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THE ASSESSMENT OF HUMAN EXPOSURE TO RADIONUCLIDES FROM A URANIUM MILL TAILINGS RELEASE AND MINE DEWATERING EFFLUENT

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Abstract—This study provides an assessment of human exposure to radiation from a river system contaminated by radionuclides of the ^{238}U decay series released through a dam break at a uranium mill tailings pond and by the continuous discharge of dewatering effluent from 2 uranium mines. The *in vivo* analyses of radionuclides in 6 Navajo Indians who lived near the river indicate no detectable elevations above background concentrations. Dose estimates for inhalation of suspended river sediment indicate a maximum annual 50-yr dose commitment of 204 mrem to the endosteum. Estimates of doses (50-yr dose commitments) from the ingestion of livestock range between 1 mrem (to liver) and 79 mrem (to bone) suggest that the major contribution to human exposure is from mine dewatering effluent that has been continuously released into the river system for many years. Although the estimated exposures do not exceed existing state or federal regulations, their magnitude justifies further measurement of radionuclides in animals and in the natural environment and the consideration of strategies to reduce radiation exposure to humans and animals.

INTRODUCTION

THE CLARIFICATION of radiation exposure to humans following the accidental release of radionuclides into the environment is a matter of great concern to the nuclear industry, the public health sector and the general public alike. This paper describes initial efforts to estimate public radiation exposure after a dam break at a uranium mill tailings pond. Investigations of

accidents such as this provide insight into the effects of acute and chronic exposure to industry-related releases of radionuclides.

Early on the morning of 16 July 1979, a breach occurred in the earthen retaining dam of a tailings pond of the United Nuclear Corporation's (UNC's) Church Rock uranium mill. The water (approx. 94 million gal of acidified effluent) and tailings slurry (approx. 1100 tons) spilled through the damaged portion of the retaining wall into the pipeline arroyo that is a tributary to the Rio Puerco River system. The Rio Puerco runs through Gallup, NM (population 18,000), and eventually crosses the New Mexico-Arizona border (Fig. 1). On its way to

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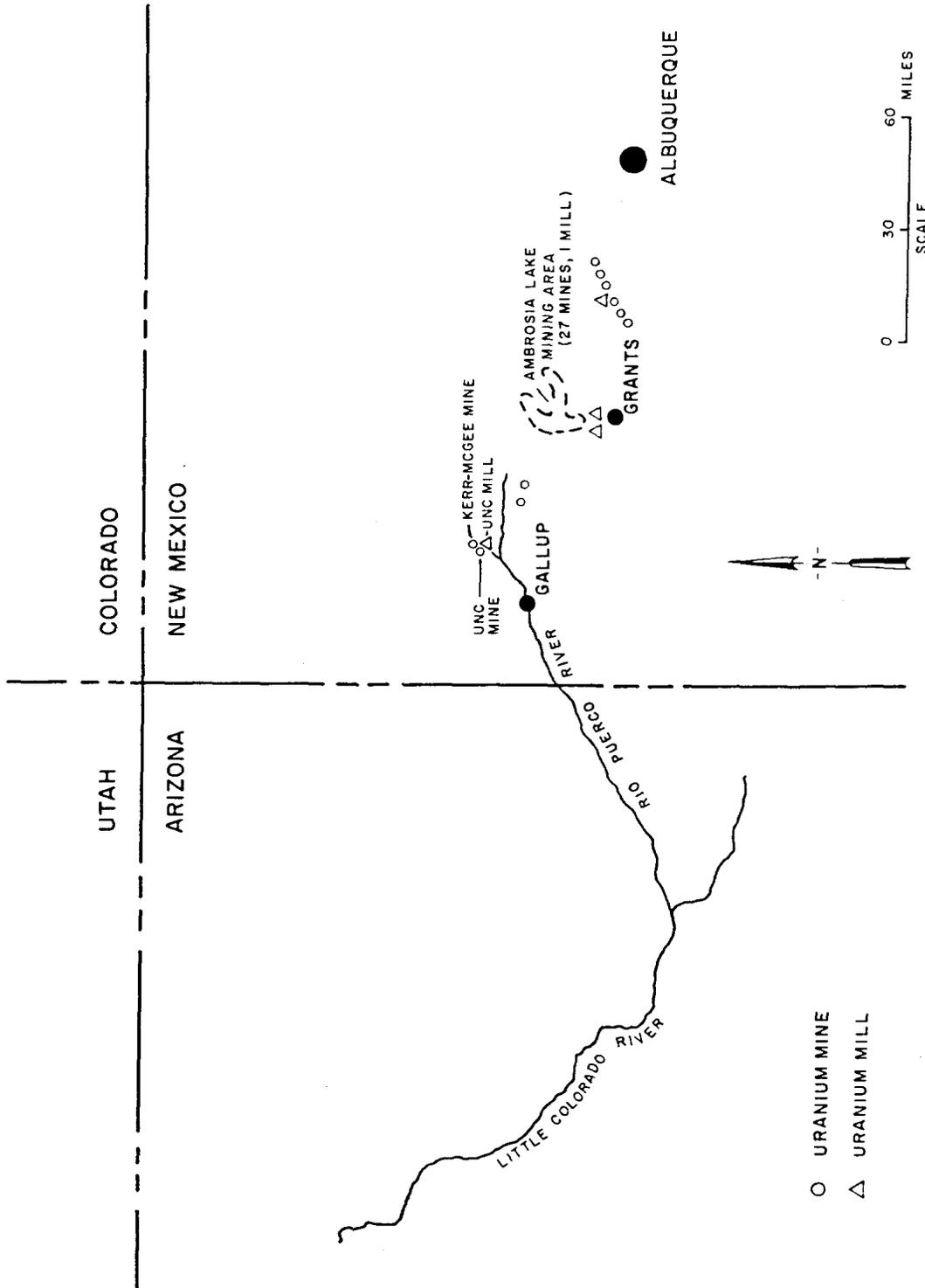


FIG. 1. Location of United Nuclear Corp. mine and mill, New Mexico, 1979.

Gallup, the Rio Puerco and its tributaries pass through land with a checkerboard pattern of ownership, with portions owned or leased by Navajo Indians, private individuals, the Bureau of Land Management, and the state. No other tailings impoundments exist along the Rio Puerco between its junction with the pipeline arroyo and the town of Gallup.

In terms of tailings liquid volume, the UNC spill ranks as one of the largest spills to date (NRC79). The mass of solids released in the slurry appears to be close to the median for accidents of this kind, however. The circumstances that resulted in the dam break have been reported elsewhere (Ru81), and the concentrations of radionuclides released into the environment have been measured by the New Mexico Environmental Improvement Division (NMEID) (report in preparation).

