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Subject: McNally Petition on Tritium (PRM-50-37) (references cited in denial)

Harold T. Peterson, Jr.

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TRITIUM PRODUCTION, RELEASES AND POPULATION DOSES AT NUCLEAR POWER REACTORS*

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ABSTRACT

Tritium is produced in light-water-cooled reactors as a product of ternary fission and by nuclear reactions with the coolant and with neutron-absorbing materials used for reactor control. Pressurized water reactors (PWRs) have greater amounts of tritium produced in or released into the coolant than boiling water reactors (BWRs). Consequently, tritium releases to the environment from PWRs [29 GBq/MW(e)-y (0.78 Ci/MW(e)-y)] are about 6½ times greater than from BWRs [4.4 GBq/MW(e)-y (0.12 Ci/MW(e)-y)]. Most of the tritium released from PWRs appears in the liquid effluent (about 85%), whereas 75% of the tritium released from BWRs is as airborne effluents. Radiation doses from these tritium releases are small; the average site collective (population) dose in 1981 was less than 0.002 person-sieverts per year (0.2 person-rem/ year). The total collective dose from all tritium releases was 0.08 person-sieverts (8 person-rem).

I. TRITIUM PRODUCTION IN LIGHT-WATER REACTORS

A. Fission

Tritium was identified as a product of the ternary fission of uranium-235 in 1959[1] and subsequently has been identified as a product of the fission of most fissile materials. A nominal fission yield of 1 triton per 10,000 ^{235}U fissions[2] would produce approximately 14 terabecquerels (380 curies) of tritium per metric ton of fuel having a burnup of 30,000 megawatt(t)-days. Fissions occurring in plutonium-239 and uranium-238 raise the total production to about 19 TBq (515 curies) per metric ton of fuel. This results in a total annual production of approximately 600 TBq (16,000 curies) per year for a 3,000 MW(t) [1,000 MW(e)] reactor operating for 300 full-power equivalent days.

The amount of ternary fission-product tritium which is released to the environment depends upon the fraction which escapes from the fuel rod into the reactor coolant. This fraction is affected by the materials used for the fuel rod cladding and its integrity. The zirconium alloy (Zircaloy) fuel rad cladding used in most current light-water reactors exhibits greater retention of tritium than the stainless steel cladding used in early reactors. Stainless steel is highly permeable to tritium. Early boiling water reactors (BWRs) with stainless steel cladding had normalized tritium releases between 0.02 to 0.03 TBq per electrical megawatt-year. (0.5 to 0.8 Ci/MW(e)-y)[3]; whereas a zirconium-clad BWR had tritium releases of 0.0015 to 0.0026 TBq/MW(e)-y (0.04 to 0.07 Ci/MW(e)-y)[3]. This improved retention appears to be due to the formation on the surface of the zirconium cladding of a hydrogen- impermeable oxide layer.[4,5] Approximately half of the total tritium inventory in the fuel may be bound to the cladding[6].

B. Boron Reactions

Boron is a neutron absorber and is used in several forms in LWRs for control.. In General Electric boiling water reactors and Combustion Engineering pressurized water reactors boron carbide (B_4C), is used in moveable control rods. (Most PWRs use AgInCd control rods which are not a source of tritium.) Boron is also used in the form of either a borosilicate glass (Westinghouse) or B_4C (Babcock & Wilcox and Combustion Engineering plants) in fixed "burnable" absorbers to compensate for fuel depletion and fission-product "poison" buildup. Pressurized water reactors also use a soluble boron

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compound in the primary coolant to compensate for fuel depletion and, by changing the concentration through removal or addition of boron, to make changes in power levels.

The primary boron reactions which yield tritium are:

$^{10}B\ (n,\alpha)\ ^{7}Li\ (n,\ n\alpha)\ ^{3}H$,

$^{10}B\ (n,\ 2\alpha)\ ^{3}H$, and

$^{11}B(n,\ n2\alpha)^{3}H$.

The ^{10}B reaction yielding ^{7}Li is the primary neutron absorbing reaction having a thermal neutron absorption cross-section of approximately 3.8 E-25 m^2 (3,800 barns). The other two reactions have small cross-sections (less than 1E-30 m^2 or 0.1 barns.) The ^{11}B reaction has a threshold of 14 MeV. It is not an important source of tritium due to the small number of high-energy neutrons present in light-water reactors as well as its small reaction cross-section.

The amount of tritium produced by reactions with boron (B_4C) in the moveable control rods of a 1,000 MW(e) boiling water reactor is relatively large, about 115 TBq (3,100 Ci).[5] Due to differences in the use of the control rods (differences in the time the rods are kept in the reactor core) Combustion Engineering PWRs produce less tritium in the B_4C control rods ~ 33 TBq/y (~ 900 Ci/y).[5] About 37 TBq (1,000 curies) of tritium are produced per year in the fixed absorbers and 11-18.5 TBq (300-500 Ci) of ^{3}H are produced by the boron in the coolant of PWRs[2,3,5,6,8,9].

All of the tritium produced by reactions with soluble boron is produced directly in the coolant. Only about 0.2% of the tritium generated in the B_4C control rods appears to be released to the coolant.[5,7]. Releases from fixed absorbers using B_4C are also less than 1%. Borosilicate glass absorber plates appear to have less retention and releases have been estimated to be between 10 and 50% of the production [5,9].

C. Lithium Reactions

Lithium salts are added to the coolant of pressurized-water reactors to control acidity (pH). In operation the lithium concentration in the coolant varies between 0.2 and 2 parts per million. Natural lithium has an isotopic composition of 7.4% ^{6}Li and 92.6% ^{7}Li. Tritium is produced by neutron capture reactions with both isotopes:

$^{6}Li\ (n,\ \alpha)\ ^{3}H$ and

$^{7}Li\ (n,\ n\alpha)^{3}H$.

The ^{6}Li reaction has a considerably larger neutron absorption cross-section for tritium production than ^{7}Li. The ^{6}Li thermal neutron cross-section is approximately 9.5E-26 m^2 (950 barns).[10] The ^{7}Li reaction has a 3-MeV threshold and a cross-section of about 8.6E-30 m^2 (0.086 barns).

Commercial light-water reactors have employed lithium salts which are enriched to 99.9% ^{7}Li in order to reduce tritium production. The lithium used for pH control yields approximately 0.3 TBq/MW(e)-y (0.01 Ci/MW(e)-y) in an equilibrium fuel cycle.[6]

D. Deuterium Activation

Deuterium (^{2}H or D) occurs naturally, comprising approximately 0.015 per cent of natural hydrogen. Tritium can be formed by neutron activation of deuterium by the reaction:

$$^{2}H\ (n,\ \gamma)^{3}H$$

with a thermal neutron cross-section of 4.6 E-32 m^2 (0.46 millibarns).[10] Although this reaction is the major source of tritium in heavy-water moderated reactors, the low natural abundance of deuterium and its small activation cross-section make this a minor source in light-water reactors with an estimated production of 0.2 TBq (slightly less than 10 curies) per year in a 1,000 MW(e) reactor.[10]

E. Summary of Tritium Production

Estimates of the tritium production rates in 1,000-MW(e) light-water-cooled nuclear power reactors are summarized in Table 1. As indicated in the table, a boiling water reactor is estimated to produce slightly (10%) more tritium than an equivalent pressurized water reactor. However, due to production of tritium from boron in the coolant, pressurized water reactors are estimated to have more tritium available for releases to the environment.

II. TRITIUM RELEASES TO THE ENVIRONMENT

The amount of tritium released to the environment from commercial light-water reactors is highly variable as the data in Table 2 indicate. Typically, pressurized water reactors had tritium releases between 0.01 to 0.055 TBq per electrical megawatt-year (0.3 to 1.5 Ci/MW(e)-y). Boiling water reactors had lower releases 0.74 GBq to 0.013 TBq per MW(e)-year (0.02 to 0.34 Ci/MW(e)-y). This variation reflects not only differences in reactor type, design and materials of construction; but also differences in power generating history and waste management actices at individual sites. The higher ritium release from pressurized water reactors [average: 0.029 TBq/MW(e)-y (0.78 Ci/MW(e)-y)] compared to boiling water reactors [average: 4.4 GBq/MW(e)-y (0.12 Ci/MW(e)-y)] reflects the greater tritium production in and release into the coolant of PWRs. The higher tritium releases through stainless steel clad fuel is evident from the normalized (Ci/MW(e)-y) data for the LaCrosse BWR and the Haddem Neck and San Onofre PWR plants.

The mode of tritium release to the environment as well as the magnitude of activity released is highly variable from site to site and differs between pressurized water and boiling water reactors. The total tritium release to the environment from U.S. BWRs in 1981 was approximately 44 TBq (1,200 curies). Of this total, approximately 10 TBq (280 Ci) were released as liquid effluents and 34 TBq (920 Ci) were released as airborne effluents. Pressurized water reactors released approximately 768 TBq (20,710 Ci) of tritium; 690 TBq (18,600 Ci) as liquid effluents and 78 TBq (2,110 Ci) as discharges to the atmosphere. The activity of tritium per unit energy production released as airborne emissions from the two reactor types is similar (about 3.7 GBq/MW(e)-y (~0.1 Ci/MW(e)-y). However, the airborne releases comprise 80% of the tritium released at a typical BWR site but only about 15% of the tritium released from pressurized water reactors, the remaining 85% being released with liquid effluents.

The release mode for tritium can be modified to accommodate local environmental conditions of the site such as the availability of dilution water for liquid effluents. Thus, it is possible to have BWRs releasing 90% of the tritium via liquid effluents (Oyster Creek) and PWRs releasing over 60% of the tritium via airborne effluents (Rancho Seco).

III. RADIATION DOSES FROM TRITIUM RELEASES TO THE ENVIRONMENT

The models used for the dose calculations have been described elsewhere.[11,12] The total population doses from releases of radionuclides in 1981 have also been published[13]; however, the doses from tritium were not evaluated separately in that report.

