Technical Note
ORP/LV-80-3

# RADIOACTIVE EMISSIONS FROM YELLOWCAKE PROCESSING STACKS AT URANIUM MILLS

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# PREFACE

This study was conducted by the Office of Radiation Programs-Las Vegas Facility of the U.S. Environmental Protection Agency. The Office of Radiation Programs carries out a national program designed to evaluate population exposures to radiation, and to promote the development of controls necessary to protect the public health and safety. The purpose of this study was to evaluate the emissions of uranium concentrate (yellowcake) from processing stacks at uranium mills.

We would appreciate receiving any comments on this report that readers may have to offer.

Donald W. Hendricks

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# INTRODUCTION

The recent expansion of the uranium milling industry, coupled with increasing awareness of the potential health effects of radioactivity releases from uranium milling sites, has created increased concern about the current and future radiological environmental impact of the industry. The regulations and licensing requirements for the industry, issued by the Federal and State agencies, reflect this concern. For example, the U.S. Environmental Protection Agency's (EPA) uranium fuel cycle standard, 40 CFR 190, limits the annual radiation exposure to any member of the public, from a uranium fuel cycle facility, to 25 mrem. This standard becomes effective for uranium mills in December 1980. It does not include exposure from (1) radon and its progeny, including lead-210 and polonium 210, (2) radioactive contamination from milling operations prior to December 1980, or (3) radioactivity from uranium mining operations.

Several reports have described the sources of radioactive material releases in and around a uranium mill, the quantity of material released per unit time (the source term), the pathways of the released material to people living near the mill, and the dose received by these people. (See, among others, Glauberman and Breslin [1964], Sears et al. [1975], EPA [1976], Harward [1977], Sill [1977].) The source terms and pathways are qualitatively well defined by these reports. They, as well as numerous environmental reports and environmental impact statements prepared as mill licensing documents, also assign quantitative values to the source terms and pathway transfer parameters, and estimate radiation doses to individuals and population groups. However, these quantitative values are necessarily often based on estimates and assumptions, because few actual measurements of the source terms and transfer parameters have been made and published in the scientific literature.

Considering the shortage of documented technical data about yellowcake* emissions from uranium mills, and the increasing need for such data, the EPA's Office of Radiation Programs-Las Vegas Facility (ORP-LVF) has undertaken studies to provide such data. Excepting tailings piles, the source with the highest potential for human exposure appears to be particulate emissions from the yellowcake drying and packaging stacks. (The dose from radon and its progeny are not included in the 40 CFR 190 standard.) Although emissions from these stacks are small in terms of mass release rate, the concentrated nature of the yellowcake product (usually reported to be about 85-90% $U_3O_8$) provides a relatively high radionuclide release rate. Consequently, EPA's studies gave the yellowcake stacks high priority.

Planning for these studies began in late 1976. We then learned that Argonne National Laboratory (ANL) was beginning similar studies for the Nuclear Regulatory Commission. After discussion, the two groups joined forces to reduce duplication of effort. As study plans evolved, ANL assumed the lead role in defining radon source terms, while ORP-LVF concentrated on measuring releases from the yellowcake stacks and on measurements of ambient levels of radon and airborne particulates. In early 1977, we approached several uranium milling companies to establish a cooperative basis for studies to be conducted at their mills. Several companies agreed to cooperate in these studies.

This report describes the results obtained from a series of tests on yellowcake drying and packaging stacks at six mills. The tests were conducted from April 1977 to August 1978.

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* Yellowcake is the generic name given the packaged uranium concentrate produced by uranium mills. The chemical form of the initial uranium concentrate at all mills prior to drying or calcining is ammonium (or sodium) diuranate which has a bright yellow color-hence the name yellowcake. However, the uranium concentrate as packaged at the various mills will have a variety of chemical forms including ammonium (or sodium) diuranate, $U_3O_8$, and $UO_2$, depending on each mill's process and drying or calcining temperature. The color of these forms will vary from bright yellow through olive green to black. The uranium content of yellowcake is typically expressed as an equivalent mass of $U_3O_8$ regardless of the actual chemical form. The chemical forms of the yellowcake reported on are unknown and it is important for the reader to realize that these materials may not be $U_3O_8$ or exhibit the solubility characteristics of $U_3O_8$.

The six mills that participated in this study are:

| | |
|---|---|
| Anaconda | Bluewater, New Mexico |
| Kerr-McGee | Ambrosia Lake, New Mexico |
| Sohio | Seboyeta, New Mexico |
| United Nuclear-Churchrock | Gallup, New Mexico |
| United Nuclear-Homestake Partners | Milan, New Mexico |
| Union Carbide | Uravan, Colorado |

The ORP-LVF staff and the responsible staff at each mill agreed that the tests would be conducted with full company approval and cooperation and would not interfere with mill operations.

## SAMPLING EQUIPMENT AND METHODS

Stack sampling for particulates consists of extracting from within the stack samples of stack gas and collecting any entrained particulates on a filter. The air velocity through the opening of the sample nozzle is equal to the stack gas velocity at the sample point. Sampling this way is isokinetic and ensures that all particles approaching the plane of the sample nozzle opening will be collected without bias due to size.

### Stack Sampling Equipment

The stack testing equipment used for these tests was manufactured by Research Appliance Company. Essentially, the equipment is a meter console and a sample case connected by sampling hose, power cables and pitot lines. The meter console monitors stack gas conditions (temperature and velocity pressure) and allows the operator to maintain an isokinetic sampling rate by adjusting the sampling velocity through the sampling nozzle. The sample case holds the sampling probe for in situ extraction of the stack gas sample, and also contains a filter in a heated compartment with temperature control and a series of four glass impingers in an ice bath compartment for moisture condensation and removal.

The meter console consists of an oil-type fiber vane positive displacement pump; a 0.1-cubic foot per revolution totalizing dry gas meter; an orifice meter for sampling rate determinations; a dual 0-10" $H_2O$ incline/vertical manometer (incline portion = 0 to 1" $H_2O$ with 0.01" $H_2O$ minor divisions) to measure the orifice meter differential pressure ($\Delta H$) and stack gas differential velocity pressure ($\Delta P$); a vacuum gauge to indicate filter loading as testing progresses, or to indicate the occurrence of a major leak in the sampling train; coarse and fine adjustment valves to set isokinetic flow rates; and a rheostat for adjusting the temperature of the sampling probe. An optional feature of this unit is a six-button digital temperature indicator using special grade (±2°F) "J" thermocouples for temperature measurements of

the stack gas, probe liner, filter compartment, silica gel inpinger outlet, and inlet and outlet of the dry gas meter.

The sampling probe consists of a three foot stainless steel liner wrapped with heat tape protected by a stainless steel sheath. A gooseneck nozzle is attached to the inlet end of the probe. A thermocouple to measure probe temperature is located underneath the heat tape near the exit of the probe. The heat tape lead and thermocouple lead are terminated with connectors just outside the sheath. A Type S pitot for $\Delta P$ measurements and a thermocouple for stack gas temperature measurements are mounted on the sheath surface and extended to a point in-line with the gooseneck sample nozzle. The pitot and thermocouple are positioned to conform with the specifications of EPA Method 2.

A Hewlett-Packard programmable calculator (HP-65 or HP-67) was used for all tests instead of the standard stack testing nomograph. The calculator offers small size, accuracy, speed, and each of the ten variables in the isokinetic equation are used in the isokinetic sampling rate calculations. This is unlike the nomograph with its several potentially faulty assumptions. Ultimately, HP-67 programs were used in all phases of a stack test, and reference materials were unnecessary. These programs included pre-test stack gas moisture fraction estimation, sampling nozzle size selection, number of sampling points, location of sampling points for a circular stack, stack gas moisture fraction by condensation, dew point temperature and the post-test determination of the isokineticity of the test. An EPA publication (Ragland, 1976) is an excellent source of calculator programs for use in air pollution measurements at stationary industrial sources.

The pre-test estimation of stack gas moisture fractions calculated using the HP-67 program proved extremely reliable, providing care was taken in determining stack gas wet and dry bulb temperatures and stack gas dynamic pressure. The program was so developed that reference charts or tables were unnecessary. Appendix A describes this program.

Stack Sampling Methods

All yellowcake stack tests employed the EPA Reference Methods 1 through 5 as defined in "Standards of Performance for New Stationary Sources" (CFR, 1977). Reference Methods 1 through 4, respectively, define the techniques for (1) locating and defining the number of sample or velocity traverse points, (2) determining stack gas velocity and volumetric flow rate, (3) analyzing stack gas carbon dioxide and oxygen concentrations and determining the dry molecular weight of the stack gas, and (4) determining the stack gas moisture content. Reference Method 5 combines these techniques in determining the particulate emissions from a stack.

Since yellowcake stacks are not subject to the EPA standards of performance, these methods are not officially prescribed for these stacks. However, these methods (with slight modifications to Method 5 reflecting the unique material being sampled) are the best available. Since tests at all six mills were conducted on a non-interfering and non-interrupting basis, they were performed under existing operating conditions without consideration for a stable or optimum operating level as would be required for compliance testing. Therefore, parameters affecting emission rates were not controlled except that tests were usually conducted when yellowcake was being actively dried or packaged. Once a test was started, it was completed even though drying or packaging may have ceased or a scrubber malfunction may have occurred. Attempts were made to determine amounts of yellowcake dried or packaged during tests.

