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MASTER

UPDATING THE TRITIUM QUALITY FACTOR - THE ARGUMENT FOR CONSERVATISM

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ABSTRACT

Estimated doses resulting from tritium releases to the environment are linearly dependent upon the quality factor (Q) chosen for tritium beta radiation. In 1969 the International Commission on Radiological Protection (ICRP) recommended using 1 as the Q for all low energy beta radiation. Considerable improvements have been made in evaluating exposures to tritium at very low dose rates and in refining physiological and biological endpoints since the 1969 ICRP recommendations.

This study summarizes recent experiments to determine the relative biological effectiveness of tritium. Based upon our study of published data related to quality factor, its importance in the calculation of dose, and the currently accepted conservative philosophy in radiation protection, it is concluded that a value of 2 would seem to be more defensible for environmental assessments and that a reevaluation of the tritium quality factor by the ICRP is needed.

INTRODUCTION

The concept of dose equivalent (DE) was introduced by the ICRP (1) and the International Commission on Radiation Units and Measurements (ICRU) (2) in order to define a uniform scale of biological damage resulting from exposure to different types and energies of radiation. The unit of dose equivalent is the rem† which is calculated using the following expression:

$$DE = D \times Q \times (RDF)_1 \times (RDF)_2 \dots$$

where

DE = dose equivalent in rem,†

D = absorbed dose in rad,

Q = quality factor (dimensionless), and

$(RDF)_1, (RDF)_2 \dots$ = radionuclide distribution factors (dimensionless).

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† In the International System of Units (SI), the conventional unit rem has been changed to the sievert where one rem is equivalent to 10^{-2} sievert; the unit rad has been changed to gray where one rad is equivalent to 10^{-2} gray. Conventional units only are used in this text.

The term quality factor (Q) has been accepted for use in the calculation of dose and is related to linear energy transfer (LET) on a common scale for all ionizing radiation. A radionuclide distribution factor (RDF) is applied in the calculation of DE to express the modification of biological effectiveness due to nonuniform distribution of internally deposited isotopes. Since tritium is essentially uniformly deposited in body tissue due to its association with body water, its distribution factor is assumed to be 1. Some disagreement still exists, however, as to the best value of quality factor for tritium betas as well as for other radionuclides emitting low LET radiations. This value affects the dose equivalent linearly and therefore could significantly alter the calculated dose equivalent.

In ICRP Publication 9 (1), the Commission recommended a value of 1.7 be used as the quality factor for β^- , β^+ , and e^- radiation with maximum energies ≤ 0.03 MeV. In an amendment to ICRP Publication 9, the Commission reduced the quality factor to 1 for all β^- , β^+ , e^- , γ , and x rays. Following a review of the biological and physical evidence related to relative effectiveness of low energy beta radiation, it was concluded that a value of unity is appropriate within the degree of precision required for the purposes of radiological protection. The decision to reduce the quality factor from 1.7 to 1 was also based upon insufficient scientific evidence to support the higher value as well as the variability in physiological endpoints and reference radiations reported in the literature.

The ICRP decision in 1969 followed a review of the experimental literature by Vennart (3). He concluded that in view of experimental evidence on the quality factor of β^- particles from tritiated water, a value different from unity could hardly be justified. Vennart also based his conclusion on the fact that since the ICRP in Publication 8 (4) recommended expressing risk per unit dose only in terms of orders of magnitude, those factors included in the calculation of dose should be rounded to whole numbers. A later review of the literature by Rohrer (5) stated that most of the information on tritium exposure did not justify the value of 1.7, although he pointed out that this was an area needing further study and evaluation. This conclusion regarding further study was also reached in a recent review by Carsten (6).

The National Council on Radiation Protection and Measurements (NCRP) recently reviewed the

tritium quality factor issue in a study of the significance of compounds incorporated into genetic material (7). It concluded that there is sufficient data to assign an RBE* of 1 when reference radiation is in the order of 60 to 80 kVp x rays. This reference radiation is similar to that of tritium betas in terms of energy deposition per unit track length. Some ambiguity exists in their argument, however, since the reference radiation utilized for determining RBE is ordinarily 220 to 250 kVp x rays or ^{60}Co γ rays.

