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Polonium-210 and Lead-210 in Food and Tobacco Products: A Review of Parameters and an Estimate of Potential Exposure and Dose

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Health and Safety Research Division

POLONIUM-210 AND LEAD-210 IN FOOD AND TOBACCO PRODUCTS: A REVIEW OF PARAMETERS AND AN ESTIMATE OF POTENTIAL EXPOSURE AND DOSE

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ABSTRACT

Food-chain transport of Pb-210 and Po-210 from soil to edible plant parts and from animal feed to meat and milk were evaluated from a review of literature. The degree of transfer was characterized by estimating concentration factors (unweighted arithmetic means) as well as the transfer coefficients B_v, B_r (unweighted geometric means), f_m and f_f (unweighted arithmetic means). Global dietary intake of Pb-210 and Po-210 was also summarized, and 50-year dose estimates to target organs calculated. The greatest estimated ingestion doses were those to populations with large dietary complements of animal protein in the form of seafood (Japan) or caribou/reindeer muscle and organ meats (Arctic Eskimos and Lapps). The magnitude of this latter source illustrates the importance of simple food chains in generating significant exposures to populations dependent upon them.

The origin and magnitude of inhalation exposure and dose from tobacco products was also assessed. For the majority of internal organs evaluated, the dose resulting from smoking commercially available tobacco products is comparable to or greater than the dose estimates for ingestion of naturally occurring dietary Pb-210 and Po-210.

1. INTRODUCTION

The mining and milling of phosphate ores in Southwest Florida has produced large volumes of processed ore and tailings materials that contain quantities of the naturally occurring isotope U-238 ($T_{1/2} = 4.5 \times 10^9$ y) (Kocher, 1977). This isotope and the principal members of its decay series, Ra-226 ($T_{1/2}$ = 1600 y), Pb-210 ($T_{1/2}$ = 22.3 y), and Po-210 [$T_{1/2}$ = 138.4 d (Kocher, 1977)] have been identified as potential sources of radiation exposure to local residents via food ingestion, and have therefore been chosen as candidates for evaluation. The transport and presence of Ra-226 in drinking water and food has been assessed in a previous report by the author and her colleagues (Watson, Etnier, and McDowell-Boyer, 1983); Pb-210 and Po-210 will be addressed in the current evaluation.

2. Pb-210 AND Po-210 SOURCES

Lead-210 and Po-210 are directly produced during radioactive decay of the parent radionuclide Ra-226; which is, in turn, a daughter of U-238 decay. Lead-210 and Po-210 are found at elevated concentrations in association with a Ra-226 source, but are often detected at some distance from supposed sites of release due to the extreme mobility of the noble gas Rn-222 [$T_{1/2}$ = 3.8 d (Kocher, 1977)], an intermediate product of Ra-226 decay. The rapid decay of Rn-222 in the atmosphere generates Pb-210 and Po-210, which are adsorbed by aerosols and returned to earth as surficial deposition or rainout. The degree of potential long-range atmospheric transport in North America has been previously demonstrated for Rn-222 (Travis et al., 1979). Other intermediates in the decay series between Ra-226 and Po-210 exist, but are all relatively short-lived and are thus not important to food-chain transport.

Radium-226 is found in the presence of uranium-bearing ores such as in phosphate and uranium mines. It and its daughters also occur naturally in other minerals (shale, coal, granite, and lead), soil, geothermal sites, and well water (particularly those draining granitic aquifers). Technologically enhanced sources of these isotopes, i.e., from an "activity not expressly designed to produce radiation" (Gesell and Prichard, 1975) include operation of coal-fired power plants, combustion of gasoline containing anti-knock compounds composed of lead, and application of phosphate fertilizers to agricultural soils (Moore, Martell, and Poet, 1976; Jaworowski and Grzybowska, 1977; Jaworowski and Kownacka, 1976; Eisenbud and Petrow, 1964; Breslin and Glauberman, 1970; Jaworowski, Bilkiewiez, and Zylicz, 1971; Moore and Poet, 1976). Human intake of Pb-210 and Po-210 from these sources via food and tobacco products will be the focus of this report.

3. ROUTES OF UPTAKE

3.1 FOOD

Ingestion of food is considered the single greatest source of naturally occurring lead among individuals consuming a modern, Western-style diet in a major metropolitan area. In a study of New York City residents, as much as 85.7% of total daily Pb-210 intake was supplied by food when compared with intake via air (12.9%) and water (2.9%) (Bogen, Welford, and Morse, 1976). Inhalation of cigarette smoke was not considered in their survey. Similar results were obtained in a controlled diet study of 12 institutionalized adult males in Illinois (Spencer et al., 1977). When ingestion of food/water, inhalation of air/cigarette smoke and decay of inhaled Rn-222 were all taken into consideration, 78.1% of the total daily Pb-210 intake was estimated to be supplied in food, 69% in water, 5.0% in air, and 0.4% by Rn-222 decay. Cigarette smoke inhalation supplied the remaining 9.4% (Spencer et al., 1977).

Food ingestion also represents the major source of naturally occurring Po-210 intake. Additional data collected by Spencer and her colleagues (1977) indicates that 77.3% of an adult male's daily Po-210 intake is supplied by food, 4.7% by water and 0.6% from air. Inhalation of cigarette smoke provides more Po-210 (17.1%) than drinking water and air combined. A similar distribution was observed in a market basket survey of typical food items comprising an adult diet in Bombay, India (Khandekar, 1977). With no correction for cigarette smoking sources, food in Bombay was estimated to supply 86.4% of the total daily adult Po-210 intake, while 9.3% was supplied by water, and 4.3% by inhalation.