The modifications of Method 5 referred to above consisted of (1) using distilled water or 0.1N $HNO_3$ as the probe and glassware rinse solution instead of acetone, (2) using a pressurized spray nozzle to clean the sampling probe instead of a probe brush, (3) reducing the filter and probe heat from 248 $\pm$ 25°F to 20-30°F higher than stack gas dew point when the higher temperature could not be achieved, (4) deleting the filter weighing requirements, and (5) removing the sampling holder filter gasket. These changes are minor but resulted in increased accuracy and ease of operation noted below.

An early test used acetone, as prescribed by Method 5, as a wash solution.

However, the acetone tended to leave a yellowcake film that was not easily removed, and it was therefore not acceptable as a washing agent. Distilled water or 0.1N $HNO_3$ was adopted as the wash solution and worked very well. A probe brush was initially used to clean the probe. It smeared the yellowcake over the internal probe surface, and the particles adhering to the brush were not easily removed. To overcome this difficulty, a standard garden sprayer nozzle, adjusted to a conical spray, was fitted to a brass pipe and flexible hose attached to a plastic pressure tank containing water or 0.1N $HNO_3$. The tank was pressurized and the nozzle was inserted into the stain-less steel probe liner and run its length for three passes. A wash bottle was used to rinse the surface of the nozzle into the probe wash after the final pass. The pressurized spray removed all particles from the probe wall and washed them ahead of the nozzle into the collection jar. By advancing the spray nozzle slowly on the first pass through the probe, surface contamination of the nozzle was minimized.

The filter and probe temperature requirement of EPA Method 5 is directed at compliance testing to define the particulate or gaseous nature of the stack gas components. If, at this temperature, any of the stack effluent components exist as a gas, they obviously will not be collected by the filter and therefore, at least by EPA standards, are not considered particulates. Since the purpose of the tests reported herein was not to determine a mass emission rate, this temperature requirement was not considered binding. Initially polonium-210 was considered as existing as a gas and for several tests the Method 5 temperature was rigidly maintained as a reference temperature should there be volatilized polonium. After determining that this was not the case (polonium was not detected in impinger catches) the Method 5 temperature was used when achievable. Otherwise, a filter and probe temperature at least 20-30°F above the stack gas dew point temperature was maintained only to prevent moisture condensation upstream of the filter.

Deletion of the filter weighing requirement was an obvious and timesaving simplification of Method 5. Since these tests were designed to measure radioactivity emission rates instead of mass rates, there was no need for a gravimetric determination.

The filter holder was modified by removing the rubber gasket that normally fits between the filter and the glass inlet cap, and adding a second glass fiber filter. This eliminated cleaning the gasket after each test, and the tandem filters were less likely to tear at the edges when removed. Leaks were never detected using this technique and the change caused only a slight increase in flow resistance.

For these studies, the stack testing team consisted of two people a probe operator to position the probe at the traverse (or sampling) points within the stack as testing progressed, and a control meter operator to observe changes in stack gas velocity pressure and temperature, and to adjust and maintain isokinetic sampling rates as these parameters changed. The control meter was at ground level, connected to the sample case at the stack by 50-100 feet of cable and tubing. The probe operator at the stack and the control meter operator used portable radios to communicate.

Basically, the stack testing routine consisted of moving the sample nozzle from traverse point to traverse point at predetermined intervals (usually 5 minutes) until all points across the stack diameter had been sampled. After the first traverse, the sample case with probe was repositioned to a second port at 90° from the first and the sampling routine repeated. Velocity pressures and temperatures were recorded at each traverse point and, when different from the previous point, entered into the calculator to compute the adjustment needed at the control meter to maintain an isokinetic flow rate through the sample nozzle.

At the conclusion of a test, the sampling probe was dismantled from the filter holder, and the probe, filter holder, and intermediate glassware were taken to a clean area for washing. The filter was removed and placed into a covered petrie dish. All parts of the sampling train forward of the filter were then washed into a glass jar. The filter plus the wash solution constituted the sample. Water condensed in the impingers during the test was weighed to calculate the stack gas moisture fraction.

All tests were conducted for a minimum of one hour. Typically, at least 30 dry standard cubic feet of stack gas (at 68°F and 29.92 in. Hg) were

sampled during each test.

## Yellowcake Grab Samples

The project collected six yellowcake grab samples, not associated with stack tests from four mills. These samples were analyzed to measure the concentration of the uranium daughter radionuclides in yellowcake from different mills, and coincidentally to cross-check the analytical capabilities of different laboratories. Grab samples were used rather than stack test filters to provide a larger quantity of yellowcake for analysis, and for convenience in sending replicate aliquots to the different laboratories.

## Sample Analysis and Data Calculation

Several laboratories analyzed the stack test samples from these studies. Early in the study, the EPA laboratory in Las Vegas analyzed the samples using standard radiochemical procedures as described by Johns (1975). Later in the study, Eberline Instrument Corporation analyzed samples using similar procedures. EPA, Eberline, and LFE Corporation Environmental Analysis Laboratories analyzed the grab samples discussed above and functioned as laboratory cross-checks. These three laboratories analyzed the samples for isotopic uranium, isotopic thorium, radium-226, lead-210, and polonium-210. Uranium-West Laboratories analyzed one set of stack test samples for uranium, using a technique counting delayed neutrons from the fission of uranium-235 (Gale, 1967). The respective company laboratories also analyzed aliquots of some of the stack test samples, using fluorimetric or colorimetric procedures, as an analytical cross-check procedure between them and the EPA (or EPA contractor) laboratories.

The stack test samples consisted of the 2.5-inch glass fiber filter and one to three liters of the probe wash solution, which was either distilled water or dilute nitric acid. They were prepared for analysis by evaporating the probe wash solution to near dryness and combining the remaining solution with the filter. This combined sample was treated with nitric and hydrofluoric acids to dissolve the glass fiber filter for subsequent radiochemical analysis. If inter-laboratory cross-check analyses were to be made on the

sample, aliquots of this solution were taken.

The analytical laboratory reported the stack test sample results as total picocuries of each nuclide in the sample. The radiochemical analysis measures the three uranium nuclides (U-238, U-234, and U-235) separately by alpha spectroscopy. These uranium nuclide results were converted to total uranium ($U_{tot}$) activity by summing the activities of the three nuclides. This total uranium activity was converted to total uranium mass using the conversion factor of 0.677 uCi $U_{tot}$/gram $U_{tot}$ (CFR, 1975). The uranium mass was converted to $U_3O_8$ using the conversion factor of 0.848 gm $U_{tot}$/gm $U_3O_8$.

## DESCRIPTION OF MILLS AND YELLOWCAKE PROCESSING FACILITIES

Five of the mills involved in this study employ the acid leach process, while the United Nuclear-Homestake Partners (UNHP) uses an alkaline leach process. A general description of each mill and the yellowcake exhaust air cleanup system(s) in use at the time these tests were made is given below.

Kerr-McGee: Nominal ore processing capacity 6500 tons per day (tpd). Three exhaust stacks vent the plant, each served by a Joy Microdyne wet scrubber. The three scrubbers are referred to as the Main, East, and West. The Main Microdyne scrubber and stack is the primary exhaust pathway from the dryer, and the East and West systems primarily serve the packaging area, although there is some cross connection.

Union Carbide-Uravan: Capacity 1300 tpd. The exhaust streams from both the dryer and packaging area are combined and discharged through a single rectangular stack. The exhaust air is cleaned by a three-tiered wet scrubber.

Anaconda: Capacity 2000 tpd at the time of the test, subsequently expanded to 6000 tpd. The exhaust stream from the packaging area is cleaned by a Joy Micordyne; the dryer exhaust by the "Anaconda scrubber," a wet impaction scrubber.

Sohio L-Bar Mill: Capacity 1500 tpd. The dryer exhaust is cleaned by a venturi scrubber; the packaging area exhaust by a Mikro-Pulsaire Dust Collector bag filter system.

United Nuclear Churchrock: Capacity 4000 tpd. Dryer exhaust cleaned by Impi-Jet scrubber, packaging area exhaust by Joy venturi wet scrubber.

United Nuclear-Homestake Partners: Capacity 2700 tpd. Vanadium roaster stack has a Hydrofilter followed by a Type W Rotoclone; the packaging stack has a Hydrostatic precipitator followed by a Type W Rotoclone; the dryer stack has a Joy-Denver scrubber followed by a Type W Rotoclone.

## SAMPLING AND ANALYTICAL RESULTS

Table 1 shows individual stack test results. Each sample is identified by mill, date of sampling, and stack sampled. The calculated uranium emission rate for each sample is shown, expressed as microcuries of each uranium nuclide released per hour (radioactivity emission rate), and as pounds of $U_3O_8$ released per hour (mass emission rate). The table also shows volumetric flow rate of the stack at the time of sampling.

Table 2 shows the emission rates for radionuclides other than uranium (i.e., thorium-230, radium-226, polonium-210, and lead-210) for the stack test samples collected at the United Nuclear-Homestake Partners and the United Nuclear Churchrock mills. Eberline analyzed these samples, the only stack test samples analyzed for the nuclides other than uranium. Table 3 shows the ratios of these nuclides to the U-238 parent for these samples.

