidation states of uranium are U^{4+} and U^{6+} , the U^{4+} being readily oxidized to U^{6+} state. Minerals containing uranium as U^{4+} are generally dark colored and do not exhibit fluorescence in ultraviolet light. These ores characteristically contain numerous other elements such as Th, Ca, Zr, Ce, and other rare earths, substituting for the U^{4+} ion.

Uranium in the U^{6+} state forms two series: uranate and uranyl compounds. The uranate series has a general formula $M_2^{+}U^{6+}_XO_{3X+1}$. These minerals are generally red, orange, or brown in color. The uranyl series contains $(UO_2)^{2+}$ ion, which is sufficiently stable in solution to retain its identity. The uranyl colors are generally yellow or green and uranyl compounds fluoresce in similar hues.

After extraction from the ore the uranium is precipitated, dewatered by centrifugation, vacuum filtration and pressing, and dried at temperatures exceeding $125^{\circ}C$. The color of the product is highly dependent upon the temperature of the drying (or roasting) process. At low temperatures ($< 150^{\circ}C$) the product is the bright yellow color of ammonium diuranate $((NH_4)_2 \cdot U_2O_7)$ or magnesium diuranate, while at higher temperatures it changes color to the olive green of U_3O_8 and eventually forming black UO_2 . As a result, the color of the final product may be yellow, green, brown, or black, depending on many process parameters.

The purpose of this study was to characterize yellowcake by its radioisotopic composition and estimate the release rate from stacks and packaging operations. Accordingly, samples were obtained from the following uranium mills: Anaconda Company (Grants, NM), Kerr-McGee Nuclear Corporation (Grants, NM), Union Carbide (Uravan, CO) and Exxon (Douglas, WY). These mills process the major portion of uranium mined in the United States and also represent the different geographical uranium-producing regions. Their processing characteristics are summarized below and have been previously reported in detail by Merritt (1971), Guccione (1974) and Coleman (1978).

Anaconda Company, Bluewater, NM

Leaching: Sulfuric acid and MnO_2

Uranium Separation: Resin-in-pulp ion exchange (IRA-430 anion exchange resin with $FeSO_4$ and MgO) to January 1, 1978. The process for extraction was changed to solvent extraction after an expansion in 1978.

Yellowcake Precipitation: Addition of MgO

Yellowcake Drying: 200 to 250°F (steam dryer)

Product Grade: Average of 87% U_3O_8 , yellow color

Kerr-McGee Nuclear Corporation, Ambrosia Lake, NM

Leaching: Sulfuric acid and $NaClO_3$

Uranium Separation: Solvent extraction with Alamine as extractant and $NaCl$ as stripping agent.

Yellowcake Precipitation: Ammonia and air at 140°F

Yellowcake Drying: 320 to 360°F (steam dryer)

Product Grade: 88 to 90% U_3O_8 , yellow color

Union Carbide Uravan Mill, Uravan, CO

Leaching: Sulfuric acid and $NaClO_3$

Uranium Separation: Column ion exchange with anion exchange resin

Yellowcake Precipitation: Ammonia at 120°F

Yellowcake Drying: 500-700°F (multiple-hearth roaster).

Product Grade: Not reported, yellow-brown color

Exxon Highland Mill, Douglas, WY

Leaching: Sulfuric acid with oxidant

Uranium Separation: Solvent extraction

Yellowcake Precipitation: Ammonia

Yellowcake Drying: > 900°C (UO₂ formed as product)

Product Grade: Not reported, gray-black color

EXPERIMENTAL PROCEDURES

Composite samples representing average monthly production of yellowcake were obtained from each mill. The composites were prepared by the staff of each mill using a mechanical sampling procedure. An aliquot portion of each yellowcake lot* was taken and then combined into a single monthly composite. The yellowcake composites were dissolved in concentrated nitric acid and hydrogen peroxide (30%) was added to convert all uranium to the hexavalent state. Aliquots were taken for isotopic analysis and the specific activities of U-238, U-235, U-234, Th-230, Ra-226 and Pb-210 were determined. Only a single analysis per composite for each radionuclide was possible within our time-scheduled protocol. For each extraction, a run of 8 to 14 samples, a blank was also included in the analysis.

