NRC Research and/or Technical Assistance Rept

PDR

ORNL/HASRD-151

INTERIM REPORT

Contract Program or Project Title:	Analysis of Proposed New IAEA Basis for Transportation Regulatory System
Subject of this Document:	Technical Analysis
Type of Document:	Special Report
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Date of Document:	June 30, 1982
NRC Individual and NRC Office or Division to Whom Inquiries Should be Addressed:	Dr. D. R. Hopkins Division of Risk Analysis

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Prepared for
U. S. Nuclear Regulatory Commission
Washington, D.C. 20555
Under Interagency Agreement DOE #40-550-75
NRC FIN No. B0810-2

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Oak Ridge, Tennessee 37830
operated by
Union Carbide Corporation
for the
U. S. Department of Energy

INTERIM REPORT

REVIEW OF "Q SYSTEM OF LIMITS FOR TYPE A PACKAGE CONTENTS" PROPOSED BY MACDONALD AND GOLDFINCH

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1. INTRODUCTION

The International Atomic Energy Agency (IAEA) is developing a revision to their 1973 Safety Series 6 "IAEA Regulations for the Safe Transport of Radioactive Materials" (IAEA, 1979). The intent is to publish the revision in 1984. The IAEA regulations are intended to apply to international shipments and may also serve as guidance for intranational shipping regulations.

E. P. Goldfinch and H. F. Macdonald (1981a, 1981b) and Goldfinch and Macdonald (1982) have developed and proposed a revision to the "A_1/A_2" system used in 1973. The Oak Ridge National Laboratory (ORNL) has recently contracted with the U. S. Nuclear Regulatory Commission (NRC) to review the new proposals critically.

The new proposals purport to have the following advantages over the current IAEA regulations:

1. They state the radiation protection criteria employed more clearly.
2. They incorporate the data and conclusions on metabolic pathways as given recently by the International Commission on Radiological Protection (ICRP, 1977, 1979, 1980, 1981).
3. They include dosimetric routes not previously considered.

4. They harmonize IAEA regulations with ICRP recommendations in Publications 26 and 30.
5. They introduce a "Q" System in place of the previous "A_1/A_2" system.
6. They are couched in S.I. units.

Understanding of these proposals requires understanding the definitions of the 1973 A Limits and new Q limits. A_1 limits are for packages containing special form material and A_2 for other forms of material, with special form material being material which is non-dispersible when subjected to certain specified tests (IAEA, 1979). These limits are to be replaced by new Q_1 and Q_2 limits. The Q_1 and Q_2 are (generally) taken as the least of new limits Q_A, Q_B, . . . Q_F which apply for specific modes of exposure.

Q_A results from direct external gamma or X-ray exposure; Q_B from direct external beta particle radiation; Q_C from inhalation exposure; Q_D from skin contamination; Q_E from submersion in gaseous materials; and Q_F from alpha particle radiation. Q_A and Q_B alone apply to Q_1, while Q_A through Q_F apply for Q_2.

2. INDIVIDUAL Q LIMITS

2.1 Q_A Limits

Macdonald and Goldfinch (1981a), in developing their formulation for Q_A limits for direct photon exposures, assume an accident which results in complete loss of shielding; accept as the limiting criteria an effective dose equivalent of 50 mSv (5 rem) which is the ICRP recommended annual limit for radiation workers; assume a 30-minute exposure

at one meter from a point source; calculate the radiation exposure rate at one meter from a photon source as 0.53 × quantity of radioactivity (curies) × mean photo energy (MeV) released per disintegration; and convert from exposure to an average dose equivalent using a factor of 0.8 Sv/Gy × 0.00867 Gy/R for a sphere with a 30-cm radius.

The combination of proposed changes of dose equivalent limit, distance and duration of exposure from the previous values (IAEA, 1979) of 30 mSv (3 rem), 3 m and 3 h to the new values would increase Q_A limits by about 10%, assuming no attentuation in air. However, we recommend the following time-distance accident scenario as more realistic: 4 hours exposure at 3 meters and 5 minutes at 1 meter (for recovery operations). This recommendation yields Q_A limits midway between the two other values and thus has no appreciable impact on Q_A limits (see Table 1) but it does impact Q_B values (Section 2.2). Although calculations for exposure rate per unit of source activity and conversion from roentgens to sieverts may supress weak dependencies on photon energy and the representation of the human body as equivalent to a sphere may be an oversimplification, these methods are considered adequate for the purpose.