Table 4 shows the inter-laboratory cross-check results for uranium on several stack test samples. Since the industry laboratories used a fluorimetric analysis (United Nuclear-Churchrock used a colormetric procedure) with results expressed as mass per unit volume, the EPA, Uranium-West, and Eberline radionuclide results have been converted to mass units for comparison. The table reports the results as milligrams of uranium per liter of solution and refers to the solution used to dissolve the stack test filter. Since the purpose is to compare the results obtained by different laboratories, the results have not been converted to total uranium per sample or to an activity or mass emission rate.

Table 5 shows the radionuclide concentrations in several grab samples of yellowcake from four mills. EPA, Eberline and LFE analyzed six of these samples. Due to a misunderstanding, the Eberline results were reported as picocuries per total sample, rather than as picocuries per gram of yellowcake. Consequently, the Eberline analytical results cannot be compared directly to the EPA or LFE results.

Each analysis shows the percentage of each of the nuclides of the U-238 parent. (Note the Eberline results for this value are comparable to EPA and LFE analyses.) The right hand column shows the average percentage from the three analyses for each nuclide.

TABLE 1. URANIUM EMISSION RATES FROM YELLOWCAKE PROCESSING STACKS

| Date | Stack | Volumetric Flow Rate $10^5$ DSCFH* | EMISSION RATE uCi/hr U-238 | U-235 | U-234 | Pounds of $U_3O_8$/hr |
|---|---|---|---|---|---|---|
| **Kerr-McGee (samples analyzed by EPA-Las Vegas)** | | | | | | |
| 4-15-77 | Main | 6.7 | 39.2 | 1.4 | 38.4 | 0.30 |
| 4-16-77 | East | 2.4 | 12.8 | 0.5 | 12.8 | 0.10 |
| 4-17-77 | East | 2.3 | 9.1 | 0.3 | 8.4 | 0.067 |
| 9-08-77 | West | 2.0 | 4.4 | 0.2 | 4.4 | 0.034 |
| 9-09-77(am) | West | 2.0 | 7.3 | 0.3 | 7.4 | 0.058 |
| 9-09-77(pm) | West | 2.4 | 30.7 | 1.2 | 29.7 | 0.24 |
| 9-12-77 | East | 2.4 | 6.4 | 0.2 | 6.4 | 0.050 |
| 9-13-77 | Main | 4.5 | 113 | 4.0 | 109 | 0.87 |
| **Union Carbide-Uravan (samples analyzed by EPA-Las Vegas)**** | | | | | | |
| 10-13-77 | Dryer-Package | 2.1 | 18 | 0.8 | 17.7 | 0.14 |
| 10-14-77 | Dryer-Package | 2.0 | 14.2 | 0.6 | 13.5 | 0.11 |
| 10-15-77 | Dryer-Package | 2.0 | 22.8 | 1.1 | 22.7 | 0.18 |
| 10-16-77 | Dryer-Package | 1.9 | 32.1 | 1.1 | 33.0 | 0.25 |
| 10-17-77 | Dryer-Package | 1.9 | 24.2 | 0.9 | 22.0 | 0.18 |
| 10-18-77 | Dryer-Package | 1.9 | 40.6 | 1.7 | 39.6 | 0.31 |
| 10-19-77 | Dryer-Package | 1.9 | 36.9 | 1.4 | 34.5 | 0.28 |
| **Anaconda (samples analyzed by EPA-Las Vegas)** | | | | | | |
| 4-18-77 | Package | 1.1 | 9.3 | 0.3 | 9.9 | 0.075 |
| **United Nuclear-Churchrock (samples analyzed by Eberline)**,*** | | | | | | |
| 6-16-78 | Dryer | 3.5 | 50.5 | 2.6 | 47.0 | 0.38 |
| 6-17-78(am) | Dryer | 3.6 | 29.2 | 1.6 | 27.7 | 0.22 |
| 6-17-78(pm) | Dryer | 3.5 | 61.9 | 3.7 | 59.4 | 0.48 |
| 6-18-78(am) | Package | 1.0 | 30.0 | 1.6 | 28.2 | 0.23 |
| 6-18-78(pm) | Package | 1.0 | 30.4 | 2.1 | 28.3 | 0.23 |
| 6-19-78 | Package | 1.0 | 15.4 | 0.7 | 15.4 | 0.12 |

TABLE 1 URANIUM EMISSION RATES FROM YELLOWCAKE PROCESSING STACKS (continued)

| Date | Stack | Volumetric Flow Rate $10^5$ DSCFH * | EMISSION RATE uCi/hr U-238 | U-235 | U-234 | Pounds of $U_3O_8$/hr |
|---|---|---|---|---|---|---|
| United Nuclear-Homestake Partners (samples analyzed by Eberline)** | | | | | | |
| 3-13-78 | Dryer | 0.4 | 5.2 | 0.2 | 5.2 | 0.041 |
| 3-14-78 | Package | 3.1 | 2.7 | 0.1 | 2.7 | 0.021 |
| 3-15-78 | Vanadium Roaster | 0.9 | 2.7 | 0.1 | 2.7 | 0.021 |
| (samples analyzed by Uranium-West Labs)**,*** | | | | | | |
| 8-15-78 | Vanadium Roaster | 1.0 | 4.6 | 0.2 | 4.6 | 0.036 |
| 8-16-78(am) | Vanadium Roaster | 1.0 | 4.2 | 0.2 | 4.2 | 0.033 |
| 8-16-78(pm) | Vanadium Roaster | 1.0 | 2.1 | 0.1 | 2.1 | 0.016 |
| 8-17-78(am) | Vanadium Roaster | 0.9 | 4.3 | 0.2 | 4.3 | 0.033 |
| 8-17-78(pm) | Dryer | 0.4 | 1.5 | 0.07 | 1.5 | 0.012 |
| 8-18-78(am) | Dryer | 0.4 | 3.8 | 0.2 | 3.8 | 0.030 |
| 8-18-78(pm) | Package | 2.8 | 110 | 5.1 | 110 | 0.87 |
| 8-19-78(am) | Package | 2.8 | 82.6 | 3.9 | 82.6 | 0.65 |
| 8-19-78(pm) | Package | 2.8 | 118 | 5.5 | 118 | 0.93 |
| Sohio (samples analyzed by Eberline)** | | | | | | |
| 5-17-78 | Package | 0.3 | 45.2 | 1.9 | 49.0 | 0.37 ++ |
| 5-18-78 | Package | 0.4 | 0.6 | 0.02 | 0.7 | 0.0052 |
| 5-21-78 | Dryer | 1.3 | 0.5 | 0.03 | 0.4 | 0.0039 |
| 5-22-78 | Dryer | 1.0 | 0.2 | 0.008 | 0.2 | 0.0013 |
| 5-23-78 | Dryer | 1.0 | 0.8 | 0.03 | 0.8 | 0.0061 |
| 5-24-78 | Package | 0.4 | 2.6 | 0.09 | 2.6 | 0.020 |

* DSCFH = Dry Standard Cubic Feet per Hour (at 68°F and 29.92 in. Hg).
** Cross-comparison analyses were also performed on aliquots of these samples. (Table 4)
*** See Table 2 for Ra-226, Th-230, Pb-210, and Po-210 emission rates.
++ One bag filter was ruptured during this test. This condition occurred the day of the test and was corrected that evening.

TABLE 2. RADIUM, THORIUM, LEAD, AND POLONIUM EMISSION RATES FROM YELLOWCAKE PROCESSING STACKS*

| Date | Stack | EMISSION RATE, nCi/hr** | | | |
|---|---|---|---|---|---|
| | | Ra-226 | Th-230 | Pb-210 | Po-210 |
| **United Nuclear - Homestake Partners** | | | | | |
| 3-13-78 | Dryer | 18.5 | 37.8 | 132 | NA*** |
| 3-14-78 | Package | 19.1 | 32.2 | <50**** | NA |
| 3-15-78 | Vanadium Roaster | 10.3 | 10.4 | 105 | NA |
| 8-15-78 | Vanadium Roaster | 77.4 | 56.6 | 176 | 328 |
| 8-16-78(am) | Vanadium Roaster | 15.9 | 52.0 | 133 | 780 |
| 8-16-78(pm) | Vanadium Roaster | 10.8 | 24.8 | 108 | 299 |
| 8-17-78 | Vanadium Roaster | 20.2 | 77.8 | 72.0 | 285 |
| 8-17-78 | Dryer | 11.9 | 151 | 57.3 | 77.8 |
| 8-18-78 | Dryer | 41.2 | 46.0 | 94.4 | 55.7 |
| 8-18-78 | Package | 52.3 | 3780 | 222 | 161 |
| 8-19-78(am) | Package | 54.4 | 2580 | 165 | 76.9 |
| 8-19-78(pm) | Package | 25.7 | 4050 | 274 | 93.0 |
| **United Nuclear - Churchrock** | | | | | |
| 6-16-78 | Dryer | 4.41 | 142 | 114 | NA |
| 6-17-78(am) | Dryer | <1 | 79.1 | 36.1 | NA |
| 6-17-78(pm) | Dryer | <1 | 185 | 29.4 | NA |
| 6-18-78(am) | Package | 1.41 | 102 | 13.6 | NA |
| 6-18-78(pm) | Package | 3.49 | 46.4 | 12.6 | NA |
| 6-19-78 | Package | 0.59 | 49.7 | < 5 | NA |