Since 1968, the UNC Church Rock mine, adjacent to the UNC mill, has continuously released dewatering effluent into the pipeline arroyo at a rate of 1400 gal/min. Before 1975 this effluent was not treated; after 1975 it received precipitation treatment for removal of ^{226}Ra . Radionuclides are also released into the river system through the dewatering of the Kerr-McGee uranium mine located 1 mile north of the UNC mill (Fig. 1). During usual mining operations, approx. 3600 gal/min of effluent are released into the pipeline arroyo and then into the Rio Puerco. The continuous release of Kerr-McGee dewatering effluent began in January 1972.

The effluent from both mines has transformed the downstream portion of the Rio Puerco from a sporadically dry riverbed to a continuously flowing stream. Radionuclides in dewatering effluent from both mines have contributed to the current levels of background radiation along the river system. In 1974 the Kerr-McGee mine began ^{226}Ra precipitation treatment of its dewatering releases, but data obtained by NMEID indicate that treatment has been incomplete on many occasions.

An initial review of environmental data collected after the mill tailings spill suggested potential human exposure from inhalation of re-suspended dry tailings and ingestion of domestic animals that watered from the Rio Puerco. Other potential exposure routes were considered

but ruled out because no residents of the Church Rock area (between the mill and Gallup) used Rio Puerco water for personal consumption, and only a few small gardens were adjacent to the river. Contamination of regional groundwater was also evaluated but considered unimportant as test wells showed no chronically elevated radionuclide concentrations in groundwater for the 10 months following the spill. However, data from 1980 and 1981 suggest that some of the UNC tailings ponds have contaminated local groundwater through defects in their linings (NMEID data, 1981). During the initial evaluation of environmental data, it became apparent that radionuclide concentrations in the river water were also influenced by the dewatering effluent from the UNC and Kerr-McGee uranium mines. Because pre-spill background levels of certain radionuclides had not been adequately measured, determination of relative magnitudes of radionuclide contribution was impossible. The data in this report, therefore, reflect the combined effects of the mill tailings accident and the chronic dewatering releases.

METHODS

Atmospheric monitoring

Airborne particulates were collected for radionuclide analysis with a high-volume air sampler located on a bank of the Rio Puerco near Gallup, NM (Fig. 2). The sampler operated continuously between 22 August and 28 September 1979, and between 21 November 1979 and 4 January 1980. The filters (99.9% efficient for 0.3- μm dia. particles) were analyzed for natural U (U-nat), ^{230}Th , ^{226}Ra , ^{210}Pb and ^{210}Po . One 7-hr sample was collected with a cascade impactor to measure the particle-size distribution of the airborne particulates. This sample was taken 13 November 1979, downwind from where a UNC cleanup crew was working (near station 6, Fig. 2), and represents worst-case conditions for occupational exposure.

In vivo analysis

In vivo monitoring was used as a screening technique for evaluating acute radiation exposure from the tailings spill. Aerial photographs and information from representatives of the Church Rock community were used to select

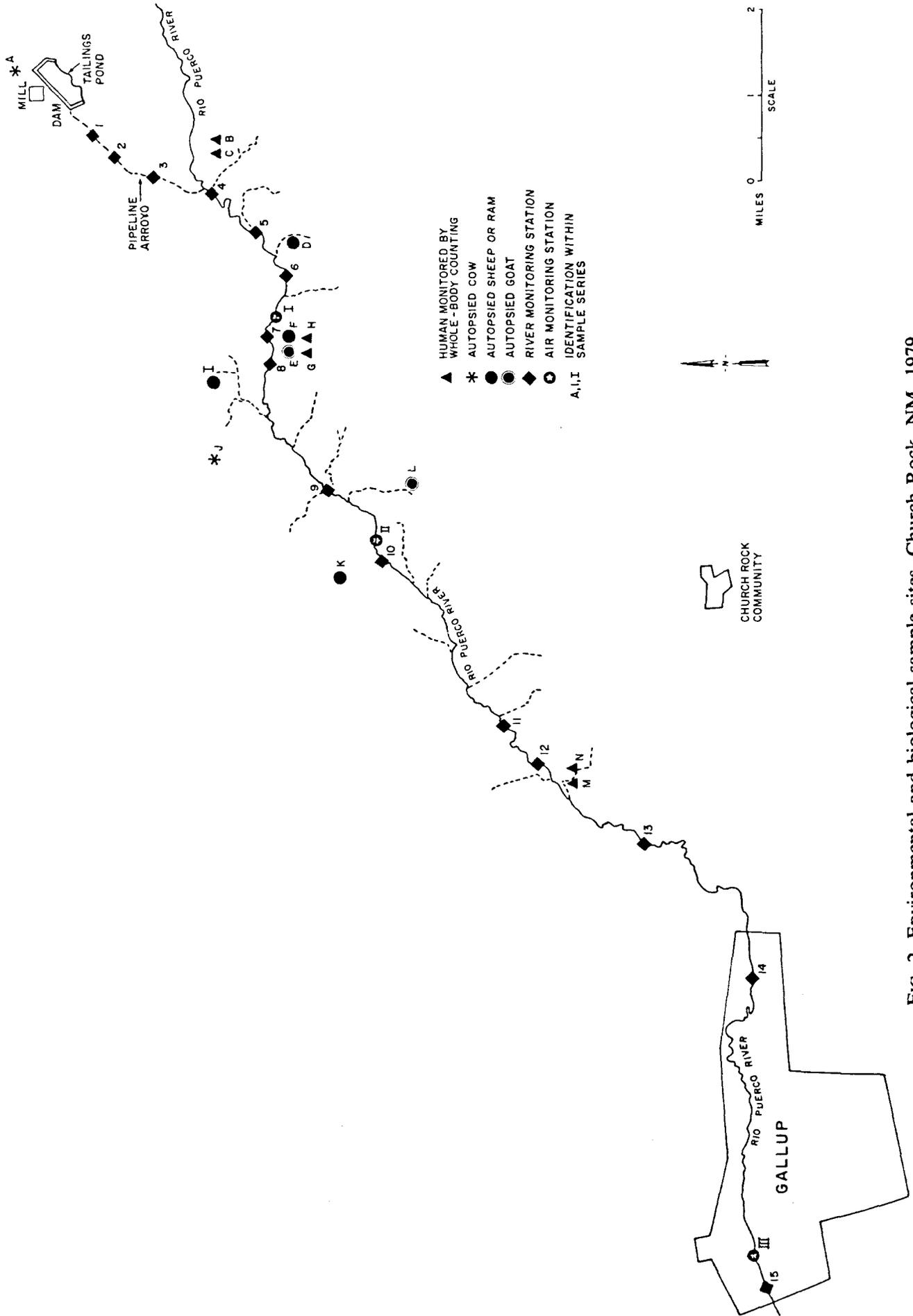


FIG. 2. Environmental and biological sample sites, Church Rock, NM, 1979.

a small group of individuals who were most likely to have been exposed to the tailings contamination and who would consent to *in vivo* monitoring. Only 6 persons (5 children and 1 adult) admitted to being near the banks of the Church Rock portion of the Rio Puerco during the 2 weeks following the spill, and consented to having *in vivo* analyses performed. These individuals underwent whole-body counting for 30 min each at the Los Alamos Scientific Laboratory (LASL) on 23 August 1979. The locations of their residences are shown in Fig. 2.

