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**ESTIMATED DOSE TO MAN FROM URANIUM MILLING
VIA THE TERRESTRIAL FOOD-CHAIN PATHWAY**

by

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FOREWORD

Literature pertinent to the movement of uranium-series radionuclides through the terrestrial food chain has been reviewed in this report in an effort to assess the dose received through this pathway by human populations living in the vicinity of uranium mill tailings piles. As part of this effort, the consistency of the data reported by various investigators has been examined, and the adequacy of such data for use in dose estimation has been evaluated.

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ESTIMATED DOSE TO MAN FROM URANIUM MILLING VIA THE TERRESTRIAL FOOD-CHAIN PATHWAY

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ABSTRACT

One of the major pathways of radiological exposure to man from uranium milling operations is through the terrestrial food chain. Studies by various investigators have shown the extent of uptake and distribution of U-238, U-234, Th-230, Ra-226, Pb-210, and Po-210 in plants and animals. These long-lived natural radioisotopes, all nuclides of the uranium decay series, are found in concentrated amounts in uranium mill tailings. Data from these investigations are used to estimate the dose to man from consumption of beef and milk contaminated by the tailings. This dose estimate from this technologically enhanced source is compared with that from average normal dietary intake of these radionuclides from natural sources.

INTRODUCTION

Processing of uranium ore began in the United States during the early 1950s with the establishment of production mills by such companies as Anaconda and Climax.¹ U.S. production of radium from uranium-containing ore predates uranium milling by more than 35 years, the first radium-processing plant having been founded in 1914 at Denver.² The tailings piles produced by these operations are located throughout the country, but most are in the Southwest.

Recently, attention has been directed toward estimating the potential health hazards due to the presence of these exposed tailings, particularly hazards to human populations living in the vicinity of these piles. Among the many considerations has been the contamination of the terrestrial food chain with the long-lived radionuclides of the uranium series: uranium-238, uranium-234, thorium-230, radium-226, lead-210, and polonium-210, all present in uranium tailings.

Several computer codes have been developed to make theoretical predictions of the dose received by man due to radionuclide transport from nuclear facilities. Some of these codes, including AIRDOS-EPA,³ FOOD,⁴ UDAD,⁵ and MILDOS,⁶ propose models of food-chain contamination with uranium-series nuclides as at least part of their overall scheme. In addition, measurements have been made of the parameters that control man's exposure to radionuclides in the uranium decay chain via the meat/milk food-chain pathway, including uptake coefficients (soil to forage plant), transfer coefficients (forage to meat and milk), and conversion factors relating food ingestion by humans to internal dose.

Results of a search of the literature pertinent to the food-ingestion pathway from uranium mill tailings are presented in this report. Data from various investigators have been examined in an attempt to estimate the dose to man from terrestrial food-chain transport of tailings radionuclides as shown in Figure 1. Contamination of this food chain through groundwater, wild game, locally grown vegetables, livestock other than cattle, and direct ingestion of "forage" plants by humans has not been considered because of a lack of data.

The first step in the food-chain movement of tailings radionuclides is the uptake of these radioelements from tailings (or tailings-contaminated soil) to forage plants. Recently, the U.S. Environmental Protection Agency (EPA) has proposed that the Ra-226 concentration in soil at abandoned uranium mill sites not exceed 5 pCi/g subsequent to remedial action such as burial.⁷ At this time, cleanup operations are being outlined by the Department of Energy (DOE), with the intention of performing remedial action on many of the inactive uranium mill tailings sites in the United States within the next several years. The 5 pCi/g concentration has therefore been used in this paper to estimate the amount of radionuclide activity taken up by forage vegetation at such sites. In this, equilibrium with Ra-226 for Th-230, Pb-210 and Po-210 is assumed.

Tracking the transport of radionuclides from tailings to forage plants, and subsequently to cattle and human beings, is complicated by the fact that there are two modes of vegetation contamination: (1) root uptake of radionuclides from tailings, or from soil contaminated with tailings, and (2) foliar deposition and subsequent foliar absorption of airborne radionuclides from tailings, ore, yellowcake, or particulates containing radon decay products. Data are lacking from the latter mode, since plants are generally washed prior to radiochemical analysis. Thus, the concentrations of radionuclides in forage reported in this paper are reflective only of root uptake and any foliar absorption that may take place. The "dusting" effect may, nevertheless, be a significant means of radionuclide transfer in the tailings-to-forage-to-cattle pathway.

Following the accumulation of tailings radionuclides in forage, some plants may be consumed by grazing cattle. It has been conservatively assumed in this report that cattle graze throughout the entire year; however, it is recognized that the grassland ranges in the Southwest sometimes are not available for grazing during the winter or during severe drought. Only a small amount of data characterizing the transfer of uranium-series radionuclides from ingested forage to cattle muscle (beef) and milk has been reported. These data (transfer coefficients) were used in estimating the activity concentrations of tailings radioisotopes in beef and milk as a result of their transfer via the tailings-forage-cattle pathway.