* All analyses were by Eberline

** nCi = $10^{-3}$ μCi

*** NA = no analysis

**** < indicates "less than" value at 95% confidence level

TABLE 3. RATIOS OF DAUGHTER RADIONUCLIDES TO U-238 PARENT FOR STACK TEST SAMPLES*

| | | Percent of U-238 Activity | | | |
|---|---|---|---|---|---|
| Stack | Date | Ra-226 | Th-230 | Pb-210 | Po-210 |
| United Nuclear-Homestake Partners | | | | | |
| Dryer | 3-13-78 | 0.354 | 0.723 | 2.53 | NA** |
| Package | 3-14-78 | 0.700 | 1.18 | < 2 *** | NA |
| Vanadium Roaster | 3-15-78 | 0.378 | 0.383 | 3.87 | NA |
| Vanadium Roaster | 8-15-78 | 1.86 | 1.36 | 4.21 | 7.86 |
| Vanadium Roaster | 8-16-78(am) | 0.458 | 1.50 | 3.83 | 22.5 |
| Vanadium Roaster | 8-16-78(pm) | 0.621 | 1.43 | 6.21 | 17.2 |
| Vanadium Roaster | 8-17-78 | 0.539 | 2.08 | 1.92 | 7.62 |
| Dryer | 8-17-78 | 0.162 | 2.06 | 0.779 | 1.06 |
| Dryer | 8-18-78 | 1.17 | 1.31 | 2.69 | 1.59 |
| Package | 8-18-78 | 0.055 | 4.00 | 0.235 | 0.171 |
| Package | 8-19-78(am) | 0.071 | 3.36 | 0.214 | 0.100 |
| Package | 8-19-78(pm) | 0.024 | 3.70 | 0.250 | 0.085 |
| United Nuclear-Churchrock | | | | | |
| Dryer | 6-16-78 | 0.0087 | 0.282 | 0.226 | NA |
| Dryer | 6-17-78(am) | < 0.004 | 0.271 | 0.123 | NA |
| Dryer | 6-17-78(pm) | < 0.002 | 0.290 | 0.046 | NA |
| Package | 6-18-78(am) | 0.005 | 0.341 | 0.045 | NA |
| Package | 6-18-78(pm) | 0.015 | 0.153 | 0.041 | NA |
| Package | 6-19-78 | 0.004 | 0.324 | < 0.035 | NA |

* All analyses were by Eberline

** NA = no analysis

*** < indicates "less than" value at 95% confidence level

TABLE 4. INTER-LABORATORY COMPARISON OF URANIUM ANALYTICAL RESULTS ON STACK TEST SAMPLES +

| UC - Uravan Samples | | Sohio Samples | | Kerr-McGee Samples | | UNC Samples | | UNHP Samples | |
|---|---|---|---|---|---|---|---|---|---|
| Analyses by: | | | | | | | | | |
| EPA | Union Carbide | Eberline | Sohio | EPA | Kerr-McGee | Eberline | UNC | Eberline | U-West |
| 157 ± 12 | 182 | 1478 ± 120 | 1591 | 38 ± 2 | 54* | 241 ± 10 | 261 ± 8* | 42 ± 2 | 47 ± 2 |
| 129 ± 9 | 144 | 28 ± 2 | 31 | 64 ± 3 | 68* | 140 ± 6 | 152 ± 5* | 36 ± 2 | 44 ± 2 |
| 205 ± 15 | 225 | 9 ± 0.8 | 13* | 259 ± 11 | 317* | 296 ± 11 | 313 ± 9* | 17 ± 0.4 | 21 ± 1 |
| 291 ± 20 | 288 | 3 ± 0.3 | 5* | 54 ± 3 | 56* | 332 ± 16 | 354 ±11* | 39 ± 2 | 45 ± 2 |
| 202 ± 14 | 216 | 15 ± 1 | 24* | 378 ± 20 | 320* | 337 ± 13 | 355 ±11* | 203 ± 6 | 41 ± 2 |
| 357 ± 26 | 377 | 98 ± 7 | 102 | | | 175 ± 8 | 197 ± 6* | 86 ± 4 | 94 ± 5 |
| 315 ± 39 | 326 | | | | | | | 512 ± 21 | 600 ± 29 |
| | | | | | | | | 422 ± 21 | 454 ± 22 |
| | | | | | | | | 602 ± 21 | 651 ± 32 |

+ All results are expressed in milligrams of uranium per liter of solution. Error terms, where shown are the 2-sigma counting error.

* Average of duplicate analyses.

TABLE 5. RADIONUCLIDE CONCENTRATIONS IN YELLOWCAKE GRAB SAMPLES

| | EPA | | LFE | | Eberline** | | |
|---|---|---|---|---|---|---|---|
| | pCi/gram ± 2-Counting Error | Percent of U-238 Activity | pCi/gram ± 2-Counting Error | Percent of U-238 Activity | pCi/sample ± 2 Counting Error | Percent of U-238 Activity | Percent of U-238 Equilibrium (Mean ± 1 ) |
| **United Nuclear-Homestake Partners - EPA Sample 188941** | | | | | | | |
| U-238 | 253,000 ± 12,900 | - | 274,000 ± 27,400 | - | 178,000 ± 20,000 | - | - |
| U-234 | 264,000 + 13,300 | 104.35 | Not reported | - | 176,000 ± 20,000 | 98.9 | 102 ± 3.85 |
| U-235 | 11,000 ± 1,580 | 4.35 | Not reported | - | 6,200 ± 1,600 | 3.48 | 3.92 ± 0.62 |
| Th-230 | 6,780 ± 523 | 2.68 | 7,330 ± 733 | 2.68 | 5,300 ± 1,200 | 2.98 | 2.78 ± 0.173 |
| Ra-226 | 840 ± 50.0 | 0.332 | 938 ± 93.8 | 0.342 | 590 ± 4 | 0.331 | 0.335 ± 0.006 |
| Pb-210 | 263 ± 20.2 | 0.104 | 219 ± 21.9 | 0.080 | 160 ± 8 | 0.090 | 0.091 ± 0.012 |
| Po-210 | 213 ± 15.9 | 0.084 | 159 ± 15.9 | 0.058 | No analysis | - | 0.071 ± 0.018 |
| **United Nuclear-Homestake Partners - EPA Sample 188942** | | | | | | | |
| U-238 | 253,000 ± 11,500 | - | No analysis | | No analysis | | - |
| U-234 | 252,000 ± 11,500 | 99.6 | No analysis | | No analysis | | 99.6 |
| U-235 | 10,400 ± 1,200 | 4.11 | No analysis | | No analysis | | 4.11 |
| Th-230 | 6,860 ± 566 | 2.71 | No analysis | | No analysis | | 2.71 |
| Ra-226 | 880 ± 30.0 | 0.348 | No analysis | | No analysis | | 0.348 |
| Pb-210 | 247 ± 18.4 | 0.098 | No analysis | | No analysis | | 0.098 |
| Po-210 | 166 ± 9.07 | 0.066 | No analysis | | No analysis | | 0.066 |
| **United Nuclear-Homestake Partners - EPA Sample 188943** | | | | | | | |
| U-238 | 255,000 ± 14,500 | - | 270,000 ± 27,000 | - | 182,000 ± 33,000 | - | - |
| U-234 | 251,000 ± 14,400 | 98.4 | Not reported | - | 179,000 ± 32,000 | 98.4 | 98.4 ± 0.0 |
| U-235 | 10,200 ± 2,050 | 4.00 | Not reported | - | 7,800 ± 3,000 | 4.29 | 4.15 ± 0.025 |
| Th-230 | 6,600 ± 478 | 2.59 | 6,650 ± 665 | 2.46 | 3,600 ± 500 | 1.98 | 2.343 ± 0.321 |
| Ra-226 | 630 ± 27.0 | 0.247 | 793 ± 79.3 | 0.294 | 530 ± 4 | 0.291 | 0.277 ± 0.026 |
| Pb-210 | 230 ± 20.4 | 0.090 | 218 ± 21.8 | 0.081 | 128 ± 10 | 0.070 | 0.080 ± 0.010 |
| Po-210 | 178 ± 15.4 | 0.070 | 177 ± 28.3 | 0.066 | No analysis | - | 0.068 ± 0.003 |

