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Environmental Radon and Radon Daughter Dosimetry in the Respiratory Tract

R. B. McPherson

April 1979

Prepared for the U.S. Nuclear Regulatory Commission

Pacific Northwest Laboratory Operated for the U.S. Department of Energy by Battelle Memorial Institute

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ENVIRONMENTAL RADON AND RADON DAUGHTER DOSIMETRY IN THE RESPIRATORY TRACT

R. B. McPherson

April 1979

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Pacific Northwest Laboratory Richland, Washington 99352

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SUMMARY

This report describes work performed by Pacific Northwest Laboratory (PNL) for the Nuclear Regulatory Commission's program to determine safety and costs related to decommissioning nuclear fuel cycle facilities. Individual dose factors for the inhalation of radon and its daughter products are calculated for use in environmental dose assessments. The calculated committed dose equivalent factors for ²²²Rn and its daughters are given below. An activity median aerodynamic diameter of 0.1 µm was used. The dose to an individual is calculated by multiplying the estimated intake from inhalation for a particular radionuclide by the corresponding dose factor.

		rem/pCi Inhaled		
Radionuclide	Pulmonary, Lung	Tracheobronchial Region (epithelium)(a)	Other Organs	
$^{222}Rn + D^{(b)}$	9.1 × 10 ⁻¹⁰	3.6 x 10 ⁻⁹	8.2 × 10 ⁻¹⁰	
²¹⁸ Po + D	1.8 x 10 ⁻⁸	3.2×10^{-7}	negligible	
²¹⁴ Pb + D	9.2 x 10 ⁻⁸	5.1 x 10 ⁻⁸	negligible	
214Bi + D	7.0 x 10 ⁻⁸	1.2 x 10 ⁻⁷	negligible	

Calculated Committed Dose Equivalent Factors for $^{\rm 222}\rm Rn$ and its Daughters

(a)Dose to the epithelial tissue of the tracheobronchial (T-B) region.
 (b)+ D indicates that after inhalation the decay energy of the short lived daughters is included with the parent.

A working level month-to-dose conversion factor is calculated to be 1 rad/WLM to the epithelial tissue of the T-B region, assuming 100% daughter equilibrium and 10% free ²¹⁸Po ions. This value is in reasonable agreement with recently reported values.

CONTENTS

ACKNOWLEDGMENT		* 1	*	*	*	·	•	•	•	•		iii
SUMMARY .	÷. '											V
INTRODUCTION		•								•		1
PHYSICAL PROPER	RTIES									•		1
RESPIRATORY SYS	STEM											3
BIOLOGICAL HALF	-TIME	S										4
ENVIRONMENTAL S	STATE											5
DEPOSITION											•	5
ABSORBED ENERGY	1							•				7
LUNG CANCERS AN	ND BIO	LOGI	CAL	TARGET								8
DOSE FACTORS		•			1							9
WORKING LEVEL									 1			11
DOSE EQUIVALENT							92			12		12

TABLES

1	²²² Rn Decay Series	•	2
2	Potential Alpha Energy per Atom Deposited		2
3	Deposition Fractions for Rn Decay Products for Nose Breathing at a Ventilation Rate of 15 ℓ/min		6
4	Absorbed Alpha Energy in Tissue for Radon Daughters		8
5	Tissue Mass Assumptions		9
6	Calculated Absorbed Dose Commitment Factors for Radon Daughters		10
7	Reported WLM to Absorbed Dose Comparisons	÷	12
8	Calculated Committed Lose Equivalent Factors for ²²² Rn and Its Daughters		13

INTRODUCTION

A review of the current literature has led us to the conclusion that little information exists on dose calculations for inhalation of individual radon daughters. However, there are many reported values for the conversion of working level month to dose. These reported values cover a wide range. This study presents a review of the reported working level month-to-dose conversions and of the parameters used to calculate dose from inhalation of radon and its daughter products. Individual dose factors for radon daughters are calculated using the International Commission on Radiological Protection Task Group Lung Model and selected parameters from the literature. These dose factors are suggested for use in estimating the environmental consequences to people from exposure to ²²²Rn and its daughters, such as from exposure to a uranium mill tailings pile. A working level month-to-dose conversion factor is calculated to compare the dose calculations in this study to other reported values for radon and its daughters.

