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REVIEW AND RECOMMENDATIONS OF DOSE CONVERSION FACTORS AND ENVIRONMENTAL TRANSPORT PARAMETERS FOR ^{210}Pb AND ^{226}Ra

Final Report

L. M. McDowell-Boyer
A. P. Watson C. C. Travis

Oak Ridge National Laboratory

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**REVIEW AND RECOMMENDATIONS OF
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Final Report

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ABSTRACT

Methods of determining dietary exposure and dose to man from environmental concentrations of ^{210}Pb and ^{226}Ra were investigated. Environmental transport of these nuclides from soil and air to vegetation, meat, and milk was quantitatively analyzed following a review of literature sources. Implications of soil ingestion by livestock as a source of ^{210}Pb or ^{226}Ra in animal products were also examined. Inhalation and ingestion dose commitment factors were derived for human exposure to environmental ^{226}Ra and ^{210}Pb .

The recommended equilibrium concentration factor (CF) for ^{210}Pb transport from soil to vegetables, fruits, and grains is 1.0×10^{-2} (fresh weight), while the related value for forage is 2.0×10^{-1} (dry weight). Recommended values for transfer coefficients, f_m and f_f , describing transport of ^{210}Pb from forage to milk and meat on a daily consumption basis, respectively, are 2.5×10^{-4} day/liter and 8.0×10^{-4} day/kg. Corresponding recommended values for radium include a vegetable, fruit, and grain CF of 2.0×10^{-2} and a forage CF of 9.0×10^{-2} . Equilibrium transfer of ^{226}Ra from forage to milk may be estimated with a recommended transfer coefficient of 5.9×10^{-4} day/liter. A recommended transfer coefficient for ^{226}Ra in beef is 5.1×10^{-4} for cattle.

Until more direct data are made available, the degree of ^{210}Pb and ^{226}Ra aerosol deposition onto edible crops and forage is best estimated by the model contained in recent drafts of the Regulatory Guide 1.109 provided by the U.S. Nuclear Regulatory Commission.

Fifty-year dose commitment factors for ingestion and inhalation of ^{210}Pb and ^{226}Ra are provided based on calculations from recent dosimetry models. The critical organ for ^{210}Pb inhalation and ingestion is bone. For ^{226}Ra , the critical organ for inhalation is lung, while for ingestion, the critical organ is bone.

Appendix B includes development of dose commitment factors for inhalation of ^{222}Rn and ^{210}Po , nuclides which are likely to be present in many aerosols containing ^{210}Pb and ^{226}Ra .

Appendix B includes development of dose commitment factors for inhalation of ^{222}Rn and ^{210}Po , nuclides which are likely to be present in many aerosols containing ^{210}Pb and ^{226}Ra .

1. INTRODUCTION

The evaluation of ingestion exposure and dose to man from specific radionuclides in the environment requires that dietary concentrations of each isotope be determined. When actual site-specific measurements of these concentrations are not available, it is possible to predict dietary exposure through the use of environmental transport models to quantify food-chain transport.

This document reviews literature pertinent to ^{210}Pb and ^{226}Ra transfer through food chains to define and evaluate parameters necessary to the implementation of these predictive models. These nuclides are of interest mainly in the assessment of radiological impacts associated with the operation of uranium mills and decommissioning of mill sites, although many other sources of environmental ^{210}Pb and ^{226}Ra exist.

Both soil and air were recognized as potential sources of ^{210}Pb and ^{226}Ra in vegetation. Nuclide transport from soil-to-plant tissues and plant-to-animal products important in the human diet were evaluated on the basis of concentration factors (CF), which are defined as the dimensionless ratios of nuclide concentrations in one compartment to concentrations in the supporting compartment. Transfer coefficients f_m and f_f characterizing nuclide transport from forage to milk (f_m in day/liter and beef (f_f in day/kg) were also calculated. Coefficients were normalized to the total daily isotopic ingestion by cattle. Soil ingestion by cattle was considered a potential source of nuclides in animal products.

Aerosol deposition was evaluated on the basis of a model provided in the U. S. Nuclear Regulatory Commission's (U.S. NRC) Regulatory Guide 1.109 (drafts, March 1976 and October 1977). Implications of the use of this model for predicting dietary concentrations in a variety of food items are discussed.

Transport parameters were derived for those portions of plants or animals that constitute the diet of cattle or humans. Parameters for fish, poultry, or eggs were not evaluated due to the lack of available information concerning the associated food chains.

In calculating concentration factors for soil-to-plant and forage-to-muscle transfer of ^{210}Pb or ^{226}Ra , the amount of the nuclide accumulated in the tissues may depend on the time interval between planting and

crop harvest, and between birth and slaughter of the animals. Because of this temporal dependence, only mature plants and animals were considered in this evaluation. However, variability in these time intervals should be noted when applying the CF values recommended in this study. For forage-to-milk transfer of ^{210}Pb or ^{226}Ra , equilibrium concentrations in the milk compartment were used or estimated in deriving CF's and transfer coefficients (f_m).

Data from research utilizing either stable or radioactive isotopes were incorporated into this review. Any variation in transport mechanisms due to isotopic effects was considered insignificant. We consider chemical form and physical state of the nuclides to influence uptake to a much greater extent. Chemical and physical descriptions were seldom complete for each study, but have been incorporated when available. A brief discussion of radium availability for root uptake is presented in Sect. 3.1.1.1.

Recommended concentration factors and transfer coefficients for use in predicting dietary concentrations of ^{210}Pb and ^{226}Ra are included in this report along with appropriate inhalation and ingestion dose commitment factors. Rationale behind the recommended values is discussed. A summary of the recommended values for transport parameters is given in Sect. 4. Comparisons between measured dietary concentrations of ^{210}Pb and ^{226}Ra and those predicted through implementation of recommended CF's and models were made. The results of these comparisons are discussed in pertinent sections of this report.

The appendices of this report include a discussion of dose commitment factors appropriate for estimating dose due to inhalation of ^{222}Rn and ^{210}Po . Environmental transport parameters for these nuclides have not been reviewed.

2. RECOMMENDATIONS OF ENVIRONMENTAL TRANSPORT PARAMETERS AND DOSE CONVERSION FACTORS FOR ^{210}Pb

To assess radiation doses associated with dietary ingestion of ^{210}Pb , several environmental transport and internal dosimetry parameters must be quantified. This chapter addresses these needs and presents

recommendations of values believed to be most appropriate for use in dose assessments based on information currently available.

2.1 Environmental Transport Parameters

Human dietary concentrations of ^{210}Pb are related to the extent of elemental transport through food chains. To quantify this transport, it is necessary to consider both foliar deposition of aerosol ^{210}Pb and root uptake of ^{210}Pb from soil, with subsequent translocation to edible plant portions. In addition, transfer of the isotope to animal products, following ingestion of contaminated feed or forage must be considered. A review of lead dynamics in food chains based on data for ^{210}Pb and stable lead isotopes follows. Recommendations of uptake parameters for various modes of ^{210}Pb entry into food chains are included in the discussions.

2.1.1 The source of ^{210}Pb in edible crops

The natural source of ^{210}Pb in the environment is its naturally occurring gaseous precursor ^{222}Rn , which is present in soil and air due to decay of ^{226}Ra in soil. Technologically enhanced sources of ^{210}Pb include other sources of ^{222}Rn , fly ash from coal-fired power plants, gasoline combustion products, and nuclear explosions (Moore *et al.*, 1976; Jaworowski and Grzybowska, 1977; Jaworowski and Kownacka, 1976). Sources of stable lead in the environment include natural lead in soil and air, smelters, pesticides, coal-fired power plants, and leaded gasolines (Jaworowski and Kownacka, 1976; Koeppe, 1977). All of these sources provide aerosol as well as soil concentrations of lead isotopes (Sharma and Shupe, 1977).

Results of various empirical studies on plant uptake of stable or radioactive lead have led to diverse conclusions regarding the relative significance of soil and aerosol lead in plant lead concentrations. Much of the apparent disparity, however, may be attributed to variability in environmental conditions between studies, and to differences in morphological characteristics of the plant parts assayed. If soil is artificially enhanced with stable lead or ^{210}Pb , as is the case in many investigations, the significance of aerosol-derived lead with respect to the total plant lead may be masked.

In studying lead uptake by crops, it is important to distinguish the edible portion from the remaining plant parts. In some cases, this edible portion of the plant may be well-protected from aerosol deposition, as with corn kernels or root crops or; well-exposed, as with lettuce leaves or pasture grass. Therefore, it is expected that the relative importance of aerosol vs soil lead will be species dependent.

In summary, it is apparent that both foliar deposition and root uptake of ^{210}Pb must be evaluated. The relative importance of each source in a particular situation will vary depending on the conditions of ^{210}Pb environmental contamination, and on the food items determined to be important in the human diet.

2.1.2 Root uptake and distribution of ^{210}Pb by vegetation

In order to quantify root uptake and subsequent translocation of ^{210}Pb to edible portions of crops, concentration factors may be calculated. These unitless factors express the ratio of the ^{210}Pb concentration in plant portions to that in supporting soil.

A large number of studies have been undertaken to determine these soil-plant relationships for lead. However, only a portion of the studies reviewed here were designed to discriminate between the atmospheric and soil contribution of lead to "edible portions" of plants (Dedolph *et al.*, 1970; Rabinowitz, 1972; Ter Haar, 1970; John and Van Laerhoven, 1972a, b). Some of the experiments which discriminate between soil and atmospheric lead involved the use of growth chambers or greenhouses with filtered air (Dedolph *et al.*, 1970; Ter Haar, 1970; John and Van Laerhoven, 1972a). In one study of lead uptake by lettuce in growth chambers, it is not clear if the intake air was filtered (John and Van Laerhoven, 1972b). Since soil lead content was experimentally enhanced to 1000 ppm in this study, natural aerosol lead should not have contributed significantly to plant lead. Rabinowitz (1972) estimated lead uptake by lettuce and oats from soil in the presence of atmospheric lead by preparing soil with a lead isotopic composition different from that of normal atmospheric lead.

Table 2.1 lists the soil-to-plant concentration factors (CF_{sp}) derived from those studies in which aerosol contributions of lead were

Table 2.1. Concentration factors representing soil-to-plant transfer of ^{210}Pb ($[\text{}^{210}\text{Pb}]_{\text{plant}} : [\text{}^{210}\text{Pb}]_{\text{soil}}$) without aerosol ^{210}Pb contribution

Edible plant portion	Mean concentration factors (CF_{sp}) ^a ($\times 10^{-3}$)	Number of derived values	Range ($\times 10^{-3}$)	Reference
Vegetables (fresh weight)				
Bean	7.8	1		b
Broccoli	0.8	2	0.6-1.0	e
Cabbage	4.8	1		b
Carrot	15	3	2.0-24	b,c
Cauliflower	0.5	2	0.4-0.5	e
Corn	4.2	1		b
Lettuce	2.9	15	0.5-9.9	b-e
Potato	4.2	1		b
Radish	3.6	4	0.3-7.1	e,f
Spinach	2.9	2		c
Tomato	2.5	1		b
Unweighted average	4.5	33	0.3-24	
Grain (fresh weight)				
Oats	5.5	2	1.8-9.1	c
Wheat	9.1	1	9.1	b
Unweighted average	7.3	3	1.8-9.1	
Forage, hay, feed (dry weight)				
Oat tops	90	12	20-290	d,e
Fodder	40	1	40	b
Forage	140	2	90-190	f
Unweighted average	90	15	20-290	

^aValues express ratios of fresh weight ^{210}Pb concentrations in plants to dry weight concentrations in soil for all food crops directly edible by man. Dry-weight concentrations in both plants and soil were used for forage, hay, and feed calculations.

^bTer Haar, 1970.

^cJohn and Van Laerhoven, 1972(a).

^dRabinowitz, 1970.

^eJohn and Van Laerhoven, 1972(b).

^fDedolph *et al.*, 1970.

insignificant. Thus, the CF_{sp} 's represent soil-to-plant transfer alone. Vegetable and grain CF_{sp} 's are given on a fresh weight basis per dry weight concentration in soil, while forage, feed, and hay CF_{sp} 's are presented on a dry weight basis. In this way, lead concentrations in crops directly consumed by man may be readily estimated on a fresh weight basis, which is usually desirable. Concentrations in forage, feed, and hay are more directly useful if given on a dry weight basis (see Sect. 2.1.4).

By comparing lead concentrations in edible portions of crops to those in soil, root uptake and translocation within the plant are simultaneously quantified. These processes are very dependent on properties such as soil pH, organic and phosphorous content of the soil, plant species, and the physiologic condition of the plant in question (Menzel, 1965; MacLean *et al.*, 1969; Dedolph *et al.*, 1970; Rains, 1971; Cox and Rains, 1972; John, 1972; John and Van Laerhoven, 1972a; Zimdahl and Foster, 1976; Koeppe, 1977). Thus, the CF_{sp} values in Table 2.1 not only represent averages over various isotopes and chemical forms of lead but also averages over a range of soil and plant conditions.

Translocation of lead isotopes within plants following root uptake is of a restricted nature. Thus, one would expect edible root crops to accumulate more lead than above ground edible crops (Hevesy, 1923; MacLean *et al.*, 1969; Miller and Koeppe, 1970; Rains, 1971; John and Van Laerhoven, 1972a; Rabinowitz, 1972; Jones *et al.*, 1973; Koeppe, 1977). This expectation appears to be reinforced with the data given in Table 2.1, where the CF_{sp} for carrots is greater than the CF_{sp} for other crops. However, the limited number of values used to obtain average CF_{sp} values precludes a statistical test of significant differences between CF_{sp} 's of different vegetable species.

An unweighted average CF_{sp} for each food category of vegetables, grains, and forage, feed, and hay was determined and is given in Table 2.1. More complete statistical analysis of the distribution of values around a mean was not possible since the extent of the data base comprising a derived CF_{sp} value is not clear in some literature sources. The average fresh weight CF_{sp} 's determined for vegetables and grains appear to differ by less than an order of magnitude. Thus, these food categories

were combined when recommending an unweighted average CF_{sp} to be used in predicting dietary concentrations of ^{210}Pb with a given soil concentration of the isotope.

If we assume an average CF_{sp} for vegetables and grains of 4.9×10^{-3} , obtained by averaging all mean CF_{sp} 's in both categories, and an average U.S. soil concentration of 1.5 pCi/g dry soil (Ter Haar *et al.*, 1967; Fisenne, 1968), dietary concentrations of 7.4 pCi/kg for fresh vegetables and grains may be predicted. It is not clear, however, if the soils assayed included fertilized soils in which ^{210}Pb concentrations may be enhanced. Measured concentrations of ^{210}Pb in various food items have been reported for a New York City market survey (Morse and Welford, 1971), and are listed in Table 2.2. The predicted value of 7.4 pCi/kg does not differ greatly from the measured values, although it is consistently greater. This difference may be much greater in reality, since deposition of aerosol ^{210}Pb may have contributed significantly to the ^{210}Pb concentrations measured in New York City food items. Some lead would be expected to be lost during food preparation and processing.

As was indicated earlier, adequate data do not exist to generate a sample distribution about a mean with a certain degree of statistical certainty. Therefore, it is recommended to use a CF_{sp} for all vegetables and grains which represents a conservative average of derived values obtained in this review, such that the probability of underestimating dietary concentrations of ^{210}Pb for the total diet is reduced. A value of 1.0×10^{-2} for a fresh-weight CF_{sp} for crops directly edible by man is thus recommended. This value is lower than the similarly defined value of B_{iv} given in the March 1976 draft of the U.S. NRC's Regulatory Guide 1.109 (1976) of 6.8×10^{-2} . This latter value is derived from data provided by Ng *et al.* (1968).

Using the same rationale as that described in selecting a value for vegetables and grains for forage, hay, and feed, a dry-weight CF_{sp} of 2×10^{-1} is recommended. Assuming factors for dry- to fresh-weight conversion based on data provided in Appendix A of this report for forage, hay, and feed, the recommended dry-weight CF_{sp} corresponds to fresh-weight CF_{sp} 's of 4×10^{-2} , 1.8×10^{-1} , and 7.7×10^{-2} , respectively. These fresh-weight values for ruminant diets closely approximate the

Table 2.2 Measured dietary concentrations of ^{210}Pb in selected New York City food items

Food category	^{210}Pb concentrations (pCi/kg) ^a
Fresh vegetables	1.1
Canned vegetables	0.44
Root vegetables	0.21
Potatoes	1.5
Flour	1.3
Whole grain products	2.2
Milk	0.29
Meat	0.49

^aMorse and Welford, 1971.

fresh-weight B_{iv} value of 6.8×10^{-2} given in the 1976 draft of the U.S. NRC regulatory guide.