TABLE 5. RADIONUCLIDE CONCENTRATIONS IN YELLOWCAKE GRAB SAMPLES (continued)

| | EPA | | LFE | | Eberline** | | |
|---|---|---|---|---|---|---|---|
| | pCi/gram ± 2-Counting Error | Percent of U-238 Activity | pCi/gram ± 2-Counting Error | Percent of U-238 Activity | pCi/sample ± 2 Counting Error | Percent of U-238 Activity | Percent of U-238 Equilibrium (Mean ± 1 ) |
| **Union Carbide - EPA Sample 188927 (Referee Sample)** | | | | | | | |
| U-238 | 250,000 ± 12,000 | - | 253,000 ± 25,300 | - | 431,000 ± 48,000 | - | - |
| U-234 | 240,000 ± 12,000 | 96.0 | Not reported | - | 438,000 ± 48,000 | 101.6 | 98.8 ± 4.0 |
| U-235 | 8,800 ± 1,300 | 3.52 | Not reported | - | 15,000 ± 3,000 | 3.48 | 3.50 ± 0.03 |
| Th-230 | 2,150 ± 144 | 0.860 | 2,040 ± 204 | 0.806 | 2,900 ± 500 | 0.673 | 0.780 ± 0.096 |
| Ra-226 | 81 ± 6.9 | 0.032 | 150 ± 15 | 0.059 | 137 ± 3 | 0.032 | 0.041 ± 0.016 |
| Pb-210 | 92 ± 7.6 | 0.037 | 101 ± 10.1 | 0.040 | 183 ± 11 | 0.042 | 0.040 ± 0.003 |
| Po-210 | 99 ± 6.1 | 0.040 | 91.5 ± 9.15 | 0.036 | No analysis | - | 0.038 ± 0.003 |
| **Union Carbide - EPA Sample 188924 (Finished Product)** | | | | | | | |
| U-238 | 250,000 ± 14,000 | - | 262,000 ± 26,200 | - | 188,000 ± 36,000 | - | - |
| U-234 | 240,000 ± 13,000 | 96.0 | Not reported | - | 187,000 ± 35,000 | 99.5 | 97.8 ± 2.5 |
| U-235 | 8,500 ± 1,800 | 3.40 | Not reported | - | 7,900 ± 3,100 | 4.20 | 3.80 ± 0.57 |
| Th-230 | 1,730 ± 164 | 0.692 | 1,460 ± 146 | 0.557 | 1,100 ± 200 | 0.585 | 0.611 ± 0.071 |
| Ra-226 | 37 ± 4.8 | 0.015 | 28.4 ± 2.84 | 0.011 | 40 ± 1 | 0.021 | 0.016 ± 0.005 |
| Pb-210 | 41 ± 6.2 | 0.016 | 45.6 ± 9.12 | 0.017 | 34 ± 7 | 0.018 | 0.017 ± 0.001 |
| Po-210 | 46 ± 6.9 | 0.018 | 38.4 ± 4.61 | 0.015 | No analysis | - | 0.017 ± 0.002 |
| **Union Carbide - EPA Sample 188925 (Feed to Dryer)** | | | | | | | |
| U-238 | 110,000 ± 8,200 | - | No analysis | | No analysis | | - |
| U-234 | 110,000 ± 7,900 | 100.0 | No analysis | | No analysis | | 100 |
| U-235 | 4,300 ± 1,100 | 3.90 | No analysis | | No analysis | | 3.9 |
| Th-230 | 717 ± 44.1 | 0.652 | No analysis | | No analysis | | 0.652 |
| Ra-226 | 18 ± 2.1 | 0.016 | No analysis | | No analysis | | 0.016 |
| Pb-210 | 22 ± 3.9 | 0.020 | No analysis | | No analysis | | 0.020 |
| Po-210 | 15 ± 1.8 | 0.014 | No analysis | | No analysis | | 0.014 |

TABLE 5. RADIONUCLIDE CONCENTRATION OF YELLOWCAKE GRAB SAMPLES (continued)

| | EPA | | LFE | | Eberline** | | |
|---|---|---|---|---|---|---|---|
| | pCi/gram ± 2- Counting Error | Percent of U-238 Activity | pCi/gram ± 2- Counting Error | Percent of U-238 Activity | pCi/sample ± 2- Counting Error | Percent of U-238 Activity | Percent of U-238 Equilibrium (Mean ± 1 ) |
| **Kerr-McGee - EPA Sample 188926** | | | | | | | |
| U-238 | 220,000 ± 16,000 | - | No analysis | | No analysis | | - |
| U-234 | 230,000 ± 17,000 | 104.5 | No analysis | | No analysis | | 104.5 |
| U-235 | 9,200 ± 1,900 | 4.18 | No analysis | | No analysis | | 4.18 |
| Th-230 | Not reported | - | No analysis | | No analysis | | - |
| Ra-226 | 5.1 ± 1.2 | 0.002 | No analysis | | No analysis | | 0.002 |
| Pb-210 | Not reported | - | No analysis | | No analysis | | - |
| Po-210 | Not reported | - | No analysis | | No analysis | | - |
| **Kerr-McGee - EPA Sample 167972** | | | | | | | |
| U-238 | 234,000 ± 15,600* | - | 247,000 ± 24,700 | - | 184,000 ± 29,000 | - | - |
| U-234 | 225,000 ± 14,900* | 96.2 | Not reported | - | 170,000 ± 27,000 | 92.4 | 94.3 ± 2.7 |
| U-235 | 7,000 ± 1,300 | 2.99 | Not reported | - | 5,800 ± 2,200 | 3.15 | 3.07 ± 0.11 |
| Th-230 | 321 ± 39.4* | 0.137 | 225 ± 22.5 | 0.091 | 121 ± 23 | 0.066 | 0.098 ± 0.036 |
| Ra-226 | 24 ± 16.7* | 0.010 | 2.95 ± 0.59 | 0.001 | 4.5 ± 0.6 | 0.002 | 0.004 ± 0.005 |
| Pb-210 | 23 ± 5.4 | 0.010 | 16.4 ± 8.2 | 0.007 | 27 ± 7 | 0.015 | 0.011 ± 0.004 |
| Po-210 | 19 ± 3.0 | 0.008 | 19.9 ± 3.18 | 0.008 | No analysis | - | 0.008 ± 0.000 |
| **Sohio - EPA Sample 188932** | | | | | | | |
| U-238 | 259,000 ± 14,000 | - | 258,000 ± 25,800 | - | 187,000 ± 26,000 | - | - |
| U-234 | 245,000 ± 13,000 | 102.5 | Not reported | - | 185,000 ± 26,000 | 98.9 | 100.7 ± 2.5 |
| U-235 | 10,800 ± 1,100 | 4.17 | Not reported | - | - | - | - |
| Th-230 | 555 ± 37.5 | 0.214 | 930 ± 93 | 0.360 | 522 ± 120 | 0.279 | 0.284 ± 0.073 |
| Ra-226 | 1.1 ± 0.58 | 0.0004 | 0.73 ± 0.31 | 0.0003 | 0.6 ± 0.3 | 0.0003 | 0.0003 ± 0.00006 |
| Pb-210 | 11.3 ± 3.18 | 0.004 | 18.4 ± 8.83 | 0.007 | 6.5 ± 6.0 | 0.003 | 0.005 ± 0.002 |
| Po-210 | 29.1 ± 2.68 | 0.011 | 58.8 ± 5.88 | 0.023 | No analysis | | 0.017 ± 0.008 |

* Average of two analyses.
** Note that Eberline results are pCi/sample, rather than pCi/gram as reported by EPA and LFE.

## DISCUSSION OF RESULTS

### Uranium Emission Rates

Table 1 shows the basic data of the study, and the uranium emission rates. At the outset of the study, we hoped to find some correlation between the uranium emission rate and one or more mill operating parameters. The emission rate should logically be a function of the amount of uranium concentrate produced by the mill per unit time, with some adjustment for the efficiency of the stack gas cleaning equipment. Such analysis is typical in the scientific literature and in environmental impact analyses. Specifically, the uranium emission rate is usually estimated using the following parameters:

- Ore processing rate (usually tons per day or year)
- Average ore grade (percent $U_3O_8$)
- Efficiency of the mill process for recovering uranium
- Appropriate assumptions of operating time per year
- Efficiency of effluent cleaning equipment

An annual average emission rate, usually expressed as curies per year, is desired for evaluating the environmental impact of the mill.

Presumably, the emission rate at any given time should be proportional to the mill operating parameters at that time. When possible during this study, the best estimate of mill operating parameters during the stack testing period was obtained, including the feed rate of yellowcake slurry to the dryer and the quantity of yellowcake packaged. However, the observed emission rates did not correlate with these two parameters. This is likely because the stack tests were conducted over a relatively short period (usually one to two hours). Consequently, mill operating changes are not likely to produce observable changes in emission rate using such short measurement periods. The Union Carbide results of Table 6 are a good example of such inconsistency. Of the seven test results, the second to the highest emission rate measured

TABLE 6. $U_3O_8$ DRYING, PACKAGING, AND EMISSION RATE PARAMETERS

| Mill | Approximate Pounds $U_3O_8$ Dried/hr | Approximate Pounds $U_3O_8$ Packaged/hr | Emission Rate Lbs $U_3O_8$/hr | Emission Rate as a Percentage of $U_3O_8$ Processing Rate |
|---|---|---|---|---|
| United Nuclear-Churchrock | 347 | | 0.38 | 0.11 |
| | 311 | | 0.22 | 0.07 |
| | 350 | | 0.48 | 0.14 |
| | | | Average = | 0.11 |
| Sohio | 180 | | 0.0039 | 0.0022 |
| | 171 | | 0.0013 | 0.00076 |
| | 50 | | 0.0061 | 0.012 |
| | | | Average = | 0.0050 |
| Union Carbide-Uravan | 224* | | 0.14 | 0.063 |
| | 159 | | 0.11 | 0.069 |
| | 243 | | 0.18 | 0.074 |
| | 231 | | 0.25 | 0.11 |
| | 261 | | 0.18 | 0.069 |
| | 343 | | 0.31 | 0.090 |
| | 159 | | 0.28 | 0.18 |
| | | | Average = | 0.094 |
| United Nuclear-Churchrock | | 3375 | 0.23 | 0.0068 |
| | | 2025 | 0.23 | 0.011(4) |
| | | 2025 | 0.12 | 0.0059 |
| | | | Average = | 0.0079 |
| Sohio | | 3600 | 0.37 | 0.01** |
| | | 450 | 0.0052 | 0.0012 |
| | | 1080 | 0.020 | 0.0019 |
| | | | Average = | 0.0016 |
| United Nuclear-Homestake Partners | | 1840 | 0.65 | 0.035 |
| | | 2760 | 0.93 | 0.034 |
| | | | Average = | 0.0345 |

* Drying and packaging operations are concurrent with a common scrubber and stack.
** Bag filter ruptured during test. Result not used in the average. See Table 1 Footnote comment.