Measurement of U-238, U-235 and U-234 Activities

A sample of yellowcake, less than 0.6 gram, was dissolved in nitric acid as a stock solution (250 ml) for separation of uranium, thorium, radium and lead. The uranium from an aliquot of this solution was extracted from the nitric acid phase into bis (2-ethylhexyl) phosphoric acid (HDEHP), and was then back-extracted with 11M HCl. The HCl was evaporated, the residue oxidized and then dissolved in an oxalic acid and ammonium chloride medium. The uranium in an aliquot of this solution (about 50 lambda) was electroplated onto stainless steel disks by means of a system similar to those described by Puphal and Olsen (1972). Samples were electroplated at a pH of 6 and a current of 2.0 amperes for about four hours. Total area of deposition on each disc was 1.13 cm². The discs were counted by alpha spectrometry for U-238 (4.20 MeV), U-235 (4.40 MeV) and U-234 (4.77 MeV) for durations of about 60

* A "lot" of yellowcake contains 72 drums, each drum has a volume of 50 gallons and contains less than 500 kg of yellowcake.

minutes. The extraction yield for each isotope sample was estimated using U-232 (5.32 MeV) as a tracer. The detection limits at the 90% confidence limit were greater than 0.02 pCi, 0.06 pCi and 0.04 pCi for U-238, U-235 and U-234, respectively. (See Appendix for details.)

Measurement of Th-230 Activity

Thorium was separated by extraction from nitric acid into a solution of 30% quaternary amine (Aliquat-336) in xylene (Sill, 1974), from an aliquot of the stock solution. The thorium was stripped from the organic phase with 10M HCl and electroplated in a manner similar to the uranium deposition. Th-230 (4.68 MeV) alpha activity was determined by alpha spectroscopy. Th-232 (4.01 MeV) and Th-228 (5.43 MeV) activities were not detectable in any of the yellowcake samples. Chemical separation yield was determined using Th-234 as a tracer. Since Th-234 is a beta emitter, its activity was determined by means of gas proportional counter. The detection limit for Th-230 at the 90% confidence limit was greater than 0.07 pCi.

Measurement of Ra-226 Activity

Radium was determined by a de-emanation method similar to that of Rushing et al. (1964). A 10-ml aliquot of stock solution of yellowcake dissolved in nitric acid was transferred to a de-emanation radon bubbler vessel. Rn-222, the 3.82-day half-life daughter of Ra-226, was allowed to grow into secular radioactive equilibrium with its parent in solution and then was swept into an evacuated Lucas cell by bubbling radon-free nitrogen through the solution. The activity in the Lucas cell was then counted with a scintillation counter for Rn-222 and its short-lived progeny (Po-218 and Po-214) after secular radioactive equilibrium was established. Each Lucas cell was calibrated for efficiency with a standard radium solution. The detection limit for Ra-226 at the 90% confidence limit was greater than 0.04 pCi.

Measurement of Pb-210 Activity

Lead was separated by the procedure of Sill and Willis (1965). It was extracted from nitric acid into 1% diethylammonium diethyldithiocarbamate in

chloroform, which isolates the Pb-210 (and its daughter, Bi-210) from other metals such as iron and aluminum. The organic phase was evaporated, the sample was oxidized and the residue dissolved in dilute HCl. The pH was adjusted to 9.5 with ammonium hydroxide and the lead and bismuth extracted into diphenylthiocarbazone (dithizone) in chloroform in the presence of citrate and cyanide ions. The lead was separated from the bismuth by back-extraction with a pH-2.71 buffer. The organic phase containing the bismuth was evaporated, then oxidized with nitric acid and 30% hydrogen peroxide. The residue was dissolved in nitric acid, evaporated on a 1.5-inch stainless steel cupped planchet and beta-counted for Bi-210 with a gas proportional counter. The activity of Pb-210 was calculated from the activity of Bi-210. Lead-210 standards were counted for each run and the yields were determined. The average yield in these determinations was about 80%. The detection limit for Pb-210 for the 90% confidence limit was greater than 3.8 pCi.

RESULTS AND DISCUSSION

Results of measurements of U-238, U-234, U-235, Th-230, Ra-226, and Pb-210 in the monthly composites from the Anaconda Mill (Bluewater, New Mexico) are given in Table 1. The range of values for uranium activity was 2.27×10^5 to 3.45×10^5 pCi U-238 per gram of dry yellowcake, with an average of 2.79×10^5 pCi U-238/g and standard deviation (ϵ) of 6.5×10^4 pCi U-238 per gram. The isotopic distribution of natural uranium is given in Table 2. Since natural uranium contains 3.31×10^5 pCi U-238/g, the yellowcake from Anaconda Mill averaged more than equivalent 99% U_3O_8 , based on the measured U-238 specific activity. Similarly, the average specific activity of U-234 in the composites was 2.71×10^5 pCi/g ($\epsilon = 6.5 \times 10^4$) with a range of 2.04×10^5 to 3.41×10^5 pCi/g. This average activity corresponds to a composition of 96.5% U_3O_8 in the yellowcake. Because of its bright yellow color, we surmised that the yellowcake from Anaconda Mill was to a great extent magnesium diuranate rather than olive green U_3O_8 . Since the U_3O_8 is about 85% uranium by weight, the observed U_3O_8 equivalent is smaller than the actual uranium content would be if it were magnesium diuranate (78% by weight). The Anaconda Company specifies their product to average 87% U_3O_8 --a value that is smaller than either the 99% or 96.5% estimates obtained from U-238 and U-234 activity measurements, respectively.

