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THE DOSIMETRY OF THE RADIOACTIVE NOBLE GASES*

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The dosimetry of the radioactive noble gases is of widespread interest. Naturally occurring radioactive noble gases are limited to isotopes of radon (element 86) but they are important contributors to the internal doses received by uranium mine workers. Radioisotopes of argon are formed by neutron activation of the stable argon present in air at low concentration. Radioisotopes of krypton and xenon are produced as fission products during the fissioning of uranium or plutonium fuels in nuclear reactors. They are generally retained within the fuel elements until the fuel is chemically reprocessed. Small amounts of fission products,however, are released from reactor facilities as a result of fuel element leaks.

The dosimetry of ^{85}Kr was discussed in detail by the previous speaker (Dr. W. S. Snyder). It is my intent to extend the dosimetry discussion to several other artificially created radioactive noble gases but in somewhat less detail than covered by Dr. Snyder.

The naturally occurring isotopes of radon will not be discussed further.

As Dr. Snyder has demonstrated, the complete dosimetry of the noble gases includes calculation of several contributions - external dose to the skin from both beta and gamma radiations, external dose to the total body, gonads and internal organs, dose to the lung from inhaled radioisotopes and dose to body from noble gases dissolved in the bloodstream and then absorbed in the various tissues.

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Doses from ingestion of noble gases are generally not addressed in the literature since noble gases have low solubility in water. Although noble gas solubility is higher in pressurized water systems, the release of the pressure results in escape of the gas. Hence, the ingestion of unusually high concentrations of noble gas in water is unlikely. The concentration of the noble gases in the water at the time of ingestion would be difficult to estimate. Adding to the uncertainty in such an estimation is the fact that xenon may be retained in the water to some degree when organic material is present.

It has been aptly demonstrated by several workers (including Dr. Snyder) that the dose from noble gases absorbed in tissue is generally small compared to the dose from direct external radiation and from inhalation.(1,3,10,12) These internal doses are, however, the ones to consider for medical applications of these nuclides. In a subsequent section some of these results will be tabulated and compared with each other and with external and inhalation doses.

External doses resulting from submersion in a large plume of radioactive gas can be easily calculated if the concentration of the material in the air is relatively uniform. Doses can be determined on the assumption that the plume is "infinite" in volume relative to the range of the emitted radiations. Under this assumption the energy absorbed per gram of material is equivalent to the energy emitted per gram. All that is required then is to convert the average energy per disintegration to dose and to correct for differences in energy absorption between air and tissue and for the physical geometry of each specific exposure situation.

Frequently the airborne concentration surrounding a person is very nonuniform; for example, when he is standing close to a tall stack. For this situation the radiation from the overhead plume must be taken into consideration when evaluating the external dose. Several computer programs have been developed for treating the required numerical integration of the contribution from each finite cloud element.(14) Such specialized situations were not included in the present work, but the calculations are manageable once the goemetry has been defined.

The dose from submersion in air is an external dose to either the skin only or the skin and the total body, depending upon the penetrating power of the radiation emitted from the airborne radioisotope.

All of the noble gases considered in this paper emit only beta and gamma radiation. For the present work, we have converted rad to rem using a quality factor of 1, except a Quality Factor (QF) of 1.7 was used for beta particles and electrons with maximum energies equal to or less than 30 keV.(2) It has been reported by H. J. Dunster that the ICRP is seriously considering the use of a QF of 1.0 for all beta and electron energies.(16)

In addition, we have chosen to calculate the skin dose at a depth of 7×10^{-3} cm (7 mg/cm^2) and the total-body dose at a depth of 5 cm as originally suggested by the NCRP.(6) Recent measurements indicate that 4 or 5 mg/cm^2 might be more appropriate than 7 mg/cm^2.(13) The dose to the male gonads was calculated at a depth of 1 cm in tissue. No separate dose calculation was made, however, for the female gonads. The total-body dose can be used as an upper bound to the female gonad dose from external radiation.

Ratio of surface to depth dose was estimated for each maximum beta energy by methods given by Loevinger, et al,(5) and summed by nuclide. Gamma radiation dose at each of the 3 depths in tissue was determined from the values of absorption coefficients for muscle and the ratio of stopping power for muscle and air tabulated by the National Bureau of Standards.(6) The decay schemes for the nuclides were taken from Lederer, et al.(4) (a)

(a)Note that the abundance values for the 2 beta transistions of Xe-137 were transposed in the Xe-137 decay diagram. The values given in the tables on pages 73 and 285 of Reference 4 were used.