Human intake of these "contaminated" foodstuffs was converted into estimated dose rates to bone and soft tissue using both National Council on Radiation Protection and Measurements (NCRP)⁸ and Oak Ridge National Laboratory (ORNL)⁹ dose conversion factors. Because of the wide variation in the reported values of food-chain transfer parameters (Table 1), minimum and maximum estimates of dose, based on the lowest and highest set of transfer parameters available from the literature, are given for each radionuclide (Table 2). Presented for comparison in Table 2 are estimates of the dose to man from (1) the transport of uranium mill tailings radionuclides through the food

Figure 1. Meat/Milk Food-Chain Exposure to Man.

Table 1. Reported Food-Chain Transfer Factors for Uranium-Series Nuclides:
Tailings-Forage-Cattle-Man Pathway

Isotope	Plant Uptake Coefficient† ¹				Transfer Coefficient† ²			
	Minimum Estimate	Reference† ³	Maximum Estimate	Reference	Minimum Estimate	Reference	Maximum Estimate	Reference
U-238/U-234	10 ⁻⁵	10	0.2	15	1.4 × 10 ⁻⁴ (milk)	17,18	6.1 × 10 ⁻⁴ (milk) 3.4 × 10 ⁻⁴ (beef)	5,24 5,24
Th-230	10 ⁻⁴ † ⁴	11	0.1	13			5.0 × 10 ⁻⁶ (milk) 2.0 × 10 ⁻⁴ (beef)	25 5,24
Ra-226	0.002	12	6.5	16	4.0 × 10 ⁻⁴ (milk) 5.1 × 10 ⁻⁴ (beef)	19 19,20	5.9 × 10 ⁻⁴ (milk) 4.0 × 10 ⁻³ (beef)	20 5
Pb-210	0.08	13	2.8	16	2.6 × 10 ⁻⁶ (milk) 2.9 × 10 ⁻⁴ (beef)	5 5	1.2 × 10 ⁻⁴ (milk) 1.0 × 10 ⁻³ (beef)	19,26,20 19
Po-210	0.03	14	0.2	14	8.9 × 10 ⁻⁵ (milk) 1.2 × 10 ⁻² (beef)	21,22,23 5	2.9 × 10 ⁻⁴ (milk) 5.0 × 10 ⁻² (meat)† ⁵	21,23,27 28

†¹ (pCi/g dry forage)/(pCi/g dry soil).

†² Day/L for milk and day/kg for beef.

†³ References are listed at the end of this report.

†⁴ Reported for Th-232.

†⁵ Reported for reindeer meat.

Table 2. Estimated Dose Rates to Man from Ingestion of Uranium-Series Radionuclides

Isotope	Dose Rate from Food-Chain Transport of Tailings Radionuclides ^{†1} (mrem/yr)		Dose Rate from Normal Dietary Intake ^{†2} (mrem/yr)	
	Minimum Estimate	Maximum Estimate	Minimum Estimate	Maximum Estimate
U-238/U-234				
Bone ^{†3}				
NCRP ^{†5}	6×10^{-4}	90	20	30
ORNL ^{†6}	2×10^{-4}	30	8	11
Soft Tissue ^{†4}				
NCRP	4×10^{-5}	6	1	2
ORNL	3×10^{-5}	5	1	2
Th-230 ^{†7}				
Bone				
ORNL	1×10^{-3}	1		0.2
Soft Tissue				
ORNL	1×10^{-4}	1×10^{-2}		3×10^{-2}
Ra-226 ^{†8}				
Bone				
NCRP	0.6	8×10^3	8	30
ORNL	0.9	1×10^4	10	40
Soft Tissue				
NCRP	8×10^{-3}	90	0.1	0.3
ORNL	1×10^{-2}	1×10^2	0.2	0.5
Pb-210 ^{†9}				
Bone				
Holtzman	0.3	50		1
ORNL	0.4	5×10^2		10
Soft Tissue				
Holtzman	4×10^{-3}	5		0.1
ORNL	6×10^{-3}	7		0.2
Po-210 ^{†10}				
Bone				
NCRP	2×10^2	7×10^3	50	60
ORNL	1	30	0.2	0.3
Soft Tissue				
NCRP	20	7×10^2	5	6
ORNL	1	30	0.2	0.3

Total Dose Rate from U-Series Ingestion				
Bone				
NCRP, Holtzman	200	1.5×10^4	80	120
ORNL	2.3	1.1×10^4	18	62
Soft Tissue				
NCRP, Holtzman	20	800	6.1	8.4
ORNL	1.0	140	1.4	3.0

^{†1} Calculated using transfer factors given in Table 1, 18.1 kg/day dry forage intake by cattle, food ingestion rates of 0.5 L/day (milk) and 0.2 kg/day beef per person, and both the NCRP and ORNL dose equivalent rates as given in References 8, 9, and 29. Concentrations of radionuclides in soil following remedial action of tailings were assumed to be 0.7-2.1 pCi/g (U-238/U-234) and 5 pCi/g (Th-230, Ra-226, Pb-210, Po-210), according to the proposed standards set by the EPA for cleanup (Ref. 7).