2.1.3 Deposition of aerosol ^{210}Pb onto vegetation

Deposition of aerosol lead on crops may contribute to the concentration of this element in edible portions. The degree of contribution is dependent on environmental conditions and plant species (Sect. 2.1.1). The resultant dietary lead concentrations due to deposition may be further influenced by the handling of the food item before consumption. For example, washing of fresh vegetables may remove some ^{210}Pb which has deposited on the food item but has not been incorporated into plant tissues.

Some investigators have conducted lead uptake studies for edible crops in environments where aerosol lead was present in significant amounts. These studies were conducted outdoors, where natural and technologically enhanced sources could contribute to the crop concentrations. Values of CF_{ep} (where the subscript "ep" designates the total environmental source, rather than soil alone) derived from these studies are listed in Table 2.3. These soil-to-plant ratios were calculated in the same manner as those for Table 2.1. Each mean CF_{ep} value is identified as to whether the samples were washed or not when possible. Unfortunately, the information was not sufficient to allow a comparison between washed and unwashed samples, although it seems possible that washing could remove some of the deposited lead before analysis.

Comparison of data in Table 2.3 with that in Table 2.1, where aerosol lead did not contribute to the plant concentrations, indicates that aerosol lead may have a more significant role in determining edible plant concentrations for forage and some grains than for vegetables. To compare fresh-weight CF_{ep} 's, the average forage, feed, and hay CF_{ep} may be converted from 3.5×10^{-1} to 7.0×10^{-2} , 1.3×10^{-1} , and 3.2×10^{-1} for these crops, respectively, using information available in Appendix A (see Sect. 2.1.2). It seems reasonable that the CF_{ep} for crops which have a relatively large surface-to-volume ratio, such as grasses and possibly grains, would exceed that for more protected crops with lower surface-to-volume ratios. The CF_{ep} for edible portions of vegetable

Table 2.3. Concentration factors representing soil-to-plant transfer of ^{210}Pb ($[\text{Pb}]_{\text{plant}} : [\text{Pb}]_{\text{soil}}$) including aerosol ^{210}Pb contribution

Edible plant portion	Mean concentration factors (CF_{ep}) ^a	Number of derived values	Range ($\times 10^{-3}$)	Reference
	($\times 10^{-3}$)			
Vegetables (fresh weight)				
Bean	3.7	3 ^b	3.3-4.4	c
Cabbage	1.5	3 ^b	0.7-2.2	c
Carrot	4.7	2 ^b	2.4-7.1	c
Corn	3.7	3 ^b	1.3-7.9	c
Lettuce	7.3	17 ^d	1.6-14	c, e, f
Potato	3.6	3 ^b	1.6-4.4	c
Radish	1.3	6 ^d	0.6-1.9	f, g
Tomato	1.9	9 ^d	<0.1-3.5	e, f
Unweighted average	3.5	46	<0.1-14	
Grain (fresh weight)				
Oats	11	2 ^b	3.6-18	c
Wheat	12	3 ^b	9.0-18	c
Unweighted average	12	5	3.6-18	
Forage, hay, feed (dry weight)				
Oat tops	370	8 ^b	110-1300	e
Fodder	250	9 ^h	11-510	c
Forage	420	29 ^d	140-1400	g, i, j
Unweighted average	350	46	11-1400	

^aValues express ratios of fresh weight ^{210}Pb concentrations in plants to dry-weight concentrations in soil for all food crops directly edible by man. Dry-weight concentrations in both plants and soil were used for forage, hay, and feed calculations.

^bWashed samples.

^cTer Haar, 1970.

^dCombination of washed and unwashed samples.

^eRabinowitz, 1970.

^fRoberts *et al.*, 1974.

^gDedolph *et al.*, 1970.

^hUnknown if samples were washed.

ⁱChow, 1970.

^jSharma and Shupe, 1977.

crops is quite similar to the CF_{sp} for these same crops. One might expect lettuce to intercept significantly more aerosol lead per unit weight than other non-leafy vegetable types, as is indicated in a comparison of CF_{sp} and CF_{ep} for this crop.

It is important to quantify aerosol deposition of lead on some crops on the basis of this data. Empirical data relating lead concentrations in edible portions of crops to aerosol concentrations alone is not available since the soil in which crops are grown always contains some concentration of the element due to the airborne lead. Therefore, a factor expressing the ratio of atmosphere lead concentrations to those in crops cannot readily be derived.

The U.S. NRC Regulatory Guide 1.109 (drafts, March 1976 and October 1977) provides a model to estimate the concentration of radionuclides in table-ready food crops, C_i^v , due both to direct aerosol deposition on vegetative tissues and to deposition on soil with subsequent root uptake. This model is expressed, for the location (r, θ) , as:

$$C_i^v(r, \theta) = d_i(r, \theta) \left\{ \frac{r[1 - \exp(-\lambda_{Ei} t_e)]}{Y_v \lambda_{Ei}} + \frac{B_{iv}[1 - \exp(-\lambda_i t_b)]}{P \lambda_i} \right\} \exp(-\lambda_i t_h) .$$

where

d_i = the deposition rate of nuclide i ($\text{pCi}/\text{m}^2\text{-hr}$),

r = the fraction of deposited activity retained on crops (dimensionless),

λ_{Ei} = the effective removal rate constant for radionuclide i from crops (hr^{-1}), for all loss processes except radioactive decay,

t_e = the time period that crops are exposed to contamination during the growing season (hours),

Y_v = the agricultural productivity (kg/m^2 wet weight),

B_{iv} = the concentration factor for uptake of radionuclide i from soil by edible parts of crops (fresh weight ratio),

λ_i = the radioactive decay constant of nuclide i (hr^{-1}),

t_b = the period of time for which soil is exposed to contaminated water (hours),

P = the effective surface density of soil (kg/m^2 dry soil),

t_h = the time interval between harvest and consumption of the food (hours).

The first term in this model is of interest in quantifying aerosol deposition of ^{210}Pb alone, and thus the term

$$B_{iv} \frac{[1 - \exp(-\lambda_i t_b)]}{P\lambda_i}$$

will be ignored in the calculation for the present purposes.

Using the values of r , λ_{Ei} , t_e , and Y_v suggested in the guide for forage and other crops, the dietary concentration of ^{210}Pb may be predicted in these items if a deposition rate d_i is given.

If a total deposition velocity, including wet and dry processes, of 3×10^{-3} m/sec is assumed, which corresponds to a 4-day aerosol residence time for ^{210}Pb (Travis *et al.*, 1978), a deposition rate of 3×10^{-3} pCi/m²-sec may be determined. Using this value and the U.S. NRC model, values of 1.1×10^3 pCi/kg and 4.9×10^2 pCi/kg per pCi $^{210}\text{Pb}/\text{m}^3$ in air, for forage and other crops, respectively, may be calculated. If the average U.S. air concentration of ^{210}Pb is assumed to be 2×10^{-2} pCi/m³ (Magno *et al.*, 1970), the concentration of ^{210}Pb in dietary foodstuffs due to aerosol deposition is 22 pCi/kg for forage (fresh weight), and 9.8 pCi/kg for other crops (fresh weight). Again, comparing these values to measured values reported in Table 2.2 (Sect. 2.1.2), the predictions based on the NRC model for aerosol deposition overestimate the actual concentrations even more so than the root uptake predictions. However, because information to quantify deposition more accurately is lacking, it is recommended that this model be used for the present.

2.1.4 Uptake and distribution of ^{210}Pb by grazing animals

The distribution of ^{210}Pb in tissues of certain herbivores following ingestion is important with respect to the potential for human consumption of the contaminated tissue. The transfer of stable lead to the muscle and milk of cattle ingesting contaminated feed and forage has

been investigated for a variety of lead sources including lead mines and smelters (Hammond and Aronson, 1964; Donovan *et al.*, 1969; Djuric *et al.*, 1971; Kerin and Kerin, 1971), automobile exhaust (Bovay, 1971), and sewage sludge containing lead (Nelmes *et al.*, 1974). In addition, the transfer of ^{210}Pb to caribou and reindeer muscle from rumen contents has been studied as a potential source of human exposure in certain populations (Holtzman, 1966; Blanchard and Kearney, 1967). The results of these studies have been used in the following sections to derive factors describing the relationship between ^{210}Pb in meat and milk and ruminant's dietary intake of the isotope.

2.1.4.1 Distribution in milk. Concentrations of lead in milk resulting from ingestion of contaminated feed by dairy cattle (Donovan *et al.*, 1969; Bovay, 1971; Djuric *et al.*, 1971; Kerin and Kerin, 1971; Nelmes *et al.*, 1974) or acute doses of lead (Stanley *et al.*, 1971) have been measured. Concentration factors, CF_m , expressing the equilibrium lead concentration of milk as a fraction of the chronically ingested concentration in feed were derived from these measurements, along with the transfer coefficients, f_m , or the fraction of the cow's daily ingestion of lead secreted per liter of milk at equilibrium. The values are presented in Table 2.4. Data from the study involving an acute intake of lead by dairy cows (Stanley *et al.*, 1971) were utilized to calculate an f_m , as suggested by Ng, *et al.* (1977), by dividing the total activity recovered in milk by the daily rate of milk secretion. A CF_m was subsequently calculated by multiplying the transfer coefficient by the daily dry-weight intake in kg/day assumed for cattle (Garner, 1971; Ng *et al.*, 1977). The CF_m 's and f_m 's for the "control" in two of the studies differ slightly from those of animals exposed to lead in auto exhaust or sewage sludge (Table 2.4). It is possible that this difference results from exposure to different chemical forms of lead present in contaminated and uncontaminated environments, rather than from natural variation alone.

The average f_m of 1.2×10^{-4} day/liter obtained in this review is only slightly less than the f_m of 2.59×10^{-4} day/liter given by Ng *et al.* (1977), and is of the same order of magnitude as the f_m of 6.2×10^{-4} provided in the March 1976 draft of the U.S. NRC Regulatory Guide

Table 2.4. Concentration factors for ^{210}Pb transfer from diet to milk ($[\text{Pb}]_{\text{milk}} : [\text{Pb}]_{\text{diet}}$) in dairy cattle

Lead source	Mean CF _m ^a ($\times 10^{-3}$)	Mean f _m ^b ($\times 10^{-4}$ day/liter)	Number of derived values	Reference
Individual studies				
Lead mine	0.31	0.31	4	<i>e</i>
Auto exhaust	0.56	0.56	2	<i>d</i>
Control	2.0	2.0	1	<i>d</i>
Sewage sludge	1.3	1.3	1	<i>e</i>
Control	1.6	1.6	1	<i>e</i>
Lead smelter	2.5	2.5	4	<i>f</i>
Acute lead dose	0.11 ^g	0.11	1	<i>h</i>
Unweighted average	1.2	1.2	14	
Range ⁱ	0.018-4.3	0.018-4.3		

^aConcentration factors CF_m derived from lead concentrations in diet (dry weight) and milk (fresh weight); soil ingestion not evaluated as part of diet.

^bTransfer coefficients (f_m) defined as the fraction of the element ingested daily that is secreted per liter of milk at equilibrium.

^cDonovan *et al.*, 1969.

^dBovay, 1971.

^eNelmes *et al.*, 1974.

^fKerin and Kerin, 1971.

^gCalculated from the derived f_m by multiplying by 10 kg/day average dry-weight ingestion.

^hStanley *et al.*, 1971.

ⁱRange of individual values over all studies.

1.109. It is recommended that a value of 2.5×10^{-4} day/liter be used, which represents the upper limit of the mean values obtained here. Using this value, a dry matter intake of 10 kg/day, the CF_{sp} of 0.2 (dry-weight) recommended in Sect. 2.1.2, and an average U.S. ^{210}Pb soil concentration of 1.5 pCi/g, a ^{210}Pb concentration in milk of 0.75 pCi/kg may be predicted due to ^{210}Pb in soil. This value is approximately 2.5 times that measured for milk in the New York City food items (see Table 2.2). Therefore, the f_m recommended does not appear to be greatly inaccurate, although aerosol contributions to the ruminant's diet were neglected in this prediction, which would result in an even greater overprediction if included.

2.1.4.2 Distribution in meat. A concentration factor CF_f representing the ratio of lead concentrations in muscle of mature animals to those in feed or rumen contents, and a transfer coefficient f_f defined as the fraction of the total daily lead ingestion by the herbivore which is present per kg of flesh, in day/kg, were calculated from data supplied by several pertinent studies on lead ingestion by cattle and caribou. The values of these parameters are presented in Table 2.5. The f_f 's are based on dry-weight intake values of 10 and 2.8 kg/day for cattle and caribou, respectively, (Garner, 1971). The overall mean muscle CF_f 's for cattle and caribou, were calculated to be 7.1×10^{-3} and 1.1×10^{-3} , respectively, and the respective mean f_f 's for cattle and caribou were determined to be 7.1×10^{-4} and 3.9×10^{-4} day/kg. For cattle, dietary lead concentrations were based on those determined for feed or forage, while for caribou, the concentrations were based on the lead measurements made on rumen contents.

The average f_f of 7.1×10^{-4} day/kg for cattle derived in this study compares with a value of 2.9×10^{-4} day/kg given in the March 1976 draft of the U.S. NRC regulatory guide. A value of 8.0×10^{-4} day/kg, which is a conservative estimate of the average f_f derived here for cattle, is recommended.

Using this recommended value, and the assumptions stated in Sect. 2.1.4.1, a meat concentration of 2.4 pCi ^{210}Pb /kg may be predicted. The concentration of ^{210}Pb in meat measured in New York City (see Sect. 2.1.2) was 0.49 pCi/kg. Thus, the f_f recommended may result in over-estimates of ^{210}Pb concentrations in meat, although the degree of

Table 2.5. Concentration factors for ^{210}Pb transfer from diet to muscle
 ($[\text{}^{210}\text{Pb}]_{\text{muscle}} : [\text{}^{210}\text{Pb}]_{\text{diet}}$) in beef cattle and caribou

Animal	Lead source	Mean CF_f^a ($\times 10^{-3}$)	Mean f_f^b ($\times 10^{-4}$ day/kg)	Number of derived values	References
Individual studies					
Cow	Auto exhaust	2.4	2.4	1	<i>c</i>
	Control	8.0	8.0	1	<i>c</i>
Cow	Sewage sludge	11	11	1	<i>d</i>
	Control	7.1	7.1	1	<i>d</i>
Caribou	Natural fallout	0.97	3.5	12	<i>e</i>
Caribou	Natural fallout	1.2	4.3	1	<i>f</i>
Unweighted average					
Cow		7.1	7.1	4	
Caribou		1.1	3.9	13	
Range ^g					
Cow		2.4-11	2.4-11		
Caribou		0.25-2.9	0.89-10		

^aConcentration factors (CF_f) derived from lead concentrations in diet (dry weight) and animal muscle (fresh weight); soil ingestion not evaluated as part of diet for cows only.

^bTransfer coefficients (f_f) defined as the fraction of the element ingested daily which appears in each kg of flesh.

^cBovay, 1971.

^dNelmes *et al.*, 1974.

^eBlanchard and Kearney, 1967.

^fHoltzman, 1966.

^gRange of individual values over all studies.

certainty that this single measured value is representative of an average meat concentration is extremely low.

2.1.4.3 Soil ingestion. Soil has not been considered as a direct source of lead for cattle in these studies, although it is known that animals may ingest significant amounts of soil while grazing (Field, 1964; Healy, 1968; Healy *et al.*, 1970; Mayland *et al.*, 1975; and Suttle *et al.*, 1975). Depending on pasture conditions and other factors, soil ingestion can constitute up to 14% of the dry-matter intake by cattle (Suttle *et al.*, 1975) but averages less than 2% over the year (Healy, 1968; Healy *et al.*, 1970; Mayland *et al.*, 1975). Thus, if soil lead is directly ingested on a regular basis but not accounted for along with the forage lead ingested by grazing animals, the calculated values of CF_m , CF_f , f_m , and f_f may overestimate the true relationship between tissue lead and forage lead. By measuring rumen contents, as was done for caribou, the dietary lead concentrations account for ingested soil lead.

Many physical forms of lead found in soil may differ in their digestibility from forms of lead which have been incorporated into plant tissues, even though it has been suggested that the chemical solubility of the ingested lead compound exerts little influence on gastrointestinal (GI) absorption of lead (Stanley *et al.*, 1971). Field (1964) assumes soil to be indigestible except for some nutritionally important minerals, while Healy (1968) assumes a 70% soil digestibility. Thus, it is difficult to estimate the contribution of ingested soil lead to the lead actually incorporated into muscle or milk in cattle. However, since the concentration of lead in soil tends to be much greater than that in forage crops, soil ingestion by cattle is potentially a significant source of dietary lead for cattle, and ultimately, for humans.