For a person standing on the ground surrounded by a very large hemisphere of radioactive gas the geometry for gamma radiation is obviously 2π. For beta radiation with its shorter range in air, the physical arrangement approaches the infinite volume (4π geometry). However, since the beta is of limited penetrating power, it will irradiate the skin from only one side and again the geometry will be 2π.[a]

The resulting equation for calculation of the dose from air submersion is:(11)

$$(\text{D.F.})_{\text{air sub}} = 0.887\ (\overline{E}_\beta + \overline{E}_\gamma) \qquad (1)$$

where $(\text{D.F.})_{\text{air sub}}$ is the dose factor in units of mrem/hr per μCi/m^3,

$\overline{E}_\beta$, $\overline{E}_\gamma$ are the effective energies of the beta and gamma radiations, respectively, calculated at the depth of interest (7×10^{-1} or 5 cm) and corrected for relative stopping power.

The constant takes into account the density of air (1.2×10^{-3} g/cc @ 20°C), the conversion from MeV to mrem and the factor of 1/2 for 2π geometry.

The dose to the lung from the air within it can be calculated from the following equation:

$$(\text{D.F.})_{\text{inhalation}} = 2.13\ \overset{V}{V_L}\ \varepsilon/m \text{ mrem/hr per } \mu\text{Ci/m}^3 \qquad (2)$$

where V_L is the volume of air within the lung, 4 liters,(10)

ε is the effective energy deposited in the lung, MeV per disintegration,

m is the mass of the lung, 1000 g.(2)

The concentration of the noble gas in the air within the lung is taken to be the same as that in the inspired air. The effective energy, ε, is calculated from the formula of the ICRP(2) using an effective radius of 10 cm for the lung.

The results obtained from applying Equation 1 to calculation of the external dose rates to skin, testes and total body from 14 radioactive noble gases are

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[a] Minor exceptions can occur in thin membranes, such as a protruding ear which could receive beta radiation from both sides, approaching a 4π geometry.

summarized in Table I. Also included in Table I are the dose rates to lung tissue from inhalation of these noble gases as well as the combined dose to the lung from both inhalation and external gamma irradiation.

Two important daughter radionuclides, Rb-88 and Cs-138, have been included in Table I. These two nuclei make significant contributions to the dose rates from their parent activities. Rb-88 has a short half-life (18 min) and will nearly always be present at equilibrium with its parent Kr-88 (2.8 hr). The total dose rate from the parent-daughter combination is from 30% to 300% higher than from the parent alone, depending upon tissue depth.

Both Xe-138 and Cs-138 have relatively short half-lives, 14 min and 32 min, respectively, and the daughter will contribute a varying amount to the total dose from the Xe-138, depending upon the time since release of the noble gas to the atmosphere. The dose rate per unit concentration of Cs-138 is 150% to 200% of that from its parent Xe-138 but its longer half-life may reduce its relative contribution slightly.

Precise lung dosimetry would require that these two daughter radionuclides be treated as attached to particles. As such there would be a buildup in the lung with an attendant increase in the dose calculated. On the practical level, this buildup may be disregarded because of the short radioactive half-lives of the daughters.

The detailed results for the beta and gamma contributions to the dose to the skin and testes are given in Table II - A and B, where they are compared to similar data presented by Schaeffer(9) at the recent IRPA Congress in Washington, D.C. Dr. Schaeffer calculated the beta dose at a point 'P' in tissue by integrating the contribution from all of the beta particles which were able to reach that point. All of the beta particles were assumed to travel in a straight line through air and tissue. There is excellent agreement between Dr. Schaeffer's results and those obtained with the empirical formula of Loevinger for all nuclides except perhaps Kr-87 where our values are 30% higher.

Dr. Schaeffer's calculations indicated that no beta rays from any of the noble gases were energetic enough to penetrate through the 1 cm of tissue assumed to be over the male gonads. The Loevinger formula, however, implies a small beta contribution to the doses to the testes, especially for the Rb-88 daughter of Kr-88 and for Kr-87 and Xe-137.

Table II-C lists the results obtained by us for the total-body dose rate and compares them with those of Schaeffer(9) and Russell and Galpin.(10) The latter authors employed the reciprocity theorem(5) for their calculation wherein:

> "the internal gamma (total-body dose from an external cloud equals the dose to the cloud from the radioactivity in the body if the concentrations are equal. .... This dose is then the difference between the dose to a point in an infinite medium with uniform source distribution and the dose absorbed in the body from a uniform source in the body with the same concentration as in the infinite media."

This method of calculation yields an average dose over the entire body rather than a dose at a single depth in tissue. Nevertheless, the results obtained by Russell and Galpin are in reasonable agreement with those obtained by Dr. Schaeffer and in the present work utilizing the half infinite cloud calculation.