Table 1. Radioactive Composition of Yellowcake Measured in Samples
of Composites Obtained from Anaconda Mill

Composite Sample	Grams Analyzed	Concentration (pCi/g dry wt) $\pm$ Standard Error of the Mean					
		U-238	U-235	U-234	Th-230	Ra-226	Pb-210
Dec. 1977	0.2086	$2.51 \times 10^5 \pm 1.4 \times 10^4$	$1.35 \times 10^4 \pm 2.5 \times 10^3$	$2.31 \times 10^5 \pm 1.4 \times 10^4$	-----	39.2 ± 2.3	155.5 ± 24.8
Jan. 1978	0.3127	$2.90 \times 10^5 \pm 1.7 \times 10^4$	$1.48 \times 10^4 \pm 3.5 \times 10^3$	$2.81 \times 10^5 \pm 1.7 \times 10^4$	$3.15 \times 10^3 \pm 2.1 \times 10^2$	18.7 ± 1.0	115.7 ± 16.6
Feb. 1978	0.2715	$3.05 \times 10^5 \pm 1.9 \times 10^4$	$1.22 \times 10^4 \pm 3.8 \times 10^3$	$2.99 \times 10^5 \pm 1.9 \times 10^4$	$4.58 \times 10^2 \pm 4.0 \times 10^1$	54.6 ± 2.9	32.9 ± 18.5
Mar. 1978	0.2296	$2.52 \times 10^5 \pm 1.4 \times 10^4$	$1.21 \times 10^4 \pm 2.8 \times 10^3$	$2.64 \times 10^5 \pm 1.4 \times 10^4$	$3.48 \times 10^2 \pm 4.3 \times 10^1$	83.8 ± 5.5	21.8 ± 21.9
Apr. 1978	0.2808	$2.93 \times 10^5 \pm 2.0 \times 10^4$	$1.56 \times 10^4 \pm 4.9 \times 10^3$	$3.13 \times 10^5 \pm 2.0 \times 10^4$	$3.79 \times 10^2 \pm 5.6 \times 10^1$	5.0 ± 5.6	-52.7 ± 21.5
May 1978	0.1825	$2.27 \times 10^5 \pm 1.6 \times 10^4$	$1.05 \times 10^4 \pm 3.1 \times 10^3$	$2.04 \times 10^5 \pm 1.5 \times 10^4$	$8.57 \times 10^2 \pm 6.6 \times 10^1$	4.6 ± 1.8	25.0 ± 27.4
June 1978	0.2091	$2.30 \times 10^5 \pm 1.5 \times 10^4$	$9.84 \times 10^3 \pm 2.9 \times 10^3$	$2.16 \times 10^5 \pm 1.4 \times 10^4$	-----	48.4 ± 1.9	11.3 ± 24.8
July 1978	0.1918	$3.36 \times 10^5 \pm 2.2 \times 10^4$	$1.64 \times 10^4 \pm 5.3 \times 10^3$	$2.57 \times 10^5 \pm 1.9 \times 10^4$	$4.33 \times 10^2 \pm 5.2 \times 10^1$	5.2 ± 1.8	-29.6 ± 31.8
Aug. 1978	0.2247	$3.45 \times 10^5 \pm 2.7 \times 10^4$	$1.71 \times 10^4 \pm 5.3 \times 10^3$	$3.41 \times 10^5 \pm 2.6 \times 10^4$	$3.23 \times 10^2 \pm 4.1 \times 10^1$	-4.3 ± 3.6	31.4 ± 22.3
Sep. 1978	0.2270	$2.67 \times 10^5 \pm 1.4 \times 10^4$	$1.28 \times 10^4 \pm 3.1 \times 10^3$	$2.59 \times 10^5 \pm 1.3 \times 10^4$	$1.97 \times 10^2 \pm 6.8 \times 10^1$	2528 ± 3.6	-12.5 ± 27.0
Oct. 1978	0.2298	$2.64 \times 10^5 \pm 1.8 \times 10^4$	$1.19 \times 10^4 \pm 3.7 \times 10^3$	$2.43 \times 10^5 \pm 1.7 \times 10^4$	$2.6 \times 10^2 \pm 4.1 \times 10^1$	4.3 ± 3.5	-----
Nov. 1978	0.1793	$2.67 \times 10^5 \pm 2.0 \times 10^4$	$1.20 \times 10^4 \pm 3.5 \times 10^3$	$2.74 \times 10^5 \pm 2.0 \times 10^4$	-----	-1.57 ± 2.4	-----
Dec. 1978	0.2558	$3.00 \times 10^5 \pm 1.9 \times 10^4$	$1.45 \times 10^4 \pm 4.9 \times 10^3$	$3.20 \times 10^5 \pm 1.9 \times 10^4$	-----	3354 ± 7.2	-----
Average		$2.79 \times 10^5 \pm 1.8 \times 10^4$	$1.33 \times 10^4 \pm 3.9 \times 10^3$	$2.71 \times 10^5 \pm 1.8 \times 10^4$	$7.12 \times 10^2 \pm 7.2 \times 10^1$	$4.72 \times 10^2 \pm 2.4$	$2.99 \times 10^1 \pm 2.1 \times 10^1$