Concentrations of Pb-210 and Po-210 in food categories of mixed diets consumed in cities of the U.S.A., U.S.S.R., southern India, and Japan are summarized in Table 1. Lowest values were observed in milk and dairy products, which do not comprise a major portion of the non-Western diet. The greatest values were determined for Pb-210 and Po-210 in the meat/fish category of Finland, Japan, and Kalpakkam, India (Kauranen and Miettinen, 1969; Okabayashi et al., 1975; and Iyengar et al., 1980). Animal protein utilized in these regions is comprised of caribou and seafood, respectively. Maximal values such as these are illustrative of the role that local availability and ethnic/cultural preferences can

Table 1. Average concentrations (pCi/kg fresh weight) of naturally occurring Pb-210 and Po-210 in some common food categories[a]

Country/City	Cereals/ grains		Leafy vegetables		Other vegetables		Potatoes		Grain products		Milk/milk products		Meat/ fish		Citation
	Pb-210	Po-210	Pb-210	Po-210	Pb-210	Po-210	Pb-210	Po-210	Pb-210	Po-210	Pb-210	Po-210	Pb-210	Po-210	
U.S.A.															
New York City	0.8	-	-	-	0.7	-	1.5	-	1.6	-	0.3	-	1.2	-	Morse and Welford, 1971
U.S.S.R.															
Rostov-on-Don	5.7	4.0	-	-	3.5	1.5	6.2	3.1	2.2	1.7	0.6	0.2	4.7	6.0	Ladinskaya et al., 1973
India															
Bombay	-	1.4	-	5.7	-	1.6	-	0.4	-	-	-	0.4	-	8.0	Khandekar, 1977
Bombay	3.5	-	-	-	-	-	-	-	-	-	-	-	-	-	Lalit, Ramachandran, and Rajan, 1990
Kalpakkam	6.3	9.1	44.5	6.3	3.3	4.5	3.9	3.2	-	-	-	-	10.8	68.0	Iyengar et al., 1980
Finland															
Inari, Lapland	-	-	-	-	-	-	-	-	-	-	-	-	5.9 to 674	122.7 to 2773	Kauranen and Miettinen, 1959
Japan															
National composite	1.5	-	17.0	-	1.6	-	0.7	-	4.2	-	0.3	-	91.1	-	Takata, Watanabe, and Ichikawa, 1963
Chiba and Okayama Prefecture	-	1.1	-	3.5	-	-	-	0.8	-	3.5	-	-	-	242.4	Okabayashi et al., 1975
Tokyo	5.0	-	10.8	-	3.0	-	2.7	-	-	-	0	-	40.1	-	Kametani et al., 1981

[a] Unweighted means.

make in determining the quantity of radionuclides ingested; the degree of dietary exposure is a direct consequence of the consumption rates for each category in Table 1.

In regions of harsh climate, where the food supply is governed by the productivity of simple food chains, the concentration of Pb-210 and Po-210 available for consumption may exceed "normal" U.S. rates by more than two orders of magnitude. Such is the case in the Arctic, where the lichen-caribou foodchain is exploited by native hunters and their families, who consume muscle and organ meats supplied by their herds (see the Pb-210 and Po-210 values for meat consumed by Lapp herders, Table 1). Polonium-210 in muscle from winter-killed caribou has averaged 248.3 pCi/kg; muscle from summer-killed caribou has contained 85.3 pCi (Holtzman, 1966a). These values illustrate a second point; human dietary content may be highly seasonal.; In this case, lichens, upon which Pb-210 and Po-210 are deposited as natural fallout, possess a roughened surface from which deposited particles are not readily removed by wind and rain, This feature, coupled with the fact that lichens are slow-growing perennials, permits these species to passively accumulate elevated concentrations of dust. Caribou and reindeer graze almost exclusively on lichens during the winter; summer pastures provide a mixed forage of annual grasses and sedges as well as lichen. Annual species are less effective accumulators than lichens.

Kalpakkam is a coastal town on the Bay of Bengal, where fishing is a commercial activity. It is also an area of high natural radiation background, as the beach sands containmonazite. This factor, plus the understanding that unsupported Po-210 (i.e., Po-210 in the absence of its parent Pb-210) is preferentially taken up by marine organisms, is thought to account for the elevated fish muscle values found at this site (Iyengar et al., 1980). The possibility of surface deposition from the monazite sands is also a likely explanation for the high levels of Pb-210 found associated with leafy vegetables at this site.

3.1.1 Distribution and Uptake in Edible Crops

These two isotopes enter the human food chain via two mechanisms: (1) plant uptake from soil and/or water, and (2) particle deposition onto plant surfaces. The Pb-210 and Po-210 content of edible foliage and reproductive structures (seeds, berries, fruits, etc..) may be the result of either one or both mechanisms. Careful studies which discriminate between each source are rare. Actual root uptake and translocation into edible portions of plants used as human food has been observed to equal <1%. The dominant mechanism is considered to be surface deposition of particulate matter on which Pb-210 and Po-210, originating as decay daughters of Rn-222 gas, are adsorbed (Hill, 1960; Berger, Erhardt, and Francis, 1965). The overwhelming source of plant Po-210 is the deposition of its precursor, Pb-210, on plant surfaces during rainfall events (Francis, Chesters, and Erhardt, 1968).