The γ -spectroscopy counting system used for whole-body counting was housed in a low-background chamber and included the following arrangement of equipment. Twin Phoswich detectors (each composed of a sandwich of sodium and cesium iodide phosphors) were placed over the chest for monitoring low-energy photon emitters such as ^{40}K , ^{137}Cs , U, Th and their daughters. A hyper-pure germanium (HpGe) detector was placed over the liver region for monitoring low-energy photon emitters of the U decay series. A large-volume lithium drifted germanium [Ge(Li)] detector was placed under the subject to monitor the whole-body radiation from energetic photon emitters of the U decay series, ^{40}K and ^{137}Cs .

A spot urine sample was collected from each subject on the day of whole-body counting, and a 24-hr sample on the next day. LASL analyzed these samples for gross α , β and γ concentrations, as well as for concentrations of specific radionuclides of the ^{238}U decay series. Gross γ activity was measured with a 4 \times 4-in. NaI detector over an energy range of 0–2.0 MeV. For each subject, the entire spot urine sample was counted, and a 400-cm³ aliquot of the 24-hr collection was assayed. Each sample was counted for 2000 s. The detection limit of 35,570 counts was calculated as the 99% upper confidence limit of the mean of 2 background determinations and 2 control samples.

Gross β activity was measured with a 2-pi windowless gas flow proportional counter, using planchets that were plated with 0.5-cm³ aliquots of the sample. An energy range of 0.167–1.7 MeV was examined for a counting time of 60 min, with a counting efficiency of 61–71%. The detection limit was estimated as 10% of sample and instrument background

(5000 pCi/l.). Gross α activity was measured by packaging the same planchet samples with a ZnS-coated Mylar film and counting with a scintillation counter at an efficiency of $49.4 \pm 1.1\%$ for 1000 min. The lower limit of detection (LLD) for type I and II errors of 5% for gross α counting was 50 pCi/l.

Total U was analyzed by neutron activation and delayed neutron counting of ^{235}U in 25-cm³ aliquots of each urine sample at the LASL Omega West Reactor Facility. The LLD was 1.4 pCi/l. Thorium was analyzed colorimetrically and radiometrically by using 200-ml aliquots of the 24-hr urine samples. The colorimetric method employed arsenazo-III dye as a chromagen for Th, and has a detection limit of 20 $\mu\text{g/l}$.

The radiometric analysis for specific nuclides involved precipitation of metals from aliquots (125–500 cm³) of urine from 4 individuals by the oxalate method, ion exchange, and subsequent electroplating onto stainless steel planchets. The samples from Church Rock subjects were compared with control samples composed of pooled urine from LASL employees not exposed to radionuclides and with a solution spiked with ^{230}Th and decay products of the ^{232}Th series. The planchets were counted in the α scintillation system described previously. After α scintillation analysis, planchets were counted for 1212 min in an α spectrometer gated for 2.9–7.3 MeV to detect the ^{230}Th and ^{232}Th decay series (excluding ^{212}Po). The LLD for each Th isotope (^{228}Th , ^{230}Th , and ^{232}Th) was 0.02 pCi/l. The 24-hr urine samples were also analyzed for ^{226}Ra by using a de-emanation technique. Aliquots of 200 cc were allowed to sit for 32 days to permit equilibrium between ^{226}Ra and ^{222}Rn . The ^{222}Rn was then isolated and counted for 900 min. The LLD for this technique was 0.3 pCi/l.

Estimation of radionuclide concentrations in animals

Human internal exposure to radionuclides could also result from eating local livestock that watered in the Rio Puerco. Sheep, cattle and goats that grazed along the Rio Puerco and its tributaries were the domestic animals most likely to have been exposed. Several Navajo herds have depended upon this river system as

their sole source of drinking water, even since the dam break.

Two cows, 4 sheep and 2 goats were purchased from the Church Rock area (Fig. 2) between 37 and 71 days after the spill, and autopsied within 1 week of purchase. As controls, 1 cow and 2 sheep were purchased outside the Church Rock area during the same time period. Most Church Rock animals received both acute exposure from the spill and chronic exposure from the mine dewatering effluent. Six of these animals and the 3 control animals were autopsied, and tissue samples of muscle, liver, kidney and bone were removed from each animal. The samples were analyzed for concentrations of ^{238}U , ^{235}U , ^{234}U , ^{232}Th , ^{230}Th , ^{226}Ra , ^{210}Pb and ^{210}Po .

Methods for detecting radionuclides in animal samples

The methodology for measuring radionuclide concentrations was an adaptation of a procedure developed at EPA-Las Vegas for analyzing the naturally occurring ^{238}U series on polystyrene air filters. In this procedure samples (100 g liver, kidney and muscle; 5 g bone) were first wet-ashed with a combination of nitric acid, sulfuric acid and hydrogen peroxide to prevent volatilization of Po and Pb. The bone samples were decomposed with only nitric acid and hydrogen peroxide to avoid the precipitation of large quantities of calcium sulfate. Samples of bone were also boiled in 1.5 l. of water for 8 hr to simulate leaching that might take place when bones are included in soup. After the organic material was destroyed, samples were treated with hydrofluoric acid to dissolve and volatilize silicates, and then treated with hydrochloric acid. Samples were split into 2 equal fractions for sequential U, Po and Th separation by anion exchange and for sequential Ra and Fe separation by precipitation techniques. Before decomposition, all samples were spiked with ^{232}U , ^{208}Po and ^{234}Th tracers and a stable Pb carrier to determine the chemical recoveries of these elements in the separation procedures.