PHYSICAL PROPERTIES

The important isotopes in the ²²²Rn decay series are listed in Table 1, along with their half-lives and alpha radiations. For internal radiation dosimetry, all other radiations (β and γ) from these radionuclides are negligible when compared to the alpha particles (Haque and Collinson 1967, Fry 1975, Eisenbud 1973, pp. 28-32, FRC 1967, Altshuler, Nelson and Kuschner 1964). This is due to the greater penetration of the beta and gamma radiation, resulting in the distribution of ionizing energy over much more tissue.

The long-lived daughters of radon, ²¹⁰Pb(RaD), ²¹⁰Bi(RaE), and ²¹⁰Po(RaF), are biologically eliminated from the bronchi and lung before any significant number decay (Fry 1975, Eisenbud 1973, pp. 28-32, FRC 1967, Altshuler, Nelson and Kuschner 1964). Since the half-life of ²¹⁴Po (163.7 usec) is much less than its parent, ²¹⁴Bi (19.8 min), ²¹⁴Po is considered to always be in equilibrium with ²¹⁴Bi. Thus, the alpha emission from ²¹⁴Po may be regarded as

1

a prompt alpha emission from ²¹⁴Bi (Altshuler, Nelson and Kuschner 1964, Cliff 1978). The potential alpha energy per atom deposited for radon daughters is given in Table 2. The potential alpha energy for ²¹⁴Po by itself can be ignored, due to its short half-life.

TABLE 1. 222Rn Decay Series^(a)

Radionuclide	Half-Life	Alpha	Radiation		
222Rn	3.824 days	5.49	MeV 100%		
²¹⁸ Po'RaA)	3.05 min	6.00	MeV 99+%		
214Pb(: aB)	26.8 min	β, γ			
²¹⁴ Bi(RaC) ^(b)	19.8 min	β,γ			
214Po(RaC')	163.7 µsec	7.69	MeV 100%		
²¹⁰ Pb(RaD)	22.3 yrs	β, γ			

(a)Taken from Chart of the Nuclides, Twelfth Edition (Walker, Kirouac, and Rourke 1977).

(b)The small fraction of ²¹⁴Bi which decays by alpha emission to ²¹⁰Tl (0.21%) (Lederer, Hollander and Perlman 1967) is of little consequence. Thallium-210 (RaC") is a β, γ emitter.

TABLE 2. Potential Alpha Energy per Atom Deposited

Radionuclide Index	Radionuclide	Alpha Energy (MeV)
1	²¹⁸ Po + D ^(a)	13.69
2	214Pb + D	7.69
3	²¹⁴ Bi + D	7.69

(a)+ D indicates that after deposition, the decay energy of the short-lived daughters is included with the parent.

RESPIRATORY SYSTEM

Figure 1 illustrates the principal anatomical features of the human respiratory tract. The tissue areas of concern for radon and its daughters deposited in the respiratory tract are the epithelium in the tracheobronchial





FIGURE 1. The Principal Anatomical Features of the Human Respiratory Tract (National Acadamy of Sciences - National Research Council 1961)

3

585268

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region (T-B) and the alveolar region of the pulmonary (P). The T-B region is considered to consist of the trachea and the bronchial tree down to the terminal bronchioles. Similarly, the pulmonary region includes the terminal bronchioles, respiratory bronchioles, alveolar ducts, atria, alveoli and alveolar sacs. In this study, lung dosimetry for radon daughters is performed using the Task Group Lung Model (TGLM) (ICRP 1966) with a modification for the particular mass of tissue included in the T-B and P compartments. The respiratory and terminal bronchioles are included in the pulmonary region due to their number, total circumference, total surface area, and long mucus transit time relative to other bronchioles. Almost all of the radon daughters deposited in this region undergo radioactive decay before moving to the T-B region. For these reasons and because of the relative thinness of their walls, the terminal bronchioles are treated with the alveolar region instead of with the T-B region for the purpose of dose calculations as recommended by Altshuler (1978).

Biological removal of radon daughters deposited in the epithelium of the T-B region is accomplished by ciliary action and absorption by tissue. Accumulation of radon daughters in the lymph nodes is negligible because of their short radioactive half-lives (Eisenbud 1973, pp. 28-32, ICRP 1966, Jacobi 1964).