2.2 Dose Conversion Factors for ^{210}Pb

2.2.1 Inhalation

Inhalation of airborne ^{210}Pb released to the environment from a large number of sources (see Sect. 2.1.1) will result in an internal radiation dose. The organ receiving the highest dose per unit ^{210}Pb inhaled is bone due to the relatively long effective half-time of the isotope in this organ (Hurst, 1973; Torvik *et al.*, 1974; Lloyd *et al.*,

1975). Fifty-year dose commitment factors (DCF) are provided for several organs, including bone, lung, kidney, liver, and whole body; assuming a quality factor (QF) of 10 for alpha radiation.

Dose factors were derived from use of the INREM-II computer code (Killough *et al.*, 1978), which is an implementation of the Task Group lung model (Task Group on Lung Dynamics, 1966) and other models. Retention functions and other metabolic parameters for ^{210}Pb , ^{210}Bi , and ^{210}Po were selected for use in this code from information in available literature (see Dunning *et al.*, in preparation). Redistribution of ^{210}Pb daughters initially deposited in one organ to other organs was not considered due to insufficient data, although redistribution has been suggested by several investigators (Blanchard and Moore, 1971; Ladinskaya *et al.*, 1973; Torvik *et al.*, 1974).

An activity median aerodynamic diameter (AMAD) of $0.3\ \mu\text{m}$ and solubility class W was assumed in the calculation of ^{210}Pb inhalation DCF. The value f_1 , uptake to blood from the gastrointestinal tract, was assumed to be 0.08 (International Commission of Radiological Protection (ICRP), 1975) for ^{210}Pb . Retention functions for ^{210}Pb in various tissues were adapted for implementation from Bernard (1977), who utilized empirical data from rats, dogs, and baboons to derive his values.

In calculating doses from inhalation of ^{210}Pb , buildup and dose contributions of the ^{210}Bi and ^{210}Po daughters within organs where ^{210}Pb is retained must be considered. In addition, a small amount of ^{210}Bi and ^{210}Po will be formed before ^{210}Pb is deposited in an organ. Thus, values of metabolic parameters for ^{210}Bi and ^{210}Po are necessary.

Little is known about the normal metabolism of ^{210}Bi in man (ICRP, 1975). A value of 0.05 was adopted for f_1 , representing fractional GI absorption of ^{210}Bi formed before absorption of ^{210}Pb from the GI tract is complete. This f_1 value represents a midrange value between those given in ICRP Publication 2 and in ICRP Publication 23 (1959 and 1975, respectively). A solubility class W for any bismuth produced from decay of ^{210}Pb while in the lungs was assumed; this is characteristic of most bismuth compounds (Task Group on Lung Dynamics, 1966). The retention function for ^{210}Bi in all organs was assumed to be identical to that measured for the kidney, where ^{210}Bi has a biological half-time of five

days (ICRP, 1959; Mathews *et al.*, 1964). The highest uptake of ^{210}Bi from blood has been found to occur in the kidney (Sollman, 1957; Durbin, 1960; Eridani *et al.*, 1964), which was found to contain about one-ninth of the total body burden of bismuth after intramuscular injection (Sollman, 1957). It was assumed, therefore, that 10% of ^{210}Bi activity absorbed by the blood is absorbed by the kidney. The remaining 90% was assumed to be distributed among all other organs of the body in equal portions according to organ mass.

The f_1 value for absorption of ^{210}Po formed before ingested ^{210}Pb is absorbed from the GI tract was assumed to be 0.1, based on available empirical data ranging from 3-6% (Anthony *et al.*, 1956) for rats to 10% (Fink, 1950) for man. Again, a solubility class W was chosen, which applies to oxides, hydroxides, and nitrates of polonium (Task Group on Lung Dynamics, 1966). The biological half-time of ^{210}Po in any organ was assumed to be 50 days, based on a summary of human excretion data (Jackson and Dolphin, 1966). The apparent effective half-life of ^{210}Po produced by ^{210}Pb in the skeleton may be equal to that of ^{210}Pb due to a high degree of equilibrium reached between the two isotopes in bone (Blanchard and Moore, 1971). Distribution of ^{210}Po from blood to body organs was assumed as follows: 10%, 10%, 10%, and 70% of ^{210}Po in blood is deposited in liver, kidney, spleen, and all other organs, respectively. These values approximate those recommended by ICRP Committee II (1959), except that kidney and liver depositions of ^{210}Po were not assumed equal. The assumptions involving deposition here are based on experimental data indicating the kidney:liver ratio of ^{210}Po content may be higher than presumed by ICRP (1959) when considering ^{210}Po distribution alone (Blanchard, 1966; Torvik *et al.*, 1974).

These metabolic values were used in conjunction with the INREM-II computer code to derive dose conversion factors for ^{210}Pb inhalation over a one-year period. The dose commitment factors thus calculated are listed in Table 2.6. The breathing rate was based on Reference Man data (ICRP, 1975) and was assumed to be $23 \text{ m}^3/\text{day}$. Bone was found to be the organ receiving the highest dose per unit of ^{210}Pb activity inhaled.

Physiological and anatomical differences in lungs and lung clearance rates among members of the population inhaling ^{222}Rn daughters such as

Table 2.6. Fifty-year dose commitment factors for ^{210}Pb inhalation over a year

Organ	rem/ μCi inhaled	mrem/pCi-m ³
Bone	110	940
Lung	5.6	47
Liver	1.8	15
Kidneys	2.3	19
Whole-body	8.4	71

NOTE ADDED IN PRESS -- The above dose commitment factors (DCF) are based on early drafts of a report by Dunning *et al.* (in preparation). The DCF's given above are based on a quality factor (QF) of 10, which was recommended in ICRP Publication 2 (1959). In more recent drafts of their document, Dunning *et al.* have incorporated recent recommendations from ICRP Publication 26 (Pergamon Press, 1977) regarding quality factors. DCF's based on a higher QF of 20 are given in Appendix C of the present report.

^{210}Pb may result in correspondingly different doses to various critical organs. Dose conversion factors used in this study were calculated on the basis of ICRP Reference Man data (1975), which represents average parameters for adults. It has been suggested, however, that radiation dose from ^{222}Rn decay products may be dependent upon the age of the individual exposed to specific air concentrations (Hoenes and Soldat, 1977; Hofmann and Steinhausen, 1977). This may be due to age-dependent variations in airway diameters (Desrosiers, 1977), lung masses, and/or physiological processes (Hoenes and Soldat, 1977; Hofmann and Steinhausen, 1977). An estimate that the dose to the respiratory tract may be two to three times higher for individuals between the ages of five and seven years than for adults was made by one group of researchers (Hofmann and Steinhausen, 1977). Another group estimated age-dependent factors based on differences in size and mass of certain organs (neglecting differences in biological clearance) for four age groups from inhalation of ^{222}Rn , ^{210}Pb , and associated decay products (Hoenes and Soldat, 1977). They indicated a dose conversion factor approximately 3.5 times greater for infants than adults. However, it was felt that sufficient information on age-dependency with respect to dose conversion factors was not available at this time, because all variables (including lung clearance rates) were not considered simultaneously as dose determinants in any of the pertinent studies.

2.2.2 Ingestion

Ingestion of ^{210}Pb may occur through consumption of foodstuffs exposed to air or soil concentrations of the isotope, as indicated in previous sections. The resulting internal dose commitment may again be quantified with the use of fifty-year dose commitment factors determined by Dunning *et al.* (in preparation), using the INREM II computer code (Killough *et al.*, 1978). Metabolic parameters describing fractional transfer of ^{210}Pb , ^{210}Bi , and ^{210}Po from the GI tract to various organs were again necessary in developing these factors. Documentation of these parameters is provided in Sect. 2.2.1.

Fifty-year DCF's for ^{210}Pb ingestion are listed in Table 2.7 for whole body, bone, kidney, and liver. Again, the organ receiving the greatest dose per unit ingestion is bone. The DCF's presented here are

Table 2.7. Fifty-year dose commitment factors for ^{210}Pb ingestion

Organ of reference	rem/ μCi ingested
Bone	52
Kidney	0.49
Liver	0.75
Whole-body	3.8

NOTE ADDED IN PRESS — The above dose commitment factors (DCF) are based on early drafts of a report by Dunning *et al.* (in preparation). The DCF's given above are based on a quality factor (QF) of 10, which was recommended in ICRP Publication 2 (1959). In more recent drafts of their document, Dunning *et al.* have incorporated recent recommendations from ICRP Publication 26 (Pergamon Press, 1977) regarding quality factors. DCF's based on a higher QF of 20 are given in Appendix C of the present report.

dependent on the values assumed for metabolic parameters. It has been suggested that lead metabolism in children may differ significantly from that in adults (Blanchard, 1966; Alexander *et al.*, 1973; Karhausen, 1973; Kolbye *et al.*, 1973); although the present metabolic values are based on the average adult. It has been suggested also that absorption and/or retention of ingested lead in tissues and organs may be greater for children (Alexander *et al.*, 1973; Karhausen, 1973; Kolbye, *et al.*, 1973). However, it was felt that accurate numbers to express age dependency of dose were not available.

2.3 References

- Alexander, F. W., H. T. Delves and B. E. Clayton, "The Uptake and Excretion by Children of Lead and Other Contaminants," pp. 319-330 in *Proceedings - International Symposium on the Environmental Health Aspects of Lead*, organized jointly by CEC and EPA, published by CEC, Luxembourg (May 1973).
- Anthony, D. S., R. K. Davis, R. N. Cowden, and W. P. Jolley, "Experimental Data Useful in Establishing Maximum Permissible Single and Multiple Exposures to Polonium," pp. 215-218 in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy*, Vol. 13, United Nations, New York (1956).
- Blanchard, R. L., "Relationship Between ^{210}Po and ^{210}Pb in Man and His Environment," pp. 281-296 in *Radioecological Concentration Processes, Proceedings of an International Symposium held in Stockholm, 25-29 April 1966*, Pergamon Press, pp. 281-96, 1966.
- Blanchard, R. L. and J. W. Kearney, "Natural Radioactivity and ^{137}Cs in Alaskan Caribou and Reindeer Samples," *Environ. Sci. Technol.* 1, 932-939 (1967).
- Blanchard, R. L. and J. B. Moore, "Body Burden, Distribution, and Internal Dose of ^{210}Pb and ^{210}Po in a Uranium Miner Population," *Health Phys.* 21, 499-518 (1971).
- Bovay, E., "Accumulation of Pb on Vegetation along Expressways. Feeding Tests on Dairy Cows with Forage Contaminated with Pb," pp. 101-117 in Switzerland Federal Committee for Air Hygiene Investigations: The Problem of Leaded Gasoline, *Bull. der Eidg. Gesundheitsamtes*, Suppl. B, No. 3 (1971).
- Chow, T. J., "Pb Accumulation in Roadside Soil and Grass," *Nature* 225, 295-296 (1970).
- Cox, W. J. and D. W. Rains, "Effect of Lime on Pb Uptake by Five Plant Species," *J. Environ. Qual.* 1(2), 167-171 (1972).
- Dedolph, R., G. Ter Haar, R. Holtzman and H. Lucas, Jr., "Sources of Pb in Perennial Ryegrass and Radishes," *Environ. Sci. Technol.* 4(3), 217-225 (1970).
- Desrosiers, A. E., "Alpha Particle Dose to Respiratory Airway Epithelium," *Health Phys.* 32, 192-195 (1977).

- Djuric, D., Z. Kerin, L. Graovac-Leposavic, L. Novak, and M. Kop, "Environmental Contamination by Lead from a Mine and Smelter," *Arch. Environ. Health* 23, 275-279 (1971).
- Donovan, P. P., D. T. Feeley, and P. P. Canavan, "Lead Contamination in Mining Areas in Western Ireland II - Survey of Animals, Pastures, Foods and Waters," *J. Sci. Fd. Agric.* 20, 43-45 (1969).
- Dunning, D. E., Jr., S. R. Bernard, P. J. Walsh, G. G. Killough, and J. C. Pleasant, *Estimates of Internal Dose Equivalents to 22 Target Organs for Radionuclides Occurring in Routine Releases from Nuclear Fuel Cycle Facilities*, Vol. II, ORNL/NUREG/TM-190/V2, NUREG/CR-0114/V2 (in preparation).
- Durbin, P. W., "Metabolic Characteristics Within a Chemical Family," *Health Phys.* 2, 225-238 (1960).
- Eridani, S., M. Balzarini, D. Taglioretti, M. Romussi, and R. Valentini, "The Distribution of Radiobismuth in the Rat," *Br. J. Radiol.* 37, 311-314 (1964).
- Field, A. C., "The Intake of Soil by the Grazing Sheep," (abstract) in *Proceedings of Nutrition Society* 23, xxiv (1964).
- Fink, R. M., ed., *Biological Studies with Polonium, Radium and Plutonium*, McGraw-Hill Book Company, Inc., New York, pp. 77-84 and 140-148 (1950).
- Fisenne, I. M., "Distribution of ^{210}Pb and ^{226}Ra in Soil," pp. 145-158 in USAEC Report UCRL-18140 (1968).
- Garner, R. J., "Transfer of Radioactive Materials from the Terrestrial Environment to Animals and Man," *CRC Critical Reviews in Environmental Control* 2, 337-385 (1971).
- Hammond, P. B. and A. L. Aronson, "Pb Poisoning in Cattle and Horses in the Vicinity of a Smelter," *Ann. N.Y. Acad. Sci.* 111(2), 595-611 (1964).
- Healy, W. B., "Ingestion of Soil by Dairy Cows," *N. Zealand J. Agric. Res.* 11, 487-499 (1968).
- Healy, W. B., W. J. McCabe and G. F. Wilson, "Ingested Soil as a Source of Microelements for Grazing Animals," *N. Zealand J. Agric. Res.* 13, 503-521 (1970).
- Hevesy, G., "The Absorption and Translocation of Pb by plants," *Biochem. J.* 17, 439-445 (1923).

- Hoenes, G. R., and J. K. Soldat, *Age-Specific Dose Commitment Factors for a One-Year Chronic Intake*, NUREG-0172 (1977).
- Hofmann, W. and F. Steinhausler, "Dose Calculations for Infants and Youths Due to the Inhalation of Radon and Its Decay Products in the Normal Environment," IRPA Publication 4, p. 497 (1977).
- Holtzman, R. B., "²²⁶Ra and the Natural Airborne Nuclides ²¹⁰Pb and ²¹⁰Po in Arctic Biota," pp. 1087-96 in *Proceedings of the First International Congress of Radiation Protection*, Part 2 (1966).
- Hurst, J. B., "Retention of ²¹⁰Pb in Beagle Dogs," *Health Phys.* 25, 29-35 (1973).
- International Commission on Radiological Protection, *Report of Committee II on Permissible Dose for Internal Radiation*, ICRP Publication 2 (1959).
- International Commission on Radiological Protection, *Report of the Task Group on Reference Man*, ICRP Publication 23 (1975).
- Jackson, W. and G. W. Dolphin, "The Estimation of Internal Radiation Dose from Metabolic and Urinary Excretion Data for a Number of Important Radionuclides," *Health Phys.* 12, 481-500 (1966).
- Jaworowski, Z. and D. Grzybowska, "Natural Radionuclides in Industrial and Rural Soils," *Sci. Total Environ.* 7, 45-52 (1977).
- Jaworowski, Z. and L. Kownacka, "Lead and Radium in the Lower Stratosphere," *Nature* 263, 303-304 (1976).
- John, M. K., "Lead Availability Related to Soil Properties and Extractable Lead," *J. Environ. Qual.* 1(3), 295-298 (1972).
- John, M. K. and C. J. Van Laerhoven, "Lead Distribution in Plants Grown on Contaminated Soil," *Environ. Lett.* 3(2), 111-116 (1972a).
- John, M. K. and C. Van Laerhoven, "Lead Uptake by Lettuce and Oats as Affected by Lime, Nitrogen, and Sources of Lead," *J. Environ. Qual.* 1(2), 169-171 (1972b).
- Jones, L. H. P., C. R. Clement and M. J. Hopper, "Lead Uptake from Solution by Perennial Ryegrass and Its Transport from Roots to Shoots," *Plant and Soil* 38(2), 403-414 (1973).
- Karhausen, L., "Intestinal Lead Adsorption," in *Proceedings - International Symposium in the Environmental Health Aspects of Lead*, organized jointly by CEC and EPA, published by CEC, Luxembourg (May 1973).