The degree of soil-to-plant uptake of any nuclide may be characterized by mathematical expressions involving several parameters. The soil-to-plant concentration factor (CF_{sp}) is defined as the unitless ratio of fresh weight specific activity in plants to dry weight specific activity in soil at harvest or equilibrium (McDowell-Boyer, Watson, and Travis, 1980). Further discrimination into edible plant parts permits evaluation of the ratio for vegetative and reproductive organs, i.e., B_v

(ratio of soil-to-plant transfer for vegetative portions of food crops and feed plants) and B_r (ratio of soil-to-plant transfer for reproductive and storage organs of food crops and feed plants) (Baes et al., in review). Reasonable values for these parameters are needed for operation of equilibrium food-chain models to estimate potential exposure when site-specific transfer factors are unknown (Ng, 1982). Since only a very few published studies of Pb-210 and Po-210 contain data appropriate for estimating transfer coefficients for these specific isotopes, the CF_{sp}, B_v and B_r values reported here (Tables 2 and 3) are element-specific and include data on several isotopes.

These parameters are thought to be lognormally distributed (Ng, 1982); thus, geometric means are an appropriate statistic by which to summarize results. Both arithmetic and geometric means are presented in Tables 2 and 3. These estimates also represent an average over a range of soil and plant conditions; soil pH and organic/phosphorus content, plant species, and the physiologic condition of the plant are all known to govern uptake and translocation (Dedolph et al., 1970; John and Van Laerhoven, 1972a; Menzel, 1965; MacLean, Halstead, and Finn, 1969; Rains, 1971; Cox and Rains, 1972; John, 1972; Zimdahl and Foster, 1976; Koeppe, 1977). To determine a more accurate estimate of soil-plant transfer, Tables 2 and 3 include values derived from only those studies in which aerosol contributions were considered insignificant. Values are presented by main crop type as recommended by Ng (1982), who considers a single soil-to-plant transfer factor for all plant material as unnecessarily adding to the uncertainty already inherent in the summarization process.

Table 2. Parameters representing soil-plant transfer of Pb-210

Transport mode	Mean CF_{sp}[a] ($\times 10^{-3}$)	Number of derived values	Range ($\times 10^{-3}$)	Mean B_v[b] ($\times 10^{-3}$)	Mean B_r[b] ($\times 10^{-3}$)	Reference
Leafy vegetables (fresh weight)	2.9	21	0.6 to 9.7	2.3	-	
Beet greens	1.1	3	0.8 to 1.5			Tracy, Prantl, and Quinn, 1983
Cabbage	4.9	1	-			Ter Haar, 1970
Lettuce	3.6	13	0.8 to 9.7			Ter Haar, 1970; John and Van Laerhoven, 1972(a); Rabinowitz, 1972
Spinach	1.7	2	0.6 to 2.9			John and Van Laerhoven, 1972(a)
Swiss chard	1.5	2	0.9 to 2.0			Tracy, Prantl, and Quinn, 1983
Fruits and storage organs (fresh weight)	2.2	30	0.1 to 15	-	1.0	
Beans (green)	3.1	3	0.2 to 7.8			Ter Haar, 1970; Tracy, Prantl, and Quinn, 1983
Beets	0.5	2	0.4 to 0.5			Tracy, Prantl, and Quinn, 1983
Carrot	3.5	7	0.1 to 15			Ter Haar, 1970; John and Van Laerhoven, 1972(a); Tracy, Prantl, and Quinn, 1983
Onion	1.2	3	0.7 to 2.5			Tracy, Prantl, and Quinn, 1983
Potato	1.9	3	0.2 to 4.8			Ter Haar, 1970; Tracy, Prantl, and Quinn, 1983
Pumpkin	0.2	1	-			Tracy, Prantl, and Quinn, 1983
Radish	3.4	4	2.2 to 4.7			John and Van Laerhoven, 1972(a); Dedolph et al., 1970
Raspberries	0.8	2	0.2 to 1.4			Tracy, Prantl, and Quinn, 1983
Strawberries	0.5	1	-			Tracy, Prantl, and Quinn, 1983
Sweet corn	4.1	1	-			Ter Haar, 1970
Tomato	0.9	3	0.1 to 2.5			Ter Haar, 1970; Tracy, Prantl, and Quinn, 1983
Grains (dry weight)	12.1	3	4.9 to 2.2	-	10.0	
Oats	13.5	2	4.9 to 22			John and Van Laerhoven, 1972(a)
Wheat	9.1	1	-			Ter Haar, 1970
Forage, hay, feed (dry weight)	91.1	23	11.6 to 370	73.1	-	
Fodder	40.0	1	-			Ter Haar, 1970
Grass hay	84.7	11	11.6 to 188.2			Dedolph et al., 1970; Milösevič et al., 1980; Rayno, Momeni and Sabau, 1980
Oat tops	102.3	11	77 to 370			John and Van Laerhoven, 1972(b); Rabinowitz, 1972

[a] Unweighted arithmetic mean; concentration factor.

[b] Unweighted geometric mean; transfer coefficients.

