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Radiation Hazards

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Route List

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DATE 1-14-46

SUBJECT In Re: Uranium Tolerance

TO Health Physics Surveyors etc.

FROM K. Z. Morgan

COPY No [redacted]

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1/14/46

To: Health Physics surveyors and other interested persons

From: K. Z. Morgan

In Re: Uranium Tolerance

A. Tolerance of Uranium Depleted of U^{238}

Ordinary uranium consists of three isotopes of composition:

~~U^{238} = 99.28+%~~
U^{235} = .71+%
U^{234} = .006
Perhaps traces of U^{233}, U^{236}, U^{237}, U^{239} etc.

In the magnetic separation of U^{238} if there is a small depletion of U^{234}, we might expect to end up with:

U^{238} 5.00%
U^{235} 94.40%
U^{234} .60%
100.00%

One curie of this mixture of isotopes would be distributed as follows:

Isotope	Activity	Mass	Half Life	Energy (Mev)
U^{238}	.046%	1370 gm	4.51×10^9 y	4.15
U^{235}	5.61	25900	7.07×10^8 y	4.52
U^{234}	94.4	165	2.69×10^5 y	4.71
	100.0%	27435 gm		weighted average 4.70

I have shown in CH-2801 that if a radioactive substance is uniformly concentrated in a body organ, the tolerance may be expressed as,

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$$P = 4.94^{*} \frac{RfB\rho 10^5}{TE\left(t + \frac{T}{.693}e^{-.693\, t/T} - \frac{T}{.693}\right)} \mu\text{curies} \quad (1)$$

fixed in the body organ per second which will give a radiation exposure R (roentgens) in time t (sec).

In this equation,

- P = μcuries/sec fixed in the body organ
- R = roentgens exposure to the body organ in time t sec
- f = fraction the organ is of the total body
- B = total body weight in pounds
- fB = weight of body organ in pounds
- ρ = density of body organ in gm/cm^3
- T = effective half-life of radioactive element in seconds

$$T = \frac{TeTr}{Te+Tr}$$

- Te = body elimination half-life
- Tr = radioactive half-life
- t = time of exposure

Assumptions

1. That the lungs are among the body organs receiving the greatest radiation exposure from inhaled uranium.
2. The medical groups agreed in June 1945 that a pair of lungs has an average weight of ~1000 gms (fB = 2.2 lbs.).
3. Density of lung tissue, ρ = 1.
4. Of the inhaled uranium it is assumed that 25% is retained by the lungs (f_4 = 0.25), 25% is exhaled and 50% is deposited in the nose and large respiratory passages. The biological half-life is assumed to be two months (Te = 60 days or T → 60 days).
5. Tolerance radiation level of an α emitter when fixed in the body is assumed to be 0.01 roentgens equivalent physical/day.
6. The weighted average energy is 4.7 Mev.
7. The average breathing rate of a man is taken as 200 cc/sec. (The average when standing is 130 cc/sec. The average when walking at a rate of 2 mi/hr is 230 cc/sec).
8. A curie is defined as 3.7 x 10^{10} dis/sec and the half-life of radium is taken as 1590 years.

*The constant is written here as 4.94 instead of 4.38 because there are ~36 ev/ip in the case of α particles (Proc. Cam. Phil. Soc., March 1944).

~~This document contains information affecting the national defense of the United States within the meaning of the [illegible] 31 and 32. Its transmission [illegible] its contents [illegible] to an unauthorized person [illegible] by law.~~

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Differentiating equation (1), placing dR/dt ≡ roentgens/day and T_d ≡ half-life in days, we have:

$$P = \frac{dR/dt\ (6.62\times10^{-5}) fB\rho}{S' T_d\ (1-e^{-.693t/T_d})}$$ μcuries/sec is tolerance rate at which radioactive material enters the body. (2)

Substituting the above values in equation (2) we have,

$$P_1 = \frac{P}{200(.25)} = \frac{P}{50}$$ μcuries/cc is tolerance concentration in the air. (3)

$$Q = 2.74 \times 10^4\ P_1$$ μgms/cc is tolerance concentration in the air of uranium depleted of U^{238} (4)

Table I shows the time required to obtain the tolerance concentration of this uranium mixture in the lungs.