2.2 Q_B Limits

In calculating Q_B limits for direct beta exposures, Macdonald and Goldfinch again assume complete loss of shielding by the package and 30-minute exposure at 1 meter from a point source. In addition, they use a series of four intrinsic shielding factors (150 mg/cm^2 of source) for non-overlapping ranges of initial beta particle energies and accept ICRP guidance on a skin dose limit of 0.5 Sv (50 rem). They give Q_B

Table 1. Comparison of scenarios for duration of exposure, distance and photon dose limits

Scenario proposed by	Values used in scenario			Radiation dose rate limit for source at 1 m mSv/h (rem/h)
	Duration h	Distance m	Dose limit mSv (rem)	
IAEA, 1973-79	3	3	30 (3)	90 (9)
Macdonald-Goldfinch	0.5	1	50 (5)	100 (10)
This MS	1/12	1		
	4	3	50 (5)	95 (9.5)

values for beta particles with maximum energies of less than 0.3 MeV, 0.3–0.5 MeV, 0.5–1 MeV and greater than 1 MeV. To allow for bremsstrahlung, an additional limit of 40 TBq (1000 Ci) is set for evaluating Q_1.

Aside from the different scenario which we propose (see Section 2.1) and discuss further subsequently, there are at least four weaknesses in the procedure of Macdonald and Goldfinch:

1. The bremsstrahlung limit of 40 TBq is based on historical precedent rather than rational justification. For materials with very low specific activities (i.e., long radioactive half-lives), this limit is probable unnecessarily restrictive.
2. For nuclides with multiple beta spectral components (e.g., beta energy groups with different maximum energies), calculation for the maximum beta energy is likely to be inappropriate and of course is completely inappropriate for conversion electrons. Summation of calculations for each component weighted by its probability of occurrence seems to be the best procedure.
3. In the work of Macdonald and Goldfinch (1981a) attentuation by 1 meter of air is built in through their Figure 1a (based on calculations by Cross, 1967), which gives no points below maximum beta energy of 0.4 MeV. Since the range of a 0.3 MeV electron is but about 75 cm in air at atmospheric pressure at 20° C, we suggest that Q_B for beta energies of 0.3 MeV or less, at one meter should be limited only by consideration of bremsstrahlung.

4. The Macdonald-Goldfinch assumption of a self-absorption thickness of 150 mg/cm^2 implies an unexpressed mass- and hence activity-limit for direct exposure by beta emitters. We conservatively estimate these by envisioning a spherical body of 0.5-m radius surrounded by a source at 1.5 m radius and thickness of 0.15 g/cm^2. The maximum source mass is then 4π (2.25 m^2) $\times$ 1.5 $\times$ 10^3 (g/m^2) or about 42 kg and the maximum source strength in (TBq) of about 42 $\times$ 1.33 $\times$ 10^7/At where A is the atomic number of the radioisotope and t is its half-life in years. If this source strength is less than the calculated Q_B, the effective values of Q_B should revert to that controlled by bremsstrahlung.

From the scenario of Macdonald and Goldfinch calculations for direct exposures are made assuming a distance of 1 meter of 30-minute duration. As mentioned in Section 1.1, we consider more realistic and propose a 4-hour exposure at 3 meters and 5-minute exposure at 1 meter. For Q_B the shorter exposure at 1 meter gives one-sixth of that for the Macdonald-Goldfinch scenario. The extra 2 meters air absorption are important since the range of electrons with initial energies up to 0.8 MeV is less than 3 meters. Thus at 3 meters only the bremsstrahlung consideration applies at less than 0.8 MeV.

We note that for very high energy beta particles the 0.15 Sv (15 rem) limit as recommended by the ICRP for the lens may need to be discussed. Because of shielding by the head, this consideration can probably be dismissed except when facing the source. We also note that the self-shielding factor for very low energy betas may be underestimated.