Table 3. Parameters representing soil-plant transfer of Po-210[a]

Transport mode	Mean CF_{sp}[b] (x10^{-4})	Number of derived values	Range (x10^{-4})	Mean B_v[c] (x10^{-4})	Mean B_r[b] (x10^{-4})	Reference
Leafy vegetables (fresh weight)	3.6	5	0.4 to 8.0	2.3	-	
Celery (leaves)	1.0	2	0.4 to 1.5			Watters, Johnson, and Hansen, 1969
Spinach	5.3	3	2.3 to 8.0			Watters, Johnson, and Hansen, 1969
Fruits and storage organs (fresh weight)	2.2	10	3.7×10^{-2} to 6.2	-	0.8	
Onion	0.4	2	0.4 to 0.5			Watters, Johnson, and Hansen, 1969
Pea	0.3	2	0.3 to 0.4			Watters, Johnson, and Hansen, 1969
Potato (whole)	4.4	2	2.5 to 6.2			Watters, Johnson, and Hansen, 1969
Radish	5.8	2	5.7 to 5.8			Watters, Johnson, and Hansen, 1969
Tomato	0.1	2	3.7×10^{-2} to 1.6×10^{-1}			Watters, Johnson, and Hansen, 1969
Grains (dry weight)	2.6	4	1.7×10^{-1} to 7.4	-	0.9	
Barley	5.0	2	2.6 to 7.4			Watters, Johnson, and Hansen, 1969
Wheat	0.2	1	-			Watters, Johnson, and Hansen, 1969
Corn	0.2	1	-			Watters, Johnson, and Hansen, 1969
Forage, hay, feed (dry weight)	39.1	13	0.4 to 129	21.3	-	
Fodder	5.4	3	0.4 to 12			Watters, Johnson, and Hansen, 1969
Grain straw	54	3	14 to 97			Watters, Johnson, and Hansen, 1969
Pasture hay	47	7	22 to 129			Milösević et al., 1980

[a] Values are nuclide-specific for chloride, oxide and naturally occurring forms of Po-210 only.

[b] Unweighted arithmetic mean; concentration factor.

[c] Unweighted geometric mean; transfer coefficients.

The CF_{sp}'s, B_v's and B_r's are given in dry weight concentrations for forage, hay, feed and grain; and in fresh weight concentrations for vegetables and fruits (i.e., moisture content as normally consumed). When necessary, conversion of literature data to fresh or dry weight was made with the use of information supplied in the text of each paper cited or by standard conversions documented in Spector (1956). The unweighted arithmetic average of mean CF_{sp}'s, geometric means of B_v's and B_r's, and range of individual values are given for each food category. The ranges indicate that much uncertainty is involved in determining a single value for any transfer parameter; much of this uncertainty is probably due to the variability in experimental conditions among the studies cited as well as the wide range of soil, climatic, and nutritional conditions evaluated.

Edible plant portions appear to take up less Po-210 from soil than Pb-210; the difference is an approximate order of magnitude for leafy vegetables, fruits and storage organs; and approximately two orders of magnitude for grains and animal hay or forage. For either nuclide, the degree of uptake by fruits and vegetables is nearly the same; among dry foodstuffs, uptake is greater in hay and animal forage than grains.

The actual fraction of total grain Pb-210 or Po-210 present in the grain hull versus that present within the grain kernel is not known; most of the available studies do not mention whether analyses were performed on whole or de-hulled grain. The one study that described grain processing analyzed Po-210 in wheat "chaff" (Watters, Johnson, and Hansen, 1969). A CF_{sp} of 1.1×10^{-3} was determined for chaff, as compared to a CF_{sp} of 1.7×10^{-5} for the kernel. On the basis of this single determination, it appears that approximately 98% of the wheat Po-210 may be incorporated into the hull. It is assumed that all remaining data presented for grain Pb-210 and Po-210 are for whole grain.

3.1.2 Distribution and Uptake in Animal Products

Coefficients developed for evaluating transfer to meat and milk incorporate daily intake rather than concentration ratios and thus consider accumulation in muscle and milk according to ingestion rates. The transfer coefficient, f_m, represents the fraction of the element or nuclide ingested daily that is secreted per liter of milk at equilibrium (day per liter); f_f represents the fraction of the element ingested daily that appears in each kilogram of meat at the time of slaughter (day per kilogram).

Very little work specifically designed to measure transfer of naturally occurring Pb-210 and Po-210 from feed sources to meat and milk of domestic animals has been performed. Those studies that exist are the result of acute controlled feeding exposure (Stanley, Mullen, and Bretthauer, 1971), Po-210 introduction directly to the rumen of individual cows (Watters and McInroy, 1969; Watters, Johnson, and McInroy, 1971), or sample collection from animals feeding near sources of industrial emissions (uranium mines and/or mills) (Holtzman et al., 1979). As a result of the sparse record of nuclide-specific data, information collected during longer-term studies of stable isotope transfer to muscle and milk were also included in the summary presented as Table 4. Sources included lead mines and smelters (Donovan, Feeley, and Canavan, 1969; Kerin and Kerin, 1971; Hammond and Aronson, 1964; Djuric et al., 1971),

Table 4. Parameters representing transfer of Pb-210 and Po-210 to meat and milk

Transport mode	Unweighted mean	Range	Reference
Pb-210			
Milk, f_m[a]	1.2×10^{-4} (n=14)	1.8×10^{-6} to 4×10^{-4}	Donovan, Feeley, and Canavan, 1969; Kerin and Kerin, 1971; Bovay, 1971; Nelmes et al., 1974, Stanley et al., 1971
Beef, f_f[b]	1×10^{-3} (n=4)	2×10^{-4} to 2×10^{-3}	Nelmes et al., 1974; Holtzman et al., 1979
Po-210			
Milk, f_m	1.7×10^{-4} (n=3)	8.9×10^{-5} to 2.9×10^{-4}	Watters and McInvoy, 1969; Watters, Johnson, and McInvoy, 1971; Johnson and Watters, 1972
Beef, f_f[c]	2.2×10^{-3} (n-2)	3.0×10^{-4} to 4.0×10^{-3}	McDowell-Boyer and Baes, 1980; Baes et al. (in review)

[a] f_m expresses the estimated fraction of the element ingested daily that is secreted per liter of milk at equilibrium (d/L).