Dr. Schaeffer's values do not include a correction for attentuation in the first few centimeters of tissue. The attenuation becomes important for the very low energy radiation such as the bremsstrahlung from Kr-85.

For this nuclide the dose calculated in the present work is 2/3 of that calculated by Dr. Schaeffer. Neither of these two sets of calculations, however, include a "build-up factor" which could compensate somewhat for the ommission of the attenuation corrections in Dr. Schaeffer's calculations. In addition to the results shown in the table, Dr. Snyder has reported values of 1.6×10^{-3} for Kr-85(10) and 0.020 for Xe-133.(15)

The calculated internal dose to the lungs from inhalation of noble gases is summarized in Table III along with the values calculated by Russell and Galpin. The latter authors assumed a lung volume of 5.6 liters rather than the 4 liters employed in the present work. As a result our values are generally but not always lower than those of Russell and Galpin. The two exceptions are Kr-88 and Xe-135.

Table III also tabulates internal doses to lung and other tissues calculated by Mrs. Whitton(12) for Kr-85 and Xe-133. The value used by Mrs. Whitton for the lung volume was not stated, but her calculated values for lung dose agree well with ours. As can be seen in the table the doses from noble gases absorbed in tissue are significantly lower than those from external gamma radiation.

Table IV lists the results obtained by several workers for internal and external doses from Kr-85.

The assumptions used by the various authors were not always the same. This is especially true of the depth at which the "skin" dose was calculated and the lung volume. The value for the lung dose attributed to Dr. Schaeffer was not actually given in his paper but was estimated by us from the value of the $(MPC)_a$ which he calculated for lung as the critical organ. As such the value of 5.8×10^{-3} probably includes the contribution from external radiation.

When the variations in assumptions are taken into account there is reasonable agreement among the values obtained by the different authors. It is also obvious that the internal doses are insignificant compared to the external total-body and skin doses. For noble gas nuclides with relatively little penetrating radiation, the critical organ will be the skin even considering its less restrictive dose standard. For the other noble gases, the total body will be the critical organ.

If one were to calculate the $(MPC)_a$ values for these noble gases based upon the dose rates presented in this paper, the values obtained would be much less restrictive than the ICRP values currently accepted. For example, the ICRP $(MPC)_a$ for 168-hr occupational exposure to Kr-85 is 3 $\mu Ci/m^3$. Dr. Schaeffer calculated corresponding values of 20, 170, and 290 for skin, total body and lung (internal and external), respectively, as critical organs. As a result one should always rely on first principles when calculating radiation doses, rather than to simply multiply fractional MPC by the dose standard.

TABLE I

DOSE RATES TO SELECTED TISSUES FROM A SEMI-INFINITE CLOUD OF NOBLE GASES

(mrem/hr per μCi/m^3[a])

	External			Internal	
Nuclide	Skin[b]	Testes[c]	Total Body[d]	Lungs[e]	Total Lungs
Ar-39	0.12	0.00043	0.00033	0.0016	0.0019
Ar-41	1.6	1.1	1.1	0.0067	1.1
Kr-83m	0.0041	$7.2\text{x}10^{-4}$	$1\text{x}10^{-4}$	$4.3\text{x}10^{-4}$	$5.3\text{x}10^{-4}$
Kr-85m	0.32	0.13	0.13	0.0024	0.13
Kr-85	0.16	0.0022	0.0022	0.0019	0.0041
Kr-87	2.7	1.3	1.3	0.013	1.3
Kr-88	2.0	1.5	1.5	0.0067	1.5
(Rb-88)	(2.7)	(0.93)	(0.56)	(0.026)	(0.59)
Xe-131m	0.071	0.023	0.016	0.0013	0.017
Xe-133m	0.15	0.051	0.041	0.0017	0.043
Xe-133	0.069	0.025	0.025	0.0012	0.027
Xe-135m	0.50	0.35	0.35	0.0019	0.35
Xe-135	0.49	0.21	0.21	0.0033	0.21
Xe-137	1.8	0.22	0.12	0.015	0.14
Xe-138+D	1.7	1.2	1.2	0.0066	1.2
(Cs-138)	(3.1)	(1.8)	(1.8)	(0.012)	(1.8)

(a) No credit taken for attenuation by clothing.