BIOLOGICAL HALF-TIMES

Biological removal of highly soluble dust in the alveolar region of the lung is by resorption from the lung into the blood stream and passage to other organs (Eisenbud 1973, pp. 28-32, Jacobi 1964). The removal of dust particles (laden with radon daughters) by phagocytes seems to be unimportant in dosimetry calculations due to the short radioactive half-lives (Jacobi 1964). The biological half-time is the time required for the body to eliminate onehalf of a radionuclide by regular biological processes of elimination. Biological half-times for radon daughters have been reported to range from 6 to 60 hours in the pulmonary region and 10 minutes to 4.8 hours in the T-B region (Altshuler 1964, ICRP 1966, Jacobi 1964, UNSCEAR 1977, pp. 68-80, ICRP 1972, Jacobi 1972). With such short clearance in the respiratory

585269

tract, the adsorbed daughters must be able to leave the dust particles they were originally attached to soon after deposition. Adsorbed daughters are weakly attached to dust, and in a liquid medium the adsorbed atom might be easily removed, since molecular forces are greater in liquid than air. Solvation of the dust particles would also release the adsorbed atom. Thus the radon daughters appear to behave as soluble substances, probably due to their special physical state (ICRP 1966). The biological half-times for Class D material are the most appropriate to use for radon daughters. These values are 12 hours in the P region and for the T-B region, 14.4 minutes for 95% of the deposited radon daughters and 4.8 hours for the remaining 5% (ICRP 1972).

ENVIRONMENTAL STATE

RaA (218Po), formed by the decay of 222Rn gas, initially exists as a highly mobile free ion or atom. In relatively dust-free air, it may persist in this form for about 50 seconds. The mean life-time for unattached ions is 10 to 50 seconds (FRC 1967, Jacobi 1964). The free ions tend to form clusters very rapidly with water, oxygen, or other trace gases. All radon daughter products eventually attach themselves to available surfaces such as dust particles in the atmosphere or become nuclei for the concentration of water vapor, and cease to follow gaseous radon (FRC 1967, UNSCEAR 1977, pp. 68-80, Harley 1973). The average fraction of free ²¹⁸Po ions in air is reported to range from 1 to 10% (Haque 1967, ICRP 1966, UNSCEAR 1977, pp. 68-80, Jacobi 1972, Harley 1973, ICRP 1959, Budwitz 1974). However, a maximum value of 73% was reported by Craft (1966). A conservative value of 10%, as recommended by Harley (1973), ICRP-II (1959), and Budwitz (1974), is used in this paper for environmental air. The attached radon daughters are assumed to have an activity median aerodynamic diameter (AMAD) of 0.1 um (Jacobi 1972), which appears to be most representative of environmental air.

DEPOSITION

Deposition within the respiratory tract may result from inertial impaction, settling, or Brownian motion for particulates less than 0.1 µm in dia-

585270

meter. Larger particles are deposited between the nasal passageways and the lower bronchi. Small particles have a high probability of reaching the alveoli (Eisenbud 1973, pp. 28-32). Free ions impinge on the walls of the T-B region and the terminal bronchioles and are trapped with virtually 100% efficiency (Fry 1975, FRC 1967). However, free ions would not be expected to travel fully through the nose as well as particles, thus the importance of free ions is disputed (ICRP 1966). Deposition values for attached and unattached atoms are taken from Altshuler (1964) and are presented in Table 3 for the different regions of the respiratory system.

> <u>TABLE 3</u>. Deposition Fractions for Rn Decay Products for Nose Breathing at a Ventilation Rate of 15 2/min

	Deposition	n Fraction
Respiratory Region	Free Atoms(a)	Attached Atoms(b)
T-B	0.6	0.01
Р	0.2	0.5

(a)Taken from Altshuler et al. (1964) for a diffusion constant of 0.054 cm²/sec. Fractions are rounded to one significant figure.

(b)Taken from Altshuler et al. (1964) for 0.1 µm size particles (nuclei) for a diffusion constant of 1.6 x 10⁻⁵ cm²/sec. Fractions are rounded to one significant figure.

Assuming 10% free ²¹⁸Po ions, total deposition fractions for ²¹⁸Po are calculated to be 0.07 for the T-B region and 0.47 for the P region. For the rest of the radon daughters, the deposition fractions for attached atoms are used.