A. Methods

The specific activity approach is used to estimate the long-term concentrations of tritium in vegetation, and animal food products. The tritium concentrations in water and in the atmosphere are estimated using established

Table 1. Tritium Production in Light-Water-Cooled Nuclear Power Reactors

[1,000 MW(e) plant operated for 300 full-power equivalent days]

Reactor type and Tritium Source	Amount of Tritium Produced		Amount Released to Reactor Coolant		
	TBq	Ci	TBq	Ci	% of production
Boiling Water Reactor (BWR)					
ternary fission	600	16,000	3.7	100	0.6
encapsulated boron	115	3,100	0.4	~ 10	0.3
deuterium activation	0.4	< 10	0.4	< 10	100
	715	19,100	4.4	120	0.6
Pressurized Water Reactor (PWR)					
ternary fission	600	16,000	3.7	100	0.6
encapsulated boron	37	~ 1,000	11	300	30
boron/lithium in coolant	11-18	300-500	11-18	300-500	100
deuterium activation	0.4	< 10	0.4	< 10	100
	650	17,400	30	800	5

Table 2. Reported Tritium Releases to the Environment in 1981 from Nuclear Power Reactors

	1. Reactor Type and Site (number of units)	2. Energy Generation [MW(e)-h][a]	3. TRITIUM RELEASES Activity (curies)[a] Liquid	4. Airborne	5. Total	6. Activity per Electrical Megawatt (Ci/MW(e)-y)[b] Liquid	7. Airborne	8. Total	
I.	**BOILING WATER REACTORS**								
1.	Duane Arnold (1)*	2.2E+06	0.0E+00*	NR*	--*	0.0E+00*	--	--	
2.	Big Rock Point (1)*	4.7E+05	3.1E+00*	<1.0E+01*	<1.3E+01*	5.8E-02*	<1.9E-01	<2.4E-01*	
3.	Browns Ferry (3)	1.8E+07	2.4E+02	3.8E+01	6.2E+01	1.2E-02	1.8E-02	3.0E-02	
4.	Brunswick (2)	5.8E+06	2.3E+01	1.8E+01	4.0E+01	3.4E-02	3.0E-02	6.0E-02	
5.	Cooper (1)	3.8E+06	<8.4E+00	4.5E+00	<1.3E+01	1.9E-02	1.0E-02	2.9E-02	
6.	Dresden 2-3 (2)	8.6E+06	6.0E+00	3.2E+02	3.2E+02	6.1E-03	3.2E-01	3.3E-01	
7.	James Fitzpatrick (1)	4.8E+06	4.1E+00	6.6E+00	1.1E+01	7.5E-03	1.2E-02	2.0E-02	
8.	Edwin I. Hatch (2)	7.2E+06	2.1E+01	9.6E+00	3.0E+01	2.5E-02	1.2E-02	3.7E-02	
9.	LaCrosse (1)*	2.4E+05*	7.7E+01*	2.3E+01*	1.0E+02	2.8E+00*	2.5E-01*	3.7E+00*	
10.	Millstone-1 (1)	2.5E+06	2.6E+00	9.5E+01	9.7E+01	9.1E-03	3.3E-01	3.4E-01	
11.	Monticello (1)*	3.3E+06*	4.2E-03*	1.1E+02*	1.1E+02	1.1E-05*	3.0E-01*	3.0E-00*	
12.	Nine Mile Point (1)	3.3E+06	5.0E+00	6.3E+01	6.8E+01	1.3E-02	1.7E-01	1.8E-01	
13.	Oyster Creek (1)	2.6E+06	2.7E+01	3.2E+00	3.0E+01	2.7E-02	3.2E-03	3.0E-02	
14.	Peach Bottom 2-3 (2)	9.8E+06	3.7E+01	2.8E+01	6.5E+01	3.3E-03	2.6E-02	5.9E-02	
15.	Pilgrim (1)	3.4E+06	3.4E+01	7.6E+01	1.1E+02	8.7E-02	1.9E-01	2.8E-01	
16.	Quad Cities (2)	9.5E+06	1.2E+01	8.6E+01	9.8E+01	1.1E-02	7.9E-02	9.0E-02	
17.	Vermont Yankee (1)	3.6E+06	3.7E-01	2.0E+01	2.0E+01	9.1E-04	4.8E-02	4.9E-02	
	Mean ± S.E.	[Ci/reactor-y]	10. ± 0.4	38. ± 2.4	48. ± 2.4	0.022 ± 0.002	0.098 ± 0.009	0.12 ± 0.01	[Ci/MW(e)-y]
		[TBq/reactor-y]	0.38 ± 0.02	1.4 ± 0.1	1.8 ± 0.1	0.81 ± 0.06	3.6 ± 0.3	4.4 ± 0.4	[GBq/MW(e)-y]
II.	**PRESSURIZED WATER REACTORS**								
1.	Arkansas Nuclear One (2)	9.2E+06	6.9E+02	1.3E+02	8.1E+02	6.5E-01	1.2E-01	7.7E-01	
2.	Beaver Valley (1)	4.7E+06	1.4E+02	9.5E-02	1.4E+02	2.6E-01	1.8E-04	2.6E-01	
3.	Calvert Cliffs (2)	1.2E+07	1.0E+03	5.8E+00	1.0E+03	7.6E-01	4.4E-03	7.7E-01	
4.	Donald C. Cook (2)	1.3E+07	9.2E+02	5.5E+00	9.2E+02	6.1E-01	3.6E-03	6.1E-01	
5.	Crystal River (1)	4.0E+06	2.7E+02	1.6E+01	2.9E+02	5.9E-01	3.4E-02	6.3E-01	
6.	Davis-Besse (1)	4.4E+06	1.6E+02	8.6E+00	1.7E+02	3.2E-01	1.7E-02	3.3E-01	
7.	Joseph Farley (2)	5.5E+06	8.0E+02	1.9E+02	9.9E+02	1.4E+00	3.0E-01	1.6E+00	
8.	Fort Calhoun (1)	2.2E+06	2.4E+02	8.2E+01	3.2E+02	1.0E+00	3.3E-01	1.3E+00	
9.	Robert E. Ginna (1)	3.3E+06	2.4E+02	7.0E+01	3.1E+02	6.3E-01	1.8E-01	8.2E-01	
10.	Haddem Neck (1)	4.1E+06	5.3E+03*	8.6E+01*	5.4E+03*	1.1E+01*	1.9E-01*	1.1E+01*	
11.	Indian Point (3)	6.1E+06	8.8E+02	7.9E+00	8.9E+02	1.3E+00	1.1E-02	1.3E+00	
12.	Kewaunee (1)	3.8E+06	2.5E+02	3.8E+00	2.6E+02	5.8E-01	8.8E-03	5.9E-01	
13.	Maine Yankee (1)	5.2E+06	2.2E+02	4.5E+00	2.2E+02	3.7E-01	7.7E-03	3.8E-01	
14.	McGuire (1)	1.9E+04*	6.2E+00*	6.5E-02*	6.3E+01*	2.9E+00*	3.0E-02*	2.9E+00*	
15.	Millstone-2 (1)	6.1E+06	3.7E+02	1.4E+02	5.1E+02	5.3E-01	2.0E-01	7.4E-01	
16.	North Anna (2)	1.0E+07	1.3E+03	3.1E+01	1.3E+03	1.1E+00	2.7E-02	1.1E+00	
17.	Oconee (3)	1.4E+07	5.1E+02	5.8E+01	5.6E+02	3.2E-01	3.7E-02	3.6E-01	
18.	Palisades (1)	3.5E+06	2.8E+01	6.4E+00	2.8E+02	7.0E-01	1.6E-02	7.2E-01	
19.	Point Beach (2)	6.3E+06	6.5E+02	4.8E+02	1.1E+03	9.0E-01	6.6E-01	1.6E-01	
20.	Prairie Island (2)	6.9E+06	5.6E+02	7.4E+01	6.4E+02	7.1E-01	9.0E-02	8.0E-01	
21.	Rancho Seco (1)	2.6E+06	8.4E+01	1.4E+02	2.2E+02	2.8E-01	4.7E-01	7.5E-01	
22.	H. B. Robinson 2 (1)	3.5E+06	1.9E+02	1.1E+01	2.0E+02	4.7E-01	2.6E-02	4.9E-01	
23.	Salem (2)	7.8E+06	1.3E+03	3.9E+00	1.3E+03	1.5E+00	4.0E-03	1.5E+00	
24.	San Onofre (1)	7.8E+05	3.0E+02*	1.2E+01*	3.1E+01*	3.3E+00*	1.4E-01*	3.5E+00*	
25.	Sequoyah (1)	2.5E+06	7.6E+01	9.2E-01	7.7E+01	2.6E-01	3.2E-03	2.7E-01	
26.	St. Lucie (1)	5.0E+06	3.2E+02	3.7E+02	7.0E+02	5.8E-01	6.6E-01	1.2E+00	
27.	Surry (2)	7.5E+06	5.3E+02	6.3E+01	5.9E+02	6.2E-01	7.2E-02	6.9E-00	
28.	Three Mile Island (2)*	-	7.2E+01*	6.5E+01*	7.3E+01*	-	-	-	
29.	Trojan (1)	6.4E+06	1.0E+02	4.0E+01	1.4E+02	1.4E-01	5.5E-02	2.0E-01	
30.	Turkey Point 3-4 (2)	5.4E+06	2.0E+02	6.9E-01	2.0E+02	3.2E-01	1.1E-03	3.2E-01	
31.	Yankee (1)	8.8E+05	1.0E+02	3.1E+00	1.0E+02	1.0E+00	3.1E-02	1.0E+00	
32.	Zion (2)	1.1E+07	8.7E+02	NR	-	6.7E-01	-	-	
	Mean ± S.E.	[Ci/reactor-yr]	302 ± 4.	47. ± 2.	350 ± 4.	0.65 ± 0.01	0.12 ± 0.01	0.78 ± 0.02	[Ci/MW(e)-y]
		[TBq/reactor-yr]	11 ± 0.1	1.7 ± 0.07	13 ± 6.2	24 ± 0.4	4.6 ± 0.3	29. ± 0.6	[GBq/MW(e)-y]
III.	**HIGH-TEMPERATURE GAS -COOLED REACTOR**								
	Fort St. Vrain (1)	7.6E+05	2.2E+02	1.4E+01	2.3E+02	2.5E-01	1.6E-02	2.7E-01	

* not included in averages because of atypical design (LaCrosse, Monticello, Big Rock Point, Haddem Neck, San Onofre) non-operation (Three Mile Island), or low power production (McGuire) but included in total-release.

(a) Energy generation and activity release data is taken from J. Tichler and C. Benkovitz, "Radioactive Materials Released from Nuclear Power Plants - Annual report" NRC Report NUREG/CR-2907 Vol. 2 (June 1984) for 1981 releases.

(b) Calculated from the reported released activity (columns 3-5) divided by the energy generation (column 2) converted to MW(e)-y.

dispersion models. Tritium concentrations in vegetation and animal food products are determined from the calculated tritium specific activity (S.A.) in atmospheric moisture. The specific activity of atmospheric moisture is calculated as:

$$(S.A.)_{air} = [X/Q\ (r,\theta]Q/H$$

where H is the absolute humidity (taken to be 0.008 kg water/m^3 air) and $[X/Q(r,\theta)]Q$ gives the atmospheric tritium concentration (activity/ m^3) at point r,θ.