In the anion exchange separation each sample was diluted with 9 M HCl and passed through an anion exchange column, which removed U and Po. Absorbed iron was eluted with HI, and U and Po were sequentially eluted with dilute HCl

Table 1. Detection limits for radionuclide analysis of animal samples, Church Rock, NM, 1979

Radionuclide	Per Sample (pCi)	100 g Tissue (pCi/kg)	5 g Bone (pCi/kg)
U, Th	0.03	0.3	6
^{210}Po	0.3	3	60
^{210}Pb	1	10	200
^{226}Ra	0.3	3	60

and concentrated HNO_3 . The original eluate (containing thorium) was diluted with 7 M HNO_3 and passed through the column again to remove the Th. The Th was then eluted with 9 M HCl. The U and Th fractions were converted to sulfates and electroplated on stainless steel discs. The Po fraction was converted to its chloride salt and spontaneously plated on a nickel disc. Each of the 3 discs was counted on an α spectrometer to measure ^{234}U , ^{235}U , ^{238}U , ^{230}Th , ^{232}Th , ^{210}Po and the ^{232}U and ^{208}Po tracers. Thorium recovery was determined by β counting for ^{234}Th .

Radium-226 and ^{210}Pb were separated by precipitating $\text{Ba}(\text{Ra})\text{SO}_4$ in the presence of HCl and then precipitating PbSO_4 after the chloride anion was removed by evaporation. The concentration of ^{226}Ra was determined by the classical emanation technique. Lead-210 was measured by allowing its ^{210}Po daughter to ingrow for approx. 3 months, spontaneously plating the Po on a nickel disc, and analyzing the disc by α spectrometry. For ^{210}Pb analysis, the Pb recovery was determined by atomic absorption analysis of the added Pb carrier, and the recovery of the ingrown Po was determined by measuring a second ^{208}Po spike added at the time the ^{210}Pb was isolated.

Detection limits for a typical analysis can be estimated from the reagent blank analyses performed during this study. These limits are expressed per sample, per 100 g tissue, and per 5 g bone (Table 1).

RESULTS

Atmospheric monitoring

The high-volume composite air sample data and the dose conversion factors of Dunning *et al.* (Du79) were used to calculate inhalation

doses to those human tissues receiving the highest doses. To provide the most conservative estimates of inhalation doses for the residents of Gallup, all material deposited on air sampler filters was considered respirable. This assumption is supported by data from the single cascade impactor sample, which showed the diameters of all collected particulates to be between $0.3\ \mu\text{m}$ and $3.3\ \mu\text{m}$ —sizes considered respirable by humans. The inhalation rate was assumed to be $15\ \text{m}^3/\text{day}$, and the duration of inhalation, 1 yr; quality factors were selected to give the most conservative dose estimates.

The total 50-yr dose commitments from all radionuclides for this 1-yr exposure (in mrem for clearance Class W, the only clearance class with conversion factors for all nuclides of interest, Du79) were 17, 1, 15 and 4 for bone, spleen, endosteum and lungs, respectively. Thorium-230 contributed the greatest single radionuclide dose to the endosteum, ^{210}Pb the greatest dose to bone, and ^{210}Po to spleen and lungs. The use of clearance class D in calculations for U-nat and ^{210}Po raises the spleen dose to 3 mrem, lowers the lung dose to 2 mrem, and does not change doses calculated for bone and endosteum. The use of Class Y in calculations for U-nat and ^{230}Th changes lung and endosteum doses to 4 and 5 mrem, respectively, and does not appreciably alter other organ doses. It should be noted that the atmospheric concentrations used in the calculation of doses for Gallup residents reflect a combination of contributions from background, mine dewatering effluent, and the mill tailings spill. The similarity of these concentrations to background concentrations measured in other mining and milling areas suggests that the spill material did not contribute significantly to the Gallup radionuclide concentrations.

Human inhalation doses based on cascade impactor data were calculated in a similar manner, but for an occupational exposure period of 40 hrs per week. Fifty-year dose commitments (in mrem for Class W) of 50, 1, 204 and 5 were calculated for bone, spleen, endosteum and lungs, respectively. It should be emphasized that the cascade impactor data reflect high dust loading ($794\ \mu\text{g}/\text{m}^3$) and may result in an overestimation of exposure under realistic conditions. The ^{230}Th doses to bone and endosteum

were much higher than doses from any other radionuclide, while doses to other organs were similar from all radionuclides that were measured. The magnitude of the inhalation dose from ^{230}Th depends on the solubility of this radionuclide. If clearance Class Y (indicating less rapid physiological clearance) is used instead of Class W for the ^{230}Th dose calculations, doses to the endosteum and bone are much lower, and the lung dose is much higher.

In vivo analysis

All 6 subjects who underwent whole-body counting were found to have normal amounts of radioactivity, primarily ^{40}K . LASL specifically searched for radionuclides of U, Th and their daughters and found no detectable activity of these isotopes in any of the subjects. Because negative spectroscopy results indicate only the absence of concentrations above the counting system's limits of detection, these limits were computed for the deposition of ^{230}Th in the lung. The ^{230}Th detection limit was then converted to the smallest organ dose detected by the counting system. This minimum detectable dose, therefore, is the largest dose that could be incurred but not detected. LASL estimated the limit of detection for ^{230}Th deposited in the lungs of a 10-yr-old child to be 10 nCi and calculated a minimum detectable dose rate to lung of 4.0 rem/yr and a minimum detectable total lung dose of 7.9 rem. The lung dose from ^{230}Th was assumed to be the highest organ dose expected from an acute exposure to radionuclides from the tailings pond spill.

In all urine samples, gross α , β and γ activities were found to be below the detection limits of the analytical systems. Likewise, concentrations of total uranium and thorium were below detection limits. When the net α count for each radionuclide was compared with the specific detection limit, all samples from subjects and controls were below the limits; however, when total dis/min were compared, 2 of the 4 samples exceeded the detection limit, with count rates of 0.15 and 0.18 dis/min. Neither of the control samples exceeded the limit of detection. The net count data from α spectroscopy analysis suggest that ^{224}Ra , ^{228}Th and ^{216}Po may be responsible for these elevations. These radionuclides are components of natural background, and are not

specifically associated with U mining and milling.

The limit of detection for ^{230}Th in urine can be used to estimate the associated minimum detectable dose rate. The dose rate was calculated for a 10-yr-old child with a daily urinary output equal to the average for the Church Rock subjects (709 ml/24 hr) and an exposure to ^{230}Th that ceased 30 days before the urine was collected (the assumption that provides the most conservative dose estimate). It was also assumed that Th was excreted in a manner similar to Pu. Under these circumstances, radiometric analysis could detect internal deposition of ^{230}Th that would give a dose rate of 127 mrem/yr to bone surface and 25.3 mrem/yr to total bone mass.