ABSORBED ENERGY

The absorbed alpha energy per pCi deposited is calculated with Equation 1.

$$W_{i} = 2.22 E_{i} / \lambda_{ri}$$
(1)

where

- W_i the absorbed alpha energy per pCi of radionuclide i deposited, including contribution from daughter products, MeV/pCi
- 2.22 the number of disintegrations per min per picocurie, dis/min per pCi
 - E_i the absorbed alpha energy per atom deposited for radionuclide i, including contribution from daughter products, MeV/atom deposited
- the radioactive decay constant for radionuclide i, min⁻¹.

The alpha energy absorbed in tissue per atom deposited is calculated for the T-B and P region using Equation 2.

$$E_{i} = \int_{j=1}^{3} \varepsilon_{j} \frac{\pi}{k=i} [\lambda_{rk} / (\lambda_{b} + \lambda_{rk})]$$
(2)

585272

where

- the alpha energy emitted per disintegration for radionuclide j, MeV
- the biological removal constant in the region of interest, min⁻¹
- $\boldsymbol{\lambda}_{rk}$ the radioactive decay constant for radionuclide k, min^-1.

The radionuclide index for each radon daughter is given in Table 3. Allowance must be made for more than one biological removal pathway.

To calculate the absorbed alpha energy, W_i ', in MeV per pCi inhaled, W_i is multiplied by the deposition fractions. The alpha energy absorbed in tissue for each radon daughter in the T-B and P regions of the respiratory tract is given in Table 4.

Rn Daughter	Free Ion Fraction	Potential a Energy MeV/Atom	Respiratory Region	Absorbed a E per Atom Deposited, E	nergy in MeV per pCi Deposited, W	Absorbed a Energy in MeV per pCi Inhaled, W'
218po + D(RaA)	10%	6.0	P	5.98	58.4	27.5
		7.69	P	7.19	70.2	33.0
		13.69				60,5
			T-B	5.00	48.9	3.42
			T-B	1.09	10,6	0.74
						4,16
214P5 + D(RaB)	0	7.69	p	7.22	620	310
			T-8	1.10	94.2	0,94
214Bi + D(RaC)	0	7.69	P	7.49	472	236
			T-B	3.44	217	2.17

TABLE 4. Absorbed Alpha Energy in Tissue for Radon Daughters

LUNG CANCERS AND BIOLOGICAL TARGET

Observed lung carly's among uranium miners from radon daughter inhalation appear to arise primarily in the bronchi of the T-B compartment near the hilus of the lung. The integrity of the epithelial tissue depends on continued integrity of the basal cells. Thus, the relevant biological target is considered by some to be the nuclei of the basal cells in the bronchial epithelium (Eisenbud 1973, pp. 28-32, FRC 1967, UNSCEAR 1977, pp. 68-80, Parker 1969). The distance between the surface of the epithelium and basal cell nuclei (source and biological target) in the bronchus is reported to range from 27 μ m to 85 μ m (Gastineau, Walsh and Underwood 1972). The bronchioles are not included in this range since they are treated with the pulmonary region. The ranges in tissue for the 6.00 MeV alpha emitted from ²¹⁸Po and the 7.69 MeV alpha emitted from ²¹⁴Po have been calculated to be 47 μ m and 71 μ m, respectively (FRC 1967, Altshuler, Nelson and Kuschner 1964).

DOSE FACTORS

In this study, the regional dose to the bronchial epithelium of the T-B compartment is averaged over the depth of penetration to estimate the dose to the basal cells from inhaled radon daughters. It is assumed that all alpha energy emitted by deposited radon daughters is absorbed by the near and far wall of the bronchi.

Dimensions for the trachea and bronchi are reported by Altshuler (1964) for the Landahl lung model. These dimensions are used to calculate a total surface area of 417 cm² for the T-B compartment. The regional mass receiving the dose from absorption of the 6.00 MeV and the 7.69 MeV alphas is calculated by assuming a tissue density of 1 g/cm³ and using the ranges in tissue for the two alpha particles. The results are reported in Table 5. A mass of 540 g was calculated from ICRP-23 (1975) for the P region. This includes the weight of the lung tissue, associated lymph nodes, and capillary blood. The weight of the arterial and venous blood was not included since the alpha particles do not penetrate through the tissue to this depth. Thus it is assumed that the arterial and venous blood does not receive any dose.