JUSTIFICATION FOR A REEVALUATION OF Q FOR TRITIUM

Variability still exists in experimental endpoints and reference radiation reported in the literature which are used to evaluate the relative biological effectiveness for β^- radiation. Cumming et al. (8) demonstrated that radiation dose from a single injection of tritiated water can be greatly influenced by minor changes in experimental conditions. Since the review by Vennart and the decision by the ICRP to reduce the quality factor from 1.7 to 1, improvements have been made in the techniques to evaluate incorporated tritium at very low dose rates (on the order of a rad per hour or less) and in choosing physiological and biochemical endpoints leading to a significant increase in the sensitivity of the tests.

The potential genetic consequences of chronic low-level exposure to tritium may be of particular importance for environmental releases from the nuclear industry. Carsten and Commerford (9) studied mice exposed to 3 $\mu\text{Ci}/\text{ml}$ drinking water (approximately equal to 1 rad/day). Second generation females were sacrificed in late pregnancy to determine mutation frequency. Analysis of their results showed a significant reduction in the number of viable embryos resulting from mating between animals exposed to tritium, but showed no effect on breeding efficiency. The authors noted that no direct parallelism exists between man and the data deduced from mice; however, the significant effect seen in the study suggests that further investigation at lower tritium concentrations is necessary.

Another study by Dobson et al. (10) also suggests that genetic effects produced from internally deposited tritium may warrant renewed consideration. In their experiment, female germ cells in both mouse and monkey were shown to be extremely sensitive to destruction by low-level, chronic tritium exposure. Their results appear to be inconsistent with previous reports which conclude that oocytes in both monkey and man are relatively radioresistant, having lethal doses for 50% of the population (LD_{50}) for x rays up to 5,000 rads. Dobson and his colleagues suggest that the greater sensitivity they observed resulted from chronic exposure to tritium in body water that acted on cells as they passed through highly vulnerable periods of

early development. The exposure conditions established in their experiments are likely to be more representative of low-level environmental exposures from tritium such as releases by nuclear facilities than those described by most investigators; therefore, the results of their study must be given serious consideration in the design of further bioeffects research involving tritium and in current radiological assessment methodologies.

These data suggest that exposure to tritium under chronic, low-level conditions may result in greater biological damage than previously anticipated. Because of this possibility, and because significant improvements have been made in evaluating exposure from internally deposited tritium since the ICRP recommendation, an analysis of literature reporting RBE values for tritium since 1969 is needed.

EXPERIMENTAL DATA ON RBE FOR TRITIUM SINCE THE 1969 ICRP RECOMMENDATION

Table 1 summarizes experiments reported since the review published by Vennart (3). The table includes only animal and cell studies in which values for RBE are calculated. Additional experiments have been reported using plants or in which radiosensitivity of organisms is studied but where no RBE is calculated. These experiments have been omitted from the table.

Lambert (11) irradiated mouse testes using internally distributed tritium from injected tritiated thymidine ($^3\text{HTdR}$) and tritiated water (^3HOH). The criteria for damage was the inability of spermatogonia to divide twice and produce resting spermatocytes. The reference radiation was 200 kVp x rays delivered at an exponentially decreasing dose rate over a 72-hour period to give a total dose of 30 rads. The RBE of tritium as tritiated thymidine or tritiated water relative to 200 kVp x rays was in the range of 1.3-2.4. Lambert pointed out that a direct extrapolation of his results to man is not possible; however, this study is particularly important because of its use of a very low dose rate and low total dose, yet high sensitivity of the endpoint used as damage criteria.

Richold et al. (12) reported an RBE of 0.94 ± 0.11 following irradiation of aqueous solutions of ribonuclease with ^3HOH under anoxic and aerobic conditions. The reference radiation was $^{60}\text{Co}(\gamma)$. Of the studies listed in Table 1, this is the only case in which an RBE value of less than 1.0 is reported. However, a relatively high dose rate of 100 rads/hr was used. It has been shown that exposures to tritium under chronic conditions at low dose rates (comparable to "routine release" exposures) potentially produce more harmful effects than acute, high-level exposures per unit dose (13), and this "dose rate effect" may have affected the outcome of the studies by Richold et al.