Only in 1 Church Rock urine sample did the activity exceed the detection limit for ^{226}Ra . Interestingly, in 4 of the 6 Church Rock samples, the activities exceeded the mean of the control samples used to calculate the detection limit of this radionuclide.

Radionuclide concentrations in animals

Radionuclide concentrations in edible samples from animals were corrected for reagent blank activity and are presented in Table 2. The fact that radionuclide concentrations were measured in only 11 animals precludes definitive statements on representative concentrations for the animal population in the Church Rock region. The following data and discussion should therefore be regarded as a preliminary assessment of animal radionuclide concentrations and human ingestion exposure. As noted by footnote (k) of Table 2, the ^{226}Ra concentration reported in the muscle of Goat L is considered to be in error. Since the entire original sample had been wet-ashed for the initial analysis, replication of the analysis was impossible. Radon-222 was re-emanated from the bubbler and recounted with essentially the same result. However, it appears this result is anomalous and, for the following reasons, we strongly suspect that the sample was contaminated, either during collection or in the laboratory: (1) this value is more than 50 times higher than the next highest muscle concentration of ^{226}Ra , and (2) this value is more than an order of magnitude higher than the ^{210}Po and ^{210}Pb concentrations in the same sample; in only

1 other sample is Ra higher than Pb and Po, and then only slightly so.

When exposed (Church Rock) and control animals were compared, most radionuclide concentrations in exposed animals were higher than those for controls. In particular, the concentrations of ^{230}Th , ^{226}Ra , ^{210}Pb and ^{210}Po in the exposed animals were consistently higher than those in the controls. The cow from station A (above UNC tailing dam, but downstream from Kerr-McGee and UNC dewatering effluent) had higher concentrations of ^{230}Th , ^{226}Ra , ^{210}Pb and ^{210}Po in the liver and kidney than the other Church Rock cow exposed to effluent from the spill. When radionuclide levels in all edible organs were compared, the kidney had the highest concentrations, and muscle the lowest. Concentrations of ^{210}Pb and ^{210}Po were consistently higher than those of other radionuclides.

Radionuclide concentrations in bone samples are presented in Table 3. Although radionuclides in animal bones do not usually enter the human food chain (with the exception of ingested bone marrow and soup), their levels provide useful data for evaluating pathways by which animals are exposed. Additionally, many radionuclides of the U decay series are concentrated in bone to a greater extent than in other organs and are, thus, more easily detected.

For all radionuclides, exposed cows and sheep had higher bone concentrations than controls. Concentrations of ^{226}Ra in bone exceeded those of ^{210}Pb and ^{210}Po in both exposed and control cows. In sheep, bone concentrations of ^{210}Po were generally higher than those of ^{226}Ra , except for the exposed sheep that watered from the well near the old Church Rock mine. There appeared to be no consistent relationship between ^{226}Ra and ^{210}Po concentrations in the bone samples from the two goats. Lead-210 was detected in only half of the bone samples, and all of these concentrations were less than those reported for ^{210}Po . When radionuclide concentrations in bone were compared by ages of the animals, ^{210}Po and ^{226}Ra appeared to be directly related to age for cows and sheep.

Estimation of human ingestion doses

Radionuclide concentrations in animal organs (Table 2) can be used to estimate human radiation doses that result from eating these

Table 2. Corrected^a radionuclide concentrations in samples of edible animal organs, Church rock, NM, 1979

Animal ^b	Organ	Location ^c	Radionuclide Concentration (pCi/kg) ^d					
			238U	234U	230Th	226Ra	210Pb	210Po
Cow								
Exposed	Muscle	J	0.2*	0.4	ND	ND	ND	6.6
Exposed	Muscle	A	ND	0.2*	ND	ND	ND	16.
Control	Muscle	O	0.1*	0.4*	0.1	ND	ND	2.6
Control	Cube Steak ^e	P	0.1*	0.4	ND	ND	ND	3.2
Exposed	Liver	J	3.5	3.6	0.3	ND	5.4	56.
Exposed	Liver	A	4.4	4.3	1.6	0.8	44.	130.
Control-1	Liver	O	0.3*	ND	0.1	ND	1.1	29.
Control-2 ^f	Liver	O	0.2	0.5	0.9	ND	7.6	25.
Exposed	Kidney	J	11.	11.	0.3*	6.9	56.	260.
Exposed	Kidney	A	3.6	3.1	3.1	9.9	130.	650.
Control	Kidney	O	0.1*	0.1*	0.1	ND	44.	160.
Exposed	Bone (Soup) ^g	A	ND	ND	ND	1.6	0.6	0.6
Exposed	Bone (Soup)	J	ND	ND	ND	ND	0.5	0.3
Control	Bone (Soup)	O	ND	ND	0.1	ND	ND	0.3
Sheep								
Exposed	Muscle	F	0.4	0.9	0.3	ND	4.1	12.
Exposed	Muscle	D	0.2*	0.7	0.2	ND	26.	11.
Exposed	Muscle	K	0.1*	0.1*	ND	ND	ND ^h	0.1
Exposed ⁱ	Muscle	I	0.1*	0.4	0.1	1.3	ND	1.5
Control-1 ^j	Muscle	O	0.1*	0.2	ND	ND	ND	1.3
Control-2	Muscle	O	0.1	0.1*	0.1	ND	ND	0.8
Exposed	Liver	D	2.0	2.1	1.0	9.6	290.	50.
Exposed	Liver	K	1.9	2.1	2.0	0.4	43.	29.
Exposed ⁱ	Liver	I	1.6	1.5	0.1	ND	26.	17.
Control-1 ^j	Liver	O	0.4	0.4	ND	0.1	45.	28.
Control-2	Liver	O	0.1*	0.3	0.1	ND	34.	31.
Exposed	Kidney	D	4.0	5.2	4.4	4.2	710.	460.
Exposed	Kidney	K	ND	3.3	1.9	1.6	28.	150.
Exposed ⁱ	Kidney	I	3.0	2.8	27.	6.7	72.	220.
Control-1 ^j	Kidney	O	0.6	1.1	0.4	ND	80.	200.
Control-2	Kidney	O	0.5	0.8	4.0	1.1	19.	170.
Goat								
Exposed	Muscle	E	0.9	1.3	0.9	1.7	ND	8.5
Exposed	Muscle	L	0.5	1.1	0.3	93. ^k	ND	8.0
Exposed	Liver	L	0.6*	0.5*	1.1	ND	210.	72.
Exposed	Kidney	L	0.8	0.6*	0.1*	ND	470.	540.

^aConcentrations based on gross counts with reagent blanks subtracted; negative corrected concentrations are reported as ND.

^bExposed animals taken from herds known to water at Rio Puerco or pipeline arroyo; control animals taken from areas distant from the Rio Puerco.