^{†2} Average dietary intakes used to make these estimates were 0.7-1.0 pCi/day (U-238/U-234), 0.1 pCi/day (Th-230), 0.7-2.4 pCi/day (Ra-226), 1.3-1.6 pCi/day (Pb-210), and 1.3-1.6 pCi/day (Po-210) (Refs. 8, 29-38).

^{†3} Specifically osteocytes.

^{†4} Represented by gonads.

^{†5} Dose equivalent rates from the National Council on Radiation Protection and Measurements (NCRP) used (Ref. 8).

^{†6} Dose equivalent rates from ORNL used (Ref. 9).

^{†7} No NCRP rate available.

^{†8} Dose from ingested Ra-226 and Ra-222 and daughters (assuming 30% retention of Ra).

^{†9} Dose conversion factors given by Holtzman (Ref. 29) are for Pb-210 only; those from ORNL (Ref. 9) are for Pb-210 and ingrown Po-210.

^{†10} Dose conversion factors given by the NCRP (Ref. 8) are based on Pb-210 intake and retention (with subsequent ingrowth of Po-210); those from ORNL reflect only intake of Po-210.

chain, and (2) normal dietary intake of these same radionuclides in the average diet, using values of radioisotope consumption reported in the literature.*

The estimates of the doses to man have been made in an extremely conservative manner. Particularly conservative assumptions used in making these dose estimates are (1) that the cattle continually graze on rangeland contaminated with tailings and (2) that all beef in the diet of a person living in the vicinity of a tailings pile would come from these cattle. These two assumptions probably lead to an overestimation of the dose to man from food-chain movement of tailings radionuclides.

URANIUM-238/URANIUM-234

Uptake by Forage Plants from Soil and Tailings

Estimates of average uranium content in soil range from 1×10^{-4} to $3 \times 10^{-4}\%$ by weight (0.7 to 2.1 pCi/g).^{39,41} Some of this uranium is assimilated into vegetation, probably as the UO_2^{++} ion,⁴² and measurements have been made of the concentration of this radionuclide in plants growing on "natural" soils.^{43,44} According to Laul et al., the uranium content of various garden vegetables rooted in soil that contained about 2 pCi/g of uranium activity ranged from 5.6×10^{-4} to $<7.0 \times 10^{-4}$ pCi/g.⁴⁴

Much attention has been given to uranium uptake by vegetation growing in uranium-rich locations. Plants exhibiting a high uptake of uranium have been used as "uranium indicators" in botanical prospecting,^{45,49} and the assimilation of uranium by vegetation growing in the vicinity of coal mines, phosphate mills, copper ore beds, the Russian taiga region, and nuclear test sites has been studied.⁵⁰⁻⁵⁹

The absorption of uranium isotopes into edible vegetation has also been examined using soils spiked with known amounts of radioactivity.^{60,63} These studies indicate that the uptake coefficient (the ratio of radionuclide activity in the plant to that in root-zone soil)** is dependent on several variables, including the plant species and the fraction of soil uranium available for root uptake (not ionically bound). The greatest concentrations of uranium in plants tend to be in the roots.

Several studies have focused on the uptake of uranium by forage plants growing on or near uranium mine spoils or mill tailings. [U-238 and U-234 are found in lower abundance in the tailings than are their long-lived daughter

*The composition of a "normal" diet is often specific for a given geographic location. Dietary habits can also vary widely from individual to individual. The ranges of average rates of ingestion of uranium-series radionuclides in the United States (as reported in the literature) were used as the average "normal" dietary intake for this report.

**In the literature, this ratio is also referred to variously as the concentration factor (ratio), uptake factor, bioaccumulation factor, and observed concentration ratio.

products (Th-230, Ra-226, Pb-210, and Po-210) since the mill extraction processes remove about 91% to 93% of the uranium in the ore.]⁶⁴ The uptake coefficients for aboveground plant tissue obtained from these studies vary widely. Moffett and Tellier, Morishima et al., and Dreesen and Marple^{10,65,66} report estimated uptake coefficients ranging from 10^{-5} to 10^{-3} , while Rayno et al. and Winsor and Whicker^{13,15} report higher values, ranging from 0.01 to 0.2.* (Winsor and Whicker also report an average coefficient of 3.3 for phlox, but this seems exceptionally high.) As reported by Whicker,¹⁶ a wide number of parameters affect radionuclide assimilation by vegetation, including climate, soil type, plant structure, and experimental conditions. The effects of one or more of these parameters thus may be responsible for the extreme variation of uranium uptake coefficients reported by various investigators.