Table 1*

Tolerance Values for Uranium Mixture
5% U^{238}, 94.4% U^{235} and 0.6% U^{234}

Time in which the tolerance amount of uranium is built up in the lung	Tolerance rate of concentration, P in μc/sec in the lungs	Tolerance concentration in air in μc/cc (P_1)	Tolerance concentration in air in μgm/cc (Q)
1 day	4.31×10^{-7}	8.62×10^{-9}	2.36×10^{-4}
10 days	4.70×10^{-8}	9.40×10^{-10}	2.58×10^{-5}
100 days	7.56×10^{-9}	1.512×10^{-10}	4.14×10^{-6}
365 days	5.25×10^{-9}	1.050×10^{-10}	2.88×10^{-6}
∞ days	5.17×10^{-9}	1.034×10^{-10}	2.84×10^{-6}

*Multiply tolerance values in table by 4.2 to get tolerance values for a 40 hour week.

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B. Tolerance of Natural Uranium

In the case of natural uranium, one curie would be distributed as follows:

Isotope	Activity (%)	Mass (gm)	Energy (Mev)	% Present by weight
U^{238}	48.9	1,460,000	4.15	99.28±
U^{235}	2.26	10,440	4.52	.71±
U^{234}	48.9	85.6	4.71	.00583
		1,470,525gm	4.43 wt.energy	99.996

The assumptions are the same as before, except the weighted average energy is 4.43 Mev. Equations (2) and (3) are the same as before, but equation (4) becomes,

$$Q' = 1.47 \times 10^{6} P_1 \ \mu\text{gms of natural U/cc is tolerance concentration in air of natural uranium.} \quad (4)'$$

Table II shows the time required to obtain the tolerance concentration of natural uranium (freed of all other element impurities) in the lungs.

Table II*

Tolerance values for natural uranium
(99.28% U^{238}, 0.71% U^{235} and .0058% U^{234})

Time in which the tolerance amount of uranium is built up in the lung	Tolerance rate of concentration, P in μc/sec in the lungs	Tolerance concentration in air in μc/cc (P_1)	Tolerance concentration in air in μgm/cc (Q')
1 day	4.58×10^{-7}	9.16×10^{-9}	1.35×10^{-2}
10 days	5.00×10^{-8}	1.00×10^{-9}	1.47×10^{-3}
100 days	8.04×10^{-9}	1.608×10^{-10}	2.36×10^{-4}
365 days	5.58×10^{-9}	1.116×10^{-10}	1.64×10^{-4}
∞ days	5.50×10^{-9}	1.100×10^{-10}	1.62×10^{-4}

*Multiply tolerance values by 4.2 to get tolerance values for a 40 hour week.

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C. Comparison of Tolerance of Uranium with that of Plutonium

In this case the assumptions are the same as before except the energy is 5.15 Mev. The biological half-life seems in general to be about the same. Actually plutonium consists of a mixture of isotopes but for simplicity sake we will take the case of Pu^{239}. Equation (4) becomes,

$$Q'' = 16\ P_1\ \mu\text{gms of Pu/cc is tolerance concentration in the air} \quad (4)^{11}$$

Table III shows the time required to obtain the tolerance concentration of plutonium in the lungs.

Table III*

Tolerance Values for Plutonium (Pu^{239})

Time in which the tolerance amount of Plutonium is built up in the lung	Tolerance rate of concentration, P in μc/sec in the lungs	Tolerance concentration in air in μc/cc (P_1)	Tolerance concentration in air in μgm/cc (Q'')
1 day	3.93×10^{-7}	7.86×10^{-9}	1.26×10^{-7}
10 days	4.29×10^{-8}	8.58×10^{-10}	1.37×10^{-8}
100 days	6.90×10^{-9}	1.380×10^{-10}	2.21×10^{-9}
365 days	4.79×10^{-9}	9.58×10^{-11}	1.53×10^{-9}
∞ days	4.72×10^{-9}	9.44×10^{-11}	1.51×10^{-9}

D. Discussion

The values given in the above tables indicate considerable variation in radiation tolerance for these alpha emitters when expressed in micrograms per cubic centimeter of air concentration (2.88×10^{-6} for uranium depleted in U^{238}, 1.64×10^{-4} for natural uranium and 1.53×10^{-9} for Pu) but very little variation in microcuries per cubic centimeter (1.05×10^{-10} for uranium depleted in U^{238}, 1.12×10^{-10} for natural uranium, and 0.958×10^{-10} for Pu).