(b) At a tissue depth of $7\text{x}10^{-3}$ cm

(c) At a tissue depth of 1 cm

(d) At a tissue depth of 5 cm

(e) Assuming a lung volume of 4 liters

TABLE II

COMPARISON OF EXTERNAL DOSE RATES TO SELECTED TISSUES FROM A SEMI-INFINITE CLOUD OF NOBLE GASES

(mrem/hr per $\mu Ci/m^3$)

A. SKIN

Nuclide	Schaeffer(9) Beta	This Paper Beta(a)	This Paper Gamma(b)	This Paper Total
Ar-39	----	0.12	4.5×10^{-4}	0.12
Ar-41	0.37	0.40	1.2	1.6
Kr-83m	----	0	4.1×10^{-3}	4.1×10^{-3}
Kr-85m	0.17	0.18	0.14	0.32
Kr-85	0.17	0.16(0.21)(c)	2.8×10^{-3}(2.1×10^{-3})(c)	0.16(0.21)(c)
Kr-87	0.83	1.1	1.6	2.7
Kr-88	----	0.32	1.70	2.0
(Rb-88)	(----)	(2.1)	(0.64)	(2.7)
Xe-131m	----	0.044	0.026	0.071
Xe-133m	----	0.098	0.056	0.15
Xe-133	0.040	0.040(0.0075)(d)	0.029(0.026)(d)	0.069(0.034)(d)
Xe-135m	----	0.089	0.41	0.50
Xe-135	0.23	0.25	0.24	0.49
Xe-137	----	1.6	0.17	1.8
Xe-138	----	0.38	1.36	1.7
(Cs-138)	(----)	(1.0)	(2.1)	(3.1)

(a) Including conversion electrons.

(b) Including bremsstrahlung

(c) Values reported by Dr. Snyder in Reference 10.

(d) Values reported by Dr. Snyder in Reference 15.

TABLE II (Cont'd)

COMPARISON OF EXTERNAL DOSE RATES TO SELECTED TISSUES FROM A SEMI-INFINITE CLOUD OF NOBLE GASES

(mrem/hr per $\mu Ci/m^3$)

B. TESTES

	Schaeffer(9)	This Paper		
Nuclide	Beta	Beta(a)	Gamma(b)	Total
Ar-39	----	0	4.3×10^{-4}	4.3×10^{-4}
Ar-41	0	2×10^{-4}	1.1	1.1
Kr-83m	----	0	7.2×10^{-4}	7.2×10^{-4}
Kr-85m	0	0	0.13	0.13
Kr-85	0	0	$2.2\times10^{-3}(1.8\times10^{-3})$(c)	$2.2\times10^{-3}(1.8\times10^{-3})$(c)
Kr-87	0	0.047	1.3	1.3
Kr-88	----	2×10^{-3}	1.5	1.5
(Rb-88)	(----)	(0.37)	(0.56)	(0.93)
Xe-131m	----	0	0.023	0.023
Xe-133m	----	0	0.051	0.051
Xe-133	0	0	0.025(0.020)(d)	0.025(0.020)(d)
Xe-135m	----	0	0.35	0.35
Xe-135	0	0	0.21	0.21
Xe-137	----	0.10	0.12	0.22
Xe-138	----	9×10^{-4}	1.2	1.2
(Cs-138)	(----)	(0.012)	(1.8)	(1.8)

(a) Including conversion electrons

(b) Including bremsstrahlung

(c) Values reported by Dr. Snyder in Reference 10.

(d) Values reported by Dr. Snyder in Reference 15.

TABLE II (Cont'd)

COMPARISON OF EXTERNAL DOSE RATES TO SELECTED TISSUES FROM A SEMI-INFINITE CLOUD OF NOBLE GASES

(mrem/hr per μCi/m^3)

C. TOTAL BODY

Nuclide	Schaeffer(9)	Russell and Galpin(8) (a)	Snyder(10,15)	This Paper
Ar-39	-----	-----	-----	3.3×10^{-4}
Ar-41	1.16	-----	-----	1.1
Kr-83m	-----	-----	-----	1.0×10^{-5}
Kr-85m	0.16	0.076	-----	0.13
Kr-85	3.3×10^{-3}	1.2×10^{-3}	1.6×10^{-3}	2.2×10^{-3}
Kr-87	0.90	0.90	-----	1.3
Kr-88	-----	0.88	-----	1.5
(Rb-88)	(-----)	(-----)	(-----)	(0.56)
Kr-89	-----	4.2	-----	-----
Xe-131m	-----	6.8×10^{-3}	-----	0.016
Xe-133m	-----	0.041	-----	0.041
Xe-133	0.074	0.030	0.020	0.025
Xe-135m	-----	0.24	-----	0.35
Xe-135	0.24	0.14	-----	0.21
Xe-137	-----	0.090	-----	0.12
Xe-138	-----	1.7	-----	1.2
(Cs-138)	(-----)	(-----)	(-----)	(1.8)

(a) 1. The authors used the reciprocity theorem(5) to calculate total body dose.