Table 2. Isotopic Distribution of Natural Uranium

<i>Isotope</i>	<i>Natural Abundance, wt. %</i>	<i>Specific Activity of Isotope, dpm/g</i>	<i>Specific Activity of Natural Uranium</i>	
			<i>dpm/g</i>	<i>pCi/g</i>
U-234	5.4×10^{-3}	1.37×10^{10}	7.35×10^5	3.31×10^5
U-235	0.7196	4.75×10^6	3.45×10^4	1.55×10^4
U-238	99.2741	7.43×10^5	7.35×10^5	3.31×10^5

The average ratio of the specific activity of U-234 to U-238 in the Anaconda yellowcake is given in Table 3. Since fractionation of U-234 from U-238 during uranium extraction is not anticipated, the observed equality of the U-234 and U-238 activities in the yellowcake suggests that the uranium isotopes were in radioactive secular equilibrium in the ore.

Table 3. Relative Concentration of Radionuclides in Yellowcake from Various Mills as Percentage of U-238 Activity $\pm$ Standard Error (%) of the Mean

<i>Isotope</i>	<i>Anaconda</i>	<i>Exxon</i>	<i>Kerr-McGee</i>	<i>Uravan</i>	<i>Average</i>
U-235	4.8 ± 0.2	4.6 ± 0.1	5.2 ± 0.1	4.5 ± 0.1	4.8 ± 0.1
U-234	97.1 ± 0.3	93.2 ± 0.1	106.5 ± 0.1	93.2 ± 0.1	97.5 ± 0.1
Th-230	0.26 ± 0.2	No data	0.08 ± 0.1	0.46 ± 0.1	0.27 ± 0.1
Ra-226	0.17 ± 0.2	0.004 ± 0.1	0.00*	0.01 ± 0.1	0.05 ± 0.1
Pb-210	0.01 ± 0.2	No data	No data	0.03 ± 0.1	0.02 ± 0.2

* A less-than-detectable level was set at zero for determination of the average relative concentration.

Radioisotopic composition of the yellowcake composites from Exxon Mill (Wyoming), Kerr-McGee (New Mexico) and Uravan (Colorado) is given in Table 4. The average activities are 2.94×10^5 pCi U-238/g (Exxon), 2.60×10^5 pCi U-238/g (Kerr-McGee) and 2.92×10^5 pCi U-238/g (Uravan). These specific activities correspond to an equivalent U_3O_8 content of 105%, 92.6%, and 104%, respectively. The U_3O_8 equivalent content of yellowcake samples from Exxon and Uravan are higher than 100%, indicating some conversion of U_3O_8 to UO_2 because of higher drying temperatures. The average ratio U-234/U-238 of activities (Table 3) for these mills ranged from 93.2% to 106.5%, also suggesting that secular radioactive equilibrium exists between U-234 and U-238 in the ores. The yellowcake from the Highland Mill seemed to be mostly UO_2 compound, judging by its dark gray color.

The ratio of U-235 to U-238 activity in natural uranium (Table 2) is 4.7%. From our studies, the average ratio of U-235 to U-238 activity in composites from the mills (Table 3) ranged from 4.5% to 5.2%. This difference between the measured values and the expected value (4.7%) is within the estimated experimental error.

The Th-230 specific activity in the yellowcake from the Anaconda Mill is given in Table 1. The values range between a low of 197 and a high of 3150 pCi Th-230/g with an average activity of 712 pCi Th-230/g. The data from Kerr-McGee and Uravan are listed in Table 4. The ratio of Th-230 to U-238 activities for all of the mills is given in Table 3. The average for Anaconda is 0.26%, for Kerr-McGee 0.08%, and for Uravan 0.46%. These average values of Th-230 content relative to U-238 are much smaller than the value of 5% utilized in previous studies of model uranium mills (Sears, 1975; NRC, 1979).