(0.28 lbs $U_3O_8$/hr) corresponds to the lowest production rate (159 lbs $U_3O_8$/hr). Alternately, the test resulting in the lowest emission rate (0.11 lbs $U_3O_8$/hr) corresponds to a period of identical production rate (159 lbs $U_3O_8$/hr).

The study demonstrated that the emission rate at a given time (the "instantaneous" emission rate) is controlled by a complex interaction of several factors. In addition to the mill operating parameters listed above, these include, as a partial list, such things as:

- Drying and packaging ventilation design factors which, under varying operational conditions, create varying yellowcake entrainment with subsequent variations in yellowcake concentrations arriving at the stack gas cleaning system.
- The adjustment and "tuning" of stack gas cleaning systems at a particular time (i.e., the "instantaneous" efficiency of the system).
- The general condition and cleanliness of the yellowcake processing and stack gas cleaning systems, including ductwork. There may be a considerable amount of yellowcake deposited in these systems that may be available for resuspension.
- The nature of operating procedures for yellowcake packaging, and how well these procedures are being followed by the particular operator at the particular time.

Undoubtedly, the instantaneous yellowcake emission rate at the point of the stack exhaust involves still other factors, which may be subtle, intermittent, and highly variable in impact on the emission rate. These factors are also difficult to observe, and are difficult or impossible, at least within the context of this study, to quantify relative to the observed emission rate.

Table 7 shows a measure of the gross variability of the test results, expressed as a ratio of the highest to the lowest measured values for each stack tested. (The Anaconda result is not included because only one test was performed there.) For the UNHP and Sohio packaging stacks, an "alternate" calculation was made to include additional tests made under operating

TABLE 7. VARIABILITY OF MEASURED YELLOWCAKE EMISSION RATES

| Mill | Stack | No. Of Tests | Ratio-Highest To Lowest Emission Rate | Remarks |
|---|---|---|---|---|
| Kerr-McGee* | Main | 2 | 2.9 | |
| | East | 3 | 2.0 | |
| | West | 3 | 7.1 | |
| UC-Uravan | Dryer-Package | 7 | 2.8 | |
| UN-Churchrock | Dryer | 3 | 2.2 | |
| | Package | 3 | 1.9 | |
| UNHP* | Vanadium Roaster | 5 | 2.3 | |
| | Dryer | 3 | 3.4 | |
| | Package | 3 | 1.4 | Yellowcake was being packaged. |
| | Package (alternate) | 4 | 23 | Includes 3 tests above, plus one when yellowcake was not being packaged. |
| Sohio | Dryer | 3 | 4.7 | |
| | Package | 2 | 3.9 | Excludes test made when bag filter was ruptured. |
| | Package (alternate) | 3 | 71 | Includes test with ruptured filter. |

* The stacks at these mills were tested on two occasions at five-month intervals.

conditions that were known to be unusual or markedly different. The table notes these conditions. Although usually the number of tests on each stack is small, the ranges of emission rates shown in this table give an estimate of the variability to be expected from a series of tests on a given stack.

The alternate calculation for the UNHP packaging stack shows the emission rate range from a packaging stack that can occur depending upon whether yellowcake is or is not being packaged at the time of the test. The Sohio alternate calculation shows the effect of a ruptured bag filter. Certainly more data are needed to draw definitive conclusions about the effects of such variables. However, by just these two tests, the emission rate shows variation by a factor of roughly 10 to 20.

Aside from these major known variables, this study shows that the emission rates from a given stack, as measured by an individual test, can vary by roughly a factor of two to five. Again, the relatively small number of tests involved should be kept in mind when evaluating this data. The only exception is the three tests on the Kerr-McGee west Microdyne stack, which vary by a factor of seven. This may be at least partially explained by the fact that mill operating data were not obtained during these tests. Consequently, some unobserved factor (e.g., differences in the amount of yellowcake packaged) may account for the larger range of these results.

The range of variation discussed above includes sampling and analytical error. The Method 5 sampling procedure is estimated to have an overall probable sampling error of about 7% (Shigehara et al., 1970). Analytical errors may vary considerably, depending on the method used and the expertise of the laboratory performing the analysis. Estimated analytical errors for routine analyses at the uranium levels involved are as follows:

- Delayed neutron counting: 5% or less
- Radiochemistry and alpha spectroscopy: 5-20%
- Fluorimetric analysis: 25%
- Colorimetric analysis: 3%

The approximate combined sampling and analytical error could therefore range

from less than 10% to about 25%. Consequently, the combined error in measuring the emission rate is considerably less than the minimum variation of 100% (factor of two) observed in the stack test results.

Estimation of Annual Average Emission Rates

Disregarding the variability and the apparent lack of correlation, we calculated emission rates of $U_3O_8$ as a percentage of $U_3O_8$ processing rates for four of the mills where such data was available, and averaged these percentages over the number of tests conducted. The results of these calculations, listed in Table 6, show United Nuclear Churchrock (UNC) releasing an average of 0.118% of the $U_3O_8$ dried and packaged, Sohio releasing an average of 0.0066% of the $U_3O_8$ dried and packaged, and Union Carbide releasing an average of 0.094% of the $U_3O_8$ dried and packaged. The Union Carbide result is notable since it corresponds to an identical release rate calculated by Momeni et al. (1979). They were provided the Union Carbide $U_3O_8$ emission rate data from the tests reported herein, but calculated the $U_3O_8$ production rates by considering ore throughput and grade, extraction efficiency, and mill operating time. Using Union Carbide data and that from another mill, Momeni recommended using a value of 0.1% for estimating $U_3O_8$ released from annual $U_3O_8$ production estimates. Note also that the UNC data agrees with Momeni's estimation. Sohio is obviously the exception with a total $U_3O_8$ release rate of 0.0066%, and based on the data of Table 6, a 0.1% release rate applied here would over-estimate their total $U_3O_8$ release by a factor of 15.

The use of emission rates (as normally reported from stack tests viz lb or pCi $U_3O_8$/hr) in calculating annual source terms must consider the operating periods of the several sources. Since yellowcake packaging operations are typically intermittent with respect to yellowcake drying operations, it would obviously be invalid to simply add the reported hourly release rates for these two sources to arrive at a total hourly release rate in calculating annual source terms. How intermittent packaging operations are, at least at two mills, can be calculated based on the data of Table 6. These calculations show the ratio of the packaging time to drying time at UNC and Sohio to be respectively about 0.14 and 0.08. Therefore, in calculating an annual source term, the total operating times for each source must be defined after which

individual source terms may be calculated for each stack for the year. The source term for each stack is then summed to arrive at the total annual source term. Annual source term calculations based on the summation of drying and packaging stack hourly release rates, without time-weighting, at UNC and Sohio would overestimate their annual source terms by a factor of 1.5 and 2.8, respectively.

Concentrations of Uranium Daughter Radionuclides in Yellowcake

Table 5 shows the concentrations of the three uranium nuclides (U-238, -234, and -235) and of the daughter nuclides (thorium-230, radium-226, lead-210, and polonium-210). This table includes results for six samples that were analyzed by three laboratories, and for three additional samples that were analyzed only by the EPA laboratory. Each analysis shows the percent of each daughter nuclide's activity relative to the parent U-238 activity.

The agreement between the three laboratories is generally good. Usually where comparable analytical results are available (between EPA and LFE), they are within the two-sigma counting error range. (Remember that the Eberline concentration results are not directly comparable to the EPA and LFE results, due to reporting differences.) All the samples analyzed by EPA and Eberline show U-238 and U-234 in equilibrium within the limits of reported counting errors. (LFE did not report U-234 results.)

For the daughter nuclides, the results reported by all three laboratories can be compared by referring to the calculated percentage of the daughter nuclides relative to the parent U-238. The differences between these results are fairly randomly distributed between the three laboratories. For thorium-230, EPA tends to be low and Eberline high, with LFE in the middle. For polonium-210, EPA is generally lower than LFE (Eberline did not report polonium-210 results). While inter-laboratory agreement does not necessarily prove accuracy, it is encouraging that there are no strong biases between the three laboratories. Because of this, the percentage values for all analyses of all samples from each of the four mills have been averaged to arrive at an overall average daughter percentage in yellowcake from each mill. Table 8 shows these values.

Sears (1975) reported that, for an acid leach mill, about 5% of the thorium and 0.2% of the radium are carried over to the yellowcake product, although these percentages may vary according to the details of the chemistry used for uranium extraction and yellowcake purification. An alkaline leach mill is reported to carry over 1.8% of the radium, and none of the thorium. For both circuits, no other significant radionuclides are reported to be present. The results in Table 8 show that thorium-230 in the yellowcake from the three acid leach mills is considerably lower than the 5% stated above and that the radium-226 is equal to or considerably lower than the 0.2% stated. For the one alkaline leach mill, significant thorium-230 was found in the yellowcake, while the radium-226 was about 17% of the value quoted by Sears. Measurable amounts of Pb-210 and Po-210 were found in all yellowcake samples, with that from the alkaline leach mill being higher than that from the acid leach mills. Always, however, less than 0.09% each of lead and polonium was carried over to the yellowcake.