2.3 Q_C Limits

For calculation of Q_C limits for inhalation, Macdonald and Goldfinch pragmatically assume a breathing rate of 0.72 m^3/h, a 30-minute exposure in a room size of $3 \times 10 \times 10$ m^3 with four air changes per hour and that 1% of the package contents are instantaneously and uniformly dispersed in that room. They use the ICRP recommended annual limits of intake (ALI) for the most restrictive chemical forms of the nuclides as the limiting exposures. For calculations, they then use:

$$Q_C = ALI/(6 \times 10^{-6}) \text{ (Bq)} . \qquad (1)$$

We have four comments regarding this treatment: (1) the average breathing rate for adult males is given as 23 m^3/d or as 1.2 m^3/h during waking hours (ICRP, 1975), either of which is somewhat greater than the rate Macdonald and Goldfinch use; (2) justification of the air change rate seems desirable since it is appreciably greater (and hence less conservative) than the reported rate of 0.5 to 1.5 changes per hour in American homes (Handley and Barton, 1973); (3) no allowance is made for radioactive decay in the formulation but this is unlikely to be significant under the conditions of international shipping and 30-minute exposure; and (4) the formulation behind equation (1) illogically gives intake via inhalation as 6×10^{-6} Q_C, independent of duration of exposure. We suggest the following treatment.

Set

$C(t) = C(0)\ \mathrm{Exp}\ (-Rt-\lambda t)$, and equating the intake to the ALI, one has (2)

$$ALI = 0.72 \int_0^T C(t)dt \qquad (3)$$

$$= \frac{0.72\ C(0)}{R + \lambda} \left[1 - Exp\ (-RT - \lambda T)\right] \qquad (4)$$

where C(t) is the concentration of respirable nuclide at time t or initially 0.01 Q_C/300, 0.72 is the breathing rate in m^3/hour, R is the air exchange rate of 4/hour and λ is the decay constant. This yields under the assumed conditions of λ = zero and t = 0.5 hour:

$$Q_C = ALI/(5.19 \times 10^{-6}) \ . \qquad (5)$$

Although the change amounts to only a 16% increase in all Q_C values, it seems conceptually sounder should others apply the formulation for other durations of exposure.

2.4 Q_D Limits

The calculations of Q_D limits by Macdonald and Goldfinch are based on the following: the contamination of bare skin is 0.1% of Q_D per square meter continuing for 5 hours; the dose is calculated to the sensitive basal layer of the skin at a depth of 7 mg/cm^2; a skin dose limit of 0.5 Sv (50 rem) as recommended by the ICRP is accepted; and the calculations are made for five ranges of beta energies. We note that the thickness of skin to the basal layer has been given as 30–50 mg/cm^2 for the palms of the hands and soles of feet, 6–10 mg/cm^2 for

the backs of hands and 3–5 mg/cm^2 for many other areas (Whitton, 1973). Macdonald and Goldfinch have conservatively used 7 mg/cm^2. Although the dose calculation becomes increasing uncertain, perhaps by a factor of 2.5 or more at beta energies of 0.2 MeV or less, we believe the Q_D values are generally acceptable. However, as discussed in Section 2.2, for nuclides with multiple beta spectral components, calculation for the maximum beta energy is likely to be inappropriate and is completely inappropriate for conversion electrons. Summation of calculations for each component of a complex beta spectrum weighted by its probability of occurrence seems to be the best procedure.

2.5 Q_E Limits

Macdonald and Goldfinch's calculation of Q_E limits for submersion in gaseous material depends on assuming 30-minute exposure in a room with a volume of 300 m^3 and four air changes per hour; assuming an instantaneous, uniformly dispersed release of all the initial material, Q_E, regardless of the packaged pressurization; relying on the derived air concentration (DAC) limit recommended in ICRP Publication 30; and accepting the average airborne concentration of material at time t as:

$$\bar{C}(t) \doteq \frac{Q_E\ (1 + 4t)}{2400t} ,$$

$$= Q_E/400, \text{ at } t = 0.5 \text{ hour} . \qquad (6)$$

Equating the exposure from submersion at this average concentration for 0.5 h to that for 2000 h at the DAC, one obtains

$$Q_E = 1.6 \times 10^6 \text{ DAC} \tag{7}$$

As we have commented with regard to the inhalation dose limit Q_C, radioactive decay is ignored. We find that the time-dependence of the formulation for the average air concentration is unacceptable because it illogically yields $\bar{C}(0)$ = infinity, $\bar{C}(0.25\ h)$ = the expected initial concentration of $Q_E/300$, and at all great times approaches a limit of half the expected initial concentration. We suggest calculating the Q_E limit in the following way. Let the time dependent concentration be given, as previously, by:

$$C(t) = C(0)\ \text{Exp}\ (-Rt-\lambda t), \text{ where } C(0) = Q_E/300\ . \tag{8}$$