Radium-226 and lead-210 specific activities in the yellowcake are given in Tables 1 and 4, and the ratios of activities of these radionuclides to U-238 are given in Table 3. The averages for Ra-226 are 0.19% (Anaconda), 0.004% (Highland), and 0.01% (Uravan). These ratios for Ra-226 are consistent with those previously reported (Sears, 1975; Merritt, 1971). For Pb-210 the average ratios of its specific activity to the U-238 activity are 0.01% and 0.03%. Recent data from Fort (1979) indicate average specific activity ratios in yellowcake from Uravan of 0.61% Th-230, 0.015% Ra-226, 0.017% Pb-210 (all

Table 4. Radioactive Composition of Yellowcake Measured in Composites
Obtained from Uravan, Kerr-McGee and Highland Mills

Composite Sample	Mill	Grams Analyzed	Concentration (pCi/g dry wt) ± Standard Error of the Mean						
			U-238	U-235	U-234	Th-230	Ra-226	Pb-210	
June 1978	Exxon	0.5191	2.95 × 10 ⁵ ±1.4×10 ⁴	1.32 × 10 ⁴ ±2.7×10 ³	2.52 × 10 ⁵ ±1.3×10 ⁴	-----	4.7 ±0.9	-----	
July 1978	Exxon	0.3876	2.60 × 10 ⁵ ±1.4×10 ⁴	1.12 × 10 ⁴ ±2.9×10 ³	2.58 × 10 ⁵ ±1.4×10 ⁴	-----	24.1 ±1.0	-----	
Aug. 1978	Exxon	0.1819	3.27 × 10 ⁵ ±2.1×10 ⁴	1.62 × 10 ⁴ ±4.2×10 ³	3.12 × 10 ⁵ ±2.0×10 ⁴	-----	9.7 ±1.9	-----	
Average			2.94 × 10 ⁵ ±1.7×10 ⁴	1.35 × 10 ⁴ ±3.3×10 ³	2.74 × 10 ⁵ ±1.6×10 ⁴		12.8 ±1.3		
Apr. 1977	Kerr-McGee	0.3259	2.72 × 10 ⁵ ±1.8×10 ⁴	1.14 × 10 ⁴ ±3.3×10 ³	2.49 × 10 ⁵ ±1.7×10 ⁴	280.4 ±36.3	3.6 ±1.0	-----	
Apr. 1977	Kerr-McGee	0.2805	2.47 × 10 ⁵ ±1.9×10 ⁴	1.57 × 10 ⁴ ±5.1×10 ³	3.04 × 10 ⁵ ±2.1×10 ⁴	124.4 ±34.1	-8.8 ±1.0	-----	
Average			2.60 × 10 ⁵ ±1.8×10 ⁴	1.36 × 10 ⁴ ±4.3×10 ³	2.77 × 10 ⁵ ±1.9×10 ⁴	2.02 × 10 ² ±35.2	-2.6 ±1.0		
Nov. 1977	Uravan	0.2405	3.00 × 10 ⁵ ±1.9×10 ⁴	1.50 × 10 ⁴ ±4.3×10 ³	2.74 × 10 ⁵ ±1.9×10 ⁴	1004.1 ±104.5	63.8 ±4.2	88.0 ±21.2	
Dec. 1977	Uravan	0.324	2.84 × 10 ⁵ ±1.4×10 ⁴	1.11 × 10 ⁴ ±2.4×10 ³	2.70 × 10 ⁵ ±1.3×10 ⁴	1693.0 ±158.1	3.9 ±3.6	-----	
Average			2.92 × 10 ⁵ ±1.7×10 ⁴	1.31 × 10 ⁴ ±3.5×10 ³	2.72 × 10 ⁵ ±1.6×10 ⁴	1.35 × 10 ³ ±134.0	3.39 × 10 ¹ ±3.9	8.80 × 10 ¹ ±15.0	

relative to U-238) and from Kerr-McGee of 0.1% Th-230, 0.004% Ra-226, 0.04% Pb-210. Data from Frigerio (1977) and Sill (1977) also suggest similar specific activities of Th-230, Ra-226 and Pb-210 in yellowcake.

Source Terms for Combined Emissions of Drying and Packaging of Yellowcake

Uranium emissions from yellowcake drying and packaging stacks were measured by Fort (1979) using an isokinetic technique. These data suggest an average release rate of about 350 g/hr (Kerr-McGee) from drying, 100 g/hr (Uravan) from drying and packaging, and 35 g/hr from packaging (Anaconda). Since these measurements were made Anaconda and Kerr-McGee have installed new air-cleaning systems and presumably the emission rates have been reduced. During these measurements the mills processed daily about 4500 tonnes of ore (Kerr-McGee), 1500 tonnes (Uravan), 3000 tonnes (Anaconda). Anaconda Mill throughput was subsequently increased to about 6500 tonnes per day. Thus, the rates of release of yellowcake from these stacks, assuming continuous drying operation, are 1.9 g/tonne (Kerr-McGee) and 1.6 g/tonne (Uravan) of ore processed. The assumption of continuous operation of dryers is conservative since the dryers are often operated less than 16 hours per day. This shorter drying period results in a lower rate of release of yellowcake--about 1.1 g/tonne of the ore processed. The annual release rates, assuming that these mills operate continuously (365 days/yr), is about 3 tonnes (Kerr-McGee) and 0.9 tonne (Uravan). The only value available from the Anaconda mill is 0.26 g/tonne released from packaging alone. Estimated yellowcake dryer emission at Anaconda, based on an average drying of about 362 kg/hr and a 10% dust loading of the collection system, is reported (Anaconda, 1976) to be about 20 g/hr based on stack sampling for one year period (1974 to 1975). Assuming a continuous operation, this release rate corresponds to 0.16 g/tonne of ore processed from the dryer stack. The procedure for stack sampling was not isokinetic and sampling procedure errors were not reported.