In their study of the effects of soil containing elevated concentrations of uranium on the ecological system of the Los Alamos test sites, Hanson and Miera⁶⁷ suggest that the resuspension of soil (by wind or rain spattering, for example) could be an important agent of radionuclide transfer to plants through foliar deposition and possible subsequent incorporation into the plant tissue. This is particularly true in the arid Southwest (where most of the uranium mines and mills are located) since drier soil would be more susceptible to wind erosion.

Concentration in Cattle

U-238 and U-234 concentrations found in grazing livestock (sheep and cattle) have been reported in the literature.^{68,69} However, the actual transfer of these isotopes from forage to grazing animal has not been studied extensively. Data are particularly sparse for beef and dairy cattle, which are strategic links in the food-chain exposure pathway. Some measurements and estimates, however, have been made. The transfer coefficients (pCi/L or pCi/kg in milk or meat per pCi/day intake by cattle) reported are low, with estimates ranging from 1.4×10^{-4} to 6.1×10^{-4} day/L for milk.^{5,17,18,24} The only value for beef found in the literature was 3.4×10^{-4} day/kg.^{5,24} These values are based on an assumed equilibrium of radionuclide intake and excretion.

Estimated Dose to Man: Tailings-Forage-Cattle-Man Pathway

The U.S. Environmental Protection Agency (EPA) has recently recommended that after remedial action, the average radium concentration of soil at a mill-tailings site should not exceed 5 pCi/g for any 5-cm depth.⁷ If the U-238/U-234 content of tailings is assumed to be 7% of that of Ra-226,⁶⁴ then the uranium activity concentration in the soil after remedial action would be reduced to within the range of average background concentrations (0.7 to 2.1 pCi/g),^{39,41} based on an initial Ra-226 concentration in tailings of 280 pCi/g.⁷⁰ Using the lowest (10^{-5}) and highest (0.2) values of uptake coefficients for tailings forage reported above, this range of U-238/U-234 concentrations in soil would result in minimum and maximum concentrations of 7×10^{-6} and 0.4 pCi/g, respectively, in plants supported by that soil.

*All of these uptake coefficients are for dried plant matter, except that from Morishima.¹⁰

According to Comar,⁷¹ a cow consumes 8 to 20 kg/day of dry feed, while Heath et al.⁷² report that a grazing dairy cow ingests an average of 11.3 to 18.1 kg/day of dry pasture forage. Based on an intake of 18.1 kg/day and the uranium concentration values for contaminated forage given above, the range of U-238/U-234 ingested by a cow feeding on this vegetation would be 0.1 to 8000 pCi/day. By employing the previously reported transfer coefficients for milk (minimum of 1.4×10^{-4} day/L, maximum of 6.1×10^{-4} day/L), one can then calculate that these ingestion rates would result in estimated maximum and minimum U-238/ U-234 concentrations in milk of 2×10^{-5} and 5 pCi/L, respectively. Similarly, the transfer coefficient for beef reported above (3.4×10^{-4} day/kg) would result in estimated minimum and maximum uranium concentrations of 4×10^{-5} and 3 pCi/kg, respectively, in meat. According to Fisenne and Keller,⁷³ the human diet includes about 0.2 kg/day of meat and 0.5 L/day of milk; so on the basis of the above estimations, persons living in the vicinity of the mill and consuming these foodstuffs would ingest a minimum of 2×10^{-5} and a maximum of 3 pCi/day of U-238/U-234.

Intake of U-238/U-234 can be translated into an estimated dose to man by use of conversion factors supplied by the National Council on Radiation Protection and Measurements (NCRP)^{8,29} and Oak Ridge National Laboratory (ORNL).⁹ The NCRP conversion factors are based on a quality factor of 10 for alpha particles, and ORNL uses a quality factor of 20.⁹

Based on the NCRP dose-equivalent rates of 12 mrem/yr to bone (specifically osteocytes) and 0.8 mrem/yr to soft tissue (represented by gonads) from an intake of 0.4 pCi/day of U-238/U-234,^{8,29} ingestion of from 2×10^{-5} to 3 pCi/day of these isotopes (as a result of radionuclide transport through the meat-milk food chain from tailings subsequent to remedial action) can be translated into minimum and maximum estimated dose rates to man of 6×10^{-4} to 90 mrem/yr bone and 4×10^{-5} to 6 mrem/yr soft tissue. The dose rates to man can also be estimated using the ORNL dose conversion factors.⁹ The estimates for both sets of conversion factors are presented in Table 2.