In an animal study, Moskalev et al. (14) exposed rats to tritium oxide injected intraperitoneally. The calculated dose rate from tritium varied from 27.5 to 1.38 rads/hr and the reference radiation used was ^{137}Cs γ rays. This experiment used several indices for RBE determination includ-

*Relative biological effectiveness (RBE) is a term expressing the relative effectiveness of radiations that have different linear energy transfer (LET) values in producing a given biological effect. This quality is limited to use in radiobiology only but is similar to the quality factor, in purpose, used to calculate the dose equivalent.

ing change in weight of the spleen, thymus, adrenals, and liver, composition of peripheral blood, as well as numerous biochemical indices. The total cumulative dose was 340 rads. Calculated values for RBE ranged from 1.4 to 2.2 depending upon the index evaluated. Moskalev et al. (14) reported another study in which rats were again exposed to $^3\text{H}\text{OH}$ and ^{137}Cs γ rays. The criteria for damage were survival rate, peripheral blood response and thymic and splenic weight response. Although higher dose rates were used in this study than the previous, it was concluded that the RBE for tritium oxide was in the range of 1.5-1.9.

Bedford et al. (16) exposed two mammalian cell lines to $^3\text{H}\text{OH}$ and tritiated thymidine and used ^{60}Co γ rays as a reference radiation. The criterion for damage was cell survival. To prevent cell division, during exposures, irradiations were carried out with cells held in the frozen state or, in one case, at 5°C . For cells irradiated at 5°C , the efficiency of cell killing by beta particles from tritiated water or tritiated thymidine was not appreciably different, but both were more efficient than gamma radiation. For a dose rate of 20 rad/hr, the relative biological effectiveness of tritium beta particles compared to ^{60}Co γ rays was estimated to be between 1.7-1.9.

Weanling mice were exposed to $^3\text{H}\text{OH}$ by Dobson et al. (13) at low dose rates (approximately 5.2 rads/day). Surviving primary oocytes were counted microscopically in ovaries and compared to controls. Other weanlings were exposed to ^{60}Co γ rays at 5.9 rads/day. It was concluded that since the exposure was protracted, more effective microdistribution of tritium atoms may have occurred resulting in an RBE of 1.1-1.7. The investigators pointed out the significance of their results showing that $^3\text{H}\text{OH}$ becomes more damaging compared to gamma rays as the low-level exposure is protracted over longer periods of time. This finding is particularly noteworthy in considering exposures from tritium found in the environment. In another study by Dobson and Kwan (17), developing mice were used and survival of primary oocytes was observed. The dose rate was reduced to as low as 0.44 rads/day for both $^3\text{H}\text{OH}$ and the reference ^{60}Co γ rays. At effective gamma-ray doses of about 40 rads, the RBE was calculated to be 1.6. However, with lower dose rates giving effective total doses of only a few rads, the RBE for tritium rises to approximately 3. These studies by Dobson et al. (13) and Dobson and Kwan (17) are very important since they used extremely sensitive criteria for damage and observed noticeable effects at exceptionally low dose rates and total dose.

In a recent study by Russell et al. (18) transmitted gene mutations induced by tritium were observed in mice. Male mice were injected with tritiated water to give a dose rate of 0.8 rad/min. A specific-locus-mutation test was used to determine biological damage. This procedure is considerably more sensitive than earlier methods reported for determining RBE. Russell et al. point out that various uncertainties are involved in arriving at a precise value for RBE; however, they recommend that for the purpose of risk estimation, it seems more prudent to use the RBE value of 2 as the best point estimate computed from their data.

IMPACT ON ENVIRONMENTAL DOSE FROM AN INCREASE IN THE TRITIUM QUALITY FACTOR

Increasing the quality factor for tritium to a value greater than 1 would linearly increase the calculated dose from exposure to tritium in the environment. Typically, however, increasing the dose from tritium does not necessarily imply that the total dose in the vicinity of a nuclear fuel cycle facility would increase proportionately since tritium is only one of the radionuclides that ordinarily contributes significantly to total dose.

As an example of the effect of increasing the quality factor, Table 2 lists the predicted dose to a hypothetical individual living near a fuel reprocessing plant handling light water reactor fuel (19). A reprocessing facility is selected for this example since it represents a case in which tritium is a major contributor to dose. Assuming a quality factor for tritium of 1, the dose from tritium is 7.4 mrem and that for all other radionuclides is 6.7 mrem giving a total dose of 14 mrem. In this case, tritium contributes 52% of the dose. When the quality factor for tritium is increased to 2, the dose from tritium becomes 15 mrem and the total dose increases to 22 mrem.* The contribution from tritium is 66%. Therefore, increasing the quality factor for tritium from 1 to 2 increases the estimated total dose by 28%. In this example, doses remain acceptable when compared to guidelines published in 40 CFR 190 (20) even with the higher quality factor.