The specific activity in vegetation is estimated to be one-half (0.5) of the tritium specific activity in atmospheric moisture. Recently this assumption has been challenged by Murphy[14] who indicated that a value of 0.8 might be more appropriate. Dinner et al.[15] show that this value is dependent upon the type of vegetation; the factor of 0.5 being the mean for vegetation, but a factor of 0.8 would be more appropriate for leafy vegetables. Using the factor of 0.5 and an assumed average vegetation composition of 75% water, the transfer factor from air to vegetation is given by:

$$TF_{air\text{-}to\text{-}plant} = \frac{0.5(0.75\text{kg water/kg plant})}{(0.008\text{kg water/cu. meter air})}$$

$$= 47\ \frac{Bq/kg}{Bq/m^3} = 47\ \frac{m^3}{kg}.$$

Long-term transfer of tritium into meat and milk is calculated using tritium transfer factors which relate the concentration in the food product to the activity ingested by the animal. These factors are:

TF_{milk} = 0.01 Bq/L per Bq/day (=day/L)

TF_{meat} = 0.012 Bq/kg per Bq/day (=day/kg).

Doses are calculated for the average individual (adult) inhaling 8 x 10^3 m^3 of air per year, drinking 110 L of milk and 370 L of water and consuming 190 kg of produce, 95 kg of meat and poultry, 6.9 kg of fish and 1.0 kg of seafood per year. The whole body dose factors are based upon a quality factor (Q.F.) of 1.7 and are:

2.8×10^{-11} Sv/Bq (1.04×10^{-7} mrem/pCi) for ingestion and

4.3×10^{-11} Sv/Bq (1.58×10^{-7} mrem/pCi) for inhalation.

The population is assumed to be comprised solely of adults but the population distributions and sizes for the individual sites are used.[13]

B. Results

The calculated collective (population) radiation doses resulting from tritium releases from nuclear power reactors are shown in Table 3. The principal parameters that determine this dose are: the activity and mode of tritium releases to the environment, the amount of dilution available from atmospheric dispersion and water bodies receiving liquid effluent discharges, and the size and distribution of the population in the vicinity of the reactor site.

The total collective dose from tritium to the population residing within 80 kilometers (50 miles) of nuclear power reactors is small, 0.083 person-sieverts (8.3 person-rem). Most (93%) of this total is contributed by pressurized water reactors. The highest site collective dose of 0.048 person-sieverts (4.8 person-rem) may be high due to a possible underestimation of the available dilution water flow. This value accounts for almost 60% of the U.S. total collective dose. If this single value were neglected, the total dose at PWR sites would be 0.028 person-sieverts (2.8 person-rem) or 82% of the revised total of 0.034 person-sieverts (3.4 person-rem).

The highest site collective doses, 1.4 ± 0.2 (S.E.) person-millisieverts (0.14±0.02 person-rem) occurred at PWR sites located on lakes and impoundments. Over half (~ 55%) of this dose was received from drinking water ingestion. The smallest doses occurred at boiling water reactor sites also located on lakes or impoundments, (4.7±0.2)E-02 person-millisieverts or (4.7±0.2)E-03 person-rem. However, there was no reported drinking pathway at these sites, the doses being primarily from airborne emissions.

IV. SUMMARY AND NEED FOR ADDITIONAL TRITIUM CONTROLS

The collective dose from tritium released to the environment totalled 0.083 person-sieverts (8.3 person-rem) in 1981. The average collective dose was 4.3E-04 person-sieverts (0.043 person-rem) at BWR sites and 2.6E-03 person-sieverts (0.26 person-rem) at PWR sites. The Nuclear Regulatory Commission has a cost-effectiveness criterion of $1,000 per man-rem reduction ($10^5 per person-sievert reduced) for requiring additional radioactive effluent treatment at light-water reactors.[16] Using that criterion, no treatment to retain tritium would be indicated as there are no processes which could reduce tritium releases and cost less than $300 per site. Existing tritium concentration processes (electrolysis, distillation, hydrogen distillation, etc.) would cost between $30-$50 per cubic meter[6] to process the 3,000 m^3 (100,000 ft^3) of coolant or liquid wastes at large power reactors. The

Table 3. Calculated Collective (Population) Whole-body Doses from Tritium Releases from Nuclear Power Reactors in 1981

1. Reactor Type and Site	2. Collective Dose (person-rem) Liquid Effluents	3. Airborne Effluents	4. Total	5. Percentage of Total Tritium Dose from: Drinking Water	6. Aquatic Foods	7. Airborne Releases	8. Percentage of Total Dose Contributed by Tritium
I. BOILING WATER REACTORS							
1. Duane Arnold	-	-	-	-	-	-	-
2. Big Rock Point*	3.8E-05*	1.2E-03*	1.25E-03*	0.16*	2.5 *	9.7 *	0.03 *
3. Browns Ferry	6.8E-04	1.0E-02	1.1E-02	5.3	1.0	94.	0.34
4. Brunswick	6.8E-05	1.5E-03	1.5E-03	0	0.4	~100.	0.01
5. Cooper	8.4E-08	3.5E-04	3.5E-04	0	0.02	~100.	2.2
6. Dresden 2-3	0.0E+00	2.8E-01	2.8E-01	0	0	~100.	0.08
7. James Fitzpatrick	3.9E-04	9.3E-04	1.3E-03	29.	0.2	70.	0.02
8. Edwin Hatch	1.2E-04	1.1E-03	1.2E-03	0	10.	90.	0.05
9. La Crosse*	3.4E-04*	4.5E-03*	4.8E-03*	0 *	7 *	93. *	0.085*
10. Millstone-1	4.2E-07	3.8E-02	3.8E-02	0	< 0.01	~100.	96.
11. Monticello*	1.2E-05*	4.9E-02*	4.9E-02*	0.14*	< 0.01	~100.	~100. *
12. Nine Mile Point	2.5E-03	8.9E-03	1.1E-02	22.	~ 0.08	78.	0.18
13. Oyster Creek	2.0E-04	2.0E-03	2.2E-03	0	9.	91.	0.015
14. Peach Bottom 2-3	9.7E-02	4.5E-02	1.4E-01	68.	< 0.01	32.	5.2
15. Pilgrim	1.5E-05	3.1E-02	3.1E-02	0	< 0.05	~100.	31.
16. Quad Cities	4.2E-03	2.0E-02	2.4E-02	18.	0.14	92.	31.
17. Vermont Yankee	1.8E-05	2.2E-02	2.2E-02	0	< 0.01	~100.	0.036
BWR Total (person-rem)	0.10	0.46	0.56	-	-	-	-
(person-mSv)	1.0	4.6	5.6	-	-	-	-
BWR Average (±S.E.)[#] (person-rem)	(8.1±2.0)E-03	(3.5±0.6)E-02	(4.3±0.6)E-02	10.	1.5	88.	12.8
(person-mSv)	(8.1±2.0)E-02	(3.5±0.6)E-01	(4.3±0.6)E-01	-	-	-	
II. PRESSURIZED WATER REACTORS							
1. Arkansas Nuclear One (2)	3.9E-04	1.2E-02	1.2E-02	0.	3.2	97.	0.3
2. Beaver Valley (1)	1.5E-02	1.5E-04	1.6E-04	99.	0.1	0.9	92
3. Calvert Cliffs (2)	3.7E-02	3.4E-03	4.0E-02	0.	92.	8.5	6.2
4. Donald C. Cook (2)	9.3E-02	9.4E-04	9.3E-02	98.	0.6	1.0	19.
5. Crystal River (1)	4.2E-01	8.9E-04	4.2E-01	0.	~100.	0.2	2.1
6. Davis-Besse (1)	8.4E-03	2.2E-03	1.1E-02	77.	3.0	20.	1.4
7. Joseph Farley (2)	2.0E-03	3.4E-02	3.6E-02	0.	5.6	94.	0.9
8. Fort-Calhoun (1)	4.8E+00	1.5E-02	4.8E+00	~100	< 0.1	0.[illegible]	79.
9. Robert E. Ginna (1)	7.5E-02	1.7E-02	9.2E-02	81.	0.3	18.	60.
10. Haddem Neck (1)	7.0E-03*	1.4E-01*	1.4E-01*	0. *	4.8*	95. *	42. *
11. Indian Point (3)	4.9E-04	3.9E-02	3.9E-02	0.	1.2	99.	0.45
12. Kewaunee (1)	3.1E-01	1.0E-03	3.1E-01	99.	0.9	0.3	17.
13. Maine Yankee (1)	1.5E-04	2.0E-03	2.1E-03	0.	7.1	93.	25.
14. McGuire (1)	7.3E-02*	2.8E-05*	7.3E-02*	~99. *	~ 1.0*	0.04*	40. *
15. Millstone-2 (1)	6.3E-05	5.5E-02	5.5E-02	0.	0.1	~100.	90.
16. North Anna (2)	1.4E-02	5.9E-03	2.0E-02	0.	70.	30.	0.38
17. Oconee (3)	1.0E+00	1.5E-02	1.0E+00	98.	0.3	1.5	6.6
18. Palisades (1)	2.2E-02	1.1E-03	2.3E-02	87.	8.7	4.5	14.4
19. Point Beach (2)	2.7E-02	9.7E-02	1.2E-01	22.	< 0.1	78.	60.
20. Prairie Island (2)	3.1E-03	3.4E-02	3.7E-02	~ 0.	8.4	92.	89.
21. Rancho Seco (1)	6.9E-04	5.9E-02	6.0E-02	~ 0.	1.1	~100.	2.7
22, H.B. Robinson-2 (1)	1.4E-02	2.4E-03	1.6E-02	0.	89.	11.	0.9
23. Salem (2)	5.3E-03	2.5E-03	7.9E-03	0.	68.	32.	0.9
24. San Onofre (1)	1.0E-02*	-	-	0. *	-	-	1.2 *
25. Sequoyah (1)	4.0E-02	5.2E-04	4.0E-02	99.	0.1	1.3	5.9
26. St. Lucie (1)	6.2E-05	7.6E-02	7.6E-02	0.	< 0.1	~100.	11.
27. Surry (2)	4.5E-03	3.5E-02	4.0E-02	0.	11.	8[illegible]	2.1
28. Three Mile Island (2)	2.1E-03*	1.0E-01*	1.0E-01	2.0*	~ 0.1*	98. *	37. *
29. Trojan (1)	6.6E-05	1.8E-02	1.8E-02	< 0.1	0.3	~100.	7.8
30. Turkey Point 3-4 (2)	1.7E-04	1.2E-04	2.9E-04	0.	59.	41.	0.39
31. Yankee (1)	5.2E-04	1.8E-03	7.0E-03	~ 0.	22.	78.	1.9
32. Zion (2)	6.3E-05	2.1E-02	2.1E-02	0.3	< 0.1	~100.	6.2
PWR Total (person-rem)	7.0	0.79	7.8	-	-	-	-
(person-mSv)	70	7.9	78	-	-	-	-
PWR Average (±S.E.)[#] (person-rem)	0.25 ± 0.003	(2.0 ± 0.09)E-02	0.26 ± 0.03	30.	20.	50.	19.6
(person-mSv)	2.5 ± 0.03	0.20 ± 0.01	2.6 ± 0.3				
Grand Total (person-rem)	7.1	1.2	8.3	-	-	-	19.4
(person-mSv)	71	12.	83	-	-	-	

*Not included in averages because of atypical design (LaCrosse, Monticello, Big Rock Point, Haddem Neck, San Onofre), non-operation (Three Mile Island), or low Power production (McGuire), but included in total collective dose.