Table 8. DAUGHTER RADIONUCLIDES IN YELLOWCAKE AS PERCENT OF U-238 PARENT*

| | Mean ± one standard deviation (percent) | | | |
|---|---|---|---|---|
| Nuclide/Mill | UNHP (3 samples) | Union Carbide (3 samples) | Kerr-McGee (2 samples) | Sohio (1 sample) |
| Th-230 | 2.58 ± 0.309 (7)** | 0.689 ± 0.110 (7) | 0.0980 ± 0.0360 (3) | 0.284 ± 0.0731 (3) |
| Ra-226 | 0.312 ± 0.036 (7) | 0.0266 ± 0.0165 (7) | 0.0038 ± 0.0042 (4) | 0.00033 ± 0.00006 (3) |
| Pb-210 | 0.0876 ± 0.0117 (7) | 0.0271 ± 0.0119 (7) | 0.0107 ± 0.0040 (3) | 0.00467 ± 0.00208 (3) |
| Po-210 | 0.0688 ± 0.0095 (5) | 0.0246 ± 0.0124 (5) | 0.0080 ± 0.0000 (2) | 0.0170 ± 0.00849 (2) |

* Calculated from results in Table 5.
** Number in parenthesis is the number of analyses used in calculating the mean.

## SUMMARY AND RECOMMENDATIONS

The Nuclear Regulatory Commission is currently considering the monitoring and sampling procedures that should be required at uranium mills to demonstrate compliance with environmental radiation protection standards. In order to estimate dose to people near the mill, a source term, or radioactivity released per unit time, must be known or estimated. This is usually expressed as curies per year in order to average over a suitable period. The problem with determining uranium mill source terms in general, and yellowcake stack source terms in particular, is in realistically estimating or calculating this annual average emission rate.

This study shows that measured emission rates from a yellowcake processing stack can routinely vary by a factor of two to five when tests are conducted without any controls on the mill operating parameters. In addition, these tests show that major anomalies (e.g., a ruptured filter in the exhaust clean-up system) can cause variations of as much as a factor of 20 in the measured emission rate. Based on our experience, the major operational parameter that affects the emission rate is simply whether yellowcake is or is not being dried or packaged during the testing period. Typically, the cleaning systems run continuously while the operations may be performed intermittently. This is especially true for many packaging operations and to a lesser extent for drying operations. As discussed earlier, results from a single test at United Nuclear-Homestake Partners show measurable amounts of yellowcake being released from the operating packaging stack (0.02 lb $U_3O_8$/hr) even though yellowcake was not being packaged during the test. Later tests on this stack show a significant increase in the emissions during periods when packaging was under way. Consequently, an emission rate measurement (or measurements) representing only one operating condition would bias the annual average source term considerably.

Major events that affect the emission rate would also have to be considered for a calculation as described above. A serious malfunction of the

exhaust air cleanup systems is the most obvious of these, as discussed previously for the Sohio sample. Maintenance or repair work on the exhaust system might also increase the emission rate significantly by dislodging yellowcake from the system, although this would probably be short-lived. Conversely, a complete shutdown of the yellowcake systems would reduce the emission rate to zero during the down period.

Based on the results of this study and on other general information obtained during the course of the study, we can recommend a general stack testing and emission rate calculation scheme that should provide a reasonable estimate of the annual source term from yellowcake stacks. Basically, this scheme would consist of sampling each stack several times under various mill operating conditions and time-weighting these various emission rates according to the proportion of time per year during which the stack is operated under each particular condition. Our general recommendations are as follows:

1. A number of samples would be collected from each stack over a period of a year to establish the range of emission rates from that stack. These tests would be made while the yellowcake processing operations are underway with the operating conditions maintained at steady-state conditions.

2. During this year, each stack would also be tested several times when yellowcake operations are not underway, but with the air cleaning systems in operation, if this condition occurs normally at the mill. This would establish the range of emission rates under this condition.

3. The tests should be conducted according to the EPA Reference Methods using the appropriate modifications for yellowcake stack testing described earlier in this report.

4. For each stack, a record of the operating time under each of the above conditions would be maintained for a year, and summarized on an annual basis.

5. The total annual source term for each stack would then be calculated by multiplying the hourly emission rate for each condition by the hours per year the stack operates under that conditon, and summing for all known operating conditions. The annual source term for the mill would be obtained by summing the source terms for each stack.

6. The above calculation is intended to apply to uranium emission rates. The uranium daughter nuclides are a small fraction of the uranium in yellowcake, and the analyses for them are expensive and time consuming. Since the ratio of the daughter nuclides to uranium appears to be fairly constant for a given mill, we suggest that they be determined once a year from a blended sample of the yellowcake produced during the year.

7. An alternate method of estimating the source term is to relate the emission rate from the various stacks to the amount of yellowcake processed (e.g., dried or packaged) in the areas served by each stack. This technique requires an additional piece of data, the amount of yellowcake processed during a stack test. One can then calculate the uranium emission (not emission rate) normalized to the yellowcake production, expresed as pCi $U_{tot}$/pound of yellowcake processed. Since the amount of the yellowcake produced per year by the mill is known, the annual source term can then be estimated by multiplying this normalized uranium emission by the annual yellowcake production.

   We feel that this calculation method would be inherently less satisfactory than the time-weighted method discussed above because (a) it does not account for releases which occur when yellowcake is not being processed; (b) in some mills, the exhaust stacks do not serve a single process area, thus complicating the relationship between yellowcake processing and uranium releases; and (c) as shown previously, there is very poor correlation between the measured emission rates and the concurrent yellowcake processing rates. A comparison of the two methods may be more feasible when further data and experience are available.

Technically the number and frequency of samples should be determined by a statistical analysis of the data, and would depend primarily on the variability and the distribution of the results. These parameters would determine the reliability of the calculated annual emission rate and the degree of confidence associated with it. However, from a regulatory viewpoint, it may be simpler to specify a fixed number and frequency of samples.

We think it important to remember the variability between mills and the uniqueness of each mill. Also, changes in operating parameters can change emission rates over a period of time. Consequently, a single, fixed sampling regime is not necessarily always the optimum. It may not provide the most useful technical data, and it may not be the least costly way to obtain the data, if costs are to be considered.

Our approach in the above sampling scheme has been to outline a general program that should provide a realistic estimate of the annual emission rate for a mill. Logically, this would involve a larger number of samples and more frequent sampling the first year. This will allow one to develop a feel for the range of the results for the particular stacks and, hopefully, for the causes of these variations. Once this range is defined and the variables are at least partially controlled, the sampling program could be modified and used primarily to follow trends and to confirm that no major variations are occurring.

All the above discussions of stack sampling techniques and programs are based on the EPA Reference Method 5 sampling technique used in this study. As mentioned previously, the major shortcoming of this method for measuring the annual yellowcake emission rate is the short duration of the test period (one to two hours) compared to the time frame over which most of the variations in the emission rate occur. As a result, a single test, or a few tests, will likely give misleading results. As more and more tests are conducted to average out these variables, one is, in effect, approaching continuous stack sampling. This ideal situation would eliminate the need for, and uncertainty involved in, averaging and manipulating the results from a number of individual tests to obtain the annual average. Such a continuous monitor should be reliable, reasonably accurate, and inexpensive. Argonne National Laboratory

(Momeni et al., 1979) has developed, but not yet tested, such a concept and prototype. We recommend the further development of such equipment, with field tests and evaluations of its suitability in this area.

As an aside, we feel it important to mention a dramatic reduction of the packaging stack yellowcake release rate we observed at the United Nuclear-Churchrock mill, after minor modifications. During the emissions tests reported herein, the average emission rate for the packaging stack was 0.19 lb $U_3O_8$/hr (Table 6). Subsequently, a series of particle-sizing studies were performed on the same stack. The emission rates calculated from these studies averaged 0.005 lb $U_3O_8$ (Fort et al., 1980). Between these two studies, minor modifications had been made on the packaging stack ventilation and cleaning systems (Todd Miller, United Nuclear-Churchrock, personal communication, 1978). While emission rates determined from particle-sizing studies are not necessarily as accurate as those determined by the Method 5 procedures, the magnitude of the reduction indicates a definite improvement in the efficiency of the exhaust air cleaning system. Whether cleaning systems at other mills could be "fine-tuned" to achieve this magnitude of emission rate reduction is unknown and was not a goal of these studies.