It should be noted that the large difference between the maximum and minimum dose-rate values is primarily a result of the marked variability in the reported uranium uptake coefficients for vegetation. This variability, as given in various literature references, is illustrated in Table 1.

Estimated Dose to Man: Normal Dietary Intake

The uranium content of various foodstuffs has been reported in the literature.^{30-32,40,74-77} Dietary items with the highest uranium concentrations are tubers (e.g., potatoes and turnips), fruits and vegetables, shellfish, and certain cooking additives, such as salt, fat, and oil. According to Hamilton, cereals and grains, tubers, fruits and vegetables comprise 83% of the uranium intake through the human diet.³² Estimated "normal" dietary intake of uranium ranges from 1 to 1.4 μ g/day (0.7 to 1.0 pCi/day).^{31,32} Estimates of the dose to man from this daily intake of U-238/U-234 are given in Table 2.

THORIUM-230

Uptake by Forage Plants from Soil and Tailings

Less food-chain transport data are available for Th-230 than for any of the long-lived uranium series radionuclides. The background concentration of this isotope in soil ranges from 0.1 to 0.6 pCi/g.⁷⁸ Comar⁷⁹ and Russell⁸⁰ have stated that thorium is not readily assimilated by vegetation. Most investigations of the uptake of thorium into plants have focused on the most abundant isotope, Th-232, as well as Th-228, one of its daughters.* Bondietti and Sweeton calculated vegetation uptake coefficients for Th-232 ranging from 1×10^{-4} to 7×10^{-3} .¹¹

A few investigators have examined the uptake of Th-230 by plants.^{13,53,82} An uptake coefficient of 0.91 for Th-230 assimilation in kidney beans (*Phaseolus vulgaris*), as obtained by D'Souza and Mistry,⁸² demonstrates that aboveground vegetation can absorb thorium to a greater extent than indicated above. However, the beans were grown in a nutrient solution, and thus any effects of soil-binding on root uptake were excluded. According to Russell and Smith,⁸³ soil-binding may be the primary factor inhibiting the movement of thorium into plants. The small amount of data available indicates that thorium can be absorbed by roots but is not mobile within the plant structure, and thus is not readily translocated to the aboveground biomass (shoots). Of special interest however, are the only available data for Th-230 assimilation into vegetation growing on uranium mill tailings.¹³ These data indicate an uptake coefficient of 0.1 for Indian ricegrass (*Oryzopsis hymenoides*). It may be that the translocation of thorium from soil to plant is strongly species-dependent, but more data are required.

Concentration in Cattle

Few data have been reported describing the transfer of Th-230 from forage plants to cattle meat and milk. Since it is estimated that only 0.03% to 0.6% of ingested thorium is absorbed into the blood from the intestine of a dairy cow,¹⁸ Th-230 transfer coefficients are probably quite low for meat and milk. According to Ng et al.,²⁵ the equilibrium value for milk is about 5.0×10^{-6} day/L. The equilibrium transfer coefficient for meat has been estimated to be 2.0×10^{-4} day/kg.^{5,24} However, there are insufficient data to fully support these values.

Estimated Dose to Man: Tailings-Forage-Cattle-Man-Pathway

To estimate the dose rate to man due to Th-230 transport from uranium tailings via the meat-milk food chain, it was assumed that soil at a mill site would contain not more than 5 pCi/g of Th-230 following remedial action. This is in keeping with the proposed EPA standards,⁷ if one assumes equilibrium between Th-230 and Ra-226. Minimum and maximum estimates of dose rates were made using the transfer factors listed in Table 1 and the grazing and dietary assumptions used in making analogous dose estimates for U-238/U-234 ingestion (Table 2).

*See References 11, 33, 54, 59 and 81.

Estimated Dose to Man: Normal Dietary Intake

The average amount of Th-230 taken in through the normal diet is reported to be about 0.1 pCi/day.^{8,29,84} Much of this may come from root vegetables such as carrots and potatoes, since thorium movement to shoots is slight, as is any subsequent transfer to meat or milk. Specific sources of Th-230 in the diet, however, have not been identified. The estimated doses are given in Table 2.

RADIUM-226

Uptake by Forage Plants from Soils and Tailings

The translocation of Ra-226 into plants has been extensively studied. It has been reported to be one of the natural radioelements most readily incorporated into vegetation.⁴¹

According to Fisenne,⁸⁵ the average concentration of Ra-226 in soil is about 1 pCi/g. In some areas, however, the soil content of this isotope may be naturally or artificially enhanced. Increased concentrations of Ra-226 in soil and its subsequent assimilation by plants has been observed in the Soviet Union,^{53-56,59} India,⁸⁶ Brazil,³³ Iran,⁸⁷ Poland,⁸⁸ and the United States.^{78,85} The areas in the Soviet Union, Brazil, and Iran are considered to have naturally elevated amounts of Ra-226 in the soil, while the sites in India, Poland, and the United States that were studied have been contaminated with this radionuclide through industrial activity.