-	n. 1	Ph 8 7		pr-	1996 3		1.1				
Ε.	A	51.1	K	3 .	11	SSUP	Mass	ASI	SUM	371	075
5.1				M. 7			1100 0 0	1. 2. 10.	~ W1115	N. 18. 1	1411-2

Respiratory Region	Alpha Energy (MeV)	Mass (g)		
Р	a11	540		
T-B	6.00	1.96		
T-B	7.69	2.96		

The committed dose factor to tissue is calculated with Equation 3.

$$DCF_{z} = 1.602 \times 10^{-8} W_{z}'/M$$
 (3)

where

DCF_i • committed dose factor for tissue from radionuclide i, rad/pCi inhaled 1.602 x 10⁻⁸ • conversion factor, g-rad/MeV

- W_i' absorbed alpha energy per pCi inhaled in the respiratory region of interest for radionuclide i, MeV/pCi inhaled (given in Table 4)
 - M mass of the respiratory region of interest, g (given in Table 5).

The regional dose averaged over the depth of alpha penetration is calculated for each radon daughter using Equation 3 and is reported in Table 6.

TABLE 6. Calculated Absorbed Dose Commitment Factors for Radon Daughters

Radionuclide			Lur	rad/pCi Inhaled Lung (P) T-B(
218Po	+	D(p)	1.8	x	10-9	3.2	×	10-8	
214Pb	+	D	9.2	х	10-9	5.1	х	10-9	
²¹⁴ Bi	+	D	7.0	х	10- 9	1.2	x	10-8	

(a)Dose to the epithelial tissue.

(b)+ D indicates that after deposition, the decay energy of the short-lived daughters is included with the parent.

Radon gas, having a long half-life relative to the time a breath of air remains in the lungs (\approx 17 seconds using a plug flow model) tends to be exhaled without depositing a significant amount of radiation energy from daughter products in the lung. To calculate the dose to the lung from inhaled radon by itself (radon is a noble gas), a methodology similar to that presented by Soldat et al. (1973) is used. It is assumed that all alpha energy emitted from the 222 Rn gas in the lung is absorbed in the lung tissue. A lung volume of 4 liters is assumed to be contaminated to the same radon concentration as outside air.

Dose factor =
$$(1 \text{ pCi/m}^3) * (10^{-3} \text{ m}^3/2) * (42) * (2.22 \text{ dis/min-pCi}) * (5.49 \text{ MeV/dis}) * (60 \text{ min/hr}) * (8,766 \text{ hr/yr}) * (1.602 x 10^{-9} \text{ g-rad/MeV}) * (\frac{1}{540\text{g}}) * (\frac{1}{8,400 \text{ m}^3/\text{yr}}) = 9.1 x 10^{-11} \text{ rad/pCi inhaled}$$

585275

The inclusion of the contribution from deposited daughter products would increase this dose factor by about 6%.

The dose factor for the T-B compartment is calculated in the same way using an air volume of 4.95 x 10^{-2} & (Altshuler, Nelson and Kuschner 1964). The range in tissue for the 5.49 MeV alpha emitted from ²²²Rn is calculated to be 41 µm. Thus, the regional mass affected by the radon energy is 1.71 g. The dose factor for the T-B compartment is calculated to be 3.6 x 10^{-10} rad/pCi inhaled.

WORKING LEVEL

The working level (WL) is defined as any combination of short-lived radon daughters (through 214Po) in a liter of air that will result in the emission of 1.3 x 105 MeV of alpha energy. A 222Rn concentration of 100 pCi/2 with its daughters in equilibrium corresponds to a potential alpha-energy concentration of 1 WL. The working level month (WLM) is an exposure to a radon daughter concentration of 1 WL for 170 working hours. The radiation dose one would receive from a theoretical WLM, assuming 10% free ²¹⁸Po ions, and a human ventilation rate of 1.2 m3/hr (ICRP 1975), is calculated to be 0.4 rad to the lung and 1 rad to the epithelium region of the T-B compartment. Dose estimates for the working level concept have been reported to range from 0.2 to 15 rad/WLM (Haque and Collinson 1967, Eisenbud 1973, pp. 28-32, FRC 1967, Altshuler, Nelson and Kuschner 1964, Jacobi 1964, UNSCEAR 1977, pp. 68-80, Harley 1973, Parker 1969, Nelson and Parker 1974, EPA 1973, Barton, Moore and Rohwer 1973, Black et al. 1968, Auxier 1976, Harley and Pasternack 1972, Turner, Haloway and Loebl 1978, Jacobi 1973), depending on the assumptions for lung mass, free ion fraction, deposition fractions, and daughter equilibrium. Table 7 gives reported dose values for the WLM by some of these authors and a comparison with the method used in this paper. Where possible, the same free ion fraction for ²¹⁸Po and the daughter equilibrium assumptions are used for our recalculated value. Our recalculated conversion factors are in reasonable agreement with most of the reported values in Table 7. The 1972 BEIR report suggests using 0.5 rad/WLM as an average value, will an upper limit of 1 rad/WLM (National Academy of Sciences 1972).