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## APPENDIX A

### HP-67/97 CALCULATOR PROGRAM FOR ESTIMATING STACK GAS MOISTURE FRACTION

The basic equation for estimating moisture fraction is: $B_{ws} = \frac{\ell_a}{P_{abs}}$

Where: $B_{ws}$ = Moisture fraction

$\ell_a$ = Dew point saturation vapor pressure (in. Hg)

$$= \ell'' - \frac{(P_{abs} - \ell'')(T_d - T_w)}{2800 - 1.3 * T_w} \qquad \text{(The Carrier Equation)}$$

$\ell''$ = Saturation vapor pressure at $T_w$ (in. Hg)

$P_{abs}$ = Absolute stack pressure (in. Hg)

$T_d$ = Stack gas dry bulb temperature (°F)

$T_w$ = Stack gas wet bulb temperature (°F)

The HP-67/97 program calculates $\ell''$ for a given $T_w$ using a series of equations derived to define saturation vapor pressure (SVP) in inches of Hg as a function of wet bulb temperature over the range 0-159°F. These equations are given below:

for $T_w$ = 0- 29°F, $SVP = 0.03821\ e^{0.04911 * T_w}$

$T_w$ = >29- 59°F, $SVP = 0.05290\ e^{0.03840 * T_w}$

$T_w$ = >59-105°F, $SVP = 0.07590\ e^{0.03250 * T_w}$

$T_w$ = >105-159°F, $SVP = 0.14210\ e^{0.02651 * T_w}$

SVP values calculated using these equations will have a maximum error of approximately 3%. For those interested, more precise results may be obtained by reducing the temperature intervals and using the HP-67/97 exponential curve fit program to define the equation of the new data group. In this same manner, SVP equations for temperatures greater than 159°F may also be obtained. Use the goodness of fit test ($r^2$) in the program to determine the appropriate grouping for the accuracy desired.

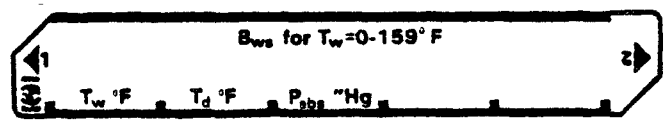
The equations have been found to be more than adequate in estimating moisture fraction for calculating isokinetic $\Delta H$. The maximum difference observed between $B_{ws}$ calculated using this method and $B_{ws}$ as determined by the condensation method for the 40 tests described in this report was approximately 10%. Typically, the agreement was a few percent or less. This method has proven sufficiently accurate for calculating $\Delta H$ with dew point temperatures as high as 141°F.

To obtain the accuracy described, care must be taken in making the wet and dry bulb temperature measurements. This amounts primarily to allowing the thermometer to reach equilibrium conditions. Of course, when using separate thermometers for the two measurements, they must be well matched.

This program is easily adapted to the HP-65 by one of two methods: (1) Perform the SVP calculations for the range 0-159°F on one magnetic card, storing SVP and $T_w$ for use with a second magnetic card to complete the calculation for $B_{ws}$. (2) Incorporate on one magnetic card the complete program for $T_w$ = 0-59°F, using a second magnetic card for $T_w$ = >60-159°F.

It should be pointed out that this method will also work to determine the moisture fraction in ambient air.

USER INSTRUCTIONS




| STEP | INSTRUCTIONS | DISPLAY |
|---|---|---|
| 1 | Input $T_w$(°F), Touch "A" | SVP,(in. Hg) |
| 2 | Input $T_d$(°F), Touch "B" | $T_d$(°F) |
| 3 | Input $P_{abs}$ (in. Hg), Touch "C" | $B_{ws}$ |

| STEP | KEY | CODE |
|---|---|---|
| 001 | *LBL4 | 21 11 |
| 002 | STO3 | 35 00 |
| 003 | X=Y? | 16-45 |
| 004 | GTO4 | 22 04 |
| 005 | 2 | 02 |
| 006 | 9 | 09 |
| 007 | RCL0 | 36 00 |
| 008 | X>Y? | 16-34 |
| 009 | GTO1 | 22 01 |
| 010 | . | -62 |
| 011 | 0 | 00 |
| 012 | 4 | 04 |
| 013 | 9 | 09 |
| 014 | 1 | 01 |
| 015 | 1 | 01 |
| 016 | 2 | 02 |
| 017 | × | -35 |
| 018 | $e^x$ | 33 |
| 019 | . | -62 |
| 020 | 0 | 00 |
| 021 | 3 | 03 |
| 022 | 8 | 08 |
| 023 | 2 | 02 |
| 024 | 1 | 01 |
| 025 | × | -35 |
| 026 | STO1 | 35 01 |
| 027 | RTN | 24 |
| 028 | *LBL1 | 21 01 |
| 029 | 5 | 05 |
| 030 | 9 | 09 |
| 031 | RCL0 | 36 00 |
| 032 | X>Y? | 16-34 |
| 033 | GTO2 | 22 02 |
| 034 | . | -62 |
| 035 | 0 | 00 |
| 036 | 3 | 03 |
| 037 | 8 | 08 |
| 038 | 4 | 04 |
| 039 | × | -35 |
| 040 | $e^x$ | 33 |
| 041 | . | -62 |
| 042 | 0 | 00 |
| 043 | 5 | 05 |
| 044 | 2 | 02 |
| 045 | 9 | 09 |
| 046 | × | -35 |
| 047 | STO1 | 35 01 |
| 048 | RTN | 24 |
| 049 | *LBL2 | 21 02 |

| STEP | KEY | CODE |
|---|---|---|
| 050 | 1 | 01 |
| 051 | 0 | 00 |
| 052 | 5 | 05 |
| 053 | RCL0 | 36 00 |
| 054 | X>Y? | 16-34 |
| 055 | GTO3 | 22 03 |
| 056 | . | -62 |
| 057 | 0 | 00 |
| 058 | 3 | 03 |
| 059 | 2 | 02 |
| 060 | 5 | 05 |
| 061 | × | -35 |
| 062 | $e^x$ | 33 |
| 063 | . | -62 |
| 064 | 0 | 00 |
| 065 | 7 | 07 |
| 066 | 5 | 05 |
| 067 | 9 | 09 |
| 068 | × | -35 |
| 069 | STO1 | 35 01 |
| 070 | RTN | 24 |
| 071 | *LBL3 | 21 03 |
| 072 | 1 | 01 |
| 073 | 5 | 05 |
| 074 | 9 | 09 |
| 075 | RCL0 | 36 00 |
| 076 | X>Y? | 16-34 |
| 077 | GTO5 | 22 05 |
| 078 | . | -62 |
| 079 | 0 | 00 |
| 080 | 2 | 02 |
| 081 | 6 | 06 |
| 082 | 5 | 05 |
| 083 | 1 | 01 |
| 084 | × | -35 |
| 085 | $e^x$ | 33 |
| 086 | . | -62 |
| 087 | 1 | 01 |
| 088 | 4 | 04 |
| 089 | 2 | 02 |
| 090 | 1 | 01 |
| 091 | × | -35 |
| 092 | STO1 | 35 01 |
| 093 | RTN | 24 |
| 094 | *LBL6 | 21 12 |
| 095 | STO2 | 35 02 |
| 096 | RTN | 24 |
| 097 | *LBL0 | 21 13 |
| 098 | STO3 | 35 03 |

| STEP | KEY | CODE |
|---|---|---|
| 099 | RCL2 | 36 02 |
| 100 | RCL0 | 36 00 |
| 101 | - | -45 |
| 102 | RCL3 | 36 03 |
| 103 | RCL1 | 36 01 |
| 104 | - | -45 |
| 105 | × | -35 |
| 106 | RCL0 | 36 00 |
| 107 | 1 | 01 |
| 108 | . | -62 |
| 109 | 3 | 03 |
| 110 | × | -35 |
| 111 | 2 | 02 |
| 112 | 8 | 08 |
| 113 | 0 | 00 |
| 114 | 0 | 00 |
| 115 | X⇄Y | -41 |
| 116 | - | -45 |
| 117 | ÷ | -24 |
| 118 | RCL1 | 36 01 |
| 119 | X⇄Y | -41 |
| 120 | - | -45 |
| 121 | STO4 | 35 04 |
| 122 | RCL3 | 36 03 |
| 123 | ÷ | -24 |
| 124 | DSP3 | -63 03 |
| 125 | RTN | 24 |

# TECHNICAL REPORT DATA
*(Please read Instructions on the reverse before completing)*

| | | |
|---|---|---|
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| 7. AUTHOR(S)<br>C. W. Fort, Jr., R. L. Douglas and W. E. Moore | | 8. PERFORMING ORGANIZATION REPORT NO. |
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15. SUPPLEMENTARY NOTES

16. ABSTRACT

A sampling program was undertaken to evaluate the releases of uranium concentrate from uranium mills. This concentrate, known as yellowcake, is the final product of the mill. It is routinely released to the environment through stacks which exhaust air from both the yellowcake drying and packaging operations. During this study, samples were taken in these stacks at six mills to evaluate the total yellowcake emission rates. This paper describes the sampling and analytical methods used, and presents the results obtained. Considerable variation in the emission rates was observed, both between mills and at the same mill over time. A total of 40 emission rate tests were made, with the measured rates ranging from 0.001 lb $U_3O_8$ (0.4 μCi $U_{total}$) per hour to 0.9 lb $U_3O_8$ (220 μCi $U_{total}$) per hour. Data are also presented on the levels of uranium daughter radionuclides (Th-230, Ra-226, Pb-210, and Po-210) found in the yellowcake. The implications of the results of this study for monitoring to meet State and Federal licensing requirements are discussed. Measurements such as these can provide specific and realistic input data to models used to estimate radiation doses to people living in the vicinity of the mill. However, the variations observed indicate the need for relatively frequent and site-specific measurements.

17. KEY WORDS AND DOCUMENT ANALYSIS

| a. DESCRIPTORS | b. IDENTIFIERS/OPEN ENDED TERMS | c. COSATI Field/Group |
|---|---|---|
| Uranium Isotopes | Yellowcake | 1802 |

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