- Kerin, D., and Z. Kerin, "Lead Contamination of Milk and Honey Caused by Industrial Aerosols," *Protectio Vitae* 2, 61-62 (1971).
- Killough, G. G., D. E. Dunning, Jr., and J. C. Pleasant, *INREM-II: A Computer Implementation of Recent Models for Estimating the Dose Equivalent to Organs of Man from an Inhaled or Ingested Radionuclide*, ORNL/NUREG/TM-84 (1978).
- Koepe, D. E., "The Uptake, Distribution, and Effect of Cadmium and Lead in Plants," *The Science of the Total Environment* 7(3), 197-206 (1977).
- Kolbye, A. C., Jr., K. R. Mahaffey, J. A. Fiorino, P. C. Corneliusen, and C. F. Jelinek, "Food Exposures to Lead," *Environ. Health Perspect.* 7, 65-74 (1973).
- Ladinskaya, L. A., D. Y. Parfenov, D. K. Popov, and A. V. Fedorova, "²¹⁰Pb and ²¹⁰Po Content in Air, Water, Foodstuffs, and the Human Body," *Arch. Environ. Health* 27, 254-258 (1973).
- Lloyd, R. D., C. W. Mays, D. R. Atherton, and F. W. Bruenger, "²¹⁰Pb Studies in Beagles," *Health Phys.* 28, 575-583 (1975).
- MacLean, A. J., R. L. Halstead, and B. J. Finn, "Extractability of Added Lead in Soils and Its Concentration in Plants," *Can. J. Soil Sci.* 49, 327-334 (1969).
- Magno, P. J., P. R. Groulx, and J. C. Apidianakis, "Pb-210 in Air and Total Diets in the United States During 1966," *Health Phys.* 18, 383-388 (1970).
- Mathews, C. M. E., W. J. Dempster, C. Kapros, and S. Kountz, "The Effect of Bismuth-206 Irradiation on Survival of Skin Homografts," *Br. J. Radiol.* 37, 306-310 (1964).
- Mayland, H. F., A. R. Florence, R. C. Rosenau, V. A. Lazar and H. A. Turner, "Soil Ingestion by Cattle on Semiarid Range as Reflected by Titanium Analysis of Feces," *J. Range. Mgt.* 28(6), 448-452 (1975).
- Menzel, R. G., "Soil-Plant Relationships of Radioactive Elements," *Health Phys.* 11, 1325-1332 (1965).
- Miller, R. J. and D. E. Koepe, "Accumulation and Physiological Effects of Lead in Corn," pp. 186-193 in *Proceedings of 4th Annual Conference on Trace Substances in Environmental Health*, Univ. of Missouri, Columbia, Mo. (1970).

- Moore, H. E., E. A. Martell, and S. E. Poet, "Sources of Polonium-210 in Atmosphere," *Environ. Sci. Technol.* 10, 586-591 (1976).
- Morse, R. S. and G. A. Welford, "Dietary Intake of ^{210}Pb ," *Health Phys.* 21, 53-55 (1971).
- Nelms, A. J., R. St. J. Buxton, F. A. Fairweather and A. E. Martin, "The Implication of the Transfer of Trace Metals from Sewage Sludge to Man," pp. 145-52 in Hemphill, D. D. (ed.), *Proceedings of 8th Annual Conference on Trace Substances in Environmental Health*, Univ. of Missouri, Columbia, Mo. (June 1974).
- Ng, Y. C., C. A. Burton, S. E. Thompson, R. K. Tandy, H. K. Kretner, and M. W. Pratt, *Prediction of the Maximum Dosage to Man from the Fallout of Nuclear Devices IV. Handbook for Estimating the Maximum Internal Dose from Radionuclides Released to the Biosphere*, UCRL-50163, Part IV (1968).
- Ng, Y. C., C. S. Colsher, D. J. Quinn and S. E. Thompson, *Transfer Coefficients for the Prediction of the Dose to Man via the Forage - Cow-Milk Pathway from Radionuclides Released to the Biosphere*, UCRL-51939 (July 1977).
- Rabinowitz, M., "Plant Uptake of Soil and Atmospheric Lead in Southern California," *Chemosphere* 1(4), 175-180 (1972).
- Rains, D. W., "Pb Accumulation by Wild Oats (*Avena fatua*) in a Contaminated Area," *Nature* 233(5314), 210-211 (1971).
- Roberts, T. M., W. Gizyn and T. C. Hutchinson, "Lead Contamination of Air, Soil, Vegetation and People in the Vicinity of a Secondary Lead Smelter," pp. 155-166 in D. D. Hemphill (ed.) *Proceedings of 8th Annual Conference on Trace Substances in Environmental Health*, Univ. of Missouri, Columbia, Mo. (1974).
- Sharma, R. P. and J. L. Shupe, "Lead, Cadmium, and Arsenic Residues in Animal Tissues in Relation to Those in Their Surrounding Habitat," *The Science of the Total Environment* 7(1), 53-62 (1977).
- Sollman, T. H., *A Manual of Pharmacology and Its Application to Therapeutics and Toxicology*, 8th ed., W. B. Saunders Co., Philadelphia, Penn. (1957).
- Stanley, R. E., A. A. Mullen, and E. W. Bretthauer, "Transfer to Milk of Ingested Radiolead," *Health Phys.* 21, 211-215 (1971).

- Suttle, N. F., B. J. Alloway, and I. Thornton, "An Effect of Soil Ingestion on the Utilization of Dietary Copper by Sheep," *J. Agric. Sci. Cambridge* 84, 249-254 (1975).
- Task Group on Lung Dynamics, "Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract," *Health Phys.* 12, 173-207 (1966).
- Ter Haar, G. L., R. B. Holtzman, and H. F. Lucas, Jr., "Lead and Lead-210 in Rainwater," *Nature* 216, 353-355 (1967).
- Ter Haar, G., "Air as a Source of Lead in Edible Crops," *Environ. Sci. Technol.* 4(3), 226-230 (1970).
- Torvik, E., E. Pfitzer, J. G. Kereiakes, and R. Blanchard, "Long-Term Effective Half-lives for Lead-210 and Polonium-210 in Selected Organs of the Male Rat," *Health Phys.* 26, 81-87 (1974).
- Travis, C. C., A. P. Watson, L. M. McDowell-Boyer, S. J. Cotter, M. F. Randolph, and D. E. Fields, *A Radiological Assessment of Radon-222 Released from Uranium Mills and Other Natural and Technologically Enhanced Sources*, NUREG/CR-0573, ORNL/NUREG-55, February, 1979.
- U.S. Nuclear Regulatory Commission, *Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I, Regulatory Guide 1.109* (drafts March 1976, October 1977).
- Zimdahl, R. L. and J. M. Foster, "The Influence of Applied Phosphorus, Manure, or Lime on Uptake of Lead from Soil," *J. Environ. Qual.* 5(1), 31-34 (1976).

3 RECOMMENDATIONS OF ENVIRONMENTAL TRANSPORT PARAMETERS AND DOSE CONVERSION FACTORS FOR ^{226}Ra

Radium-226 exists in the biosphere as a long-lived decay product ($T_{1/2} = 1620$ year) of the primordial nuclide ^{238}U . (Kocher, 1977) As such, it is present in crude uranium and yellowcake dust emissions from active uranium mills (Sears *et al.*, 1975, 1977), as well as in emissions from other anthropogenic sources. Abandoned tailings ponds and unstabilized tailings piles from inactive uranium mill sites are also known to be sources of finely divided radium-bearing dusts. Recent interest in possible transfer of this nuclide within food chains near technologically enhanced sources has led to the present evaluation of ^{226}Ra transport from air and soil to human dietary items. Recommendations for parameters to be used in environmental transport models are presented in this chapter. Fifty-year dose commitment factors for inhalation and ingestion of ^{226}Ra are also provided.

3.1 Environmental Transport Parameters

To quantify the transport of ^{226}Ra through food chains, aerosol deposition on, and root uptake and translocation by, vegetation must be considered. As with ^{210}Pb , transfer of ^{226}Ra to meat and milk of herbivores should also be evaluated. A literature review of radium dynamics in the environment was conducted, and recommendations of transport parameters appropriate for use in environmental models were derived. A discussion of the availability to plants of soil radium is included, to underscore the importance of obtaining site-specific information regarding the physical and chemical forms of the element.

3.1.1 The source of ^{226}Ra in edible crops

The relative importance of aerosol deposition versus root uptake in the contamination of edible plant parts with environmental ^{226}Ra is variable. The significance of aerosol deposition will depend on morphological and structural characteristics of the plant part in question, aerosol concentration, particle characteristics, and climate factors (Garner, 1971).

Natural sources of airborne ^{226}Ra seem to be inorganic matter originating from volcanic activity, wind transport of soil and water, forest

fires, and other meteoric material (Jaworawski *et al.*, 1971). Moore and Poet (1976) measured what they believed to be natural levels of ^{226}Ra in air and found a mean of 4.4×10^{-5} dpm/m³ or 2.0×10^{-5} pCi/m³ from 13 samples. Anthropogenic sources of ^{226}Ra , including coal-fired power plants, uranium mill tailings piles, and phosphate fertilizers, may elevate air concentrations in the vicinity of the sources (Eisenbud and Petrow, 1964; Breslin and Glauberman, 1970; Jaworawski *et al.*, 1971; Jaworawski and Kownack, 1976; Moore and Poet 1976). Breslin and Glauberman (1970) found ^{226}Ra air concentrations of 0.04 to 0.2 pCi/m³ near tailings piles. These concentrations were reduced by about a factor of 10 at 1 km from the site. Air in the vicinity of a coal-fired power plant in Alabama was found to contain 3.1×10^{-4} to 1.3×10^{-3} pCi $^{226}\text{Ra}/\text{m}^3$ (National Council on Radiation Protection and Measurements, 1975). Air concentrations in New York City have been measured by Eisenbud and Petrow (1964) to be 8.0×10^{-5} pCi $^{226}\text{Ra}/\text{m}^3$, which is somewhat greater than the previously cited natural value of 2.0×10^{-5} pCi $^{226}\text{Ra}/\text{m}^3$.

Phosphate fertilizers are probably the major anthropogenic contributors of aeriaily deposited ^{226}Ra on edible crops in most, if not all, agricultural areas. Airborne concentrations of ^{226}Ra due to resuspension of soil treated with these fertilizers, however, should not significantly exceed concentrations due to natural soil resuspension since the soil ^{226}Ra enhancement from fertilizer use is insignificant in comparison to the natural soil ^{226}Ra content (Travis *et al.*, 1979). Thus, natural airborne ^{226}Ra will probably determine the extent of foliar or surface contamination of edible plant parts in most crop production areas of the United States.

The relative importance of soil and air as sources of ^{226}Ra in edible crops will again depend on the conditions of environmental contamination, and the types of crops considered (Sect. 2.1.1). For naturally occurring ^{226}Ra , it seems likely that root uptake and translocation to edible portions is the primary contamination pathway. However, in areas directly downwind of anthropogenic ^{226}Ra aerosol sources, aerosol deposition may become a more significant source of ^{226}Ra in edible crops, especially in leafy vegetables or forage crops.

The chemistry of the ^{226}Ra isotope in soil is important in determining its mobility in soil and the degree of incorporation into biological tissues of plants (Kovaleskii, 1962; Rusanova, 1964; Taskayev *et al.*, 1977; Verkhovskaja *et al.*, 1966; Grzybowska, 1974). Although the physical and chemical forms of airborne ^{226}Ra from various sources is largely unknown, it appears that soil properties at the deposition site also greatly determine its ultimate availability to plant roots.

Soil radium has been found to exist in water-soluble, exchangeable, acid-soluble, and fixed phases (Rusanova, 1964; Verkhovskaja *et al.*, 1966, Taskayev *et al.*, 1977; Grzybowska, 1974). Available literature deals primarily with natural ^{226}Ra introduced into soil by rock weathering although radium may also have industrial sources.

There is evidence that radium migrates downward in all soils over time, but is concentrated at different depths (Rusanova, 1964; Taskayev *et al.*, 1966; Grzybowska, 1974), depending on the organic content and pH of the soil horizons encountered. Most investigators believe that radium concentrates more readily in soils with greater organic content, and that it becomes bound and accumulates in organic complexes (Rusanova, 1964; Taskayev *et al.*, 1977; Verkhovskaja *et al.*, 1966; Grzybowska 1974). The element also seems to be more prevalent in soils with a basic to neutral pH (Rusanova, 1964; Verkhovskaja *et al.*, 1966).

The concentration of other alkaline-earth elements in soils also seems to influence the form of radium in soil. It has generally been believed that radium and calcium have similar reactions in soil. More recent evidence indicates positive correlations between the chemistry of barium and radium in soil, but not between calcium and radium (Rusanova, 1964; Taskayev *et al.*, 1977; Grzybowska, 1974). Most investigators do agree, however, that the amount of mobile radium in soil tends to decrease with increasing concentrations of other mobile alkaline-earth elements (Rusanova, 1964; Taskayev *et al.*, 1977; Verkhovskaja *et al.*, 1966).

The mobile fraction of radium was found to range from 28.6 to 33.1% of total radium among different soils in an analysis by Taskayev and his colleagues (1966). Another researcher found the mobile fraction to range between 2.9 and 30.9% (Grzybowska, 1974). Thus, most radium in soil is strongly fixed in the solid phase (Taskayev *et al.*, 1977).

It was found that mobile radium can occur in the following forms:

- 1) free ionic, in which it enters plant roots,
- 2) as part of soluble mineral salts or organic complexes,
- 3) bound with carbonates, oxalates and less soluble compounds, or
- 4) bound at the surface of colloidal micelles.

Only a small fraction of the mobile radium would be directly available for root uptake in the free ionic form.

The entire mobile fraction, however, may be said to constitute the available pool, since the forms are not static. That is, previously acid-soluble or exchangeable radium may enter the water-soluble phase to re-establish equilibrium conditions as radium is depleted from the available phase. Increased organic content of soil may decrease the immediate availability of ^{226}Ra for plant uptake (Grzybowska, 1974).

Although the above parameters are essential in gaining an understanding of ^{226}Ra availability to plants, many investigators of soil-plant relations fail to measure or report this critical information. Thus, the following review of pertinent literature concerning root uptake of ^{226}Ra from soil does not lead to an evaluation of the influence of soil type on plant concentrations of the nuclide.

3.1.2 Root uptake and distribution of ^{226}Ra by vegetation

Root uptake of ^{226}Ra from soil is influenced by soil type, as well as factors such as soil pH, soil content of other alkaline-earth elements (calcium, barium, and strontium), plant species, and chemical form of the soil ^{226}Ra (Rusanova, 1964; Verkhovskaja *et al.*, 1966; Kirchmann *et al.*, 1968; Garner, 1971; and Grzybowska, 1974). Numerous studies have been performed to investigate the behavior of ^{226}Ra in the soil-plant system. Some investigators have considered the available and non-available forms of this alkaline-earth element in soil with respect to root uptake in an attempt to identify the biologically mobile forms of ^{226}Ra present. Others have restricted their studies to the quantitative analysis of ^{226}Ra in the plant compartment with respect to the total soil ^{226}Ra . Radium-226 incorporation into plant tissues from nutrient solutions has also been investigated to a lesser degree. However, studies quantifying uptake from nutrient solutions are less useful for our purposes, since retention of ^{226}Ra by soil particles and its interaction

with soil components are important elements in determining the efficiencies of transfer to plants (Rusanova, 1964; Verkhovskaja *et al.*, 1966; Garner, 1971; Grzybowska, 1974; Taskayev *et al.*, 1977). Mobility of ^{226}Ra within plant tissues appears to be high during transport from root to shoot, but low after deposition in leaf tissues. The result is acropetal concentration, and a potential for enhanced uptake by animals from forage or leafy vegetables (Rusanova, 1964; Verkhovskaja *et al.*, 1966; Grzybowska, 1974; Taskayev *et al.*, 1977). The dynamics of translocation to other non-leafy edible portions of plants has not been documented.

Since naturally occurring ^{226}Ra originates in the soil and is not a major component of natural fallout (Sect. 3.1.1), the question of direct foliar uptake has generally been neglected in studies of radium uptake by plants. From the previous discussion, however, technologically enhanced sources of radium may generate airborne concentrations of the nuclide which may be significant in some areas. In addition, radium may be resuspended with soil. Therefore, the concentration factors derived here (CF_{sp}) describing soil-to-plant transfer of radium may overestimate the actual ratio if aerosol radium was a significant source of plant radium at the study site.

Values of CF_{sp} were calculated from existing literature available for edible crops and are presented in Table 3.1 for several plant species. The CF_{sp} 's are given in dry weight (DW) concentrations for forage, hay, and feed, and in fresh weight (FW) concentrations for vegetables and grains, consistent with Sect. 2.1.2. When necessary, conversions of data to fresh or dry weight were made with the use of information supplied in Appendix A. Unweighted averages of the mean CF_{sp} 's computed for each species are given for each food category, along with the range of individual values for the various sources, as was done in Sect. 2.1.2. The ranges indicate that much uncertainty is involved in determining a single value for CF_{sp} . Much of this uncertainty is probably due to the variability in experimental conditions among the studies cited.