Equating the inhalation exposure during 2000 hours at the DAC to the exposure during time T yields

$$2000\ \text{DAC} = \int_0^T C(t)dt$$

$$= \frac{Q_E}{300\ (R + \lambda)}\ \left[1 - \text{Exp}\ (-RT - \lambda T)\right]\ . \tag{9}$$

If λ is zero, and R = 4/hour, at the time of interest (0.5 h) then $Q_E = 2.8 \times 10^6$ DAC, rather than 1.6×10^6 DAC as given by the formulation of Macdonald and Goldfinch. Thus these changes would relax (i.e., increase) the Q_E limits by a factor of at least 1.75. A survey of the Q_2 values given in the supplementary table of Q_1 and Q_2 values (called "Appendix 3," 1982) indicates that this would influence the Q_1 and Q_2 values for only Xe-131m and Xe-133, increasing all of them from 30 to 56 TBq.

2.6 Q_F Limits

For the Q_F limits for alpha emitters, Macdonald and Goldfinch merely continue using the arbitrary 1973 assumption that Q_F = 1000 times the limit, Q_C, for inhalation exposure. We suggest that this limit be replaced by a limit of 0.8 TBq (20 Ci), which is also an arbitrary limit. Either approach needs to be replaced with a more rigorous dosimetric analysis, but the time constraint of 1984 IAEA publication of new regulations may make such an analysis impractical.

3. SPECIAL CONSIDERATIONS

3.1 Neutron Sources

Direct neutron doses from (α,n), (γ,n) or Cf-252 spontaneous fission sources are dismissed by Macdonald and Goldfinch as being too small to be of concern for these regulations. Our experience with a 100 Ci Po-Be (α,n) source (Randolph et al., 1967) and calculations for a 1 mg Cf-252 confirm that the direct dose equivalent, using the distance and

duration assumptions made here and a quality factor of 20, would be at least an order of magnitude less than the limit of 50 mSv (5 rem) used here.

3.2 Tritium

Tritium is specially treated by Macdonald and Goldfinch. For organic materials, the ALI is used. Allowance is made for the more hazardous nature of inorganic materials by using ALI/50. No more than 1% of the material is assumed to escape confinement. In addition to the considerations given for other nuclei, the effects of fire in a confined space (increased water content in air and its impact on inhalation and skin contamination) and tritium concentration limits in water are developed. The paper of Macdonald and Goldfinch refers to the reduction of ALI by a factor of 50 for organic materials as recommended in ICRP Publication 30, whereas we note that ICRP 30 (1979, p. 67) states "specific values of ALI are not recommended for organic compounds of tritium but. . . they might differ considerably from those for tritiated water and. . . for tritiated thymidine might be as much as 50 times smaller. . . This matter will be kept under review." Thus Macdonald and Goldfinch have taken a conservative, but not yet recommended, approach. To us this seems a generally conservative approach, although several additional quantitative assumptions are required. The wisdom of treating one nuclide in so much more detail than the others seems uncertain.

3.3 Additional Factors

Beyond the development of the Q_A, Q_B through Q_F limits, Macdonald and Goldfinch apply some additional constraints in calculating Q_1 and Q_2. From the previous IAEA regulations, they incorporate an ad hoc allowance for bremsstrahlung by setting a limit of 40 TBq (1000 Ci) for beta emitters. For alpha-beta emitters, they set Q_1 equal to the least of Q_A, Q_B and 1000 Q_C. Furthermore for low specific activity materials, they assume the maximum possible inhalation is 10 mg. Thus if the radioactivity of 10 mg of a nuclide is less than the appropriate ALI, Q_C is listed as "unlimited" and the least of Q_A, Q_B or 40 TBq (1000 Ci) appears as both Q_1 and Q_2. These seem to be plausible rules although quantitative justifications have not been given and grams of dusts have been reported in the lungs of coal miners after years of mining. Nowhere do we find a clear statement of what was done when the daughter element was radioactive although some such rules were given in the earlier version (IAEA, 1979).