^cLocation on Figure 2; O = control animal from area not on map; P = cube steak from Las Vegas market; note that location A is above the UNC dam but downstream from dewatering effluent of UNC and Kerr-McGee mines.

^dConcentrations based on wet weight.

^eAverage between two samples.

^fDuplicate sample.

^gUnits of pCi/soup sample of approximately 1.5 l.

^hSample lost; ²¹⁰Pb concentration estimated from ²¹⁰Pb:²¹⁰Po ratio from appropriate organ.

ⁱAnimal exposed primarily to well water from old Church Rock uranium mine.

^jSheep from area not exposed to Church Rock uranium mining and milling effluent, noted for reportedly high levels of background radiation.

^kThis concentration is considered to be in error. See text for discussion.

*Estimated 3.29 sigma counting error is equal to or greater than the calculated concentration.

Table 3. Corrected^a radionuclide concentrations in animal bone samples, Church Rock, NM, 1979

Animal ^b	Sample Type	Loca- tion ^c	Age of Animal (Years)	Radionuclide Concentration (pCi/kg) ^d					
				238U	234U	230Th	226Ra	210Pb	210Po
Cow									
Exposed	Femur	J	2	72.	82.	30.	640.	72.	300.
Exposed	Femur	A	11	38.	41.	1.2 ^e	2,300.	ND	770.
Control	Femur & Tibia	0	3	0.7	1.2 ^e	33.	330.	ND	290.
Sheep									
Exposed	Unspecified	F	5	27.	27.	9.3	410.	230.	1,000.
Exposed	Femur	D	5	24.	20.	36.	540.	ND	980.
Exposed	Femur	K	2	37.	3.5	22.	580.	ND	670.
Exposed ^f	Femur	I	8	44.	42.	20. e	4,300.	-	1,200.
Control-1 ^g	Femur	0	8	2.9	3.9 ^e	35.	380.	870.	1,800.
Control-2	Femur	0	9	1.0	3.5	18.	180.	32.	760.
Goat									
Exposed	Unspecified	E	1	16.	17.	13.	260.	150.	480.
Exposed	Femur	L	3	5.2	6.3	3.8 ^e	520.	ND	260.

^aConcentrations based on gross counts with reagent blanks subtracted; negative corrected concentrations are reported as ND.

^bExposed animals taken from herds known to water at Rio Puerco or pipeline arroyo; control animals taken from areas distant from the Rio Puerco.

^cLocation on Figure 2; 0 = control animal from area not on map; note that location A is above the UNC dam but downstream from dewatering effluent of UNC and Kerr-McGee mines.

^dConcentrations based on wet weight.

^eEstimated 3.29 sigma counting error is equal to or greater than calculated concentration.

^fAnimal exposed primarily to well water from old Church Rock uranium mine.

^gSheep from area not exposed to Church Rock uranium mining and milling effluent, but noted for reportedly high levels of background radiation.

- Sample lost.

organs. Doses are calculated by assuming particular patterns of organ ingestion and applying factors that convert quantities of ingested radionuclides into doses to various human organs (Ki78; Du79). Table 4 lists human organ doses (50-yr dose commitments) calculated for the annual ingestion of 78 kg/yr of the specified animal organs (estimated average annual consumption of meat by adults) (NRC79), or 130 l. of soup prepared with bones. These conditions were chosen to reflect the highest (most conservative) doses to the human tissues that receive the highest doses from the radionuclides listed in Table 2. Since an individual probably would not consume only 1 organ exclusively for an entire year, doses have also been estimated for the more realistic consumption of animal organs in quantities proportional to each organ's percentage of the animal's total edible weight (Table 5).

The data in Table 4 indicate that calculated ingestion doses to all human organs were generally higher from exposed animals than from controls. Doses from exposed animals varied considerably, however. One should also note that doses calculated for ingested organs from 1

exposed cow and 1 exposed goat were less than those calculated for control organs. The variability between doses calculated for the duplicate samples from 1 cow liver additionally suggests that large differences must be evident before doses from exposed animals can be said to be higher than those from controls. The highest doses calculated for ingestion of a single animal organ were from the kidney. Doses from ingested muscle were generally the lowest, with the exception of the doses calculated for the goat muscle sample whose ²²⁶Ra concentration may be erroneously high.

There seemed to be no consistent relationship among human organ doses that held for all ingested animal organs, although spleen and bone doses were usually higher than other human organ doses. The highest doses to human organs calculated for ingestion of a single animal organ were to the endosteum from goat muscle, to the bone from sheep or goat kidney, and to the spleen from cow kidney. These data (Table 4) also suggested that boiled bones in soup would result in negligible radiation doses to organs of human consumers.

When raw data for ingestion doses are stud-

Table 4. Human organ doses^a (50-yr dose commitment in mrem) calculated for animal tissue ingestion, Church Rock, NM, 1979

Animal ^b	Organ	Location ^c	Total Body	Bone	Liver	Kidney	Endosteum	Spleen
Cow								
Exposed	Muscle	J	0.5	0.6	0.9	4.9	0.4	8.3
Exposed	Muscle	A	0.7	0.7	2.0	11.	0.3	19.
Control	Muscle	O	0.3	0.5	0.5	1.9	1.3	3.2
Control	Cube Steak ^d	P	0.3	0.5	0.5	2.4	0.2	4.1
Exposed	Liver	J	7.4	16.	8.5	42.	9.1	70.
Exposed	Liver	A	35.	85.	24.	100.	61.	170.
Control 1	Liver	O	2.0	3.3	4.0	21.	3.2	36.
Control 2 ^e	Liver	O	5.9	14.	5.1	19.	13.	31.
Exposed	Kidney	J	58.	140.	42.	200.	160.	330.
Exposed	Kidney	A	120.	280.	100.	490.	290.	820.
Control	Kidney	O	32.	78.	25.	120.	37.	200.
Exposed	Bone (soup)	A	2.5	6.8	0.2	0.6	27.	0.9
Exposed	Bone (soup)	J	0.3	0.9	0.1	0.2	0.8	0.4
Control	Bone (soup)	O	ND	ND	0.1	0.2	0.4	0.4
Sheep								
Exposed	Muscle	F	3.4	8.4	2.7	9.3	8.2	15.
Exposed	Muscle	D	16.	43.	4.6	10.	22.	14.
Exposed	Muscle	K	0.1	0.1	ND	0.1	0.1	0.2
Exposed ^f	Muscle	I	1.9	4.8	0.6	1.2	22.	2.0
Control 1 ^g	Muscle	O	0.1	0.2	0.2	1.0	0.1	1.6
Control 2	Muscle	O	0.1	0.2	0.2	0.7	0.8	1.0
Exposed	Liver	D	190.	520.	41.	59.	380.	70.
Exposed	Liver	K	28.	76.	11.	25.	53.	37.
Exposed ^f	Liver	I	17.	45.	5.4	15.	22.	22.
Control 1 ^g	Liver	O	27.	75.	8.6	24.	36.	36.
Control 2	Liver	O	21.	57.	7.8	25.	27.	39.
Exposed	Kidney	D	440.	1,200.	140.	390.	640.	590.
Exposed	Kidney	K	26.	61.	25.	110.	63.	190.
Exposed	Kidney	I	68.	170.	70.	170.	360.	280.
Control 1	Kidney	O	55.	140.	34.	150.	68.	250.
Control 2	Kidney	O	21.	45.	28.	120.	66.	210.
Goat								
Exposed ^h	Muscle	E	3.3	7.9	2.5	6.7	35.	11.
Exposed ⁱ	Muscle	L	120.	310.	5.7	10.	1,500.	14.
Exposed	Liver	L	120.	340.	33.	68.	170.	95.
Exposed	Kidney	L	300.	800.	120.	430.	370.	680.