11

TABLE 7. Reported WLM to Absorbed Dose Comparisons

rad/WLM	Tissue Region	Reference	Calculated Using Methodology in This Paper(rad/WLM)(a)
1	Bronchial epithelium basal cells 023 µm	UNSCEAR 1977	1
0.6	Bronchial epithelium basal cells	Harley 1973	0.9
1.6	Bronchial epithelium (basal cell nuclei @60 um)	EPA 1973	0.7
0.8 to 1.7	Bifurcation regions of bronchi	Auxier 1976	1
1.7 Nose 3.3 Mouth	Epithelium of seg- mental bronchi	Altshuler et al. 1964	0.7
1 to 2	Bronchial tissue	Black et al. 1968	0.8
2.8	Bronchial epithelium	FRC 1967	1.2
3.9	Epithelium tissue	Jacobi 1964	1.8
6.4 Living accommodations, adequate ventilation	Sronchial epithelium basal cells @ 35 µm	Haque 1967	2
3 Outdoor air	Bronchial epithelium	Turner et al. 1978	1
0.5 to 1	Basal cells	Jacobi 1973	1
0.5	Mean bronchial dose	Jacobi 1973	1

(a)Calculated using the assumptions of free ion fraction and daughter equilibrium given in the particular reference cited.

DOSE EQUIVALENT

Reported quality factors for alpha irradiation in the respiratory system range from 1 to 10 (Haque and Collinson 1967, Eisenbud 1973, pp. 28-32, Altshuler, Nelson and Kuschner 1964, Jacobi 1964, ICRP 1959, Parker 1969, EPA 1973, Barton, Moore and Rohwer 1973). A value of 10, recommended by ICRP-2 (1959), is used in this paper.^(a) Using a quality factor of 10, the dose factors in Table 6 are converted to units of rem per pCi inhaled and are listed in Table 8.



⁽a)ICRP-26 (1977) recommends a quality factor of 20 for alpha particles. However, this suggestion has not been adopted by the National Council on Radiation Protection (NCRP). If adopted, the dose equivalent factors in Table 8 should be multiplied by a factor of 2.

TABLE 8. Calculated Committed Dose Equivalent Factors for ²²²Rn and Its Daughters

	rem/pCi Inhaled		
Radionuclides	Lung (P)	T-B (epithelium)	Other Organs
$222Rn + D^{(a)}$	9.1 x 10 ⁻¹⁰	3.6 × 10 ⁻⁹	8.2 x 10-10(b)
²¹⁸ Po + D	1.8 x 10 ⁻⁸	3.2 x 10-7	Negligible ^(c)
²¹⁴ Pb + D	9.2 x 10 ⁻⁸	5.1 x 10 ⁻⁸	Negligible
²¹⁴ Bi + D	7.0 x 10 ⁻⁸	1.2 x 10-7	Negligible

(a)+ D indicates that the decay energy of the short-lived daughters is included with the parent. (b)Dose to other organs from ²²²Rn is approximately equivalent to the

dose to the lung; See Pohl (1977). Eisenbud (1973, pp. 28-32) reports an estimated dose factor for the whole body from inhaled Rn that is partially absorbed to be 8.2 x 10^{-10} rem/pCi inhaled. This is based on UNSCEAR (1966).

(c)Dose to organs other than the respiratory tract from radon daughters deposited in the respiratory tract is negligible; see Pohl (1977).

The rad per WLM conversion is not calculated for use as a dose rate estimato for exposure to environmental concentrations of radon daughter product . Nor is it appropriate or appealing from a risk standpoint (Turner, Haloway and Loebl 1978, Morken 1969). It is calculated as a means to compare the dose factors developed in this study with those developed by others (see Table 7). The rad per WLM conversion factor can also be used to convert exposure guidelines from WL to dose, such as the Surgeon General's Guidelines (1971 pp. 52-54) on use of uranium mill tailings for construction purposes. When determining environmental impact assessments, the dose factors given in Table 8 should be used instead of the WLM approach.

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