The specific behavior of radium in vegetation has been examined through laboratory and greenhouse studies. Much of the radium within most (but not all) plants is concentrated in the roots, and to a lesser extent in the leaves.^{82,90} Popova, Kodaneva, and Vavilov^{90,91} have proposed the existence of an "acropetal gradient" of Ra-226 in plants, the concentration in leaves decreasing in successive steps from oldest to youngest foliage.

The uptake of radium from soil by plants is highly dependent on the organic content of the soil, since the organic material tends to bind radium, making it unavailable for root uptake.⁹² Radium uptake by plants is to some extent species-dependent as well. Brazil nuts, for example, are known to concentrate relatively large amounts of Ra-226,^{28,80} although little of this ingested Ra-226 is absorbed through the gastrointestinal tract.⁹³

Ra-226 movement into vegetation growing on uranium mill tailings has been investigated, particularly in relation to vegetative stabilization of tailings piles.^{94,95} Reported plant uptake coefficients for Ra-226, as with the other uranium series isotopes, vary widely, ranging from 0.002 to 6.5.* (The highest coefficient observed, 6.5, was for sagebrush (*Artemisia tridentata*), a common desert browse.)

*See References 12, 13, 15, 16, 65 and 66.

Concentration in Cattle

The uptake of Ra-226 by sheep, pigs, and cattle has been reported in the literature.^{29,96,120,121} Transfer coefficient values for dairy cow milk reported in the literature range from 4.0×10^{-4} to 5.9×10^{-4} day/L while those for beef have been estimated to be between 5.1×10^{-4} and 4.0×10^{-3} day/kg, assuming an equilibrium of radium intake and excretion, and again, based on sparse data.^{5,13,28,96}

Estimated Dose to Man: Tailings-Forage-Cattle-Man Pathway

Estimates of the maximum and minimum dose rates to humans due to ingestion of Ra-226 transported from tailings through the food chain are presented in Table 2. These estimates were made in the same way as for dose rates from U-238/U-234 ingestion. The transfer factors given in Table 1 and the EPA standard of 5 pCi/g of Ra-226 in soil following remedial action were used for the calculations.

Estimated Dose to Man: Normal Dietary Intake

Concentrations of Ra-226 in various foods have been widely reported in the literature.* According to these reports, the foodstuffs that contain the highest Ra-226 concentrations are Brazil nuts, some breakfast cereals, cockles, bakery products, root vegetables, flour (particularly wheat), and eggs.

The average dietary intake of Ra-226 has been estimated for several countries of the world, as well as for various U.S. cities.** Ra-226 ingestion through the diet varies considerably from location to location, a good example of this being the reported threefold difference between Puerto Rican and United States intakes.¹⁰⁴ Even within the United States, dietary Ra-226 intake can vary by a factor of 2 to 3.²⁹

According to Holtzman,²⁹ the intake of Ra-226 from the diet ranges from 0.7 to 2.4 pCi/day. Estimates for dose rates to humans based on this average range of intake are given in Table 2.

LEAD-210

Uptake by Forage Plants from Soils and Tailings

Pb-210 originates from the α -disintegration of the gaseous decay product of Ra-226, Rn-222, and its short-lived daughters. Thus, this radioactive lead isotope is not only present in tailings, but also can be found in soil distant from the pile as a result of the atmospheric dispersion of radon. Vegetative uptake of Pb-210 can, therefore, occur by the same mechanisms involved in uptake of uranium, thorium, and radium (i.e., root uptake and dust suspension of soil, ore, tailings, etc.), plus the additional mode of atmospheric washout and subsequent foliar absorption.

*See References 8, 30, 38, 76, 79, 83, and 97-102.

**See References 8, 29, 73, 76, 83, and 103-110.

Root uptake of Pb-210 has been examined by D'Souza and Mistry using spiked nutrient solutions.^{82,111} These studies indicate that in the absence of soil binding, lead is strongly assimilated by plants, concentrating particularly in the roots. Attempts have also been made to investigate the extent of foliar absorption of Pb-210.¹¹²⁻¹¹⁵ According to Athalye and Mistry,¹¹³ over 90% of foliar-applied Pb-210 (as nitrate solution) was absorbed by tested plants.