[b] f_f expresses the estimated fraction of the element ingested daily that appears in each kilogram of muscle at time of slaughter (d/kg).

[c] Based in part on data for sheep, caribou, and reindeer.

automobile exhaust (Bovay, 1971) and sewage sludge containing lead (Nelmes et al., 1974). Even fewer data are available for Po-210, and all that could be obtained are included in Table 4.

Very little ingested lead is found in muscle, as this element is a bone-seeker. The principal sink for Pb-210 is the skeleton. The skeleton, as well as the liver and kidneys, are sinks for Po-210 (Beasley and Palmer, 1966; Hill, 1967; Kauranen and Miettinen, 1969). The wider distribution of Po-210 in soft tissues is thought to be partly the result of skeletal Pb-210 decay giving rise to daughter Po-210 which is, in turn, re-allocated by the bloodstream. The majority of Pb-210 in animal tissues is considered to be of dietary origin; Po-210 arises from both diet and internal decay of Pb-210. Animals and humans that regularly feed on bones (for broth or soup) and internal organs of grazers exposed to dietary Pb-210 or Po-210 have been observed to accumulate greater body burdens than those individuals who consume only muscle (Holtzman, 1966a,b).

3.1.3. Estimated Dietary Exposure

Global diets have been extensively sampled for the presence of naturally occurring radionuclides. Cultural food preferences, availability and anomalies of geology or atmosphere are all factors in governing the variability in world dietary exposures to Pb-210 and Po-210 summarized in Table 5. In general the smallest ingestion exposures were obtained from diets in developed countries (Federal Republic of Germany, U.K., U.S.A. and U.S.S.R) and the high grain and vegetable diet of metropolitan India. The greatest exposures were estimated for diets of Japan and Arctic dwellers in Finland, Canada, and Alaska; all of which contain animal protein in the form of seafood (Japan) or large,

Table 5. Daily intake of naturally occurring Pb-210 (pCi/d) and Po-210 (pCi/d) in the diet (excluding water and beverages)[a]

Country/City	Pb-210 (pCi/d)	Po-210 (pCi/d)	Reference
U.S.A.			
Illinois	1.8[b]	-	Holtzman, 1963
Illinois (hospital)	-	1.6[c]	Spencer et al., 1977
Boston	1.6[d]	-	Magno, Groulx, and Apidianakis, 1970
Chicago	1.7[d]	-	Magno, Groulx, and Apidianakis, 1970
Honolulu	1.5[d]	-	Magno, Groulx, and Apidianakis, 1970
Los Angeles	1.7[d]	-	Magno, Groulx, and Apidianakis, 1970
New Orleans	1.7[d]	-	Magno, Groulx, and Apidianakis, 1970
Palmer, Alaska	1.6[d]	-	Magno, Groulx, and Apidianakis, 1970
Chicago (hospital)	1.3[c]	1.6[c]	Holtzman et al., 1974
New York City	1.2	-	Morse and Welford, 1971
New York City	1.8		Cohen, Jaakkola, and Wrenn, 1973
San Francisco (middle-income)	2.7[e]	-	Michelson et al., 1962
Chicago (middle-income)	3.7[e]	-	Michelson et al., 1962
New York City (middle-income)	2.0[e]	-	Michelson et al., 1962
San Francisco (low-income)	3.9[e]	-	Michelson et al., 1962
Chicago (low-income)	3.0[e]	-	Michelson et al., 1962
New York City (low-income)	nd[e,f]	-	Michelson et al., 1962
Nine-city composite	2.4	-	Michelson, 1961; Michelson et al., 1962

Table 5. (continued)

Country/City	Pb-210 (pCi/d)	Po-210 (pCi/d)	Reference
United Kingdom	3.2	-	Hill, 1965
United Kingdom	1 to 10	1 to 10	Hill, 1967
India			
Bombay	3.4	-	Lalit, Ramachandran, and Rajan, 1980
Bombay	-	1.5[g]	Khandekar, 1977
Federal Republic of Germany	4.6	4.6	Glöbel, Muth, and Oberhausen, 1966
U.S.S.R.			
Leningrad	4.0	3.0	Yermolaeva-Markovskaya et al., 1968
Rostov-on-Don	6.2	4.1	Ladinskaya et al., 1973
Bulgaria	-	1.6 to 2.1	Keslev et al., 1975
Japan			
Tokyo	5.9	-	Kametani et al, 1981
Range, 7 regional sites	13.8 to 22.5[h]	-	Takata, Watanabe, and Ichikawa, 1968
Mean, 7 regional sites	17.0[h]	-	Takata, Watanabe, and Ichikawa, 1968
Chiba Prefecture	-	9.0 to 13.0	Okabayaski et al., 1975

Table 5. (continued)

Country/City	Pb-210 (pCi/d)	Po-210 (pCi/d)	Reference
Arctic Dwellers			
Finland (Lapps)	8.6	6.9	Kauranen and Miettinen, 1969
Canada	-	100	Hill, 1967
Alaska	10	100	Beasley and Palmer, 1966
Alaska	-	60	Blanchard, 1966
Alaska	10	100	Holtzman, 1966(a,b); Holtzman and Ilcewicz, 1971
Alaska	40	40	Ramzaev et al., 1974

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[a] Many of these values taken as cited by Holtzman, 1980.