This is probably an oversimplification of the problems involved but for general radiation tolerance values the results are as accurate as the constants used in the solution. Perhaps the most questionable assumption is that all these alpha emitters have an effective half-life of two months in the lungs. This half-life of uranium or plutonium depends on the chemical form, the size of the dust particles, the humidity, etc. Many biological

*Multiply tolerance values by 4.2 to get tolerance values for a 40-hour week inhalation experiments have been performed and some of the data is not as consistent as one would like. Dr. J. G. Hamilton has shown that the half-life of plutonium in the lungs is about two months and this value seems to be a good average for uranium dust also. However, one can show that the tolerance concentration decreases only by a factor of ten due to a change in effective half-life from two weeks to six months and since the effective half-lives of most of the compounds of plutonium and uranium do not range beyond these limits, it is believed that the two months effective half-life is satisfactory as a general value.

E. Suggested Maximum Permissible Exposure

The value used as the maximum permissible concentration at Clinton Laboratories for plutonium for the past two and a half years is 3.1×10^{-11} μc/cc of air. This value contains a factor of safety of 3 for continuous exposure and a factor of safety of 13 for a 40 hr/week exposure as determined from Table III for one year exposure. It is suggested in this paper that this value of 3.1×10^{-11} μc/cc be used also for uranium*.

F. Results of biological experiments with animals exposed to air containing various compounds and concentrations of uranium.

These experiments were performed at Rochester under the direction of A. H. Dowdy. Even a cursory review of these reports for the year 1945 reveals a wealth of information. A few examples are given below. Perhaps in most animal experiments the damage was produced by chemical toxicity rather than by radiation. However, it is of some interest to compare these exposures with the tolerance levels for natural uranium in Table II in which we found, 1.47×10^{-3} μgms U/cc as the 10 day tolerance and 1.64×10^{-4} μgms U/cc as the one year tolerance.

1. Above tolerance exposures

Inhalation of UF_4 dust in a concentration of 4×10^{-2} μgm U/cc of air (27 times 10 day tolerance) by various species of animals was lethal after a few weeks exposure. It was perceptibly toxic at a concentration of 2×10^{-2} μgm U/cc of air (14 times 10 day tolerance).

Dogs exposed to inhalation of UO_2 in a concentration of 10^{-2} μgm U/cc of air (25 times 65 day tolerance) for 65 days showed a slight weight loss. No effect was noticed when the dogs were exposed 65 days to 10^{-3} μgm U/cc of air (2½ times 65 day tolerance).

2. Near Tolerance Exposures

Inhalation of UO_2F_2 in a concentration of 7×10^{-4} μgm U/cc of air (exactly 30 day tolerance) for 30 days caused the death of 3 out of 33 rabbits and 3 out of 40 mice. The evidence is not very conclusive but this

*This tolerance value was suggested for polonium in a letter by K. Z. Morgan to M. C. Leverett on 10/6/45 and for Thorium in a letter by K. Z. Morgan to R. S. Stone on 11/29/45. If this value can be accepted for all these alpha emitters in the laboratories, it will simplify the survey efforts and procedures for measuring the air contamination. This value is believed to be on the conservative side for all these elements and it is considered within the limits of attainment without hampering operations.

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and other experiments indicate that UO_2F_2 and UF_6 are among the more hazardous uranium compounds. Even though the damage is most certainly from chemical toxicity rather than from radiation, the radiation measurements with precipitrons and filter dust collectors present one of the most convenient means of monitoring this contaminated air. If the maximum permissible level in the laboratory is maintained at 3.1×10^{-11} μc/cc of air (or 4.6×10^{-5} μgm of natural U/cc of air), this would give a factor of safety of only ~15 above the level in this animal experiment with UO_2F_2 in which deaths resulted.