From Table 3.1, it appears that grains and vegetables tend to concentrate ^{226}Ra more efficiently than fruit. This may be due to fixation in foliage before maturation of the fruit, or natural variation in the data obtained. The respective average fresh weight CF_{sp} 's for vegetables,

Table 3.1. Concentration factors representing soil-to-plant transfer of ^{226}Ra ($[\text{}^{226}\text{Ra}]_{\text{plant}} : [\text{}^{226}\text{Ra}]_{\text{soil}}$)^a

Edible plant portion	Mean concentration factors (CF_{sp}) ^b ($\times 10^{-2}$)	Number of derived values	Range ($\times 10^{-2}$)	Reference
Vegetables (fresh weight)				
Beet	1.8 ^c	2	0.6-3.0	d
Cabbage	1.6 ^c	5	<0.01-4.0	d, e
Carrot	2.0 ^c	2	1.0-3.0	d
Potato	0.3 ^c	3	0.07-0.6	d, f
Unspecified ^g	0.1	1	0.1	d, f
Unweighted average	1.2	13	<0.01-4.0	
Fruit (fresh weight)				
Unspecified ^g	0.05	1	0.05	f
Grain (fresh weight)				
Barley	3.7 ^c	2	1.4-6.0	i
Buckwheat	130	1	130	i
Flour	0.5	1	0.5	f
Millet	40	1	40	i
Wheat	100	1	100	i
Unweighted average	55	5	0.5-130	
Forage, hay, feed (dry weight)				
Alfalfa hay	7.0	24	1.0-34	j
Clover	21	6	1.1-48	d, k, l
Fescue	2.8	1	2.8	k
Grass	13	28	2.0-63	i
Herbage	0.8	1	0.8	f
Rye grass	24	24	2.0-62	d, k
Timothy hay	1.4	1	1.4	l
Vetch hay	2.1	1	2.1	k
Unweighted average	9.0	86	0.8-63	

^aAerosol ^{226}Ra was assumed to contribute insignificant amounts of ^{226}Ra to the plant.

^bValues express ratios of fresh-weight ^{226}Ra concentrations in plants to dry-weight concentrations in soil for all food crops directly edible by man. Dry-weight concentrations in both plants and soil were used for forage, hay, and feed calculations.

^cValues in (Kirchmann *et al.*, 1968) are dry-weight concentration factors as given.

^dKirchmann *et al.*, 1968.

^eVavilov *et al.*, 1964.

^fDeBortoli and Gaglione, 1972.

^gSample consisted of tomatoes, haricots, lettuce, and small pumpkins in equal amounts.

^hSample consisted of peaches, pears, apples, plums, and grapes in equal amounts.

ⁱMordberg *et al.*, 1976.

^jGrzybowska, 1974.

^kTaskayev *et al.*, 1977.

^lRusanova, 1964.

fruit, and grain are 1.2×10^{-2} , 5.5×10^{-4} , and 5.5×10^{-1} . These values appear to be somewhat greater than those previously calculated for lead isotopes, perhaps reflecting a greater absorption of radium from the soil by plants, greater translocation of radium to edible parts, the inclusion of aerially deposited radium, or a reflection of the difference in the solubility of the nuclide added to soil. Values of CF_{sp} for certain grains obtained by Mordberg *et al.* (1976) appear to be much greater than values for all other vegetation types. Information concerning empirical conditions under which these higher values were obtained was not available; it is possible that the grain crops were cultivated on soil containing radium for inordinate amounts of time, resulting in the high CF's observed.

For forage and hay, an average dry weight CF_{sp} of 9.0×10^{-2} was calculated, with a range of individual values from 8.0×10^{-3} to 6.3×10^{-1} . The value for forage, hay, and feed expressed in dry weight is most useful since herbivore ingestion studies seldom consider fresh weights. Observation of the wide range of values of CF_{sp} derived suggests that experimental conditions can greatly influence the value derived for a particular forage species. Only two forage studies cited in Table 3.1 involved vegetation grown in naturally contaminated environments (Rusanova, 1964; DeBortoli and Gaglione, 1972), results of which indicate CF_{sp} 's to be less than 3.0×10^{-2} (dry weight).

From this review, two separate values of CF_{sp} are recommended for use in environmental transport models. For vegetables, fruit, and grain, a fresh weight CF_{sp} of 2.0×10^{-2} is recommended. Results of the experiments by Mordberg *et al.*, (1976) for grain were not considered in recommending this value, due to the lack of information concerning empirical conditions in their analyses. For forage and hay, a dry weight CF_{sp} of 9.0×10^{-2} is recommended. The average value of CF_{sp} for forage, obtained in our review, is recommended in light of the large range of derived values, and the fact that naturally contaminated environments had associated CF_{sp} 's of the same order of magnitude as this average.

Dietary concentrations of ^{226}Ra in the United States have been measured for several food categories by Fisenne and Keller (1970) and

Morse and Welford (1971). Table 3.2 is a list of some of the mean values obtained by these investigators. Mean values for vegetables, fruits, and grains range from 0.24 to 2.5 pCi/kg fresh weight. Vinogradov (1959) has reported the average U.S. ^{226}Ra soil concentration to range between 0.8 to 2.8 pCi/g. Therefore, using an intermediate value of 2 pCi/g and the recommended CF_{sp} for vegetables, fruits, and grains of 2.0×10^{-2} , dietary concentrations of these food items are predicted to be on the order of 4×10^{-2} pCi/g or 40 pCi/kg. Thus, these predicted values overestimate mean measured values by more than an order of magnitude. There are many possible explanations for this discrepancy. First, the average U.S. soil concentration of 2 pCi/g assumed in the calculation may not be representative of the actual average for agricultural soils in the United States, leading to inaccurate predictions. Secondly, processing or washing of the food items before ^{226}Ra evaluations were done may have reduced the ^{226}Ra content, and decreased the measured concentration. Thirdly, the recommended CF_{sp} may overestimate the actual soil-plant relationship of ^{226}Ra . Reasons for this last possibility have been stated previously. Briefly, by ignoring aerosol ^{226}Ra contributions to ^{226}Ra concentrations in edible plant parts at the study site, or by ignoring differences between solubility characteristics of soil-applied ^{226}Ra and environmental ^{226}Ra , it is possible that the soil-plant relationships of ^{226}Ra are overestimated in the studies cited. Finally, the vegetable species for which ^{226}Ra concentrations were measured may differ significantly from the species from which CF_{sp} 's were derived in this review. Thus, if translocation of ^{226}Ra results in relatively high concentrations in vegetables such as carrots, beets, and cabbage, which were used to derive CF_{sp} 's (see Table 3.1), the recommended value based primarily on these species may overestimate the average for all vegetable species.

From Ng *et al.* (1968), a CF_{sp} value of 3.1×10^{-4} was adopted for use by the U.S. Nuclear Regulatory Commission in its Regulatory Guide 1.109 (draft, March 1976), but is labelled B_{iv} . No distinction between forage crops and other crops was made in either of these documents. Thus, it appears that our present values of 2.0×10^{-2} (FW) for vegetables, fruits and grains and 9.0×10^{-2} for forage and hay (FW) greatly exceed values commonly used to this point for soil to plant transfer of ^{226}Ra .

Table 3.2. Measured dietary concentrations of ^{226}Ra in selected food items

Food category	New York City diet (pCi/kg)		San Francisco diet (pCi/kg)
	Morse & Welford ^a	Fisenne & Keller ^b	Fisenne & Keller ^b
Fresh fruit	0.40	0.32	0.26
Fresh vegetables	1.1	1.1	0.66
Root vegetables	0.21	1.3	1.4
Potatoes	1.5	2.3	0.24
Rice	0.88	2.0	0.29
Dry beans	0.76	1.0	0.70
Flour	1.3	2.1	1.4
Whole grains products	2.2	2.5	2.2

^aMorse and Welford, 1971.

^bFisenne and Keller, 1970.

3.1.3 Deposition of aerosol radium onto vegetation

Predictions of the ^{226}Ra content of edible crops due to deposition of airborne material may be made with the use of the deposition and uptake model provided in the U.S. NRC's Regulatory Guide 1.109 (draft, October 1977) (see Sect. 2.1.3). Inadequacies of various model parameters which represent interception characteristics of the plant surfaces should be emphasized again. The regulatory guide suggests using a value of 0.2 for r , the interception fraction. The appropriateness of this value for edible plant parts, especially root crops, or other crops is unclear. It is also unclear if the value of 2.0 kg/m^2 for Y_v , the agricultural productivity, is a reasonable value to use for edible portions of all crops, although it is suggested in the guide for produce and leafy vegetables alike. In addition, the suggested t_e value of 60 days

for edible crops, representing the time of exposure of foliar components of the crops to airborne particulates, does not account for lesser exposure periods associated with some edible portions, such as tomatoes or beans. Use of this t_e value will cause overestimates for many species.

Keeping in mind these uncertainties, one may estimate dietary concentrations of ^{226}Ra for several food categories due to deposition alone utilizing the NRC model by neglecting the root uptake term in the equation (see Sect. 2.1.3). Assuming a total deposition velocity of 0.3 cm/sec , based on a mean residence time of 4 days for aerosols (Travis *et al.*, in preparation), and an average air concentration for ^{226}Ra of $2 \times 10^{-5} \text{ pCi/m}^3$ (see Sect. 3.1.1), the following predictions may be made. For all fresh vegetables, fruits, and grains, a dietary concentration of $9.6 \times 10^{-3} \text{ pCi/kg}$ may be calculated. For forage, a concentration of $2.3 \times 10^{-2} \text{ pCi/kg}$ may be calculated. It seems reasonable to assume that the forage value would more closely approximate the actual value, since the model parameters are more applicable to this type of vegetation.

Measured dietary concentrations of ^{226}Ra in the United States were given in Table 3.2 of Sect. 3.1.2. Comparing the measured concentration of ^{226}Ra in vegetables and grain products with the values predicted due to aerosol deposition and with values predicted due to root uptake alone (Sect. 3.1.2), it appears that direct deposition on edible surfaces alone may not significantly account for ^{226}Ra found in the average human

diet. However, deposition may become more important where aerosol ^{226}Ra is enhanced in areas located near anthropogenic sources (Sect. 3.1.1). To predict dietary concentrations due to aerosol deposition in these areas, it is recommended that the regulatory guide model be used at this time.

3.1.4 Uptake and distribution of ^{226}Ra by grazing animals

The number of sources documenting trophic transfer of ^{226}Ra from feed to animal products is small. Despite the presence of numerous references presenting the ^{226}Ra content of milk and meat products from market basket surveys, most of the literature does not contain the necessary information on dietary concentrations of the livestock from which the products were obtained (Morse and Welford, 1971; Belova and Lunkina, 1967; Fisenne and Keller, 1970; Hallden *et al.*, 1963; Hallden and Harley, 1964; Muth *et al.*, 1960; Turner *et al.*, 1958). As a result, many available sources were considered inadequate for inclusion in the present analysis.

3.1.4.1 Distribution in milk. A biological half-time of 50 hours for radium elimination via milk has been estimated from monitoring data on two dairy cows exposed to single oral administrations of 5 mCi ^{224}Ra (Sansom and Garner, 1966). Thus, it may be estimated that 93% of the total ^{226}Ra elimination via milk occurred during the 8 days following acute administration. At that time, sampling was discontinued. The major fate of ingested ^{224}Ra was fecal excretion (>99%), with only 0.35% of that administered being recovered in milk. It may be estimated that $0.35\%/0.93$ or 0.37% of the oral dose administered was recovered in milk. This value is expressed as an average diet-to-milk CF_m of 3.7×10^{-3} (range of 3.1 to 4.4×10^{-3}) in Table 3.3, although the isotope was not technically administered in feed. Ng and his colleagues (1977) suggest an f_m of 1.5×10^{-4} (range of 1.3 to 1.8×10^{-4}) based on 8-day recovery data (i.e., at 93% equilibrium). Thus, f_m would equal 1.6×10^{-4} at 100% equilibrium (Table 3.3).

Kirchmann and his colleagues consider forage ingestion to be a major source of radium excreted in milk (Kirchmann *et al.*, 1972). Analysis of data obtained by these investigators from individual animals exposed to either drinking water containing $^{226}\text{RaCl}_2$ or hay harvested

Table 3.3. Concentration factors (CF_m) and transfer coefficients (f_m)
for ^{226}Ra ingested by dairy animals and transferred to milk

Animal	Radium source	Mean CF_m^a milk:diet ($\times 10^{-3}$)	F_m (day/liter) ($\times 10^{-4}$)	Number of derived values	References
Cattle	Oral administration of 5 mCi ^{224}Ra	3.7	1.6	2	<i>b</i>
Cattle	Ra from industrial emissions incorporated into forage	8.4	5.9	4	<i>c</i>
Cattle	Uncontaminated herbage	32	34	2 to 4	<i>d</i>

^a CF_m defined as ratio of activity concentration in milk (fresh wt) to activity concentration in diet (dry wt.).

^bSansom and Garner, 1966.

^cKirchmann *et al.*, 1972.

^dDeBortoli and Gaglione, 1972.

from industrially contaminated pastures indicates that forage only slightly exceeds drinking water as a source. Similar results have been observed for another alkaline-earth element, ^{85}Sr (Van den Hoek, 1969). Equilibrium estimates of CF_m and f_m from the hay ingestion experiment are also presented in Table 3.3. Kirchmann suggests that the small difference between water and food sources is a function of their residence times in the intestinal tract with water being more rapidly excreted as urine.

The final entry, derived from a market basket survey performed in northern Italy, has been included for comparison (de Bortoli and Gaglione, 1972). In this study, all non-vegetable plant material was termed "herbage" and analyzed together. For the purpose of calculation, "herbage" was considered cattle forage, and its ^{226}Ra content compared with that of milk. It is also assumed that the published values existed at equilibrium. Thus, the resulting CF of 3.2×10^{-2} and f_m of 3.4×10^{-3} day/liter are not as rigorously defined as the two previous estimates.

Of these three studies, the experimental design employed by Kirchmann and his colleagues (1972) most closely approximates the classic scenario of local grazing near a polluting industry (i.e., chronic ingestion of contaminated forage in the field). As such, values of CF_m (8.4×10^{-3}) and f_m (5.9×10^{-4} day/liter) derived from their research are recommended for use in future transport models. This value of f_m compares well with the recommended stable-element radium transfer of 4.5×10^{-4} day/liter for forage to milk derived by Ng and his colleagues (Ng *et al.*, 1977). The preliminary value of 8.0×10^{-3} published in NRC Regulatory Guide 1.109 (March, 1976) is approximately one order of magnitude greater than the value derived in the present study.

Our recommended concentration factors permit us to make an estimate of ^{226}Ra activity concentrations for comparison with measured values of this isotope in table-ready milk supplies. Existing data are summarized in Table 3.4. It is known that the range of ^{226}Ra concentrations in United States soils is 0.8 to 2.8 pCi/g if areas of high background are excluded (Vinogradov, 1959). At the low end of this range and the recommended soil-to-forage CF of 9.0×10^{-2} (Table 3.1), the predicted forage content is 7.2×10^{-2} pCi ^{226}Ra /g. Multiplying this value by the

recommended forage to milk CF of 8.4×10^{-3} produces an estimated milk concentration of 6.0×10^{-1} pCi ^{226}Ra /kg milk. At the maximum soil activity, an estimate of 2.1 pCi ^{226}Ra /kg milk was obtained. Samples of U.S. milk range between 9.0×10^2 and 0.27 pCi ^{226}Ra /kg milk, with an unweighted mean of 0.22 pCi/kg (Table 3.4). The world average is 0.25, with a range extending to 1.07 pCi ^{226}Ra /kg. Thus, trophic transfer predictions based on concentration factors recommended in our analysis closely approximate existing data.

3.1.4.2 Distribution in meat. Sheep and swine have been included in the assessment of forage-to-meat transfer for several reasons:

1. the paucity of information regarding cattle ingestion of ^{226}Ra and
2. sheep are ruminants, and are grazed extensively on western lands.

All values gleaned from the literature are presented in Table 3.5. It should be noted that swine possess a digestive tract and physiology similar in many respects to that of humans (Bustad and McClellan, 1965).

The soft tissues and bones of one litter of 13 pigs and 9 crossbred lambs maintained on uncontaminated diets were periodically sampled during a study performed by Hardy and his colleagues (1969). Any ^{226}Ra in animal feed originated from natural sources. Nearly 97% of the sheep's body burden of ^{226}Ra was stored in the skeleton, while 70% of the swine ^{226}Ra burden was deposited in soft tissues. At the termination of the experiment, the sheep body burden equaled 9.2% of the total dietary radium, as contrasted to the 0.5% incorporation by swine. Soft tissues (no discrimination made between muscle and organs) of sheep and swine contained nearly equal concentrations of ^{226}Ra (7 pCi/kg and 8 pCi/kg, respectively). Thus, for both species, the CF for soft tissue approximates 3.0×10^{-3} to 4.0×10^{-3} .