This value, in combination with the 35 g/hr release rate from the packaging stack (0.28 g/tonne) suggested by Fort (1979), results in a total release rate of 0.44 g/tonne, much less than the 1.6-g/tonne yellowcake emission estimated for Uravan and 1.9 g/tonne for Kerr-McGee.

Since the yellowcake yield from each tonne of ore processed is directly related to the ore quality, normalization of the stack release rate to the ore throughput (rate of the yellowcake release/tonne of ore processed) requires adjustment to the ore quality. The annual yellowcake production rates of these mills are about 3000 tonnes (Kerr-McGee), 1000 tonnes (Uravan), and 1940 tonnes (Anaconda), assuming an average ore quality of 0.2% (2 kg yellowcake/tonne of ore) and an extraction efficiency of 90%. Thus, the rates of release of yellowcake normalized to the rates of the production of the yellowcake are 0.001 and 0.0009. The estimated releases amount to about 0.1% of the uranium produced by continuous operation and about 0.07% for a 16-hour daily operation.

The conservative annual release from the drying and packaging stacks of the Kerr-McGee mill (Tables 3 and 4) corresponds to about 0.85 Ci U-238, 0.89 Ci U-234, 4.0×10^{-2} Ci U-235, and 6.7×10^{-4} Ci Th-230. Similarly, annual release rates from Uravan are 0.25 Ci U-238, 0.23 Ci U-234, 1.2×10^{-2} Ci U-235, 1.2×10^{-3} Ci Th-230, 2.5×10^{-5} Ci Ra-226, and 7.5×10^{-5} Ci Pb-210. Assuming that Anaconda released yellowcake at a rate comparable to the other two mills, say 2 tonnes/year, the annual discharges would be 0.55 Ci each U-238 and U-234, 2.6×10^{-2} Ci U-235, 1.5×10^{-3} Th-230, 9.8×10^{-4} Ra-226, and 5.9×10^{-5} Ci Pb-210. A reasonable estimate of release rates calculated for operation 16 hr/day, 357 days/yr would be about 65% of the above values.

The model mill utilized for the GEIS (NRC, 1979, p. G-5) processed 1800 tonnes/day of an ore containing 1.6 g U₃O₈/kg (0.16% quality). It would produce 1051 tonnes of yellowcake and release 0.9 tonne of yellowcake annually, assuming the product to be 100% U₃O₈. For this model mill, a conservative estimate of the U-238 release rate would be:

$$0.9 \text{ (tonne yellowcake/yr)} \times 10^6 \text{ (g/tonne)} \times 0.85 \frac{\text{g U-nat}}{\text{g U}_3\text{O}_8} \times$$

$$3.3 \times 10^{-7} \frac{\text{Ci U-238}}{\text{g U-nat}} = 0.25 \text{ Ci/yr of U-238.}$$

An equal activity of U-234 also would be released, assuming secular radioactive equilibrium between U-238 and U-234, as would 1.2×10^{-2} Ci U-235.

The annual Th-230 emission based on the average composition of yellowcake for the mills listed in Table 3 would be:

$$2.5 \times 10^{-1} \text{ Ci/yr U-238} \times 2.7 \times 10^{-3} \text{ Th-230/U-238} = 6.8 \times 10^{-4} \text{ Ci/yr,}$$

while the annual release of Ra-226 and Pb-210 would be:

$$2.5 \times 10^{-1} \text{ Ci/yr U-238} \times 7.0 \times 10^{-4} \text{ Ra-226/U-238} = 1.8 \times 10^{-4} \text{ Ci/yr}$$

$$2.5 \times 10^{-1} \text{ Ci/yr U-238} \times 2.0 \times 10^{-4} \text{ Pb-210/U-238} = 5.0 \times 10^{-5} \text{ Ci/yr.}$$

In conclusion the release rates utilized in our previous calculations (NRC, 1979, p. G-5) for the GEIS model mill are 7.2×10^{-2} Ci/year of U-234 and U-238, 3.6×10^{-3} Ci Th-230/year, and 1.4×10^{-4} Ci/year Ra-226 and Pb-210. Comparison of these data, estimated conservatively on the basis of present field studies, suggests a higher release rate of uranium (3.5 times) and radium (1.3 times), but a reduction of thorium (5.3 times) and lead (2.8 times). Based on 357 days of milling and 16 hours of daily drying operation these values should be reduced to 65% to reflect realistic operating conditions of the mills. Our previous estimates utilized for GEIS are reasonable except for thorium, which should be reduced by a factor of 8.