[b] Based on fecal and urine analyses.

[c] Adult patients on controlled hospital diet.

[d] Assume 2 kg/d food ingestion; data collected in 1966 for children aged 9-12 years.

[e] Teenage diet.

[f] Not detected.

[g] Sixty percent of diet sample.

[h] Seven regional sites comprise the entire country, including major population centers of Tokyo and Kyoto-Osaka.

native ruminants (Arctic reindeer and caribou). The reasons for elevated Pb-210 and Po-210 concentrations in these two groups of animals have been previously discussed in Sect. 3.1.1.

3.2 TOBACCO

Inhalation of tobacco smoke is ranked second to food in providing Pb-210 and Po-210 exposure to man (Spencer et al., 1977). Among smokers, a larger portion of daily Po-210 intake (17.1%) than Pb-210 intake (9.4%) is provided by smoke. Exposure to alpha-decay products from these and other Rn-222 daughters has been established as the major risk factor in the incidence of bronchial cancer among uranium miners. It follows that exposure to alpha radiation from smoke inhalation may also be a factor in cigarette-induced bronchial cancer (Radford, 1974). Nevertheless, the reader should understand that estimates for continuous background inhalation exposure to Rn-222 and daughters (200-2000 pCi/d) greatly exceed that value estimated for Rn-222 and daughter exposure from tobacco smoke (Martell, 1974; Hamrick and Walsh, 1974).

3.2.1 Origin

The two possible sources of Pb-210 and Po-210 associated with tobacco are uptake from soils containing these isotopes as daughter Products of Ra-226 or leaf surface deposition of airborne aerosols to which naturally occurring Pb-210 and Po-210 have become adsorbed. Greenhouse experiments with artificially contaminated soils and field sampling of tobacco plantations indicate that leaf scavenging of aerosols, particularly during rainfall events is the dominant mode of Pb-210 and Po-210 entry into tobacco plants (Francis, Chesters, and Erhardt, 1968).

The time elapsed between leaf harvest and actual manufacture into a cigarette or cigar determines the Po-210 content of the consumer product, which is a function of initial Po-210 deposition plus Pb-210 decay. The activity of some domestic and imported tobacco products is summarized in Table 6.

Polonium is readily volatilized at the temperature of the burning tip of a cigarette or cigar (600 to 800°C) (Abe and Abe, 1969; Radford and Hunt, 1964) and is known to be adsorbed to submicron particles (Martell, 1974), such as are found in smoke. Lead is not sublimated at these temperatures, but is a component of the resulting smoke and ash; Pb-210 can be found electrically attached to particles <0.3 microns in diameter (Kawans and Nakatani, 1964, as quoted in Francis, Chesters, and Erhardt, 1968). Lead-210 and Po-210 aerosols attached to the sticky exudate of tobacco leaf surfaces can also be encapsulated by the exudate polymer produced as part of the curing process. During tobacco combustion, these insoluble, polymerized particles are released (Fleischer and Parungo, 1974; Martell, 1974) to be inhaled by the smoker or other persons in the vicinity. Regardless of the specific mechanism, there appears to be ample opportunity for Pb-210 and Po-210 exposure to bronchial epithelium and lung tissue during the smoking process.

3.2.2. Estimated Inhalation Exposure

Numerous variables govern the degree of exposure to tobacco smoke; geographic region where the tobacco is grown, fineness of the tobacco cut; presence or absence of a cigarette filter, size and composition of the filter, smoking habits, etc. (Black and Bretthauer, 1968; Ferri and Baratta, 1966; Marsden, 1964). Estimated ranges of daily smoke intake

Table 5. Pb-210 and Po-210 content of commercial cigarettes[a]

Country of manufacture	pCi per cigarette	pCi per g	Reference
Pb-210			
U.S.A.	0.017[b]	-	Holtzman and Ilcewicz, 1966
	0.018[b]	-	Holtzman and Ilcewicz, 1966
	0.015[b]	-	Holtzman and Ilcewicz, 1966
	0.006[b]	-	Holtzman and Ilcewicz, 1966
	0.013[b]	-	Holtzman and Ilcewicz, 1966
Po-210			
Australia	-	0.64	Hill, 1965
Brazil	0.46	0.47	Dos Santos, 1970
Bulgaria	0.38	-	Nikolova and Parfenov, 1971
Canada	0.21	0.26	Black and Bretthauer, 1968
Central and South America	-	0.67	Hill, 1965
Czechoslovakia	0.2-0.45	-	Solnicka and Bischof, 1970
Egypt	0.38	0.44	Black and Bretthauer, 1968
Federal Republic of Germany	0.41-0.60	0.41-0.50	Rajewsky and Stahlhofen, 1966
	0.52	0.56	Black and Bretthauer, 1968
Finland	0.29	0.63	Black and Bretthauer, 1968
France	0.63	0.56	Black and Bretthauer, 1968