Other ruminant data includes that for caribou and reindeer grazing on *Cladonia* lichens in Alaska and Finland (Holtzman, 1966 a,b). Due to their slow growth habit and large surface-to-volume ratio, lichens accumulate dust particles containing significant quantities of the naturally occurring isotope ^{226}Ra . Because data characterizing the radium concentration of rumen contents was unavailable, an alternate

Table 3.4. Measured values of naturally occurring ^{226}Ra in dietary meat and milk (pCi/kg fresh weight)

Site	Milk ^{a,b} (pCi/kg)	Meat ^a (pCi/kg)	References
<u>United States</u>			
New York State	0.13	0	c
Wisconsin	0.24		a
Tennessee	0.27		e
Chicago	0.23	0.55	b
	(0.22-0.24)	(0.45-0.64)	
New York City	0.25	0.46	d
	(0.24-0.25)	(0.44-0.47)	
New York City	0.25	0.02	e
	(0.19-0.30)	(0.01-0.02)	e
New York City	0.25	0.01	f
San Francisco	0.25	0.46	d
	(0.24-0.25)	(0.44-0.47)	
San Francisco	0.09	0.02	e
	(0.08-0.10)	(0.01-0.02)	
<u>Germany</u>	0.3	0.8	g
<u>United Kingdom</u>	0.10		h
	(N = 6)		h
	(0.1-0.2)		h
	0.26		h
	0.10		h
	0.18		h
	1.07		h
	0.22		h
<u>Italy</u>			
Varese, N. Italy	0.18		i
	(N = 2 to 4)		i
	(0.175-0.185)		i
<u>Puerto Rico</u>	0.15		j
	0.08		j
Unweighted average, U.S.	0.22	0.22	
Range	0.09-0.27	0-0.55	
Unweighted mean, all sites	0.25	0.29	
Range	0.09-1.07	0-0.8	

^aValues in parentheses are range and/or N samples.

^bBased on milk density of 1.028 g/cm³ (Weast, R.C. ed., *Handbook of Chemistry and Physics*, 57th Ed., CRC Press, 1976. Page F-3).

^cShandley, 1953.

^dHallden and Fisenne, 1961.

^eFisenne and Keller, 1970.

^fMorse and Welford, 1971.

^gMuth *et al.*, 1960.

^hSmith and Watson, 1963.

ⁱDe Bortoli and Gaglione, 1972.

^jHallden and Harley, 1964.

Table 3.5. Concentration factors (CF_s and CF_r) and transfer coefficients (f_f) for ^{226}Ra ingested by swine and ruminants and transferred to meat

Animal	Radium source	Mean CF_s^a , CF_r^b muscle:diet ($\times 10^{-3}$)	F_f (day/kg) ($\times 10^{-3}$)	Number of derived values	Remarks	Reference
Swine						
	Uncontaminated mothers' milk and commercial feed	4.0 (117 days old) 3.0 (177 days old)	1.7 0.9	13	Assume data given in fresh wt tissue and dry wt diet. Soft tissue only. Intestinal contents removed prior to analysis	c,d
Ruminants						
Sheep	Uncontaminated pasture and commercial feed	4.0 (168 days old) 3.0 (>224 days old)	4.0 3.0	9	Same as above	e,e
Caribou	Lichens contaminated with naturally occurring ^{226}Ra	4.7	1.7	12	Muscle analysis	f-h
Reindeer	Lichens contaminated with naturally occurring ^{226}Ra	8.6	3.1	1	Muscle analysis	f-h
	Mean for swine	3.5	1.3			
	Mean for ruminants	5.1	3.0			

^a CF_s is concentration factor for swine and defined as ratio of activity concentration in meat (fresh wt.)

^b CF_r is concentration factor for ruminants.

^cHardy *et al.*, 1969.

^dKopp, 1978.

^eBell, 1978.

^fHoltzman, 1966a.

^gHoltzman, 1966b.

^hLiden and Gustafsson, 1967.

method of estimating the concentration factor to these ruminants (CF_r) was developed. Although it is known that caribou and reindeer freely ingest annual grasses, sedges, and horsetails during the short Arctic summer, the present calculation conservatively assumes lichen ingestion as the principal source of ^{226}Ra intake. Annual means of lichen ^{226}Ra activity in Alaska and Finland were used to compute the caribou CF_r of 4.7×10^{-3} and the reindeer CF_r of 8.6×10^{-3} (Table 3.5) (Holtzmann, 1966 a,b). It must be noted that Holtzmann's data represents a special case.

Transfer coefficients were calculated on the basis of 2.3 kg and 3.4 kg dry daily intake respectively for 117- and 180-day-old pigs (Kopp, 1978), 1.0 kg and 1.2 kg dry daily feed respectively for 168- to 224-day-old lambs (Bell, 1978), and 7.0 kg fresh (2.8 kg dry) daily consumption by adult reindeer (Liden and Gustafsson, 1967). In the absence of additional ^{226}Ra data for beef cattle, estimates of f_f for swine and ruminants were derived by dividing the concentration factor by the daily dry matter intake rate (day/kg) (Table 3.5).

In previous sections of this report, transfer coefficients were derived from data specific for cattle (Sects. 2.1.4 and 3.1.4.1). Since this option is not available for developing a ^{226}Ra f_f , we have substituted the average ruminant CF of 5.1×10^{-3} (Table 3.5) and an assumed dry daily intake of 10 kg (U.S. NRC Reg. Guide 1.109, March 1976) into the above derivation procedure. The resulting beef-specific f_f of 5.1×10^{-4} day/kg is approximately two orders of magnitude smaller than the regulatory guide number of 3.1×10^{-2} day/kg. The latter value was derived by manipulation of data summarized by Ng and colleagues (1968) for human whole-body stable radium concentrations and the average radium content in vegetables and fruits consumed by man.

Comparison of predicted vs measured ^{226}Ra content of ruminant meat by the method detailed in Sect. 3.1.4.1 for milk indicates close agreement with existing data. Use of the minimum soil content of 0.8 pCi $^{226}\text{Ra}/\text{g}$ (Vinogradov, 1959) and our recommended forage-to-meat CF of 5.1×10^{-3} predicts a meat ^{226}Ra content of 0.37 pCi/kg; while the maximum soil content of 2.8 pCi $^{226}\text{Ra}/\text{g}$ (Vinogradov, 1959) gives rise to an estimated 1.3 pCi $^{226}\text{Ra}/\text{kg}$ meat. The unweighted mean for the United States is 0.22 pCi $^{226}\text{Ra}/\text{kg}$ meat with a range of 0 to 0.55 pCi/kg (Table 3.4). If

the single non-U.S. value of 0.8 pCi/kg from Germany is included in the analysis (Muth *et al.*, 1960), a mean of 0.29 pCi/kg can be calculated from this data. Use of the highest soil value predicts a meat ^{226}Ra activity approximately two times greater than the largest literature data. Otherwise, predicted values fall close to the range of available measured values.

3.1.4.3 Soil ingestion. Soil ingestion by ruminants is a normal consequence of grazing behavior (Sect. 2.1.4.3). During cropping of forage plants, quantities of soil are inadvertently consumed along with vegetation. The degree and magnitude of soil ingestion are dependent on a number of variables, including the stocking rate, type of pasture, frequency of muddy patches or earthworm casts, soil moisture, and selective grazing by individuals in the herd (Healy, 1968; Mayland *et al.*, 1970). The greatest soil intakes occur during midwinter crowding of nonlactating individuals into feedlots or protected fields (Healy, 1968). Soil commonly constitutes 10% of the dry matter intake of sheep on winter pasture, while beef cattle ingestion usually ranges between 1.3% and 18.8% with a median of 6.3% (Suttle *et al.*, 1975; Mayland *et al.*, 1970).

Sheep are known to solubilize, absorb, and utilize ingested soil as a source of trace minerals (Suttle *et al.*, 1975; Healy *et al.*, 1970). In feeding experiments with isotopically tagged soil, it was found that 34% of ^{75}Se , 14% of ^{65}Zn , 1% of ^{60}Co , and 0.4% of ^{54}Mn present in the tag were absorbed by adult sheep (Healy *et al.*, 1970). Apparently, no comparable experiments have been performed with isotopes of the alkaline earth elements, or on beef cattle.

On semiarid range, beef cattle can ingest between 0.1 and 1.5 kg soil with a median of 0.5 kg/animal-day (Mayland *et al.*, 1970). Presence of the introduced forage species, cheatgrass (*Bromus tectorum*), contributed to the incidence of soil ingestion in this investigation since the shallow-rooted plants were easily pulled from the dry soil and consumed along with roots and attached earth. In more humid climates, dairy cattle on lightly-stocked pasture have ingested between 0.5 and 0.9 kg soil/animal-day (Healy, 1968). On densely-stocked pasture during winter, this value soared to 1.75 kg/animal-day.

As discussed in Sect. 3.1.1.1, the ^{226}Ra incorporated within vegetation is in a soluble form that has been transferred across the root-cell membranes from the larger, mobile radium soil pool. Soil ingested by grazing animals will include the mobile fraction plus the larger, nonmobile "fixed" pool. Since the comparative digestibility of these radium phases by ruminants has not been investigated, the extent of soil-radium transport from the rumen to muscle and milk is not known. It is known that release of soil-ingested molybdenum from the digestive tract of sheep is related to both physical and chemical characteristics of the soil (Suttle *et al.*, 1975).

For the purpose of a worst-case calculation, it has been assumed that transfer of soil radium to muscle parallels that of forage radium. However, it should be emphasized that GI absorption of soil radium and its distribution to muscle may vary greatly from that of forage radium. Thus, for daily soil ingestion by beef cattle of 8 kg dry matter of which 0.5 kg (6.25%) is soil (Mayland *et al.*, 1970; Bustad and Terry, 1956), an average CF from soil to forage of 9.0×10^{-2} dry weight (Table 3.1), a soil content of 2 pCi/g (see Sect. 3.1.2) and the derived ruminant muscle CF of 5.1×10^{-3} (Table 3.5), a median concentration of beef muscle can be estimated. The average U.S. forage concentration may be calculated as follows:

$$(9.0 \times 10^{-2}) \times (2 \text{ pCi } ^{226}\text{Ra/g soil}) = 1.8 \times 10^{-1} \text{ pCi } ^{226}\text{Ra/g forage.}$$

Thus, the estimated ^{226}Ra concentration in beef is:

$$[(93.75\%) \times (180 \text{ pCi } ^{226}\text{Ra/kg forage}) + (6.25\%) \times (2000 \text{ pCi } ^{226}\text{Ra/kg soil})] \times (5.1 \times 10^{-3}) = 1.5 \text{ pCi/kg beef.}$$

The value represents an increase of 63% over the estimated beef concentration of 0.92 pCi/kg when no soil ingestion is included. Similar calculations for the minimum soil ingestion rate of 0.1 kg/animal-day and a maximum of 1.5 kg/animal-day (Mayland *et al.*, 1970) are 1.0 pCi/kg (increase of 9%), and 2.7 pCi/kg (increase of 193%), respectively.

Similarly, for daily dry matter ingestion of 10 kg by dairy cattle, (Ng *et al.*, 1977) a soil intake ranging from 0.5 to 1.75 kg/animal-day (Healy, 1968) and a forage to milk CF of 8.4×10^{-3} (Table 3.3), minimum and maximum milk estimates were determined. The minimum estimate is 2.3 pCi ^{226}Ra /liter, an increase of 53% over the previous value of 1.5 pCi/liter; while the maximum is 4.2 pCi/liter, an increase of 180%.

For sheep ingesting 2 kg/day dry matter of which 10% is soil, (Kopp, 1978; Suttle, 1975) use of the same assumptions and technique allows an estimate of 1.8 pCi ^{226}Ra /kg of sheep muscle. This value represents an increase of 96% over the 0.92 pCi/kg derived for ruminants from a diet lacking soil.

Thus, it appears that normal ingestion of radium-bearing soil could significantly increase muscle and milk concentrations of radium if GI absorption of soil radium parallels that of forage radium. Although presently unquantified, the rate of mineralized radium absorption from soil in the ruminant digestive tract is probably less than that from biologically available forage radium.

3.2 Dose Conversion Factors for ^{226}Ra

Factors relating the radiation dose commitment received by bone, lung, liver, kidney, and the whole body to the amount of inhaled or ingested ^{226}Ra have been determined and are presented in Table 3.6. INREM II, (Killough *et al.*, 1978), a computer implementation of both a model based on the ICRP Task Group Lung Model (Morrow *et al.*, 1966) and a recent GI tract model described by Bernard (1968) was used to develop 50-year dose commitment factors (DCF's) for ^{226}Ra and its daughter radionuclides. Current metabolic data for all isotopes in the decay chain and organ-specific retention functions from available literature were utilized by Dunning *et al.*, (in preparation) to describe and quantify the energy deposition resulting from internal ^{226}Ra .

3.2.1 Inhalation

Initial deposition fractions in the four major portions of the lung, the nasal-pharynx (N-P), tracheo-bronchial tree (T-B), pulmonary (P), and lymphatic tissue (L), were determined by the INREM II code from a graphical representation of the relationship between the activity median aerodynamic diameter (AMAD) of the inhaled particles and deposition which is given in the Reactor Safety Study (U.S. NRC, 1975). An AMAD of 0.3 μm was used for the present DCF's. Parameters for lung clearance pathways in INREM II were adapted from ICRP Publication 19 (ICRP, 1972a). A solubility class for aerosol particles of ^{226}Ra was assumed as given by Morrow *et al.*, (1966). Translocation of ^{222}Rn gas from the lung or GI.

Table 3.6. Fifty-year dose commitment factors for one year's inhalation and ingestion of ^{226}Ra

Organ	Inhalation ^a		Ingestion
	rem/ μCi	mrem/pCi-m ³ in a 1-year period ^b	rem/ μCi
Bone	24.5	210	22
Lung	49	410	6.5×10^{-5}
Liver	0.34	2.9	0.30
Kidney	0.34	2.9	0.30
Whole body	10.0	84	8.1

^a0.3 μm AMAD, Solubility Class W.

^bBased on inhalation rate 23 m³/day.

NOTE ADDED IN PRESS — The above dose commitment factors (DCF) are based on early drafts of a report by Dunning *et al.* (in preparation). The DCF's given above are based on a quality factor (QF) of 10, which was recommended in ICRP Publication 2 (Pergamon Press, 1959). In more recent drafts of their document, Dunning *et al.* have incorporated recent recommendations from ICRP Publication 26 (Pergamon Press, 1977) regarding quality factors. DCF's based on a higher QF of 20 are given in Appendix C of the present report.

tract to other portions of the body was neglected in dose calculations, since the resultant dose would be small compared to doses from other translocated daughters of ^{226}Ra (Dunning *et al.*, in preparation).

Metabolic parameters necessary in calculating an inhalation DCF include absorption fractions and retention functions for organs to which ^{226}Ra and daughters produced in the lung are translocated, and for the lung, to which ^{222}Rn gas produced in other organs is transported. Parameters used for lead, bismuth, and polonium isotopes (^{214}Pb , ^{210}Pb , ^{214}Bi , ^{210}Bi , ^{218}Po , ^{214}Po , and ^{210}Po) in the ^{226}Ra decay chain were discussed in Sect. 2.2.1 of this report. Isotopic effects were neglected. Values for f_1 , depicting GI absorption, and f_2 , depicting absorption from the blood into certain organs are listed in Table 3.7 for isotopes in the ^{226}Ra decay chain. Absorption fractions and retention functions for ^{226}Ra translocated from the lung were chosen by Dunning *et al.*, (in preparation) as follows. Fractional absorption from the GI tract into the blood (f_1) for ^{226}Ra was assumed to be 0.2, consistent with values given by the Task Group on Alkaline Earth Metabolism in Adult Man (ICRP, 1972b). Retention models were adapted from the review by this task group to characterize radium distribution after absorption from the GI tract and clearance from the lungs. From these models, the values for blood-to-organ transfer (f_2'), given in Table 3.7 were derived.

A model of the dynamics of ^{222}Rn gas was developed by Dunning *et al.*, (in preparation) to account for the amount taken into the lungs by inhalation, ^{222}Rn formed in the lungs by decay of deposited ^{226}Ra , and ^{222}Rn transported to the lungs from other portions of the body where ^{226}Ra has been translocated and deposited. The biological half-time of ^{222}Rn gas in the lungs was calculated to be 8.2×10^{-5} days from Reference Man respiratory parameters (Task Group of Committee 2, ICRP, 1975). Retention functions of ^{222}Rn , derived by Bernard and Snyder (1975), were used to determine the μCi -days residence and release of ^{222}Rn produced in other organs. All ^{222}Rn was assumed to be transported back to the lungs, and subsequently treated in the same manner as inhaled radon gas, with a removal rate consistent with the biological half-time. Percutaneous loss of ^{222}Rn formed in the body was ignored, as was radioactive decay during transport from the site of formation in the lungs.