Perspective

Establishment of a source term which can be used for radiological assessment as a result of the combined releases from drying and packaging operations requires knowledge of the temporal variation of the release rates and characterization of the effluents by size, density and physiological solubility.

Temporal variations of the stack release rates may be caused by changes in the efficiency of dust-collection system with age, in addition to the other parameters of the overall milling operations. Presently, the EPA method for isokinetic stack sampling provides little information concerning the variation of the release rate with time. The use of an integrating air sampler, as developed by our group, would provide a longer period of effluent collection resulting in a better estimate of the average release rate. The ANL prototype

stack sampler is designed for sampling stack effluents at a continuous and constant rate at multiple sites at two perpendicular axes of the stack effluent stream. The integrating stack sampler operated over a period of a week would collect sufficient effluent sample mass for subsequent measurements of radionuclide concentrations.

Dispersion of particulates in air and their ultimate deposition in the respiratory passages of individuals are functions of the size distribution of the particles in the effluents released from a plant. The transportability of particulates within the respiratory system is also a function of particle size. Physiological solubility of the nuclides of concern depends on particle size as well as the chemical nature of the particles. Because size distribution influences deposition, transportability and solubility in the respiratory tract, it follows that size distribution also has a bearing on the relocation of the nuclides in bone, lymphatic tissue, or gastrointestinal tract.

Knowledge of the transportability and physiological solubility of the compounds that make up yellowcake, based on size distribution, is not established. Indeed, "yellowcake" itself is not a well-defined substance, being a mixture of uranium compounds in various states of oxidation, including diuranates, peroxide hydrates, and basic uranyl sulfates. At present, values for the solubilities of uranium compounds--parameters that are needed to assess radiological impacts--are chosen on the basis of ICRP (1966) recommendations. However, the ICRP solubility classifications made on the basis of the particle-size variations or compound mixes that occur in yellowcake are not yet firmly established. Similarly, the values obtained from Kalkwarf's (1978) recent studies are based on passive diffusion and dissolution; that is, the results are based on the measurement of only one aspect of transportability and physiological solubility.

In order to make a reasonable estimate of the potential exposure resulting from stack releases, one needs the following site-specific data:

- a. knowledge of the temporal variation and average release rates,
- b. size distribution and density of the particulates in the effluents,
- c. the chemical characterization of the particulates, and

d. relative importance of thermal treatment of yellowcake product.

The present solubility classifications of uranium compounds are not deemed satisfactory for yellowcake inhalation studies, but until more precise values are available, they must be used.

The matter of assessing the radiological hazards associated with uranium processing plants is currently of considerable importance. The work herein reported, while adding to the very limited information in this field, leaves ample scope for further investigation.

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APPENDIX. DETERMINATION OF DETECTION LIMITS

The minimum detection limit can be estimated from the means and standard errors of blanks and sample using the t distribution. In general, confidence limits for the blank and the sample means for a confidence level α can be estimated from:

$$\bar{x}_b \pm t_{\alpha, \phi} S_{\bar{x}_b},$$

where $t_{\alpha, \phi}$ is the Student-t distribution coefficient at the chosen confidence level α and a degree of freedom ϕ . $S_{\bar{x}_b}$ is the standard error of the mean ($\bar{x}$) and is given by:

$$S_{\bar{x}_b} = \frac{s_b}{\sqrt{n_b}},$$

where n_b is the number of blanks analyzed and S_b is the standard deviation of the blanks. Similarly, for the sample:

$$\bar{x}_s \pm t_{\alpha, \phi} S_b / \sqrt{n_s}.$$

By definition the minimum detection limit is the quantity which produces a statistically significant result different from zero after correction for the blank. Thus, the difference of the means and its confidence limit from the root-mean-square error is:

$$\Delta \bar{x} = \bar{x}_s - \bar{x}_b \pm t_{\alpha, \phi} \sqrt{\frac{s_s^2}{n_s} + \frac{s_b^2}{n_b}}.$$

At the limit of detection, $\Delta \bar{x}_{lim}$ must exceed the confidence limit:

$$\Delta \bar{x}_{lim} > t_{\alpha, \phi} \sqrt{\frac{s_s^2}{n_s} + \frac{s_b^2}{n_b}}.$$

At this limit $s_s^2 = s_b^2$ (Peters et al., 1974).

Then, the minimum detectable limit for each isotope can be determined from the following expression (Peters et al., 1974):

$$\Delta \bar{x}_{\text{lim}} = t_{\alpha, \phi} S_b \sqrt{\frac{1}{n_s} + \frac{1}{n_b}}.$$

The average activity of the blanks, the standard deviation, and the number of measurements made for each radionuclide in the blanks are given in Table A.1. The background counting is a measure of both instrumental background and the activity of reagents utilized in chemical separation of the radionuclides. This background for a given counting period dictates the detection limit for the measurement of activity of a radionuclide and may not be the same for different batches of reagents. Since different batches of reagents were utilized during the chemical separations and the samples were counted for durations less than 24 hours, the detection limits reported here are only a suggestion of potential capability to measure a low level activity.