CONCLUSIONS

Tritium is a key radionuclide in the assessment of dose in the vicinity of nuclear fuel cycle facilities. A review of recent experimental data to determine the relative biological effectiveness of tritium beta particles indicates that under conditions of chronic, low-level exposure, RBE values between 0.94 and 3.0 are reported. Based on these data, we conclude that a Q greater than unity can certainly be justified and would be more in keeping with the accepted conservative approach to estimating dose. In view of this conclusion it seems reasonable to recommend use of a quality factor of two for tritium betas. Use of this value incorporates a degree of conservatism in the RBE and recognizes that under conditions of chronic, low-level exposure the RBE for tritium is likely greater than one.

It is noted that considerable conservatism already exists in models that estimate exposure from tritium released to the environment. It is also noted that increasing Q from one to two may not be justified considering the degree of precision that currently exists in these models. However, our argument is that conservatism should be maintained in present models and should only be removed with care as scientific data become available to justify it.

Existing data and biophysical considerations suggest that at low doses and low dose rates, a

*In this calculation it is assumed that the quality factor for other radionuclides emitting low LET radiations is 1 (see Conclusions).

substantial difference exists in the effect per rad of different radiations exhibiting low LET. If a quality factor for tritium betas of greater than one is accepted, it may be argued that a quality factor greater than one is needed for all radiations of comparable LET. Although this argument seems reasonable, additional research is required before this can be verified.

Adoption of a quality factor greater than unity in determining maximum permissible concentrations for tritium should also be reconsidered by the ICRP in view of scientific evidence since their 1969 decision to use 1.0. In addition, it would be helpful if the ICRP could provide better clarification for the reference radiation that should be used to determine relative biological effectiveness of low-LET radiations.

REFERENCES

1. International Commission on Radiological Protection, "Recommendation of the International Commission on Radiological Protection (Adopted September 17, 1965)," *ICRP Publication 9*, Pergamon Press, Oxford, 1966.
2. International Commission on Radiation Units and Measurements, "Radiation Quantities and Units," Report 10A, NBS Handbook 84, Washington, D. C., 1962.
3. Vennart, J., "The Quality Factor for Low-Energy β -Emitters," *Health Phys.*, 541-548, 1968.
4. International Commission on Radiological Protection, "The Evaluation of Risks from Radiation," *ICRP Publication 8*, Pergamon Press, Oxford, 1966.
5. Rohwer, P. S. and H. W. Wilcox, "Radiological Aspects of Environmental Tritium," *Nucl. Saf.* 17:216-222, 1976.
6. Carsten, A. L., "Tritium in the Environment," *Advances in Radiation Biology* (ed. J. T. Lett and H. Adler), 8:419-458, 1979.
7. National Council on Radiation Protection and Measurements, "Tritium and Other Radionuclide Labeled Organic Compounds Incorporated in Genetic Material," NCRP Report No. 63, 1979.
8. Cumming, R. B., G. A. Sega, and M. F. Walton, "Radiation Dosimetry in Experimental Animals Exposed to Tritiated Wastes Under Different Conditions," *Symp. on the Behaviour of Tritium in the Environment*, ed. by S. Freeman, 463-467, IAEA, Vienna, 1979.
9. Carsten, A. L. and S. L. Commerford, "Dominant Lethal Mutations in Mice Resulting from Chronic Tritiated Water Ingestion," *Radiat. Res.* 66:609-614, 1976.
10. Dobson, R. L., C. G. Kochler, J. S. Felton, C. Kwan, B. J. Wuebbles, and D. C. L. Jones, "Vulnerability of Female Germ Cells in Developing Mice and Monkeys to Tritium, Gamma Rays, and Polycyclic Aromatic Hydrocarbons," *Proc. Seventeenth Hanford Biology Symposium*, October 16-18, 1977, Richland, Washington.
11. Lambert, B. E., "Cytological Damage Produced in the Mouse Testes by Tritiated Thymidine, Tritiated Water, and X rays," *Health Phys.* 17:547-557, 1969.
12. Richold, P. H. C. V., J. A. B. Gibson, and M. Marshall, "The Relative Biological Effectiveness of Tritium in Aqueous Solutions of Ribonuclease," *AERE-R 6951*, 1971.
13. Dobson, R. L., J. H. Arrington, and T. C. Kwan, "Tritium Toxicity: Increased Relative Biological Effectiveness of $^3\text{H}_2\text{O}$ with Protraction of Exposure," *UCRL-76558*, March, 1975.
14. Moskalov, Y. I., V. F. Zhuravlev, V. S. Kalistratova, A. G. Istomina, J. K. Petrovich, and D. A. Kazbekova, "Evaluation of Relative Biological Effectiveness of Tritium Oxide," in *Biological Effects of Radiation from External and Internal Sources*, eds. Y. I. Moskalov and V. S. Kalistratova, *ISAE Report AEC-tr-7957*, 1972.
15. Moskalov, Y. I., V. F. Zhuravlev, and A. G. Istomina, "Relative Biological Effectiveness of Tritium," in *Tritium*, eds. A. A. Moghissi and M. W. Carter, Messenger Graphics, Phoenix, Arizona, 1973.
16. Bedford, J. S., J. B. Mitchell, H. G. Griggs, and M. A. Bender, "Cell Killing by Gamma Rays and Beta Particles from Tritiated Water and Incorporated Tritiated Thymidine," *Radiat. Res.* 63:531-593, 1975.
17. Dobson, R. L. and T. C. Kwan, "The RBE of Tritium Radiation Measured in Mouse Oocytes: Increase at Low Exposure Levels," *Radiat. Res.* 66:615-625, 1976.
18. Russell, W. L., R. B. Cumming, E. M. Kelly, and E. L. Phipps, "Introduction of Specific-Locus Mutations in the Mouse by Tritiated Water," *Symp. on Behaviour of Tritium in the Environment*, ed. S. Freeman, 489-495, IAEA, Vienna, 1979.
19. Finney, B. C., R. E. Blanco, R. C. Dahlman, G. S. Hill, F. G. Kitts, 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 - Reprocessing Light Water Reactor Fuel," *ORNL/NUREG/TM-6*, 1977.
20. U. S. Environmental Protection Agency, "Environmental Radiation Protection Requirements for Normal Operation of Activities in the Uranium Fuel Cycle," 40 CFR 190, EPA 520/4-76-016, 1976.