[#] Arithmetic mean and standard error of the collective tritium doses at individual sites. The standard error is calculated from the variations about the mean and does not include the uncertainties which might be inherent in the environmental transport, metabolic and dosimetric models.

resultant cost of $90,000 - $150,000 per site per year is clearly not commensurate with the small collective doses that result from these tritium releases. According to one study[8], even existing controls on tritium production such as the use of zirconium cladding, enriched ^{7}Li, and zirconium tritium "getters" would exceed the NRC's cost-effectiveness criterion (based upon their use only for tritium control). Reduction in liquid effluent releases by evaporation and release as airborne effluents would produce only small dose reductions and would be ranked low in terms of cost-effectiveness. The use of enriched ^{7}Li is most cost-effective (about $5,000 per person-rem reduced)[8] and is currently used.

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TRITIUM

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ENVIRONMENTAL TRITIUM CONTAMINATION FROM INCREASING UTILIZATION OF NUCLEAR ENERGY SOURCES

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Presented by B.J. Mason

Abstract

ENVIRONMENTAL TRITIUM CONTAMINATION FROM INCREASING UTILIZATION OF NUCLEAR ENERGY SOURCES. Tritium is produced in nuclear reactors by ternary fission and by neutron capture reactions in ^{10}B, ^{11}B, ^{6}Li, ^{7}Li, ^{14}N and ^{2}H. The expanding use of nuclear reactors for power production will contribute additional amounts of tritium to the existing background levels due to atmospheric testing of nuclear devices and natural production by cosmic-ray interactions in the upper atmosphere.

Production processes, both in the environment and in nuclear and thermonuclear energy sources are reviewed. US power reactor experience indicates that the primary source of tritium production in boiling light-water reactors is ternary fission, while neutron capture reactions in ^{10}B and ^{7}Li are the principal sources in pressurized-water reactors employing chemical shim. Tritium from deuterium activation in heavy-water moderated reactors far exceeds the production of tritium by ternary fission. Environmental surveillance data show no apparent increase of tritium concentrations in water sources in the vicinity of light-water reactors. Due to tritium retention by the fuel cladding, a major portion of the tritium produced by ternary fission will be released during processing of the spent reactor fuel. Estimated releases from fuel reprocessing plants are compared with discharge data from operating plants.

Forecasts of future tritium production from nuclear reactors are provided up until the year 2000. These estimates are compared with existing equilibrium levels of naturally-occurring tritium and with the decreasing inventory of tritium remaining from atmospheric nuclear testing. The implications of these projections are evaluated from a public health viewpoint. A discussion of the possible impact of new thermonuclear energy sources on environmental tritium concentrations is also included.

1.0 INTRODUCTION

Tritium is a radionuclide on which attention is currently being focused because of its radiological characteristics and its appreciable production by an expanding nuclear power industry. This industry results in tritium releases to the environment as a consequence of normal operation of both nuclear power reactors and fuel reprocessing plants. The purpose of this paper is to discuss the production of tritium by several sources and to examine the environmental consequences of this production relative to the nuclear power industry.

Although tritium is generally considered to be one of the least hazardous radioisotopes (1), its long radioactive half-life (approximately 12 years) means that tritium discharged into the environment will accumulate over a relatively long period of time. Since tritium closely follows the reactions of ordinary hydrogen, it assimilates readily into water and into biological media.

The importance of tritium as an environmental contaminant is related to the development of nuclear power. Figure 1 shows current predictions of the growth of nuclear electricity in the United States and compares it with total generation both in the United States and the world (2). In the United States, the Atomic Energy Commission has estimated that 120-170,000 megawatts of electricity will be generated by nuclear reactors by the year 1980 (3). This estimate will represent about 25% of the total electrical power generated in that year. By the year 2000 the total installed nuclear capacity in the U. S. may be as high as 1,000,000 megawatts (4).

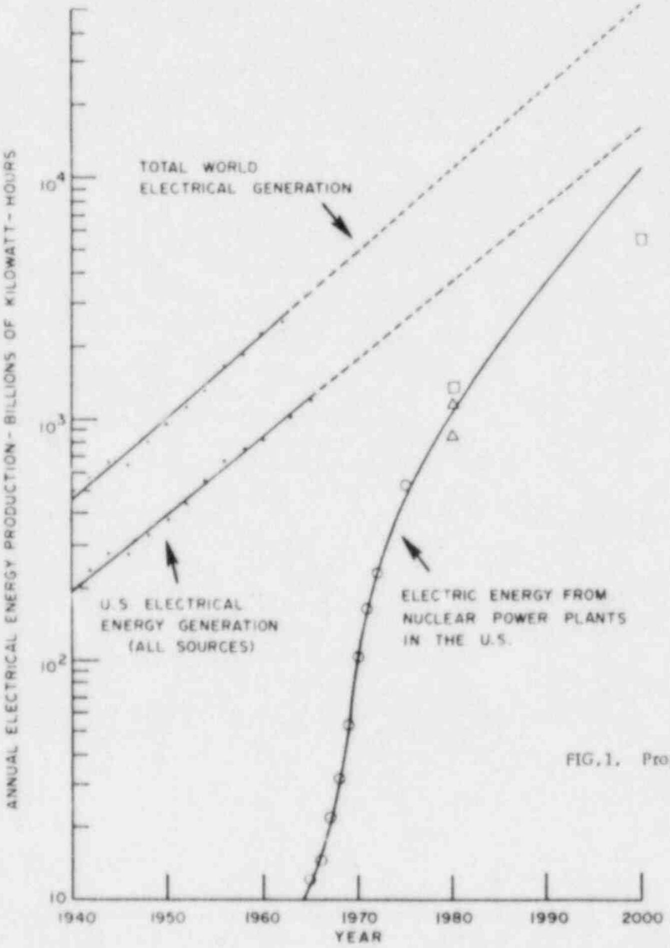

FIG.1. Projected energy requirements [2].

2.0 TRITIUM PRODUCTION IN NUCLEAR REACTORS

Tritium may be produced in nuclear reactors by several mechanisms some of which are shown schematically in Figure 2. These sources include:

1. Ternary fission having a triton as one of the fission fragments;
2. Neutron capture reactions with coolant additives such as boron, lithium and ammonia;

3. Activation of naturally occurring deuterium in light- and heavy-water moderated reactors;
4. Neutron capture reactions with poison material used in control rods and plates.

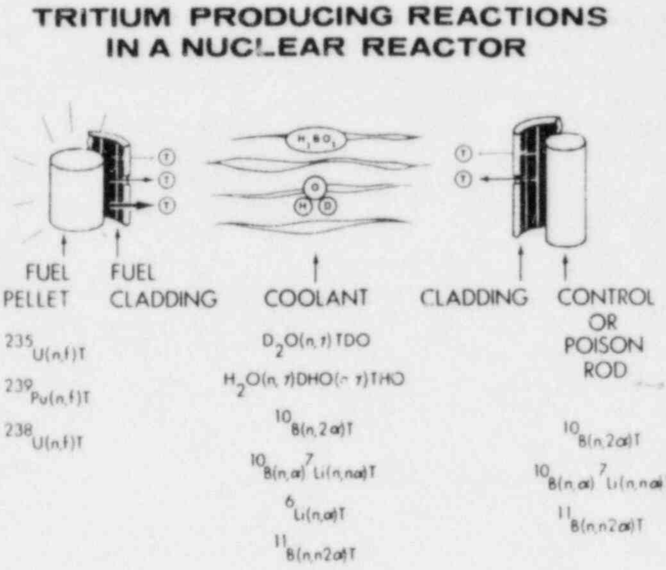

FIG.2. Schematic diagram of tritium production processes.

Other reactions with nitrogen, helium-3 or structural material may also produce tritium but the contribution to the total production rate from these reactions appears to be slight (5, 6). The relative importance of a given reaction mechanism for tritium production will depend upon the reactor type, design characteristics, operating history, and materials of construction. The manner in which tritium is released to the environment will also be a function of these parameters.

2.1 Fission Product Tritium

The discovery of tritium as a fission product was initially reported by Albenesius (7), and has since been verified by several investigators (8, 9). Measured tritium fission yields from thermal neutron induced fission of enriched or natural uranium range from 0.3 to 1.0 tritium atoms per 10^4 fissions (7-9). Theoretical fission yields have been calculated for other materials and for ^{235}U by Dudey whose results are shown in Table I (10). Measurements of the tritium released in fuel reprocessing indicate that the theoretical estimate of 1.3×10^{-4} is the most accurate (11).

Dudey has also estimated the tritium content of various reactor fuels as shown in Table II. Based upon these values we have calculated the tritium production rates shown in the last column of Table II. Since fuel burnups of 30,000 and 40,000 MW(th)-day/tonne have been forecast for replacement fuel loadings in proposed reactors and advanced reactors (3), we have extrapolated production rates for these burnups. These values are also contained in Table II.

TABLE I

ESTIMATED YIELD OF TRITIUM FROM FISSION (10)

Material	Neutron Energy (MeV)	Atoms ^{3}H Produced per Fission
^{235}U	Thermal	1.3×10^{-4}
	1.0	1.2×10^{-4}
	3.0	8.2×10^{-5}
^{238}U	Thermal	2.6×10^{-4}
	2.5	1.4×10^{-4}
^{239}Pu	Thermal	2.3×10^{-4}
	1.0	2.5×10^{-4}

Light-water nuclear reactors produce about 16.4 millicuries of tritium daily per megawatt of thermal energy. Fast reactors should produce more fission-product tritium than thermal reactors since the fission yield of plutonium-239 is higher than uranium-235 (2.5×10^{-4} and 1.3×10^{-4} atoms per fission, respectively) (10). These estimates yield tritium production rates of 18.7 Ci/yr-MW(e) for thermal reactors and 26.2 Ci/yr-MW(e) for plutonium-fueled fast reactors.

2.2 Deuterium Activation

Tritium can be formed by neutron activation of deuterium which represents about 0.015% of the atoms of hydrogen in nature (12). The thermal cross-section for the ^{2}H(n,γ)^{3}H reaction is 0.5 millibarn (13). Deuterium activation has been estimated to produce tritium in a 3295 MW(th) boiling-water reactor at a rate of about 0.15 μCi/sec or 4.45×10^{-3} Ci/MW(e)-yr. By comparison, the tritium production rate from fission was calculated to be 540 μCi/sec (14).

Because of the retention of fission-product tritium in the fuel element, the importance of tritium from deuterium activation as an environmental contaminant will be higher than its relative production rate would indicate. If only 0.1% of the fission-product tritium escaped from the fuel then deuterium activation could contribute over 27% of the activity discharged to the environment from a boiling-water reactor. In a pressurized-water reactor other sources contribute considerably greater amounts of tritium, thus deuterium activation is negligible by comparison.