The table below indicates the relative hazards presented by UF_6 and UO_2F_2:

Species	% Mortality due to 1.24×10^{-2} μgm U/cc air in form of UF_6 (~18 times 30 day tolerance)	% Mortality due to $.94 \times 10^{-2}$ μgms U/cc air in form of UO_2F_2 (~13 times 30 day tolerance)
Mouse	100	100
Rabbit	100	83
Rat	75	0
Guinea pig	45	55
Dog	40	100

Exposure of animals to 5×10^{-4} μgm U/cc (~0.7 times 30 day tolerance) of air in the form of UF_4 dust produced no deaths in 30 days but retarded the weight of rabbits, rats and guinea pigs.

In an inhalation experiment with U-nitrate in the concentration of 1.5×10^{-4} μgm U/cc (1/6 of 20 day tolerance) dogs showed slight to moderate renal damage after 500 hours exposure in 114 days.

Rats exposed to UF_6 in the concentration of 0.5×10^{-4} μgm U/cc of air for 57 days (~0.1 of 57 day tolerance) showed slight decrease in growth rate. Animals exposed to this same concentration of the nitrate showed no abnormal effects.

Respiratory Devices Tested for UF_6 and UO_2F_2 by the Rochester Division under the direction of A. H. Dowdy. The table below gives some of the results:

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Respirator	Acceptability
Willson Universal Canister with filter element	Acceptable for six hours
Mine Safety Appliance Co. All-Service Gas Mask BM-1434	Acceptable for six hours
Mine Safety Appliance Co. GMC-1 Canister	Acceptable for only 15 minutes
"Aerosol" filter with Chemical Warfare Assault Mask	Acceptable for U-component, not for HF
"Comfo" All-dust Respirator with BM-2133 Filter	Acceptable for U-component, not for F

Conclusions

One may conclude that "curie for curie" uranium presents radiation hazards of the same order of magnitude as plutonium. On a weight basis natural uranium is only about 10^{-5} times as bad as plutonium and the U^{238} depleted uranium is ~530 times as bad from the standpoint of radiation damage. The principle damage from plutonium is probably produced by the radiation. The chemical toxicity of uranium compounds (especially those containing fluorine) presents a much greater hazard than the radiation. Because of the extreme toxicity of UO_2F_2, UF_6, etc. the maximum permissible value for natural uranium in the air should not exceed 3.1×10^{-11} μcuries/cc of air (= 4.6×10^{-5} μgm U/cc of air). This gives an added factor of safety of ~3.5 above the one year radiation tolerance value for natural uranium calculated in Table II as $1.6? \times 10^{-4}$ μgm U/cc of air*. In case one is working with uranium depleted in U^{238} as shown in Table I, the tolerance value may still remain at ~1 x 10^{-11} μcuries/cc of air. This, however, would correspond to only 8.? x 10^{-7} μgms U/cc of air and would be far below the levels of chemical toxicity for any of the uranium compounds studied. There would be a factor of safety of only about 3.4 from the standpoint of radiation. In other words, the hazard for natural uranium is determined definitely by its chemical toxicity but the hazard for uranium depleted of U^{238} is determined by the radiation damage it produces in the

*The tolerance value for natural uranium used on some of the other sites is 150 μgm/cu meter of air (or 1.5 x 10^{-4} μgm U/cc). This checks with the value calculated in Table II.

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body. If one exceeds this curie tolerance of 3.1×10^{-11} $\mu c/cc$ of air for natural uranium, the chemical damage is observed in a few days or weeks and there is not much margin of safety between the amounts required to cause slight damage and death. However, on a gram basis this represents an enormous amount of natural uranium in the lungs before damage results. In the case of plutonium or uranium depleted in U^{238}, the damage is from radiation and in general the harmful effects are observed only after months or years of overexposure.

The foregoing discussion on inhalation of uranium and plutonium has considered only the radiation damage to the lungs. This is not the complete picture because, as we have assumed, 50% of the uranium or plutonium lodges in the upper respiratory passages and this is later swallowed. Some of this is absorbed from the gastro-intestinal tract into the blood stream and carried to various parts of the body where it produces damage to many body organs. For example, some of the plutonium is carried to the bone where it produces damage to the bone marrow or uranium is carried to the kidneys to cause them damage. Also, some of the materials are absorbed directly into the blood from the lungs. However, calculations on the basis of damage to other body organs seem to indicate tolerance values of the same order of magnitude (or not as bad) as given in Tables I, II and III and so will be omitted here in order not to prolong this discussion.

Karl Z. Morgan

Karl Z. Morgan

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