4. IMPACT ON U. S. SHIPPING

The impact of the new proposals on U. S. shipments of radionuclides has been done, in a preliminary way, by comparing the new Q_1 and Q_2 with the old A_1 and A_2 (Table 2). Although, the top half of Table 2 shows almost as many nuclides with less restrictive Q_1 than A_1, the bottom half shows more nuclides with significantly more restrictive Q_1 than A_1. By both criteria the number of nuclides with more restrictive Q_2 than A_2 exceeds the number with less restrictive Q_2 than A_2. About half the

Table 2. Comparisons of limits Q_1 versus A_1 and Q_2 versus A_2

	Result of comparison	Number of Nuclei	
		Special form Q_1 vs A_1	Other forms Q_2 vs A_2
1.	Q < A (more restrictive)	128	169
	Q = A	3	2
	Q > A (less restrictive)	114	77
2.	Q < 0.1A (most restrictive)	18	19
	0.1A<Q<0.5A	73	93
	0.5A<Q<2A	131	120
	2A<Q<10A	20	13
	Q>10A (least restrictive)	2	3

ratios of Q_1/A_1 and of Q_2/A_2 lie in the unimportant range of 0.5 to 2. Also a preliminary comparison (Table 3) was made of U. S. shipping of radionuclides in 1976 (Simmons et al., 1977) versus the new Q_1 limits. For about 80% of the nuclides all packages were less than the A_1 limits. From these comparisons, we conclude that adoption of the new regulations within the U. S. would be unlikely to have a major impact on internal U. S. shipments, but the impact on foreign shipments has not been specifically studied.

5. SUMMARY

The Q-system developed by Macdonald and Goldfinch is found to be generally an improvement over the existing IAEA Safety Series 6 regulations. We find:

1. Q_A limits for direct photon exposures are acceptable although we recommend a slightly different accident scenario which yields equivalent Q_A limits;
2. Q_B limits for direct beta exposures are questioned on the basis of the different accident scenario we recommend and uncertainty about treatments of beta ray spectra, beta particle ranges and source self-absorption;
3. Q_C limits for inhalation exposures are quantitatively acceptable, but we recommend a sounder formulation;
4. Q_D limits for skin contamination are generally acceptable but for beta emitters more careful treatment for complex spectra and conversion electrons is desired;

Table 3. Comparison of 1976 shipments of radionuclides in the U. S. (Simmons et al., 1976) with Q_1 limits

Result of comparison	Number of Nuclides
Quantities in all packages were less than Q_1	80
Quantities in more than 90% of packages were less than Q_1	9
Quantities in 50% to 90% of packages were less than Q_1	7
Quantities in less than 50% of packages were less than Q_1	3
Total	99

5. Q_E limits for submersion in gases should be relaxed (increased) because of a faulty formulation by a factor of 1.75, although this influences very few of the Q_1 or Q_2 limits, and the formulation for average concentration should be revised;
6. Q_F limits for special form alpha particles are arbitrarily set at 1000 Q_C in place of which we suggest an arbitrary limit of 0.8 Sv (20 Ci) although more justification or revision is needed.

Perhaps several of our comments would be better directed to revision of IAEA Safety Series 37 (IAEA, 1973) on the methodology behind setting limits. The new Q_1 and Q_2 limits proposed by Macdonald and Goldfinch are somewhat more restrictive than the old A_1 and A_2 limits but within the U. S. shipping quantities seem to be dictated more by customer convenience than by regulatory limits.

6. ACKNOWLEDGEMENTS

We are glad to acknowledge help from personal discussions with J. P. Witherspoon (ORNL), Richard Rawl (DOT), and D. R. Hopkins (NRC), the consensus on several points reached by the round table discussions at DOT Headquarters on June 22, 1982, and manuscript preparation by W. C. Minor (ORNL).

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