In heavy-water reactors the moderator and coolant can consist of 99.75% deuterium oxide. For this reason, deuterium activation represents a greater source of tritium than does fission. A comparison of the primary coolant tritium concentration for several types of reactors is shown in Table III. These data show that the tritium activity in heavy-water reactors is considerably greater than in light-water reactors. Based upon the ratio of the deuterium content in light- and heavy-water, the tritium production rate in a heavy-water reactor would be about 30 Ci/MW(e)-yr.

TABLE II

ESTIMATED FISSION-PRODUCT TRITIUM CONTENT IN VARIOUS REACTOR FUEL MATERIALS (10)

Reactor Type	Fuel Material	Fuel Burnup Rate (MW(th)-day/Tonne)	Tritium Content (Ci/Tonne)	Tritium Production Rate* (mCi/MW(th)-day)
Light-Water	^{235}U	20,000	327	16.4
		30,000	490*	
Light-Water	^{239}U	20,000	540	27.0
		30,000	810*	
Light-Water	1/2 U- 1/2 Pu fuel	20,000	440	27.0
		40,000	880*	
LMFBR**	^{235}U	100,000	1510	15.1
LMFBR	^{239}Pu	100,000	3150	31.5
LMFBR	^{239}Pu-^{235}Pu core and blanket	40,000	1900	47.5

* Estimated by the Authors

** LMFBR - Liquid Metal Fast Breeder Reactor

TABLE III

MEASURED TRITIUM CONCENTRATIONS IN REACTOR PRIMARY COOLANT WATER (20)

Reactor Type	Tritium Concentration μCi/ml
Boiling Water	0.015
Pressurized Water	1.5
Heavy Water	7200

2.3 Tritium from Boron Reactions

Boron-10 is used in some nuclear reactor control elements due to its large neutron absorption cross-section of 3840 barns (13). Both natural boron which contains 19.7% boron-10 and boron which is enriched in this isotope may be used. Boron carbide (B_4C) is usually used in boiling-water reactors (BWR) and some BWR plants in operation may contain over 20 kilograms of boron-10 in this form. Additional boron may be used as a burnable poison in the initial cores of BWRs to offset the buildup of neutron absorbing fission-products. The amount of boron-10 present in this form could be about 24 kilograms in a 1000 MW(e) reactor. This boron, in the form of borated stainless steel curtains, is removed after the initial fuel cycle burnup when fission-product poisons are near equilibrium in the reactor core.

Pressurized-water reactors (PWR) do not routinely employ control rods having an appreciable amount of boron in them and usually use an alloy of silver, cadmium and indium for the neutron absorber. Boron may be present,

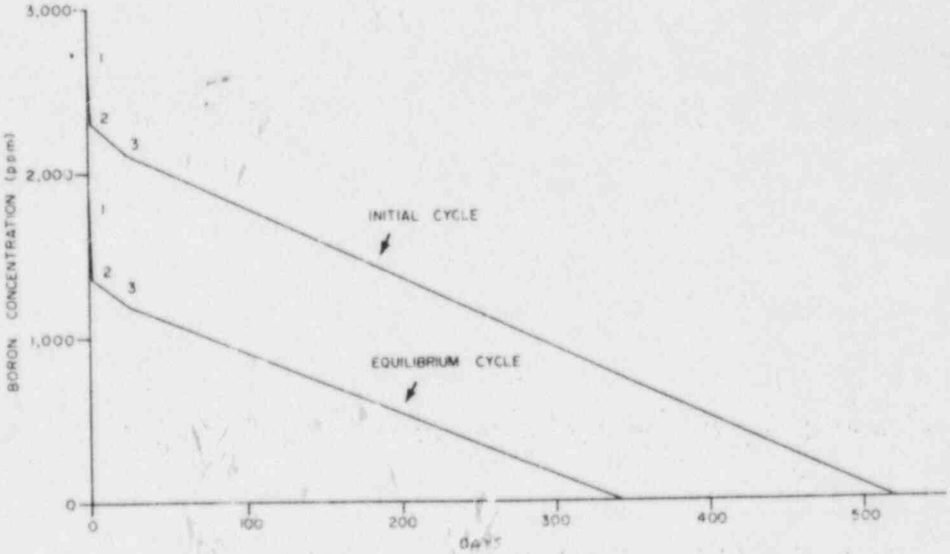

FIG.3. Variation of boron concentration over the fuel lifetime in a pressurized water reactor [16]. The regions indicated correspond to major changes in the removal rate.

TABLE IV

ESTIMATED TRITIUM PRODUCTION FROM BORON REACTIONS IN A 3391 MW(th) PRESSURIZED WATER REACTOR(17)

Source	Amount of ^{10}B (grams)	Reaction	Annual Tritium Production (Curies)	
			Initial Cycle	Equilibrium Cycle
Poison Plates	$\sim$3530	$^{10}B(n,2\alpha)^3H$	800	Not Used
		$^{10}B(n,\alpha)^7Li$ $^7Li(n.n\ \alpha)^3H$	1500	Not Used
Chemical Shim	$\sim$3400 (initial) $\sim$3150 (equilibrium)	$^{10}B(n,2\alpha)^3H$	1100	780
		$^{10}B(n,\alpha)^7Li$ $^7Li(n,n\ \alpha)^3H$	not estimated ($\sim$2100)*	not estimated ($\sim$1450)*

* Estimated by authors from ratio of production in poison curtains.

however, in the form of boric acid dissolved in the coolant (chemical shim) or as poison plates in the initial core loading. The chemical shim is used to provide a more uniform power distribution throughout the reactor core and to compensate for long-term changes in reactivity such as the depletion of uranium or the accumulation of fission-product poisons (15). The boron concentration is not maintained constant but decreases over the operating life of the fuel as shown in Figure 3.

Tritium may be formed directly from boron-10 by the $^{10}B(n,2\alpha)^3H$ reaction or by the $^{10}B(n,\alpha)^7Li(n,n\ \alpha)^3H$ reactions. Estimates of the tritium production rate for a PWR employing chemical shim are shown in Table IV. Based upon the production of 2300 curies per year from the 3.4 Kg of boron-10 in the poison plates we estimate that the 20 Kg of boron-10 in a 700 MW(th) BWR would produce 13,000 curies of tritium annually, or about 21,500 curies of tritium per year by a 1000 MW(th) BWR plant. Smith has estimated a production rate of 20,000 curies of tritium per year for a comparable plant (18).

2.4 Tritium from Lithium

Natural lithium contains 7.42 percent of ^6Li which has a thermal cross-section of 675 barns for tritium production (13). One part per million of natural lithium as an impurity in the cladding and primary coolant of a small reactor could yield approximately 50 percent as much tritium as produced by fission (6). Lithium may be intentionally added to the coolant in the form of LiOH to control the acidity of the primary system. It may also be present from ion exchange resins in Li^+ form. The importance of lithium as a source of tritium is vividly depicted in Figure 4 which shows tritium discharges from the Shippingport Atomic Power Station in the United States. The tritium discharges were significantly reduced when resin containing only ^7Li was substituted for natural lithium (19).

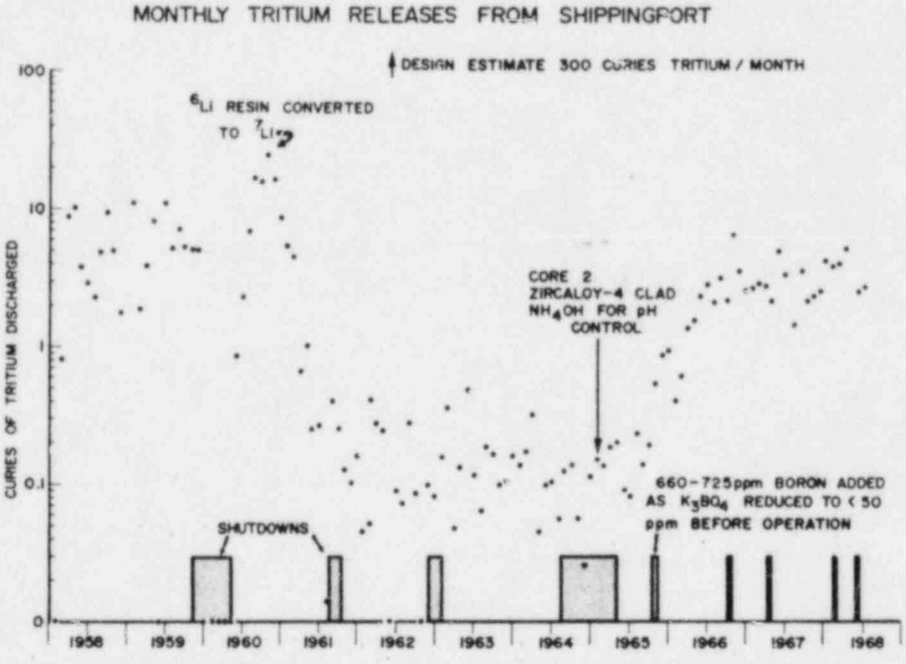

FIG.4. Tritium releases from a pressurized water reactor. The sharp decrease in the amount released after natural lithium was removed is evident [19].

Present reactor designs will employ 99.9% ^7Li in the ion exchange resins and LiOH in order to minimize tritium production (17).

3.0 TRITIUM RELEASES FROM NUCLEAR REACTORS

3.1 Retention of Fission-Product Tritium

Only a fraction of the fission-product tritium is released to the environment from a nuclear reactor. In order to reach the reactor coolant the tritium would have to diffuse out of the ceramic uranium dioxide (UO_2) fuel pellet and pass through the metallic fuel cladding as shown in Figure 2. For this reason, the bulk of the tritium is retained in the fuel elements and is not released until the elements reach the fuel reprocessing plant. Small amounts of tritium may also be produced from traces of uranium on the outer surfaces of the cladding.

Although the purpose of the metallic cladding around the fuel pellets is to contain fission products, the retention mechanisms for tritium are not well defined. These mechanisms may involve chemical reactions between tritium and the cladding material as well as physical diffusion. Three mechanisms have been proposed for the transmission of tritium through fuel cladding:

1. Direct penetration of tritons (6);
2. Effusion of tritium through minute holes or defects in the cladding material;
3. Diffusion of tritium atoms around grain boundaries and through intact cladding.

Releases of tritium by direct penetration of the cladding by tritons has been shown to be negligible (6), thus this mechanism is not as important as releases by effusion and diffusion processes.

The composition of the fuel cladding has been found to have an appreciable effect on the amount of tritium retained in the fuel (18, 20). Measurements of the tritium concentration in the coolant of boiling-water reactors indicate that the tritium release rate is approximately an order of magnitude greater in reactors employing stainless steel fuels than in those employing zirconium alloy (zircaloy) clad elements (18, 20). A limited amount of experience with zircaloy-clad fuels also indicates that there may be a three-fold difference in hydrogen retention between the two zirconium alloys, zircaloy-2 and zircaloy-4 (21).