^aDoses calculated for annual ingestion of 78 kg for specific organs (Du79;NRC79).

^bExposed animals taken from herds known to water at Rio Puerco or pipeline arroyo; control animals taken from areas distant from Rio Puerco.

^cLocation on Figure 2; O=control animal from area not on map; P=cube steak from Las Vegas market; note that location A is above the UNC dam but downstream from dewatering effluent of UNC and Kerr-McGee mines.

^dAverage between two samples.

^eDuplicate sample.

^fAnimal exposed primarily to well water from old Church Rock uranium mine.

^gSheep from area not exposed to Church Rock uranium mining and milling effluent, but noted for reportedly high levels of background radiation.

^h²¹⁰Pb sample lost; concentration of this nuclide estimated from ²¹⁰Pb:²¹⁰Po ratio from appropriate organ.

ⁱThe measurement of ²²⁶Ra in the muscle sample used for this calculation is considered to be in error. See text for discussion.

ND=not detected.

ied, inferences can be made with regard to the radionuclides that contribute the most to total organ dose. For bone, ²¹⁰Pb and ²¹⁰Po are the main contributors, with doses from ²¹⁰Pb usually exceeding those from ²¹⁰Po by a considerable margin. When present, ²²⁶Ra is a substantial contributor to endosteal dose, but many samples in our study had no detectable levels of

this radionuclide. Lead-210 concentrations consistently gave the highest total body doses and ²¹⁰Po and ²¹⁰Pb were the major contributors to liver dose. Concentrations of ²¹⁰Po provided the greatest doses to kidney and spleen. Interestingly, concentrations of ²³⁰Th contributed mainly to endosteal dose and usually to only a small extent.

Table 5. Fifty-year dose commitments (mrem) for ingestion of meat in proportion to percent of total edible weight^a, Church Rock, NM, 1979

Animal	Location ^b	Total Body	Bone	Liver	Kidney	Endosteum	Spleen
Exposed							
Cow	A	3.8	8.3	4.4	22.	7.2	38.
Cow	J	1.7	3.5	1.9	9.4	3.5	16.
Sheep	D	28.	79.	8.1	18.	44.	26.
Sheep	I	3.5	9.0	1.9	4.6	28.	7.4
Sheep	K	1.5	3.7	0.8	2.9	3.0	4.7
Goat ^c	L	120.	320.	8.6	19.	1,400.	29.
Goat ^d	-	12.	33.	5.6	16.	45.	25.
Control							
Cow	0	1.0	2.1	1.0	4.5	2.1	7.6
Sheep 1	0	2.0	5.1	1.1	4.3	2.5	6.9
Sheep 2	0	1.2	2.9	0.9	3.6	2.8	5.9

^aBased on the assumption that muscle, liver, and kidney are consumed in proportion to their contribution to total edible body weight (94.8%, 3.4%, and 1.7%, respectively). Meat ingestion is assumed to be 78 kg/yr.

^bLocation on Figure 2. 0 = control animal from area not on map.

^cThe measurement of ²²⁶Ra in the muscle sample used for this calculation is considered to be in error. See text for discussion.

^dAlternate calculation based on muscle ²²⁶Ra concentration for goat from location E, and other radionuclide concentrations for goat from location L.

DISCUSSION

We have presented these data as an example of a strategy for clarifying human radiation exposure after a documented release of radionuclides into the environment. These data provide only a general indication of human exposure and do not constitute a definitive statement on the matter. The response to accidents involving the release of radionuclides into the environment demands this differentiation, because limited time and money prevent detailed studies of all accidents. The results of the screening approach should not replace detailed studies but, rather, should suggest the direction for future studies.

Subsequent to our calculation of doses from environmental data, the conversion factors we employed were revised by their authors (Du81). The revised report includes changes in a systematic error for computation of total body dose, and a revision of dose equivalents based on improvements and corrections in metabolic parameters for ²²⁶Ra and radioisotopes of Th and U. We have compared the new dose equivalents to the ones used in our computations, and find that in most cases, the new values result in only minor changes in our calculated doses, and lead to the calculation of smaller organ doses in most cases. However, the

correction of the total body dose equivalents for ²¹⁰Pb and ²²⁶Ra leads to calculated doses that may be less than the reported doses by a factor of 4, but only for those doses accrued through the ingestion of organs with comparatively high concentrations of these two nuclides.

Radionuclides deposited in river sediment can give doses to humans by inhalation of re-suspended particulates and by ingestion of animals that consumed sediments while watering at the river. Inhalation doses are predicted to be highest in areas where tailings sediment has dried and has been scattered by wind and by activities of humans or other animals. The data for inhalation doses to humans from all radionuclides combined suggest that inhalation results in lower doses than ingestion for all tissues except respiratory lymph nodes (which are not considered to be particularly radiosensitive). If ²³⁰Th is in an insoluble form (solubility Class Y), comparatively high doses may be obtained under conditions of high dust loading. The relatively large contribution of ²³⁰Th to total inhalation dose indicates the importance of atmospheric monitoring in determining acute exposure during the days immediately after uranium mill tailings have been released into the environment. Comparatively low concentrations of ²³⁰Th measured in animal tissues

suggest that the dust from spill materials is not a major source of human or animal exposure.