2. The values presented here do not include the contribution from beta radiation as did those presented in the original paper.

TABLE III

COMPARISON OF INTERNAL DOSE RATES FROM INHALED NOBLE GASES

(units of 10^{-3} mrem/hr per μCi/m^3)

	Russell and Galpin(8)		Whitton (12) (b)				This Paper
Nuclide	Lungs(a)	Total Body(b)	Lungs(c)	Adipose Tissue	Remaining Tissue(b)	Testes(c)	Lungs(d)
Ar-39	-----	----	----	----	----	----	1.6
Ar-41	-----	----	----	----	----	----	6.7
Kr-83m	-----	----	----	----	----	----	0.43
Kr-85m	3.0	0.14	----	----	----	----	2.4
Kr-85	3.0	0.11	1.6(2.1)(e)	0.24(0.13)(e)	0.030(0.02-0.09)(e)	0.030(0.042)(e)	1.9
Kr-87	17	0.80	----	----	----	----	13
Kr-88	5.2	0.52	----	----	----	----	6.7
Kr-89	20	1.6	----	----	----	----	---
Xe-131m	1.8	0.21	----	----	----	----	1.3
Xe-133m	2.2	0.27	----	----	----	----	1.7
Xe-133	1.7	0.20	1.0(1.1)(f)	0.53(0.48)(f)	0.095(0.062-0.075)(f)	0.059(0.063)(f)	1.2
Xe-135m	4.0	0.33	----	----	----	----	1.9
Xe-135	1.5	0.54	----	----	----	----	3.3
Xe-137	18	2.1	----	----	----	----	15
Xe-138	13	2.4	----	----	----	----	6.6

(a) Based on a lung volume of 5.6 liters

(b) Used an average value of Ostwald coefficient for entire body

(c) Lung volume not stated

(d) Based on a lung volume of 4 liters

(e) Values of Dr. Snyder from Reference 10

(f) Values of Dr. Snyder from Reference 15

TABLE IV

COMPARISON OF RADIATION DOSES TO VARIOUS TISSUES FROM A SEMI-INFINITE CLOUD OF Kr-85

(mrem/hr per μCi/m^3)

Pathway	Snyder (10)	Russell and Galpin (8)	Whitton (12)	Schaeffer (9)	Kirk (3)	Hendrickson (1)	This Paper
External - Skin							
Beta	0.21(a)	1.6×10^{-3}(c)	0.19	0.17(d)	0.24(e)	0.11(d)	0.16(d)
Gamma + Bremsstrahlung	2.1×10^{-3}(b)	----	----	----	1.9×10^{-3}	2.7×10^{-3}	2.8×10^{-3}
Total	----	----	0.19	0.17	0.24	0.11	0.16
External - Total Body	1.6×10^{-3}	1.2×10^{-3}(f)	2.3×10^{-3}	3.3×10^{-3}	1.5×10^{-3}	2.7×10^{-3}	2.2×10^{-3}
- Gonads	1.8×10^{-3}	1.2×10^{-3}(f)	2.3×10^{-3}	3.3×10^{-3}	1.5×10^{-3}	2.7×10^{-3}	2.2×10^{-3}
Inhalation - Lungs	2.1×10^{-3}(g)	3.0×10^{-3}(h)	1.6×10^{-3}(i)	5.8×10^{-3} (k)	----	1.9×10^{-3}(j)	1.9×10^{-3}(g)
- Total Body	8.4×10^{-5}	1.1×10^{-4}	----	----	8.7×10^{-5}	----	----
- Adipose Tissue	1.3×10^{-4}	----	2.4×10^{-4}	----	----	----	----
- Remaining Tissue	$(2-9)\times10^{-5}$	----	3×10^{-5}	----	----	----	----
- Gonads	4.2×10^{-5}	----	3×10^{-5}	----	----	----	----

(a) Average dose to skin layer 5×10^{-3} cm thick, maximum is ∼10% higher.

(b) Average dose to a layer 0.2 cm thick.

(c) Assumed skin depth of 0.1 cm

(d) Assumed skin depth of 0.007 cm

(e) Assumed skin depth of 0.0 cm

(f) Based upon reciprocity theorem

(g) Assumed lung volume of 4 liters

(h) Assumed lung volume of 5.6 liters

(i) Lung volume not specified

(j) Assumed lung volume of 3.5 liters

(k) Estimated from Dr. Schaeffer's calculated value of $(MPC)_a$ for lung and probably includes the contribution of external radiation.

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