Table A.1. Background Activity of the Blanks, Including
Radioactive Contents of the Reagents
Used for Chemical Extraction

<i>Radionuclide</i>	<i>Average Activity, pCi</i>	<i>Standard Deviation, pCi</i>	<i>Number of Measurements</i>
U-238	0.039	0.012	4
U-234	0.08	0.025	3
U-235	0.04	0.03	3
Th-230	0.16	0.05	4
Ra-226	0.17	0.02	3
Pb-210	101.7	2.1	3

The detection limits were calculated by using factors that include efficiency of counting (ϵ), counting period Δt (24 hours), extraction yields (η), aliquot fraction taken for analysis (μ), weight of each sample (ω), and the radioactivity of the reagents. The counting error of measurements (Tables 1 and 4) are influenced by factors Δt , η , μ , and ω . For example, the counting

error for determination of U-238 in Uravan yellowcake sample (November 1977, Table 4) was calculated from the following factors: $\epsilon = 0.165$, $\eta = 0.658$, $\omega = 0.2405$ gram dissolved in 250 ml solution and an aliquot of $\mu = 50$ λ (i.e., 50×10^{-3} ml was used in electrodeposition and $\Delta t = 4000$ second. This counting period resulted in $N = 233$ alpha counts from U-238 activity. Thus the counting error for this sample is estimated from:

$$\sigma = \frac{\sqrt{N}}{2.22 \Delta t \cdot \epsilon \cdot \eta \cdot \omega \cdot \mu} ,$$

where 2.22 is the dpm-to-pCi conversion factor.

From the background measurements (radioactivity content of reagents, i.e., blanks) for the U-238, U-235, and U-234, the detection limits were calculated at the 90% confidence limit.

The U-238 detection limit for the 90% confidence level is given by:

$$\Delta \bar{x}_{lim} \geq (2.353) (0.012 \text{ pCi}) \sqrt{\frac{1}{15} + \frac{1}{4}} ,$$

$$\Delta \bar{x}_{lim} \geq 0.02 \text{ pCi for U-238} .$$

Similarly, for other isotopes of uranium we obtain:

$$\Delta \bar{x}_{lim} \geq 0.06 \text{ pCi for U-235} ,$$

$$\Delta \bar{x}_{lim} \geq 0.04 \text{ pCi for U-234} .$$

The minimum detectable limit for Th-230, determined in a similar manner, for the 90% confidence level, is:

$$\Delta \bar{x}_{lim} \geq (2.353) (0.05 \text{ pCi}) \sqrt{\frac{1}{13} + \frac{1}{4}} ,$$

$$\Delta \bar{x}_{lim} \geq 0.07 \text{ pCi for Th-230} .$$

A total of 20 yellowcake samples were analyzed for Pb-210; at 90% confidence level the minimum detection limit is:

$$\Delta \bar{x}_{lim} \geq (2.920) (2.1 \text{ pCi}) \sqrt{\frac{1}{20} + \frac{1}{3}},$$

$$\Delta \bar{x}_{lim} \geq 3.8 \text{ pCi for Pb-210}.$$

And for 20 samples analyzed for Ra-226, the detection limit for the 90% confidence limit is:

$$\Delta \bar{x}_{lim} \geq (2.920) (0.02 \text{ pCi}) \sqrt{\frac{1}{20} + \frac{1}{3}},$$

$$\Delta \bar{x}_{lim} \geq 0.04 \text{ pCi for Ra-226}.$$

The limits thus obtained are summarized in Table A.2.

Table A.2. Summary of Detection Limits

<i>Isotope</i>	<i>Detection Limit (90% Confidence Level)</i>
U-238	> 0.02 pCi (2.0 pCi/g)
U-235	> 0.06 pCi (6.0 pCi/g)
U-234	> 0.04 pCi (4.0 pCi/g)
Th-230	> 0.07 pCi (7.0 pCi/g)
Ra-226	> 0.04 pCi (4.0 pCi/g)
Pb-210	> 3.8 pCi (38 pCi/g)

It should be noted that a great deal of scattering of data occurs near or below these detection limits. This is particularly evident in the measurement Ra-226 and Pb-210 (Tables 1 and 2) where negative values (background counts larger than the gross counts) were obtained in some cases. These two isotopes were especially difficult to measure because of the high background of the system and the blanks and the low concentration of each isotope in the yellowcake samples. In the case of Pb-210, the difficulty was compounded by the high background (40 cpm or greater) characteristic of the proportional counters

used to measure the beta activity. In our opinion, the technique we have utilized for the measurement of Pb-210 concentration is inadequate for estimating low-level activities. Measurement of Pb-210 by means of other techniques will be reported elsewhere.

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