3.2 Operating Experience

The majority of the tritium in the coolant, either from fission or boron and lithium reactions, is released to the environment with the liquid waste discharge. Table V shows amounts of tritium released from several pressurized-water (PWR) and boiling-water reactors (BWR) in the United States. The appreciable difference between the releases from PWR's and BWR's is believed to be due to the presence of boron in the primary coolant of the PWR (18, 20). Tritium release rates and tritium concentrations in the coolant of the boiling-water reactors indicate that boron present in cladded control rods or alloyed in poison curtains does not contribute appreciable amounts of tritium to the effluent (20).

Liquid wastes from reactors are discharged into the condenser cooling water which provides a large amount of dilution. For typical 1000 MW(e) PWR plants releasing 5000 Ci/yr of tritium the resultant concentrations in these discharges which reach the general public would be less than 0.2% of the radioactivity concentration guide (17). Reactors which use cooling towers

TABLE V

TRITIUM RELEASED TO THE ENVIRONMENT

FROM OPERATING POWER REACTORS

Reactor	Fuel Cladding	Power MW(e)(22)	Capacity Factor(22)	Tritium Release Estimated Annual Curies (22)	 Rate Ci/MW(e)-yr
<u>Boiling Water Reactors</u>					
Big Rock Point	Stainless steel	50	0.45	20	0.89
Humboldt Bay	Stainless steel, Zircaloy	52	0.80	20	0.50
Elk River	Stainless steel	24	0.70	10	0.60
Dresden - I	Zircaloy - 2	200	0.65	5-10	0.037-0.074
<u>Pressurized Water Reactors</u>					
Indian Point - I	Stainless steel	163	0.50	500	6.1
Yankee	Stainless steel	185	0.70	1300	10.0
Connecticut Yankee	Stainless steel	483	0.71[1]	1755[1]	5.1

[1] data from Connecticut Yankee monthly operating reports for 1968 (23).

TABLE VI

ESTIMATED TRITIUM RELEASES FROM U. S. COMMERCIAL

NUCLEAR FUEL REPROCESSING PLANTS

Plant	Capacity (Tonnes/day) Design	Actual	Estimated Fuel Burnup (MWd/Tonne)	Tritium Content of Fuel (Ci/Tonne)	Design Annual Average Release Rate From Stack (Ci/day)	
Nuclear Fuel Services, West Valley, New York	1	1	22,000	200	50 130**	(37)
Midwest Fuel* Recovery Plant Morris, Illinois	1.4	0.82	NS	200	432	(38)
Barnwell Nuclear* Fuel Plant, Aiken, South Carolina	6	5	35,000 40,000	400 500	1,640	(39)

* Under construction
** Released as liquid waste to storage lagoon
NS NOT SPECIFIED

have a smaller dilution capacity than those that use once-through condenser cooling. Under certain conditions extra care in discharging tritium may be required to insure that concentration limits are not exceeded.

Atmospheric release of tritium from light-water reactors with gaseous wastes has not been extensively studied but does not appear to be appreciable. The Bureau's Nuclear Engineering Laboratory has made some measurements at a BWR which indicate that atmospheric tritium releases may be less than 0.5 curies per year. By comparison, proposed BWR's are using a hypothetical estimate (based on considerable fuel cladding defects) of approximately 0.03 to 30 Ci/yr (24-27). Gaseous tritium releases from the Yankee pressurized-water reactor are less than 100 Ci/yr (28), or approximately 6% of the total tritium discharged from the plant.

In the United States, there has not been any development of large heavy-water reactors because of the availability of enriched uranium. Canada and several other countries, however, use this concept because natural uranium can be used without enrichment (29). Tritium concentrations in the primary coolant of heavy-water reactors could be about 10-20 mCi/g after long-term operation. Past experience at heavy-water reactors indicates that D_2O losses can approach several Kg/day although there is an important economic incentive to retain this material (30-35). Holmquist has stated that no heavy-water reactor can be assumed to have a lower D_2O leakage rate than 2-3%/yr or 6 Kg/day (36). The production reactors at the Savannah River Plant in the United States are estimated to release between 1,000 and 10,000 curies of tritium per month to the atmosphere from a D_2O leakage rate of 2-3%/yr (36). Based upon the leakage experienced at Canada's 200 MW(e) Douglas Point Nuclear Station (4 Kg/day) (34), a heavy-water reactor could release over 15,000 curies of tritium per year to the environment.

4.0 TRITIUM RELEASES FROM FUEL REPROCESSING PLANTS

The largest localized concentrations of tritium in the environment will be in the vicinity of fuel reprocessing plants where 68-99.9% of the fission-product tritium will be available for release when the fuel elements are processed. Table VI lists estimated tritium discharges for three commercial fuel reprocessing plants in the United States (37-39).

Several possibilities exist for the disposal of tritium from fuel reprocessing (40). These are as follows:

1. Disposal into ground water,
2. Dilution and disposal directly to surface water,
3. Distillation and release to the atmosphere, and
4. Concentration and storage with high level waste.

4.1 Ground Water Disposal

Disposal into ground water through seepage basins or wells has been pursued for several years at the major U. S. plutonium production sites. Disposal into ground water requires hydrological and geological studies to determine the direction and rate of ground water movement and extensive monitoring programs to assure that off-site drinking water supplies are not contaminated.

4.2 Surface Water Disposal

The Nuclear Fuel Services commercial reprocessing plant at West Valley, New York disposes of tritium by a combination of surface water releases and atmospheric releases. Release rates of tritium from this plant have been estimated to be 50 Ci/day up the stack and 130 Ci/day to storage lagoons which are eventually pumped into surface waters (37). These estimated releases are considerably higher than measured discharges as shown in Table VII.

TABLE VII

ANNUAL TRITIUM RELEASES IN LIQUID WASTE FROM

A COMMERCIAL FUEL REPROCESSING PLANT (41)

Data for 1967

Measured Tritium Releases to Storage Lagoon (Ci)	4200
Tonnes of Uranium Processed	136
Percent of Plant Capacity	37
Percent of Estimated Release of 130 Ci/day	9
Percent of Estimated Release Corrected for Capacity	25

Surface water disposal requires considerable dilution and therefore is practical only for sites on large rivers. Blomeke has estimated that a least 18 million gallons of water per day would be required per ton of fuel in order to dilute the released tritium to 10^{-3} μCi/ml or less (40). For this reason this technique has limited applicability for plants with capacities on the order of 5 MTU/day.

4.3 Atmospheric Disposal

The other two commercial fuel reprocessing plants shown in Table VI are under construction in the United States. Both plants will discharge only limited volumes of liquid waste thus the major means of environmental tritium release will be from atmospheric discharges (38-39). The estimated atmospheric tritium releases from these plants are shown in Table VI to be 432 and 1,640 Ci/day. The annual average off-site air concentrations for these plants are estimated to be less than 2% of the radiation concentration guide of 2×10^{-7} μCi/ml (38-39). Cowser, et.al., analyzed the dose that would be received from atmospheric releases from a plant reprocessing fuel equivalent to 100,000 MW(th) of reactor operation. They found that the annual dose received from released HTO would be about 7 mrem/yr (42).

4.4 High-Level Waste Storage

Retention of tritium in high-level waste storage tanks is dependent on the process used. Plants that use the Purex process produce between 10,000 and 100,000 gallons of liquid waste per ton of fuel (43). The Nuclear Fuel Services plant, which uses the Purex process, is estimated to retain about 10% of the tritium in long-term storage. The rest is disposed to surface water (about 65%) or the atmosphere (about 25%) principally because of the large volume of liquid generated (37). Plants that use the fluoride volatility process, produce approximately 25-250 liters of liquid waste per ton of fuel which may have tritium concentrations of 1-10 Ci/l (43). Tank storage might be economically feasible for wastes of this activity.

5.0 TRITIUM ACCUMULATION IN BODIES OF WATER

5.1 General

All tritium, regardless of its mode of production or release, eventually deposits in the hydrosphere. Localized sources of environmental tritium such as reactors and fuel reprocessing plants may present certain public health considerations because of exposure resulting from accumulation in receiving waters. In general, these considerations are minor for streams which have a fast flushing time and sufficient dilution capability to keep concentrations low. This distinction, however, is not as apparent for facilities on lakes or impoundments where slow flushing times may result in a buildup of tritium.

In the United States some power reactors are located on a small lake or a stream which has been impounded to produce a lake large enough to provide sufficient cooling water. In some cases, several plants may be sited on a single large lake such as Lake Michigan. Tritium buildup for these two situations was examined in detail.

5.2 Tritium Buildup in Lakes

The tritium accumulated in a lake can be estimated by a model which assumes a constant outflow rate and a constant volume. The equation which governs this process is:

$$C(t) = \frac{R}{V\Lambda}\left[1 - e^{-\Lambda t}\right] + C_o e^{-\Lambda t} \tag{1}$$

where,

$C(t)$ = the concentration (μCi/cm^3) at time t,
R = the activity release rate (Ci/yr) into a lake,
r = the rate of water loss from a lake (cm^3/yr),
V = the volume of the lake (cm^3),
λ = the radioactive decay constant for tritium ($\lambda = 0.0561$ yr^{-1}),
C_o = the initial activity present in the lake from natural and fallout sources, and
Λ = $\lambda + r/V$, the effective removal constant for both radioactive decay and the physical loss of water from the lake.

The average or mean residence time of water in the lake is defined by:

$$\tau_w = V/r \tag{2}$$

while the mean residence time for tritium in the lake is given by:

$$\tau_T = 1/\Lambda \tag{3}$$

5.3 Tritium Buildup in Small Lakes

An American power company is constructing a 700 MW(e) PWR nuclear power plant on a small lake created by impounding a natural creek, a site that is reasonably typical for the United States. The volume of the lake is approximately 1.34×10^9 cubic feet and the average discharge rate is 169 ft^3/sec. Annual discharges of tritium are estimated to be about 3820 Ci/yr. It was assumed that the plant would operate 40 years and that this would be equivalent to an infinite operating time, a conservative assumption.

Tritium buildup for three conditions was examined using the model represented in equation (1). The results of these calculations for these three conditions are shown in Table VIII. It is apparent that for realistic conditions (cases I and II) accumulated tritium concentrations would not exceed established concentration guidelines. Case III represents a hypothetical maximum estimate. All calculations were conservative because water loss by evaporation was omitted.