One particular plant that has been extensively studied because of its ability to take up Pb-210 (and other radionuclides) is tobacco.¹¹⁶⁻¹¹⁹ These studies have led to investigations of the Pb-210 and Po-210 content of cigarettes and, because of their volatility, the concentration of these nuclides in cigarette smoke.^{119,122,123}

Concentrations of Pb-210 in vegetation growing in various geographical locations are given in the literature.¹²⁴⁻¹²⁷ In the arctic regions, lichens (which grow extremely slowly) are significant accumulators of fallout Pb-210 as well as other radionuclides. Several investigators have examined the contamination of the lichen-reindeer-man food chain with this radioisotope.¹²⁸⁻¹³³ (Pb-210 uptake in this ecosystem has been summarized by Holtzman.)¹³⁴

Despite these numerous Pb-210 uptake studies, the translocation of this radionuclide from tailings to vegetation has not been extensively examined. From the available literature, however, Pb-210 uptake coefficients for plants growing on tailings vary from 0.01 to 2.8,* compared to the currently accepted coefficients of 9×10^{-2} and 6.8×10^{-2} used for predictive modeling.^{5,10,26}

Concentration in Cattle

Calculated values of the transfer coefficient of Pb-210 to milk vary widely, ranging from 2.6×10^{-6} to 1.2×10^{-3} day/L.** Estimates of the transfer coefficient to beef range from 2.9×10^{-4} to 1.0×10^{-3} day/kg, based on little data.^{5,19,26}

Estimated Dose to Man: Tailings-Forage-Cattle-Man Pathway

The minimum and maximum estimates of the dose to human beings from the ingestion of beef and milk contaminated due to the food-chain transport of Pb-210 from tailings, as calculated on the basis of the transfer coefficients listed in Table 1 and assuming a maximum Pb-210 concentration of 5 pCi/g (if Pb-210 is in equilibrium with Ra-226) subsequent to tailings cleanup,⁷ are listed in Table 2.

Estimated Dose to Man: Normal Dietary Intake

Concentrations of Pb-210 in dietary foodstuffs are reported in various sources.^{30,34-37} As with Ra-226, the normal dietary intake of Pb-210 has

*See References 13, 15, 16, 66, and 65.

**See References 5, 19, 26, 135, and 136.

been extensively studied.* Most of the dietary Pb-210 is probably from vegetables and cereals.^{29,38} Data presented by Holtzman indicate that Pb-210 intake in the human diet probably ranges from 1.3 to 1.6 pCi/day.²⁹ Estimates of the dose to man from such Pb-210 intake are given in Table 2.

POLONIUM-210

Uptake by Forage Plants from Soils and Tailings

Po-210, the 138-day half-life decay product of Pb-210, is present in both the atmosphere (attached to dust particles) and the soil.¹³⁹ Thus, as with Pb-210, aerial contamination of foliage is of possible significance. Work done by Popova and Taskaev¹¹² and Athalye and Mistry¹¹³ confirms that Po-210 can be absorbed through aboveground plant tissue and subsequently distributed throughout the plant. Athalye and Mistry indicate that under laboratory conditions, about 30% of Po-210 (nitrate solution) applied to a plant surface is incorporated into the plant.

D'Souza and Mistry indicated that when assimilated through the roots, Po-210 concentrates there rather than distributing more evenly throughout the plant (as apparently occurs following foliar deposition of polonium).^{82,111} Root uptake of Po-210 from soil has been examined in plants growing under both greenhouse and outdoor conditions.** The uptake coefficients obtained from these studies ranged from 1.2×10^{-6} to 0.23. The coefficients were considerably higher for outdoor plants (from 0.03 to 0.23) than for greenhouse vegetation (from 3×10^{-6} to 3×10^{-2}).¹⁴ It is possible that this difference is due to foliar contamination of the outdoor plants with airborne Pb-210/Po-210 (either through dust deposition or rainfall), considered by some to be the principal mode of Po-210 uptake in vegetation.^{21,143}

There is little information concerning the transport of Po-210 from uranium mill tailings to vegetation, but some data are available from Holtzman.²⁹ The author did not measure uptake coefficients, but did report concentrations of 2.8 to 4.1 pCi/g of Po-210 in grass (shoots) growing on tailings piles, compared to a control grass content of 0.7 pCi/g. Moffett and Tellier reported concentrations of Po-210 ranging from 0.03 to 0.13 pCi/g in four species of grass (shoots) growing on tailings.⁶⁵ These values are probably aberrant, since the plants were dry-ashed in a muffle furnace at 750°C, a temperature that would volatilize some of the polonium.

Concentration in Cattle

Transfer of Po-210 from cattle fodder to milk has been studied by McInroy et al. and Watters and McInroy.^{22,27} Apparently, the degree of transfer depends on the chemical form of the ingested polonium. Reported Po-210 transfer coefficients for milk range from 8.9×10^{-5} to 2.9×10^{-4} day/L for PoO_2 and PoCl_4 , respectively.^{21,23,27}

*See References 29, 38, 40, 84, 137, and 138.