Table 3.7. Absorption factors f_1^a and f_2^b for nuclides in ^{226}Ra decay chain

Organ	Nuclide	f_1	f_2^b
Bone	Ra-226	0.20	0.17
	Po-218, 214, 210	0.10	0.09
	Pb-214, 210	0.08	0.56
	Bi-214, 210	0.05	0.07
Liver	Ra-226	0.20	0.05
	Po-218, 214, 210	0.10	0.10
	Pb-214, 210	0.08	0.11
	Bi-214, 210	0.05	0.03
Kidney	Ra-226	0.20	0.008
	Po-218, 214, 210	0.10	0.10
	Pb-214, 210	0.08	0.05
	Bi-214, 210	0.05	0.40
Whole body	Ra-226	0.20	1.00
	Po-218, 214, 210	0.10	1.00
	Pb-214, 210	0.08	1.00
	Bi-214, 210	0.05	1.00

f_1^a = absorption from GI tract into blood.

f_2^b = absorption from blood into organs listed.

With the use of these parameters and models, DCF's for inhaled ^{226}Ra were determined (Table 3.6). The values, given in $\text{mrem/pCi}\cdot\text{m}^3$ per year of inhalation, are based on inhalation rates of $23\text{ m}^3/\text{day}$ for Reference Man (Task Group of Committee 2, ICRP, 1975). The lungs appear to receive the highest dose per unit ^{226}Ra inhaled.

3.2.2 Ingestion

The DCF's for ingested ^{226}Ra are based on a GI tract model described by Bernard (1968) utilizing the f_1 fraction of 0.2 and f_2' values as assigned previously (Table 3.7). Retention functions for ingested ^{226}Ra and its daughters, in the various organs, are described above in the discussion of DCF's for inhaled ^{226}Ra . These functions are the same for both modes of ^{226}Ra intake considered here.

Dose conversion factors for ingested ^{226}Ra are given in Table 3.6, in mrem/pCi . Bone appears to be the organ with the greatest dose commitment per unit amount of ^{226}Ra ingested.

3.3 References

- Bell, C., Professor, Animal Science Department, University of Tennessee, Knoxville, TN 37916, personal communication to A. P. Watson, August 17, 1978.
- Belova, R. S. and Z. P. Lunkina, "The Content of Radium and Uranium in Foodstuffs and Their Accumulation in Human Teeth," *Gig. Sanit. (Hygiene and Sanitation)* 32(6), 372-375 (1967).
- Bernard, S. R., *Health Physics Division Annual Progress Report for Period Ending July 31, 1968*, ORNL-4316, pp. 283-87 (1968).
- Bernard, S. R. and W. S. Snyder, *Health Physics Division Annual Progress Report for Period Ending June 30, 1975*, ORNL-5046 (1975).
- Breslin, A. J. and H. Glauberman, "Investigation of Radioactive Dust Dispersed from Uranium Mill Tailings Piles," p. 249 in *Environmental Surveillance in the Vicinity of Nuclear Facilities*, Reinig, W. C. (ed.) Charles Thomas, Springfield, Ill. (1970).
- Bustad, L. K., and R. O. McClellan, "Use of Pigs in Biomedical Research," *Nature* 208(5010), 531-535 (1965).
- Bustad, L. K., and J. L. Terry, "Basic Anatomical Dietary, and Physiological Data for Radiological Calculations," Hanford Atomic Products Operation, Richland, WA HW-41638 (February, 1956).
- De Bortoli, M. and P. Gaglione, "Radium-226 in Environmental Materials and Foods," *Health Phys.* 22(1), 43-48 (1972).
- Dunning, D. E., Jr., S. R. Bernard, P. J. Walsh, G. G. Killough, and J. C. Pleasant, *Estimates of Internal Dose Equivalents to 22 Target Organs for Radionuclides Occurring in Routine Releases from Nuclear Fuel-Cycle Facilities, Vol. II*, NUREG/CR-0114/V2, ORNL/NUREG/TM-190/V2 (in preparation).
- Eisenbud, M. and H. G. Petrow, "Radioactivity in the Atmospheric Effluents of Power Plants that Use Fossil Fuels," *Science* 144, 288-289 (1964).
- Fisene, I. M. and H. W. Keller, "Radium-226 in the Diet of Two U.S. Cities," pp. I-2 to I-8 in *Health and Safety Laboratory, Milford Program Quarterly Summary Report (December 1, 1969 to March 1, 1970)*, HASL-224, UC-41 (April, 1970).

- Garner, R. J. "Transfer of Radioactive Materials from the Terrestrial Environment to Animals and Man," *CRC Critical Reviews in Environmental Control* 2, 337-385 (1971).
- Grzybowska, D. "Uptake of ^{226}Ra by Plants from Contaminated Soils," *Nucleonika* 19(1), 71-78 (1974).
- Halliden, N. A., I. M. Fisenne, and J. H. Harley, "Radium-226 in Human Diet and Bone," *Science* 140, 1327-1329 (1963).
- Halliden, N. A., and J. H. Harley, "Radium-226 in Diet and Human Bone from San Juan, Puerto Rico," *Nature* 204, 240-241 (1964).
- Halliden, N. A. and I. M. Fisenne, "Radium-226 in the Diet of Three U.S. Cities," pp. 90-94 in E. P. Hardy, Jr. (ed.), *Health and Safety Laboratory, USAEC Report HASL-113* (1961).
- Hardy, E., J. Rivera, I. Fisenne, W. Pond and D. Hogue, "Comparative Utilization of Dietary Radium-226 and Other Alkaline Earths by Pigs and Sheep," pp. 183-90 in M. R. Sikov and D. D. Mahlum (eds.), *Proceedings of the 9th Annual Hanford Biological Symposium*, Richland, WA (May 5-8, 1969) CONF-690501.
- Healy, W. B., "Ingestion of Soil by Dairy Cows," *N. Zealand J. Agric. Res.* 11, 487-499 (1968).
- Healy, W. B., W. J. McCabe and C. F. Wilson, "Ingested Soil As A Source of Microelements for Grazing Elements," *New Zealand J. Agric. Res.* 13, 503-521 (1970).
- Holtzman, R. B. "Natural Levels of Pb-210, Po-210, and Ra-226 in Humans and Biota of the Arctic," *Nature* 210, 1094-1097 (1966a).
- Holtzman, R. B., "Ra-226 and The Natural Airborne Nuclides ^{210}Pb and ^{210}Po in Arctic Biota," pp. 1087-1096 in Snyder, W. S., H. H. Abee, L. K. Burton, R. Maushart, A. Benco, F. Duhamel and B. M. Wheatley (eds.), *Proc. First International Congress Radiation Protection*, Rome, Italy (September 5-10, 1966) Pergamon Press, Oxford (1966b).
- International Commission on Radiological Protection, *The Metabolism of Compounds of Plutonium and Other Actinides*, ICRP Publication 19, Pergamon Press (1972a).
- International Commission on Radiological Protection, *Alkaline Earth Metabolism in Adult Man*, ICRP Publication 20, Pergamon Press (1972b).

- Jaworowski, Z., J. Bilkiewicz, and E. Zylicz, "Ra-226 in Contemporary and Fossil Snow," *Health Phys.* 20, 449-450 (1971).
- Jaworowski, Z. and L. Kownacka, "Lead and Radium in the Lower Stratosphere," *Nature* 263, 303-304 (1976).
- Killough, G. G., D. E. Dunning, Jr., and J. C. Pleasant, *INREM II: A Computer Implementation of Recent Models for Estimating the Dose Equivalent to Organs of Man from an Inhaled or Ingested Radionuclide*, NUREG/CR-0114, ORNL/NUREG/TM-84 (June 1978).
- Kirchmann, R., R. Boulenger, and A. La Fontaine, "Absorption of ^{226}Ra in Cultivated Plants," pp. 1045-1051 in Snyder, W. A., H. H. Abee, L. K. Burton, R. Maushart, A. Benco, F. Duhamel, and B. M. Wheatley (eds.), *Proceedings of the First International Congress Radiation Protection*, Rome, Italy (September 5-10, 1966). Pergamon Press, Oxford (1968).
- Kirchmann, R. A. La Fontaine, J. Van Den Hoek, and G. Koch, "Comparison of the Rate of Transfer to Cow Milk of ^{226}Ra from Drinking Water and ^{226}Ra Incorporated in Hay," *Comptes Rendus des Seances de la Societe de Biologie, et de ses Lileales*, 166(11), 1557-1562 (1972).
- Kocher, D. C., *Nuclear Decay Data for Radionuclides Occurring in Routine Releases from Light-Water-Cooled Nuclear Power Reactors*, ORNL/NUREG/TM-102 (August 1977).
- Kopp, W. J., Director, Animal Services Dept. Comparative Animal and Research Laboratory, 1299 Bethel Valley Road, Oak Ridge, TN 37830, personal communication to A. P. Watson, August 17, 1978.
- Kovalevski, A. L., "Natural Radioactive Elements in Plants," Abstracted from *Invest. Sibir. Ofdel Akad Nauk SSSR* 4, 108-114 (1962) (NSA# 16-26725).
- Liden, K., and M. Gustafsson, "Relationships and Seasonal Variation of ^{137}Cs in Lichen, Reindeer and Man in Northern Sweden 1961 to 1965," p. 193 in *Proceedings of the International Sympos., Stockholm (1966)*, Pergamon Press, Oxford (1967).
- Mayland, H. F., A. R. Florence, R. C. Rosenau, V. A. Layar, and H. A. Turner, "Soil Ingestion by Cattle on Semiarid Range as Reflected by Titanium Analysis of Feces," *J. Range Mgt.* 28(6), 448-452 (1970).
- Moore, H. E. and S. E. Poet, "Background Levels of ^{226}Ra in the Lower Troposphere," *Atmos. Environ.* 10, 381-383 (1976).

- Mordberg, E. L., V. M. Aleksandruk, G. F. Kovygin, I. I. Shevchenko, V. M. Blyumshtein and G. F. Yushkevick, "Translocation of Isotopes of the Uranium-Radium Series into the Grain of Some Agricultural Crops" (abstract from *Gig Sanit.* 2, 58-61), *Chem. Abstr.* 84: 134563 (1976).
- Morrow, P. E., D. V. Bates, B. R. Fish, R. F. Hatch, and T. T. Mercer, "Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract," *Health Phys.* 12, 173-207 (1966).
- Morse, R. S. and G. A. Welford, "Dietary Intake of ^{210}Pb ," *Health Phys.* 21, 53-55 (1971).
- Muth, H. B. Rajewsky, H. J. Hantke, and K. Aurand, "The Normal Radium Content and the $^{226}\text{Ra}/\text{Ca}$ ratio of Various Foods, Drinking Water, and Different Organs and Tissues of the Human Body," *Health Phys.* 2, 239-245 (1960).
- National Council on Radiation Protection and Measurements, *Natural Background Radiation in the United States*, NCRP Report No. 45, Washington, D.C. (1975).
- Ng, Y. C., C. A. Burton, S. E. Thompson, R. K. Tandy, H. K. Kritner, and M. W. Pratt, *Prediction of the Maximum Dosage to Man from Fallout of Nuclear Devices. IV. Handbook for Estimating the Maximum Internal Dose from Radionuclides Released to the Biosphere*, UCRL-50163, Part IV (1968).
- Ng, Y. C., C. S. Colsher, D. J. Quinn and S. E. Thompson, *Transfer Coefficients for the Prediction of the Dose to Man Via the Forage-Cow-Milk Pathway from Radionuclides Released to the Biosphere*, UCRL-51939 (July 1977).
- Rusanova, G. V., "Behavior of Radium and Calcium in the Soil Plant System," *Soviet Soil Sci.* 3, 275-280 (1964).
- Sansom, B. F., and R. J. Garner, "The Metabolism of Radium in Dairy Cows," *Biochem. J.* 99, 677-681 (1966).
- Sears, M. B., R. E. Blanco, R. C. Dahlman, G. S. Hill, A. D. Ryon and J. P. Witherspoon, *Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle for Use in Establishing "As-Low-As-Practicable" Guides-Milling of Uranium Ores*, Vol. 1, ORNL/TM-4903 (May 1975).

- Sears, M. B. R. E. Blanco, B. C. Finney, G. S. Hill, R. E. Moore and J. P. Witherspoon, *Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle - Conversion of Yellow Cake to Uranium Hexafluoride. Part 1. The Fluorination-Fractionation Process*, ORNL/NUREG/TM-7 (September 1977).
- Shandley, P. D., "The Radium Content of Common Foods," United States Atomic Energy Commission Report, UR-255 (April 1953).
- Smith, K. A., and P. G. Watson, "Radium-226 in Diet in the United Kingdom-A Preliminary Survey," pp. 90-91 in ARCRL-10 (Agricultural Research Council Radiobiological Laboratory), Wantage, Berkshire, England (1963).
- Suttle, N. F., B. J. Alloway and I. Thornton, "An Effect of Soil Ingestion On the Utilization of Diet Copper by Sheep," *J. Agric. Sci., Cambridge*, 84, 249-254 (1975).
- Taskayev, A. I., V. Y. Ovchenkov, R. M. Aleksakhim and I. L. Shuktomova, "Uptake of ^{226}Ra by Plants and Change in Its State in the Soil - Plant Tops - Litterfall System," *Pochwovedeniye* 2, 42-48 (1977).
- Task Group of Committee 2 of the International Commission on Radiological Protection, *Report of the Task Group on Reference Man*, ICRP Publication 23, Pergamon Press (1975).
- Travis, C. C., A. P. Watson, L. M. McDowell-Boyer, S. J. Cotter, M. L. Randolph and D. E. Fields, *A Radiological Assessment of Radon-222 Released from Uranium Mills and Other Natural and Technologically Enhanced Sources*, NUREG/CR-057, ORNL/NUREG-55, February, 1979.
- Turner, R. C., J. M. Radley, and W. V. Mayneord, "The Naturally Occurring α -ray Activity of Foods," *Health Phys.* 1, 268-275 (1958).
- U.S. Nuclear Regulatory Commission, *Reactor Safety Study: An Assessment of Accident Risks in U.S. Commercial Nuclear Power Plants, Appendix VI*, WASH-1400, NUREG-75/014 (1975).
- U.S. Nuclear Regulatory Commission, *Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I, Regulatory Guide 1.109* (drafts: March 1976 and October 1977).
- Van den Hoek, J., R. J. Kirchmann, J. Colard, and J. E. Sprietsma, "Importance of Some Methods of Pasture Feeding, of Pasture Type and of Seasonal Factors on ^{85}Sr and ^{139}Cs Transfer from Grass to Milk," *Health Phys.* 17, 691-700 (1969).

- Vavilov, P. P., O. N. Papora and R. P. Kodaneva, "The Behavior of Radium in Plants," Abstracted from *Dokl Akad. Nauk. SSSR* 157, 992-994 (1964) (NSA# 19 (2), 2016).
- Verkhovskaja, I. N., P. O. Vavilov, and V. I. Maslov, "The Migration of Natural Radioactive Elements Under Natural Conditions and their Distribution According to Biotic and Abiotic Environmental Components," pp. 313-28 in *Radioecological Concentration Processes*, B. Aberg and F. P. Hungate (eds.), Pergamon Press (1966).
- Vinogradov, A. P., *The Geochemistry of Rare and Dispersed Elements in Soils*, 2nd ed. Consultants Bureau, Inc., New York (1959).
- Weast, R. C. (ed.), *Handbook of Chemistry and Physics*, 57th Edition, CRC Press (1976).

4. SUMMARY OF RECOMMENDATIONS

Lead-210 and ^{226}Ra transport from air and soil to vegetables, fruits, grain, forage, milk, and beef have been evaluated through analysis of available literature sources and an atmospheric deposition model. Concentration factors (CF's) and transfer coefficients (f_m and f_f) were calculated to describe and quantify transfer of these nuclides from soil to edible portions of crops, forage, and animal products. Values for these parameters were recommended for use in environmental transport models on the basis of the information reviewed and are summarized in Table 4.1. Use of these recommended values may produce overestimates, since recommendations are based on the upper limits of the range of derived values in most cases.

For aerosol deposition on edible portions of crops and on forage, use of a model provided by the U.S. Nuclear Regulatory Commission, in recent drafts of the Regulatory Guide 1.109, is recommended at this time. More direct data relating the concentration of ^{210}Pb or ^{226}Ra in air to the resulting concentration in crops or forage were not available.

During the present evaluation, the authors have identified a number of poorly researched areas in the environmental transport literature for ^{210}Pb and ^{226}Ra . If it is important to obtain a more realistic quantification of food chain transport of these nuclides than currently possible in the absence of site-specific data, the following needs should be addressed.

1. A principal problem area is the lack of experimentation specifically designed to determine milk and meat transfer coefficients for these two isotopes in cattle. Not only should transfer coefficients for various chemical forms of ^{210}Pb and ^{226}Ra be evaluated, but releases from tailings piles should also be simulated as nearly as possible by offering contaminated feed. In addition, sufficient numbers of animals should be included to allow statistical comparison of results.
2. The importance of ingested soil 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 ^{210}Pb and ^{226}Ra solubilization from mineral soil in the digestive tract, and the subsequent internal transport.