5.4 Tritium Accumulation in a Large Lake

Large lakes have a considerable dilution capacity for tritium released to them. However, this capacity may not be sufficient if several nuclear power plants are located on one lake. Lake Michigan, one of the Great Lakes in the United States is a good example of this situation. As shown in Figure 5 there will be seven nuclear power sites on the Lake by 1973 comprising some ten reactors, or about 7030 MW(e). The total U. S. generating capacity provided by nuclear power is scheduled to be 45,600 MW(e) by 1973; thus about 15% of this total will be located on Lake Michigan. Most of this capacity will be furnished by PWR's, a situation not typical of the national trend which tends to be an even distribution between BWR and PWR plants. The tritium release rate into Lake Michigan is assumed to be 5 Ci/yr-MW(e) because of this preponderence of PWR plants. This value was obtained using the average release shown in Table V and an average generating capacity of 70 percent of the design value.

Estimates of nuclear generating capacity on Lake Michigan are shown in Figure 6. The nuclear capacity in the United States in the year 2000 will be about 1,000,000 MW(e); about 18.75% of this capacity is assumed to be located on Lake Michigan.

The water retention time in Lake Michigan has been estimated by Rainey (44) and Stigall (45) to be 30.5 years and 75 years, respectively. The mean-life of tritium is 17.8 years, which is short enough to offset variations in these values, consequently the mean residence time for tritium in the Lake only varies between 11.2 and 14.5 years. The latter value was used in the projections of tritium concentrations for Lake Michigan that follow.

The activity of tritium in the Lake at time t, A(t), is given by:

$$A(t) = A(t-1)e^{-\Lambda} + \frac{RP(t)}{\Lambda} (1-e^{-\Lambda}) \qquad (4)$$

where Λ is the removal constant, R is the tritium release rate (R = 5 Ci/MW(e)-yr), V is the lake volume, and P(t) is the anticipated power generation rate (MWe) estimated at midyear intervals. The first term accounts for the decay and removal of tritium produced during the previous year; the second term represents tritium produced during the year under consideration. The total tritium activity in the Lake in the year 2000 is estimated by this equation to be about seven megacuries.

TABLE VIII

TRITIUM ACCUMULATION IN A SMALL MAN-MADE LAKE FROM A SINGLE NUCLEAR PLANT

Case	Outflow Rate (ft^3/sec)	Volume (ft^3)	Mean Residence Time Water Days	 Tritium Days	Estimated Equilibrium Tritium Concentration (μCi/ml)
I. Annual Average	169	1.34×10^9	92	90	7.1×10^{-5}
II. Low Flow	21	1.34×10^9	740	664	1.8×10^{-4}
III. No Outflow	0	1.34×10^9	∞	6510	1.8×10^{-3}

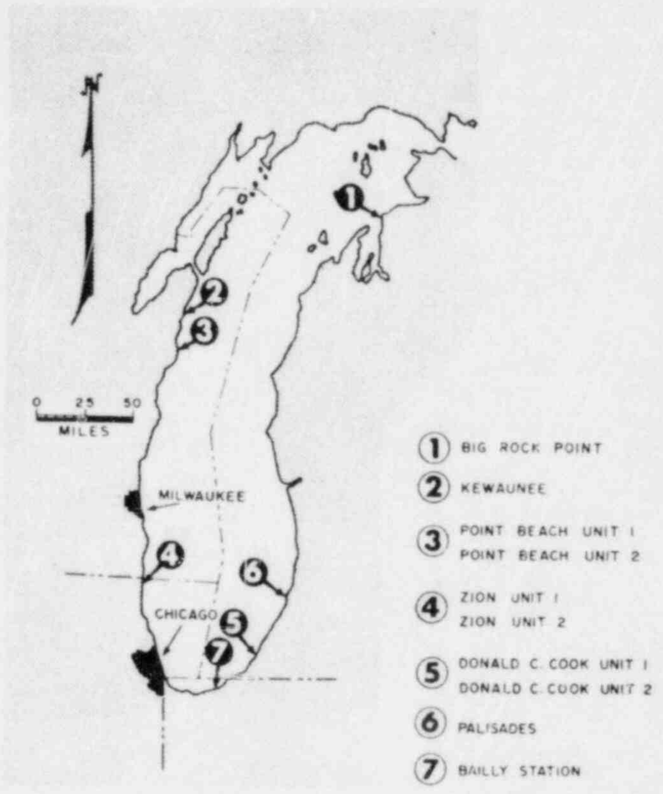

FIG.5. Nuclear power reactors at present planned for operation on Lake Michigan.

Tritium buildup in the Lake was calculated using equation (4) to account for a varying production rate due to increases in nuclear generating capacity. The concentration was calculated from:

$$C(t) = A(t)/kV \qquad (5)$$

where $C(t)$ is the average tritium concentration in the Lake, k is the fraction of the Lake volume available for dilution, and V is the total volume of the Lake.

The resulting tritium accumulation in Lake Michigan was determined by equation (5) for two conditions: 1) mixing with the total lake volume of 4781 km^3, and 2) mixing with 36.5 percent of the lake volume ($\sim$1780 km^3), a value estimated by Stigall from fallout data on ^{90}Sr as the effective dilution volume. The results of this calculation are shown in Figure 7. These data show that although the estimated tritium activity accumulated in the Lake by year 2000 is over seven megacuries, the concentration in the Lake would be less than 0.2% of the radioactivity concentration limit of 3×10^{-3} μCi/ml used by the United States Atomic Energy Commission (46).

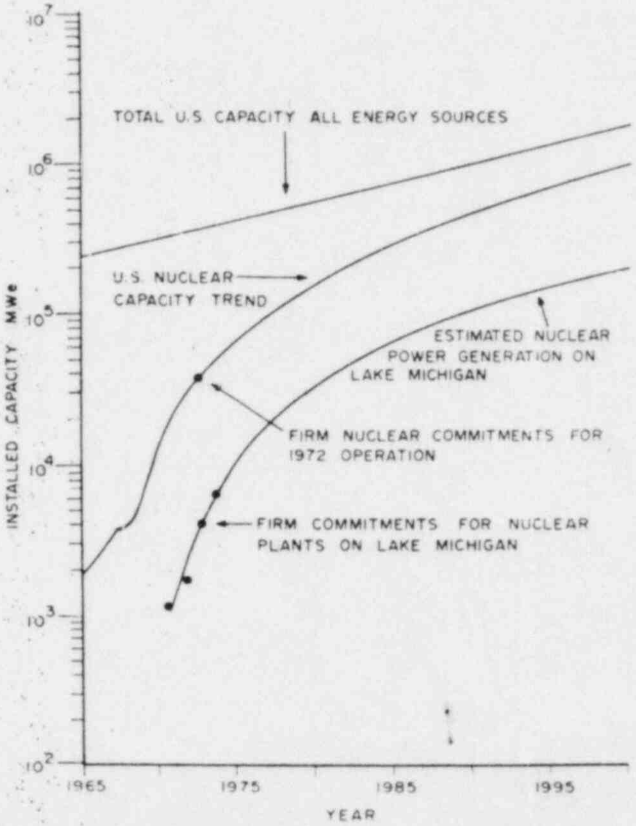

FIG.6. Projected power generating capacity from nuclear reactors discharging into Lake Michigan in comparison with total US power projections [4].

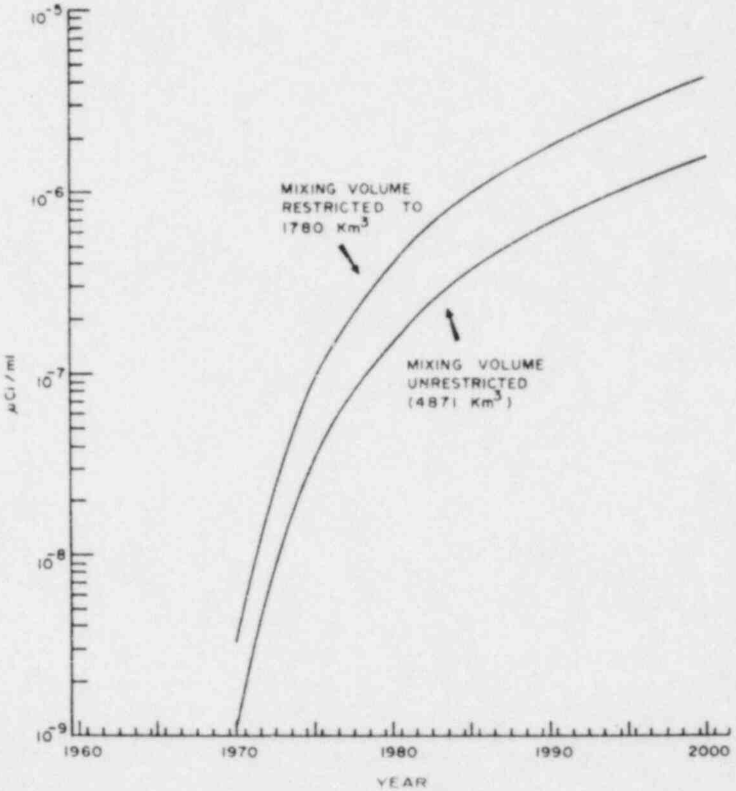

FIG.7. Estimates of future tritium concentration in Lake Michigan from nuclear reactor operation.
These values are in addition to tritium from fall-out and natural sources.

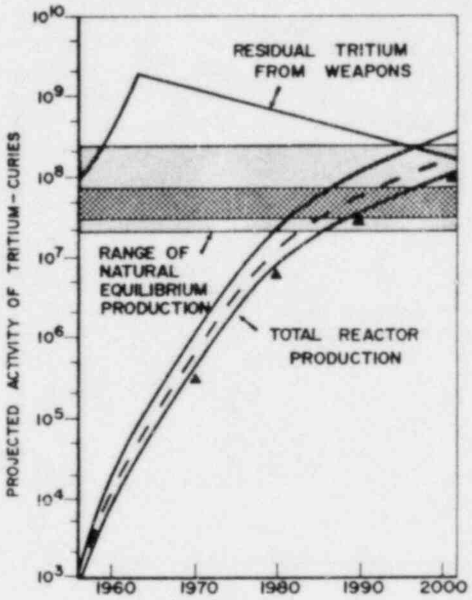

FIG.8. Comparison of tritium activity from: (a) natural production (wide range = all estimates; narrow range = most probable estimate); (b) residual weapons fall-out; (c) US reactor production; and (d) world-wide reactor production (▲) estimated in Ref.[56].

6.0 PROJECTED ENVIRONMENTAL TRITIUM LEVELS

6.1 General

The buildup of tritium in the environment from the nuclear power industry will be superimposed on tritium already present from natural production processes and from atmospheric testing of thermonuclear weapons. The relative contribution by the power industry in the United States is compared to these other sources in Figure 8. At present the nuclear power industry production is small compared to the other two sources and will not reach natural production levels until about 1985. The major source of tritium in the environment is from past nuclear weapons tests. This will continue to be the case up until about 1995 at which point reactor production will become the predominant source if the present trend continues and there are no further thermonuclear explosions.