Table 1. RBE^a values for tritium from selected animal and cell studies^b

Description of experiment	Reference radiation	Reported RBE	Reference
Irradiated mouse testes using internally distributed tritium from injected tritiated thymidine and tritiated water. Used inability of spermatogonia to divide twice as damage criteria.	200 kVp x rays	1.3 - 2.4	Lambert (11)
Irradiated aqueous solutions of ribonuclease using tritiated water. Measured residual enzymatic activity and observed survival.	⁶⁰ Co γ rays	0.94 ± 0.11	Richold et al. (12)
Exposed rats internally to tritiated water. Damage criteria were change in weight of spleen, thymus, adrenals, and liver, as well as biochemical indices.	¹³⁷ Cs γ rays	1.4 - 2.2	Moskalev et al. (14)
Exposed rats internally to tritiated water. Damage criteria were survival rates, peripheral blood response and thymic and splenic weight changes.	¹³⁷ Cs γ rays	1.4 - 1.9	Moskalev et al. (15)
Exposed two mammalian cell lines to tritiated water and tritiated thymidine. Measured cell survival.	⁶⁰ Co γ rays	1.7 - 1.9	Bedford et al. (16)
Exposed weanling mice to tritiated water and observed survival of primary oocytes.	⁶⁰ Co γ rays	1.1 - 1.7	Dobson et al. (13)
Exposed developing mice from conception to 14 days after birth. Observed primary oocyte survival.	⁶⁰ Co γ rays	1.6 - 3.0 (with varied rate)	Dobson and Kwan (17)
Exposed mice to tritiated water, observed gene mutations transmitted.	⁶⁰ Co γ rays	2.2	Russell et al. (18)

^aRelative biological effectiveness (RBE) is a factor expressing the relative effectiveness of radiations that have different linear energy transfer (LET) values, in producing a given biological effect. The unit is limited to use in radiobiology but is similar to the value of quality factor used in the calculation of dose.

^bStudies published since the literature review by Vennart (1968).

Table 2. Effects of increasing the tritium quality factor on the estimated dose to total body for a maximally exposed individual living near a nuclear fuel reprocessing plant^a

Tritium quality factor	Dose to maximally exposed individual (mrem)		
	Tritium	All other radionuclides	Total
1	7.4	6.7	14
2	15	6.7	22

^aTaken from data published by Finney et al. (19), base case, 1.5 miles from the point of release.