Table 6. (continued)

Country of manufacture	pCi per cigarette	pCi per g	Reference
Po-210 (continued)			
India and Pakistan	-	0.41	Hill, 1965
India	0-0.4	-	Athalye et al., 1972
Indonesia		0.24	Hill, 1965
Italy	-	0.37-0.53	Carfi and Lonati, 1966
Japan	-	0.31-0.84	Okabayashi et al., 1975
Japan	-	0.54	Suzuki, 1967
Japan	0.61	0.60	Black and Bretthauer, 1968
Norway	0.23	0.22	Black and Bretthauer, 1968
Philippines	0.22	0.18	Black and Bretthauer, 1968
Rhodesia	-	0.65	Hill, 1965
Turkey and Greece	-	0.24	Hill, 1965
U.S.S.R.	0.38	0.60	Black and Bretthauer, 1968
United Kingdom	0.47	0.46	Black and Bretthauer, 1968
U.S.A.	0.39-0.48	-	Radford and Hunt, 1964
U.S.A.	0.32-0.34	0.32-0.34	Rajewsky and Stahlhoffen, 1966
U.S.A.	0.54	0.52	Black and Bretthauer, 1968
U.S.A.	-	0.51	Hill, 1965

Table 6. (continued)

Country of manufacture	pCi per cigarette	pCi per g	Reference
Po-210 (continued)			
U.S.A.	0.023[b]	-	Holtzman and Ilcewicz, 1966
	0.027[b]	-	Holtzman and Ilcewicz, 1966
	0.016[b]	-	Holtzman and Ilcewicz, 1966
	0.018[b]	-	Holtzman and Ilcewicz, 1966
	0.020[b]	-	Holtzman and Ilcewicz, 1966
Yugoslavia	-	0.40-0.60	Kilibarda et al., 1966

[a] Partly derived from Parfenov (1974).

[b] In cigarette smoke.

of Pb-210 and Po-210 are presented in Table 7. Many of these results indicate that inhalation exposure via smoking may exceed normal atmospheric inhalation exposure by at least a factor of two in the case of Pb-210 and two orders of magnitude for Po-210 (Spencer et al., 1977).

4. ESTIMATED DOSE FROM DIETARY AND TOBACCO SMOKE EXPOSURES

4.1 DIETARY INGESTION

The range of total exposure estimated from global dietary studies (Table 5) was converted to 50-year dose commitments by use of the dose conversion factors recommended by Dunning et al. (1981). These estimates are presented in Table 8.

A comparison of the estimated maximum doses indicates that adults ingesting an "ordinary," mixed Western-style diet in the U.S.A., U.K., and Federal Republic of Germany would receive between 10-50% the adsorbed radiation to target organs that adults ingesting observed diets in Japan and the Arctic would receive. Nevertheless, even the maximum estimated dietary dose (Po-210 dose to the spleen of Arctic dwellers; 0.23 to 0.58 annual rems) is less than 40% of the annual permissible dose of 1.5 rem to the public for non-blood-forming organs (U.S. DOE, 1981). To date, there are no available estimates of Pb-210 and Po-210 dietary exposures specific to residents of the phosphate-bearing areas of Southwest Florida. It would be particularly useful to compare dietary exposure between populations ingesting foods grown locally on reclaimed or mine-adjacent properties and those ingesting a mixed diet largely derived from non-local sources. It is expected that such exposures would not be comparable to the maximally exposed populations of Japan or the Arctic.

Table 7. Estimated daily inhalation exposure of Pb-210 and Po-210 to smokers

Radionuclide	Estimated exposure (pCi/d)	Reference
Pb-210	0.76	Nikolova and Parfenov, 1971
	2.1	Yermolaeva-Makovskaya, Pertsov and Popov, 1969
	0.52-1.1	Ferri and Christiansen, 1967
	0.12-0.36	Holtzman and Ilcewicz, 1966
	0.15	Spencer et al., 1977
Po-210	16.0	Turner and Radley, 1960
	1.6-7.2	Parfenov, 1974
	8.0	Marsden, 1964
	2.0	Rajewsky and Stahlhoffen, 1966
	1.4	Nikolova and Parfenov, 1971
	1.5	Little and McGandy, 1966
	0.36	Spencer et al., 1977

Table 8. Estimated range of adult 50-year dose commitments (rem) for one year's dietary intake of Pb-210 and Po-210 from natural sources[a,b]