**See References 14, 124, 125, and 140-143.

Based on an estimate of 0.05 day/kg given by Garner for reindeer, the transfer coefficient for Po-210 from forage to meat appears to be higher than that for the other uranium-series radionuclides.²⁸ The only transfer coefficient estimate reported for beef, however, is 0.012 day/kg.⁵

Estimated Dose To Man: Tailings-Forage-Cattle-Man Pathway

Minimum and maximum estimates of the dose rate to man from food-chain transport of Po-210 are presented in Table 2. These estimates were made using the data of Table 1 and assuming a concentration limit of 5 pCi/g in site soil after remedial action (if Po-210 is in equilibrium with Ra-226).⁷

Estimated Dose to Man: Normal Dietary Intake

The Po-210 content in a variety of common foods is reported in the literature.^{30,34,35,144} As with Pb-210, it appears that most dietary intake of Po-210 is from cereals and vegetables.^{29,38} Although some meats and seafoods contain concentrations of Po-210 similar to those of cereals and vegetables, the former foodstuffs are consumed in smaller quantities. An estimate of the range of dietary intake of Po-210 is reported as 1.3 to 1.5 pCi/day.¹²⁸ Maximum and minimum dose estimates are given in Table 2 for this range of daily Po-210 intake.

However, most of the dose from Po-210 results from that which grows in from Pb-210 present in the body, not from intake of external Po-210.¹⁴⁵ In Table 2, the dose rates due to Po-210 intake in a normal diet, using NCRP dose-conversion factors, are based on intake and retention of Pb-210 and subsequent ingrowth of Po-210. These estimated dose rates are much higher than the analogous estimates made using ORNL dose-conversion factors, which were based solely on "normal" dietary intake of Po-210.

SUMMARY

An effort was made to relate radioactive contamination of foodstuffs near uranium mills to radiation doses to people living in the vicinity of those mills by tracking radionuclide transfer through successive levels of the food chain and then converting the estimated amounts of contaminants ultimately consumed by man into dose equivalents by use of NCRP and ORNL dose conversion factors. The major portion of this effort consisted of an extensive literature search to glean data on food-chain uptake and transport coefficients for long-lived nuclides in the uranium series. Because of the lack of information in the literature, this process could be carried out only for direct uptake/consumption of radionuclides from mill tailings piles through the crucial "forage vegetation → cattle → meat/milk → man" pathway (Table 1).

The doses to man estimated in this report are considered highly conservative for the meat/milk pathway. However, these estimates did not take into account any other food-chain exposures attributable to mill tailings (e.g., drinking of contaminated water or consumption of locally grown vegetables contaminated with tailings nuclides). Even for the beef/milk pathway, the available data on uptake and transfer coefficients (particularly the former) were so variable (ranging over several orders of magnitude in some cases) and the methods by which they were determined were so inconsistent,

that a minimum and maximum range of possible doses to man were calculated (Table 2). Estimated doses from average total dietary intake of the same radionuclides were presented for comparison.

Based on the data in Table 2, the minimum dose rates from food-chain transport of U-238/U-234, Th-230, Ra-226, and Pb-210 in tailings are below the expected dose from a normal diet. However, the minimum dose rates from food-chain transport of Po-210 in tailings exceed that predicted for normal dietary intake. The primary reason for this is that the transfer coefficients (meat) reported for Po-210 are higher than for the other uranium-series radionuclides discussed in this paper (see Table 1). The maximum estimates for dose rates from food-chain transport of tailings radionuclides in nearly all instances exceed those expected via a normal diet, substantially so for Po-210, as can be seen in Table 2.

The great variability in the vegetation uptake coefficients reported in the literature was a principal reason for the wide range of the final minimum and maximum estimated doses to man. Among the many factors that influence radionuclide uptake into plants are plant species, composition of soil, and climate. In addition, the methods used to determine uptake coefficients can have considerable bearing on the findings. For example, greenhouse studies of the intake of uranium-series radionuclides by plants do not account for "out-door" effects such as tailings resuspension by wind and rain, and the deposition of radon progeny on foliage. Even in reported determinations of the uptake coefficients of field vegetation, these effects are often neglected.

The uptake of uranium series radionuclides specifically from uranium mill tailings by forage has not been extensively studied. The small amount of data available from the literature, however, indicate that uptake coefficients for plants growing in tailings or soil contaminated with tailings are higher than those for other vegetation. This is possibly a result of dusting of above-ground foliage by windblown ores, tailings, yellowcake or radon decay products, followed by incorporation of the radionuclides into the plant structure. The effect has not been verified by measurements, however.

In summary, the dearth of field data characterizing forage-plant assimilation of uranium-series radionuclides from mill tailings, as well as the high degree of variability in the data that are available, makes it difficult to predict the dose to man due to meat/milk food-chain transport of these isotopes. Data pertinent to other exposure pathways, such as consumption of locally grown vegetables contaminated with tailings nuclides and drinking of contaminated water, are even more scarce. Thus, there is at present insufficient information available to completely assess the consequential dose to man as a result of food-chain contamination by radionuclides from mill tailings.

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