The whole-body counts and urinalyses of Church Rock residents suggest that in this instance acute human exposure to radionuclides was minimal. Urinalysis for ^{226}Ra in New York residents (Sp73) indicates that these subjects excrete approx. one-tenth of the ^{226}Ra Church Rock subjects excrete ($\bar{x} = 0.15$ pCi/day). However, daily urinary excretion of ^{226}Ra by Brazilians living in areas of high natural background radiation (Pe65) is similar to that of Church Rock residents. Data from areas of the United States with high background levels of radiation indicate urine concentrations of ^{226}Ra that are much higher than those from Church Rock subjects (St56). In the light of these comparisons and the aforementioned detection limits, additional whole-body counting and urinalyses were not recommended for Church Rock residents.

Four cows (2 exposed to mill tailings and 2 controls) from the Anaconda Mill area (approx. 10 miles northwest of Grants, NM) had concentrations of ^{226}Ra , ^{210}Pb and ^{210}Po in edible tissue that were higher than those measured in Church Rock cows but similar to the highest concentrations detected in the Church Rock goats and sheep (Ho79). Rabbits collected in the Anaconda Mill area had concentrations of ^{226}Ra , ^{210}Pb and ^{210}Po in edible tissue that were generally lower than those in Church Rock animals (Ho79). Concentrations of ^{210}Pb and ^{210}Po in rural German cows (with no industrial exposure) indicate levels in liver similar to those in Church Rock cows and levels in kidney lower than those in Church Rock and control cows (Bu79). These reports suggest that radionuclide concentrations in Church Rock animals may come from a combination of environmental routes, including soil, vegetation and the atmosphere.

Chronic exposure from sources other than the tailings spill is supported by the fact that the cow from location A and the sheep from location 1 (Fig. 2) had comparatively high radionuclide concentrations in edible tissues, but these animals were exposed to mine effluent rather than to spill materials. Since cows from Germany and the Anaconda Mill area had concentrations of radionuclides in edible tissue that were comparable with or in excess of those found in edible tissue of exposed animals in

Church Rock, these concentrations apparently can result from an animal's exposure to radionuclides in soil and air alone. The tendency of radionuclide concentrations in bone to increase with an animal's age also suggests that ^{226}Ra and ^{210}Po have been assimilated chronically, rather than from the comparatively short exposure to the tailings spill. Radionuclide uptake from chronic exposure is also supported by calculations that show annual dewatering effluent from Kerr-McGee and UNC mines (with 5 pCi/l. of ^{226}Ra) to contain an amount of ^{226}Ra similar to that released in the tailings spill. Years of chronic exposure to dewatering effluent, therefore, may lead to radionuclide levels in animals that would exceed those expected from the pulse of tailings liquid released in the spill.

The 2 animals with the highest bone concentrations of ^{226}Ra were the cow exposed only to mine dewatering effluent and the sheep that drank from a well on the site of the old Church Rock U mine. These animals also had comparatively high bone concentrations of ^{210}Po —a finding which, along with the ^{226}Ra levels, suggests that chronic ingestion of mine dewatering effluent could lead to the reported radionuclide concentrations in Church Rock animals that drank from the Rio Puerco. When the bone concentrations in cows exposed to Anaconda Mill tailings (Ho79) are expressed in terms of wet weight, these animals are seen to have lower ^{226}Ra concentrations than the Church Rock animals that drank mine dewatering effluent, but higher concentrations than the Church Rock animals exposed to Rio Puerco water contaminated by the spill. The bone concentrations of ^{210}Pb and ^{210}Po were higher in Anaconda cows than they were in any Church Rock animals.

In spite of the likelihood that the mine dewatering effluent is a major route of exposure, we did not clearly identify the primary route by which Church Rock animals were exposed. The clarification of exposure routes has been particularly hampered by the absence of data on concentrations of ^{210}Pb and ^{210}Po in the dewatering effluent of the two mines. More environmental and autopsy data are, therefore, needed to establish the extent of the ingestion and inhalation exposures to both humans and domestic animals.

Regardless of the route of exposure, the main contributions to human ingestion dose appear to come from ^{210}Pb and ^{210}Po and not from ^{226}Ra . The current treatment of mine water before it is released into the river system removes only ^{226}Ra . Before considering efforts toward reduction of ^{210}Pb and ^{210}Po in animals, the relative contributions to animal radionuclide burden of inhalation, of soil and water ingestion, and of foraging should first be clarified. Human intake from food crops or from eating bone marrow should also be more thoroughly evaluated.

A review of state and federal regulations that pertain to the ingestion doses calculated from the Church Rock data indicated that no exposure limits were exceeded by the spill, or through chronic exposure to mine dewatering effluent (IC60; FRC61; EP77; RPB80). Some of the calculated doses were high enough to suggest further monitoring, however. A sampling program designed to clarify the origin of animal radionuclide concentrations has been recommended, and may lead to technically feasible reductions in animal exposure, particularly if mine dewatering effluent is the major source of exposure. We also suggested that Church Rock residents' exposure to radiation can be reduced by eating less liver and kidney from local animals, by not drinking from wells known to contain elevated levels of radionuclides, and by avoiding the banks of the Rio Puerco when the weather is dry and windy. The reported radionuclide levels and human doses, as well as the uncertainty over the sources of animal radionuclide concentrations, underscore the need for thorough radionuclide monitoring and dose estimation both before and during the operation of all nuclear facilities.

Some unique conditions illustrated by the UNC mill accident are discussed in greater detail elsewhere (Ru81). These conditions include the contamination of a river system by both mines and a mill, the siting of tailings ponds near a river system, and the watering of local animal herds with uranium mine dewatering effluent. These situations point to the limitations of "generic" radiobiological impact statements designed to reflect conditions at all uranium mills. They also suggest inconsistencies in the current federal population exposure regu-

lations which exempt uranium mines, exposure from radon and its daughters (including ^{210}Pb and ^{210}Po), and exposure from accidental releases.

The need for a cancer registry of persons exposed through the Church Rock spill came up often during our investigation. This issue has been explored by Rutenber and Kreiss (Ru81) and has been widely discussed in the recent scientific literature (Go75; HB80; La80). Briefly, the usefulness of a registry depends on an accurate assessment of radiation doses for each registrant, the likelihood that these doses are sufficient to produce detectable morbidity or mortality, and a population of adequate size for detecting possible elevations in cancer mortality. Though the first condition may be met for Church Rock residents if an extensive environmental monitoring program is established, the estimated population at risk for exposure is less than 400. In the light of the currently known cancer incidence and mortality risks associated with levels of radionuclides measured at Church Rock and at Gallup, we conclude that the exposed populations are too small for investigators to detect increases in cancer mortality with acceptable levels of statistical power. In fact, it may be misleading to establish a registry with the foreknowledge of a low probability of detecting mortality increases.

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