Target organ	Region						
	U.S.A.	U.K.	India	Federal Republic of Germany	U.S.S.R.	Japan	Arctic
Pb-210							
Bone	9.2E-3 to 3.0E-2	7.7E-3 to 7.7E-2	2.6E-2	3.5E-2	3.1E-2 to 4.8E-2	4.5E-2 to 1.7E-1	6.6E-2 to 3.1E-1
Endosteal tissue	4.2E-3 to 1.4E-2	3.5E-3 to 3.5E-2	1.2E-2	1.6E-2	1.4E-2 to 2.2E-2	2.1E-2 to 7.9E-2	3.0E-2 to 1.4E-1
Red marrow	4.4E-4 to 1.4E-3	3.7E-4 to 3.7E-3	1.2E-3	1.7E-3	1.5E-3 to 2.3E-3	2.2E-3 to 8.2E-3	3.1E-3 to 1.5E-2
Kidney	4.1E-4 to 1.3E-3	3.4E-4 to 3.4E-3	1.2E-3	1.6E-3	1.4E-3 to 2.1E-3	2.0E-3 to 7.7E-3	3.0E-3 to 1.4E-2
Liver	6.1E-4 to 2.0E-3	5.1E-4 to 5.1E-3	1.7E-3	2.4E-3	2.0E-3 to 3.2E-3	3.0E-3 to 1.2E-2	4.4E-3 to 2.0E-2
Whole body	7.4E-4 to 2.4E-3	6.2E-4 to 6.2E-3	2.1E-3	2.9E-3	2.5E-3 to 3.8E-3	3.7E-3 to 1.4E-2	5.3E-3 to 2.5E-2
Po-210[c]							
Spleen	9.3E-3	5.8E-3 to 5.8E-2	8.8E-3	2.7E-2	9.8E-3 to 2.4E-2	7.6E-2	2.3E-1 to 5.8E-1
Kidney	5.4E-3	3.4E-3 to 3.4E-2	5.1E-3	1.6E-2	5.4E-3 to 1.4E-2	4.4E-2	1.4E-1 to 3.4E-1
Liver	9.3E-4	5.8E-4 to 5.8E-3	8.8E-4	2.7E-3	9.3E-4 to 2.4E-3	7.6E-3	2.3E-2 to 5.8E-2
Whole body	2.4E-4	1.5E-4 to 1.5E-3	2.2E-4	6.9E-4	2.4E-4 to 6.1E-4	2.0E-3	6.0E-3 to 1.5E-2

[a] Using dose conversion factors developed by Dunning et al. (1981).

[b] Using range of exposures as presented in Table 5.

[c] Direct ingestion only; does not include contribution of internal Pb-210 decay.

When compared to previous estimates for U.S. ingestion of the parent Ra-226 (Watson, Etnier, and McDowell-Boyer, 1983), Pb-210 appears to provide a comparable dose for bone, kidney, liver, and whole body. The upper values of the dietary dose ranges for Pb-210 exceed those for Ra-226 in the kidney and liver by approximately an order of magnitude. The liver doses for dietary Po-210 and Ra-226 are approximately equal; the estimated Po-210 dose in the kidney exceeds that for Ra-226 by an approximate order of magnitude; the whole body Po-210 dose is less than that for Ra-226 by an approximate order of magnitude.

4.2 TOBACCO SMOKE INHALATION

Exposures additional to those received from ingestion and unique to smokers inhaling Pb-210 and Po-210 (summarized in Table 7) were converted to internal dose commitment by applying the dose conversion factors recommended by Eckerman, Ford, and Watson (1981). An activity median aerodynamic diameter (AMAD) of 0.3 µm was assumed. Estimated annual dose commitments are summarized in Table 9.

Tobacco smoking appears to provide a dose equal to or greater than that provided by dietary ingestion for both Pb-210 and Po-210 in bone tissues, liver and kidneys; and for Po-210 in the spleen for the three Western-style diets addressed in Sect. 4.1. The smoking dose estimates are most comparable to those obtained for dietary intake by Arctic dwellers.

Table 9. Estimated range of committed dose equivalent (rem) for one year's inhalation exposure to tobacco smoke containing natural concentrations of Pb-210 and Po-210[a,b]

Target organ	Dose commitment (rem)
Pb-210	
Bone surface	9.2E-3 to 1.6E-1
Liver	2.6E-3 to 4.5E-2
Kidneys	1.2E-3 to 2.1E-2
Lungs	5.3E-5 to 9.3E-4
Po-210[c]	
Spleen	1.1E-2 to 4.8E-1
Kidneys	6.3E-3 to 2.8E-1
Liver	1.1E-3 to 4.9E-2
Lungs	4.9E-4 to 2.2E-2

[a] Using internal dose conversion factors developed by Eckerman, Ford, and Watson, 1981.

[b] Using range of values reported in Table 7.

[c] Does not include contribution of internal Pb-210 decay.

During the present evaluation, the author has identified a number of research areas that should. be addressed in order to obtain a more realistic quantification of food chain transport of Pb-210 and Po-210 than is currently possible in the absence of site-specific data:

(1) For the majority of internal organs evaluated, the dose resulting from smoking commercially available tobacco products is comparable to or greater than the dose estimates for ingestion of naturally occurring dietary Pb-210 and Po-210.

(2) No data are readily available to characterize dietary exposures from Pb-210 and Po-210 for the residents of southwestern Florida ingesting locally grown food. These data could be readily collected from market-basket or whole meal surveys of the local populace.

(3) A principal problem area is the lack of experimentation specifically designed to determine milk and meat transfer coefficients in cattle. Not only should transfer coefficients for the chemical forms of Pb-210 and Po-210 specific to background, historical, and current phosphate mining areas of southwestern Florida be determined; but releases from tailings piles should also be simulated as nearly as possible in feeding studies by offering contaminated feed. In addition, sufficient numbers of animals should be investigated to allow statistical comparison of results.

(4) The importance of ingested soil and water as a source of contamination for milk and meat has not been critically examined. Data from feeding studies are needed, to determine the degree of Pb-210 and Po-210 solubilization from ingested mineral soil in the digestive tract, and the subsequent internal transport.

(5) For most food items and tobacco, aerosol deposition seems to be the principal mode of Pb-210 and Po-210 entry. This feature is of particular concern for leafy vegetables. As a result, only fruit-bearing crops such as citrus, berries, and cane fruits should be grown on phosphate-reclaimed land.

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