3. Experimental data are needed to distinguish between the contributions of soil and atmospheric ^{226}Ra to total plant ^{226}Ra content. Particular attention should be paid to simulation of field conditions by the use of ^{226}Ra compounds which may occur in soil or air naturally, or as a product of technological activities.

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Table 4.1. Summary of recommended values for ^{210}Pb and ^{226}Ra concentration factors and transfer coefficients

Transport mode	CF		f_m (day/liter)		f_f (day/kg)	
	^{210}Pb	^{226}Ra	^{210}Pb	^{226}Ra	^{210}Pb	^{226}Ra
Soil-to-plant (CF_{sp})						
vegetables, fruits, grains ^a	1.0×10^{-2}	2.0×10^{-2}				
Forage and hay ^b	2.0×10^{-1}	9.0×10^{-2}				
Forage-to-animal products (CF_m or CF_f) ^c						
Milk	2.0×10^{-3}	8.4×10^{-3}	2.0×10^{-4}	5.9×10^{-4}		
Beef	8.0×10^{-3}	5.1×10^{-3}			8.0×10^{-4}	5.1×10^{-4}

^aValues express ratios of fresh-weight ^{226}Ra concentrations in plants to dry-weight concentrations in soil for all food crops directly edible by man.

^bDry-weight concentrations in both plants and soil were used for forage, hay, and feed calculations.

^cDry-weight concentrations in forage and fresh-weight concentrations in beef and milk were used in CF_m and CF_f calculations.

APPENDIX A

Table A.1. Ratios of fresh to dry weights (FW:DW) for food crops available for human or herbivore consumption^a

Food crop	% water	FW:DW
Vegetables (edible portion)		
Bean	88.9	9.0
Beet	87.6	8.1
Broccoli	89.9	9.9
Cabbage	92.4	13.2
Carrots	88.2	8.5
Cauliflower	91.7	12.1
Corn	73.9	3.8
Cucumber	96.1	25.6
Lettuce	94.8	19.2
Peas	74.3	3.9
Potatoes	77.8	4.5
Pumpkin	90.5	10.5
Radish	93.6	15.6
Spinach	92.7	13.7
Squash	88.6	8.8
Tomato	94.1	17.0
Fruit (edible portion)		
Apple	84.1	6.3
Apricot	85.4	6.8
Banana	74.8	4.0
Grape	81.9	5.5
Lemon	89.3	9.3
Peach	86.9	7.6
Pear	82.7	5.8
Plum	85.7	7.0
Grain (edible portion)		
Barley	11.1	1.1
Oats	8.3	1.1
Rice	12.0	1.1
Wheat	12.5	1.1
Forage, hay, feed ^b		
Grain (barley, corn, oats, wheat, Cottonseed meal)	8.8 (7.0-10.7)	1.1
Silage (alfalfa, corn, grass)	75.5 (74.7-76.2)	4.1
Hay (alfalfa, bluegrass, oats, grass, prairie, wheat)	11.0 (9.3-15.7)	1.1
Forage (grass) ^c	80.0	5.0

^aSpector, 1956.

^bAverage and range of percent water given for different species in Spector, 1956.

^cHealy, 1968.

REFERENCES -- APPENDIX A

- Healy, W. B., "Ingestion of Soil by Dairy Cows," *N. Zealand J. Agric. Res.* 11, 487-499 (1968).
- Spector, W. S. (ed.), *Handbook of Biological Data*, W. B. Saunders Company, Philadelphia and London (1956).

APPENDIX B

B.1 Fifty-Year Dose Commitment Factors for Inhalation of ^{222}Rn and Short-lived Daughters

The calculation of dose commitment resulting from the inhalation of ^{222}Rn and short-lived daughters, including ^{210}Po (RaA), ^{214}Pb (RaB), ^{214}Bi (RaC), and ^{214}Po (RaC'), must usually rely on assumptions regarding aerosol characteristics which may not be directly applicable to actual exposure conditions for a particular site. Factors such as radon-daughter equilibrium conditions, the fraction of daughter ions attached to surfaces of aerosols, and sizes of the composite daughter and carrier particle will affect dose calculations, yet vary over a wide range of possible conditions (Johnson *et al.*, 1973). Therefore, an attempt was made to assume reasonable, or average, values for these variables to determine a dose commitment factor (DCF) for ^{222}Rn , when supporting literature was available. When documentation was lacking, values representing the upper limits of estimated values were chosen for the variable in question, to decrease the probability of producing underestimates of dose commitment when using the DCF derived here.

The dose rate (in rem per unit time) for radon and daughters varies mainly with the amount and composition of the progeny breathed, the fraction of that material deposited on the respiratory region of interest, and the fraction of radiation emitted after deposition that penetrates the critical cells. This last value, the fraction penetrating critical cells, was assumed to be equal to unity in this assessment.

For doses resulting from inhalation of ^{222}Rn and its short-lived daughters, the critical tissue has been well-documented as being a portion of the tracheobronchial (TB) tree (Jacobi, 1972; Walsh and Hamrick, 1977). According to a literature review by Walsh (1970), the calculated dose averaged over the bronchial epithelium of the entire TB tree does not differ greatly from the highest doses calculated for specific TB regions from a uniform deposition of activity in each region. The critical tissue, with respect to ^{222}Rn and short-lived daughters, was assumed here to be the entire bronchial epithelium of the TB region of the lung.

The dose commitment factor for ^{222}Rn and short-lived daughters also will vary with the degree of equilibrium of radon progeny with ^{222}Rn . For this study, it was assumed that radon daughters are in equilibrium

with ^{222}Rn , such that, on the average, for both indoor and outdoor exposures, for every 100 pCi/liter of ^{222}Rn present in the atmosphere, there is an associated 0.5 working level (WL)* of short-lived radon daughters. This assumption was made based on equilibrium values found in the literature for typical dwellings, for which calculated WL concentrations of radon progeny seldom exceed 0.50 per 100 pCi/liter of ^{222}Ra (Johnson *et al.*, 1973). A condition of secular equilibrium, where 100 pCi/liter of ^{222}Ra results in a 1 WL concentration of daughters, is doubtful due to removal processes for progeny in most environments (Jacobi, 1972; Johnson *et al.*, 1973). The degree of disequilibrium for outdoor exposures would be expected to vary greatly due to wide variations in atmospheric stability (Schlein, 1963).

Other variables affecting dose received due to inhalation of radon and short-lived daughters are aerosol particle sizes and the fraction of "free" ions, or those daughters not attached to aerosol particles, both of which affect deposition in the organ of concern. Median diameters of particles with which activity is associated, as measured in uranium mine atmospheres by Harley and Pasternack (1972), are reported to range between 0.2 μm and 0.4 μm . However, Hamrick and Walsh (1974) report that the major fraction of radon daughter activity is associated with particles less than 0.1 μm diameter. In this assessment, it was assumed that all particles were less than 0.1 μm in diameter, which will tend to maximize dose calculations with respect to this parameter, since these sizes deposit with greater efficiency in the region of concern, the TB region of the lung (Morrow *et al.*, 1966). The most accurate assessment would involve dose calculations based on a continuous distribution of particle sizes, but this was not considered necessary due to uncertainties involved with other assumptions made concerning the DCF. A value for the unattached fraction of total potential alpha activity was chosen to be 0.1, which may tend to overestimate dose calculations. This value will actually vary with differences in aerosol characteristics such as humidity and ventilation rates (Johnson *et al.*, 1973). The unattached fraction in this study refers to the uncombined fraction of the total potential alpha activity, rather than that fraction of ^{218}Po atoms, as

* 1 WL = any combination of radon daughters which will result in the emission of 1.3×10^5 MeV of alpha energy in decay through ^{214}Po , per liter of air.

is usually considered, since this designation is relatively independent of ventilation rates (Jacobi, 1973).

Clearance from the TB region of the lung was neglected for radon progeny based on evidence provided by Holleman *et al.* (1968) that it is insignificant for uranium miners. Clearance half-times for the TB are probably much greater than the short radioactive half-lives involved in the ^{222}Rn decay chain (Walsh and Hamrick, 1977). The effective quality factor (QF) used in determining a dose commitment factor for radon progeny was 4 rem/rad, a value based on epidemiological data provided by the committee on the Biological Effects of Ionizing Radiation (National Academy of Sciences, 1972), as derived by Walsh (Walsh, 1976), and on indications that the QF for alpha radiation may be lower than 10, and closer to 3 (Johnson *et al.*, 1973; Walsh, 1976) under these circumstances.

Under these assumptions, a dose factor of 5 rem/WLM* may be derived. Using slightly different assumptions, Jacobi (1973) calculated a factor of 4-5 rem/WLM under normal conditions in mines, and the BEIR report (National Academy of Sciences, 1972) provides a value of 5 rem/WLM (0.5 rad/WLM with a QF of 10 rem/rad). These numbers were based on breathing rates of 20 liters/minute for uranium miners while at work. Since breathing rates averaged over a 24-hour period would be approximately 80% of the miner's 8-hr/day rate (20 liter/minute), calculated by assuming 20 liter/minute for 16-hr/day and 7.5 liter/minute for 8-hr/day (Task Group of Committee 2, 1975), the dose commitment factor should be decreased in the same proportion. Therefore, it seems that the value of 4 rem/WLM is reasonable in the calculation of population organ doses from inhalation of radon and short-lived progeny. Using the previously discussed value for radon daughter equilibrium (i.e., 0.5 WL of radon daughters: 100 pCi/liter of ^{222}Rn), a dose commitment factor of 1.0 mrem/pCi-m³ was derived, for continuous exposures, in pCi/m³ of ^{222}Rn , over a one-year period.

B.2 Fifty-Year Dose Commitment Factors for Inhalation of ^{210}Po

Polonium-210, the alpha-emitting daughter of ^{210}Pb , may contribute to the total inhalation dose received as a consequence of ^{222}Rn released to the atmosphere. Fifty-year dose commitment factors have been derived

*1 WLM = exposure to a concentration of 1 WL for 170 hours (8 hours per day, 5 days per week). Continuous exposure to 1 WL for a month would result in about 4.3 WLM equivalents.

on the basis of available metabolic data for ^{210}Po , as detailed in Sects. 2.2 and 3.2 of this report, and the INREM-II computer code (Killough *et al.*, 1978). Table B lists these factors derived for ^{210}Po in this manner by Dunning *et al.* (in preparation). From these values, it appears that lung is the critical organ for ^{210}Po inhalation.

Table B. Fifty-year dose commitment factors for ^{210}Po inhalation over one year

Organ	rem/ μCi inhaled ^a	mrem/pCi-m ³
Bone	2.0	17
Lung	40	340
Liver	1.2	10
Kidneys	7.1	60
Whole-body	1.0	8.4

^aAn activity median aerodynamic diameter of 0.3 μm was assumed.

NOTE ADDED IN PRESS — The above dose commitment factors (DCF) are based on early drafts of a report by Dunning *et al.* (in preparation). The DCF's given above are based on a quality factor (QF) of 10, which was recommended in ICRP Publication 2 (Pergamon Press, 1959). In more recent drafts of their document, Dunning *et al.* have incorporated recent recommendations from ICRP Publication 26 (Pergamon Press, 1977) regarding quality factors. DCF's based on a higher QF of 20 are given in Appendix C of the present report.

References — Appendix B

- Dunning, D. E., Jr., S. R. Bernard, P. J. Walsh, G. G. Killough, and J. C. Pleasant, *Estimates of Internal Dose Equivalents to 22 Target Organs for Radionuclides Occurring in Routine Releases from Nuclear Fuel-Cycle Facilities, Vol. II*, NUREG/CR-0114/V2, ORNL/NUREG/TM-190/V2 (in preparation).
- Hamrick, P. E. and P. J. Walsh, "Environmental Radiation and the Lung," *Health Perspectives* 9, 33-52 (1974).
- Harley, N. H. and B. S. Pasternack, "Alpha Absorption Measurements Applied to Lung Dose from Radon Daughters," *Health Phys.* 23, 771-782 (1972).
- Holleman, D. F., K. J. Schiager and A. H. Dahl, *Radiation Dosimetry for the Respiratory Tract of Uranium Miners*, Colorado State University, AEC Project Report AE (11-1)-1500 (1968).
- Jacobi, W., "Relations Between the Inhaled Potential α -Energy in the Bronchial and Pulmonary Regions," *Health Phys.* 23, 3-11 (1972).
- Jacobi, W., "Relation Between Cumulative Exposure to Radon Daughters, Lung Dose, and Lung Cancer Risk," in *Proceeding of Nobel Gases Symposium*, Las Vegas, Nevada (September 1973).
- Johnson, P. H., D. E. Bernhardt, N. S. Nelson and H. W. Colley, Jr., "Assessment of Potential Radiological Health Effects from Radon in Natural Gas," pp. 22-24 in *Environmental Protection Agency*, EPA-520/1-73-004 (November 1973).
- Killough, G. G., D. E. Dunning, Jr., and J. C. Pleasant, *INREM-II: A Computer Implementation of Recent Models for Estimating the Dose Equivalent to Organs of Man from an Inhaled or Ingested Radionuclide*, ORNL/NUREG/TM-84 (1978).
- Morrow, P. E., D. V. Bates, B. R. Fish, R. F. Hatch, and T. T. Mercer, "Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract," *Health Phys.* 12, 173-207 (1966).
- National Academy of Sciences, Division of Medical Sciences, *Biological Effects on Populations of Exposure to Low Level of Ionizing Radiation (BEIR report)*, National Academy of Sciences, Washington, D.C. (1972).
- Shleien, B., "The Simultaneous Determination of Atmospheric Radon by Filter Paper and Charcoal Adsorptive Techniques," *J. Am. Ind. Hyg. Assoc.* 24, 180-187 (1963).

- Task Group of Committee 2 of the International Commission on Radiological Protection, *Report of the Task Group on Reference Man*, ICRP Publication 23 (1975).
- Walsh, P. J., "Radiation Dose to the Respiratory Tract of Uranium Miners -- A Review of the Literature," *Environ. Res.* 3, 14-36 (1970).
- Walsh, P. J., "Dose to the Tracheobronchial Tree Due to Inhalation of Radon Daughters," *Proceedings of the 10th Midyear Topical Symposium of the Health Physics Society*, Saratoga Springs, New York (October 1976).
- Walsh, P. J. and P. E. Hamrick, "Radioactive Materials -- Determinants of Dose to the Respiratory Tract," pp. 233-242 in *Handbook of Physiology*, American Physiological Society, Bethesda, Maryland (1977).

APPENDIX C

(added in press)

The dose conversion factors (DCF's) presented here are tentative. These tentative values are based on a report by Dunning *et al.* (in preparation) which is still under review. The DCF's given in this appendix are based on a quality factor (QF) of 20 for alpha emitters, which was recently recommended in ICRP Publication 26 (International Commission on Radiological Protection, 1977), rather than on the QF of 10 which has been used extensively in the past.

Table C.1. Fifty-year dose commitment factors for ^{210}Pb inhalation over a year (QF = 20)

Organ	rem/ μCi inhaled	mrem/pCi-m ³
Bone	220	1800
Lung	11	92
Liver	3.4	29
Kidneys	4.4	37
Whole-body	16	130

Table C.2. Fifty-year dose commitment factors for ^{210}Pb ingestion (QF = 20)

Organ of reference	rem/ μCi ingested
Bone	100
Kidney	0.96
Liver	1.44
Whole-body	7.5

Table C.3. Fifty-year dose commitment factors for one year's inhalation and ingestion of ^{226}Ra (QF = 20)

Organ	Inhalation ^a		Ingestion
	rem/ μCi	mrem/pCi-m ³ in a 1-year period ^b	rem/ μCi
Bone	49	410	43
Lung	95	800	1.3×10^{-4}
Liver	0.68	5.7	0.60
Kidney	0.68	5.7	0.60
Whole body	20	170	16

^a0.3 μm AMAD, Solubility Class W.

^bBased on inhalation rate 23 m³/day.

Table C.4. Fifty-year dose commitment factors for ^{210}Po inhalation over one year (QF = 20)

Organ	rem/ μCi inhaled ^a	mrem/pCi-m ³
Bone	3.9	33
Lung	78	650
Liver	2.4	20
Kidneys	14	120
Whole-body	2.0	17

^aAn activity median aerodynamic diameter of 0.3 μm was assumed.

REFERENCES — APPENDIX C

- Dunning, D. E., Jr., S. R. Bernard, P. J. Walsh, G. G. Killough, and J. C. Pleasant, *Estimates of Internal Dose Equivalents to 22 Target Organs for Radionuclides Occurring in Routine Releases from Nuclear Fuel-Cycle Facilities, Vol. II*, NUREG/CR-0114/V2, ORNL/NUREG/TM-190/V2 (in preparation).
- International Commission on Radiological Protection, *Recommendations of the International Commission on Radiological Protection*, ICRP Publication 26, Pergamon Press (1977).