6.2 Tritium Production in Nature

The production of tritium in nature is caused by cosmic-ray interactions with nitrogen and oxygen in the upper portion of the atmosphere. The principal reactions which produce tritium are high energy (E >100 MeV) proton spallation reactions and $^{14}N(n,t)^{12}C$ and $^{16}O(n,t)^{14}N$ reactions with secondary neutrons from cosmic radiation (47-52).

Nir, et.al., reviewed all previous calculations of tritium production and estimated that the average production rate is 0.19 ±0.09 ^3H atoms/cm^2-sec and that the total decay rate is 0.5 ± 0.3 ^3H atoms/cm^2-sec (50). This latter value corresponds to a total equilibrium activity inventory of approximately 69 megacuries of tritium. Estimates of the rate of production vary considerably, ranging from 0.09 - 2.0 ^{3}H atoms/cm^{2}-sec. This variation corresponds to equilibrium levels of 12 and 275 megacuries, respectively, as shown in Figure 8. Other natural processes such as spontaneous fission in thorium and uranium and reactions with boron, lithium, deuterium and helium-3 also produce tritium but the production rate is insignificant compared to the oxygen and nitrogen reactions (51-52).

6.3 Tritium from Nuclear Testing

Tritium is formed from fission in atomic weapons at a rate of 0.7 curies per kiloton of TNT explosive yield. Small amounts of tritium may also be formed by neutron interactions with nitrogen and oxygen (53). The largest source of tritium in the environment at the present time is from the detonation of thermonuclear (fusion) weapons in which tritium was produced by $^{2}H(n,\gamma)^{3}H$ and $^{6}Li(n,\alpha)^{3}H$ reactions with lithium deuteride in the devices. The yield of these reactions is estimated to be 6.7 megacuries per megaton equivalent of TNT (54). Nuclear testing has contributed about 1700 megacuries of tritium to the environment (55). This contribution is considerably higher than the equilibrium value from natural production which is about 69 megacuries (see Figure 8).

6.4 Tritium from Nuclear Energy

Projections of the environmental inventory from nuclear energy sources are dependent upon forecasts of power consumption rates and developments in reactor technology as indicated by the curves in Figure 8. Cowser's estimate (▲) of worldwide reactor production (56) is lower than the total U. S. estimate (2) shown by the three curves in the Figure. The curves of U. S. production in Figure 8 represent the contribution from fission (lower curve), fission plus soluble boron in PWR reactors (middle curve), and fission plus total boron in all reactors (upper curve). Boron control rods and curtains should not represent an appreciable source of tritium in the environment since the fraction of tritium which escapes from encapsulated boron is small and these elements are generally disposed intact as high-level solid waste.

An increase in the utilization of large heavy-water power reactors could substantially increase environmental tritium levels. It has been estimated that the tritium inventory would be doubled if 10% of the power production was from heavy-water reactors (57). Development of thermonuclear power sources could also increase tritium levels in the environment. The tritium production rate from a thermonuclear reactor has been estimated to be over 100,000 times greater than in a fission reactor (57); however, most of this material would be contained in the reactor for use as fuel material. Present information is insufficient to project future estimates of tritium from this source.

7.0 PUBLIC HEALTH SIGNIFICANCE

The principal form of tritium in reactors is tritiated water and conventional concentration processes such as ion exchange and evaporation do not remove appreciable amounts of tritium from liquid effluents. For this reason tritium may comprise between 50 and almost 100% of the total activity in reactor discharges to the environment (20). The Radioactivity Concentration Guide (RCG) for tritium, however, is much higher than for most common fission products. The RCG of tritium is 10^{-3} μCi/ml and because of its short biological half-life (12 days), its low disintegration energy, and uniform distribution in the body, its relative health significance is smaller than for other fission products such as ^{90}Sr and ^{137}Cs.

The estimated tritium release from a 1000 MW(e) pressurized-water reactor is approximately 7000 curies per year (average of Table V). Plants employing single-pass condenser systems to remove excess heat have circulation rates of about 1.5×10^{15} cm^3 per year and the resulting concentration would be about 5×10^{-6} μCi/ml or 0.5% of the RCG for exposure of the general population (10^{-3} μCi/ml). The volume of water discharged from reactors employing cooling towers is considerably smaller and may be only 2000 gallons per minute. Under these circumstances the effluent concentration could approach 20% of the guideline value. Under certain circumstances it may be necessary to restrict discharges or to provide supplementary dilution for these plants.

The Public Health Service operates a tritium surveillance network in the United States to provide information on nationwide trends and tritium concentrations in water downstream from large nuclear installations. The results reported by this network are published periodically (59-62). Present levels are 0.2-10 nCi/l and the dose contribution from the maximum concentration has been estimated to be less than 1.6 mrem/year (62).

The buildup of tritium in small confined bodies of water may approach concentrations approximately 20% of the RCG which suggests that environmental surveillance would be indicated to monitor this accumulation. Our predictions for Lake Michigan show that the tritium concentrations in the Lake will be about 40 nCi/l, most of which will be contributed by the reactors on the Lake. Continuous consumption of this water for a fifty-year period would result in a total integrated dose of only 350 mrem, which is considerably less than that from natural radiation sources.

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DISCUSSION

J. TADMOR: I should like to ask whether the figures presenting the projection of tritium production in nuclear reactors up to the year 2000 include possible future fusion reactors.

A recent study made by F.L. Parker indicates that tritium waste from reactors of this type would present a major problem from a disposal point of view.

Another remark I would like to make concerns the possibility of tritium release into the atmosphere following the distillation of liquid waste. One of the problems encountered in this method of disposal is fog formation due to the large amounts of water distilled, and the consequent deposition of tritium in the neighbourhood of the release point. Have you any comment to make on this problem of fog formation?

B.J. MASON: In answer to your first question, I do not believe that the possibility of the development of fusion reactors has been taken into consideration. It would certainly not seem to be the case in the light of Mr. Parker's paper.

With regard to the fog question, I have no comment to make, but I have raised the matter of a 'rain-out' effect which I believe to be related to fog in that either process would result in fairly high levels of tritium in local environments. Apparently no consideration has been given to the possibility of such localized concentrations.

S.O.W. BERGSTRÖM: A tritium release of 15 000 Ci/yr from heavy-water reactors seems to be an upper limit. The Ågesta heat and power reactor has been operating for a long time with a tritium leakage of some 100 Ci/yr. This corresponds to 5-10 Ci/MW(e)/yr, a rate which it should be possible to reduce in the case of larger reactors. The dominating source of environmental tritium is therefore likely to be fractions other than those from deuterium activation in cooling water. In the power plants the higher water leakage rate from the light-water reactor results in tritium levels which may in fact be in excess of those which will be caused by the heavy-water type.

B.J. MASON: 15 000 Ci/yr was an estimate of what could result from the heavy-water reactor. The data used for the estimate are those available from one particular Canadian plant.

J.K. MIETTINEN: I understand that when calculating the radiation dose values for humans you used the biological half-life (12 days) given by the ICRP for tritiated water and a homogeneous distribution in the body.

In nature, in a lake containing 40 nCi T_2O/litre, part of this tritium will be present in plankton and fish, for example in the form of tritiated amino acids, and will be taken up as such by people consuming fish. It will then be built up into nucleic acids in the body and will certainly have a much longer biological half-life and give a genetic dose several orders of magnitude higher than if homogeneously distributed in the body water.

Have you calculated the contribution of this biologically bound tritium to the radiation doses received by human beings?

B.J. MASON: The authors have not made this calculation since data are not readily available. Evans, at the Savannah River plant, has indicated that the dose calculations based upon ICRP recommendations should be increased by a factor of 1.4 because of tissue-bound tritium. I agree with you that some effort must be made to take this tissue-bound tritium into consideration.

A. LAFONTAINE: In studying the problem of the radiological capacity of rivers and lakes in respect of tritium provision must be made for an adequate safety factor that allows for changes (i.e. an increase in strictness) in the maximum permissible concentrations recommended by the ICRP.

The biological cycle of tritium does in fact appear to indicate that it does not behave solely as an element of tritiated water, but that it can be concentrated in certain elements of the biological cycle, as has been indicated by Mr. Miettinen. Moreover, the distribution of tritium in the body is not as regular as was believed.

Account should also be taken of the possibility of tritium release partly in the form of molecules different from tritiated water, notably in fuel reprocessing plants.

B.J. MASON: All I can say is that I agree with you.

TABLE IV

ESTIMATED TRITIUM PRODUCTION FROM BORON REACTIONS IN
A 3391 MW(th) PRESSURIZED WATER REACTOR(17)

Source	Amount of ^{10}B (grams)	Reaction	Annual Tritium Production (Curies)	
			Initial Cycle	Equilibrium Cycle
Poison Plates	~3530	$^{10}B(n,2\alpha)^3H$	800(510)	Not Used
		$^{10}B(n,\alpha)^7Li$ $^7Li(n,n\alpha)^3H$	1500(960)	Not Used
Chemical Shim	~3400 (initial) ~3150 (equilibrium)	$^{10}B(n,2\alpha)^3H$	1100(346)	780(480)
		$^{10}B(n,\alpha)^7Li$* $^7Li(n,n\alpha)^3H$* $^6Li(n,\alpha)^3H$**	8.8 ~~not estimated~~ 8.8 ~~(~2100)*~~	8.8 ~~not estimated~~ 8.8 ~~(~1450)*~~

~~* Estimated by authors from ratio of production in poison curtains.~~

* ^7Li concentration controlled to less than 2.2ppm

**^6Li impurity in 99.9% ^7Li

NOTE: Westinghouse has advised the authors that a design change is presently being made which will reduce tritium production to the values shown in parenthesis in the table. These values would also change the text in Sec. 2.3

Table VI

Plant	Capacity (Tonnes/day) Design	Actual
Nuclear Fuel Services, West Valley, New York	1	0.82

Table VII

Percent of Plant Capacity	45
Percent of Estimated Release Corrected for Capacity	20

Section 4.3 Atmospheric Disposal, last line

annual dose received from released HTO would be about 9 mrem/yr (42).

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Tritium in the Environment From Nuclear Powerplants

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TRITIUM, an isotope of hydrogen with an atomic mass number of three, is produced in nuclear reactors in substantial quantities. Although tritium is one of the least hazardous radioactive nuclides, its continued production and long half-life for radioactive decay may lead to increased levels in the environment. Because tritium is an isotope of hydrogen, it can be metabolized in the form of tritiated water and incorporated into body fluids and tissues. This source of population exposure requires that public health agencies be cognizant of the significance of tritium as an environmental contaminant.

The nuclear power industry has expanded rapidly during the past few years, and the U.S. Atomic Energy Commission predicts that this high rate of growth will continue during the next decade. Nuclear power reactors and fuel reprocessing plants release tritium to the environment under normal operating conditions.

Due to the stratospheric fallout from previous atmospheric testing of nuclear weapons, measurable levels of tritium are already present in the environment. An analysis of the effect of an expanding nuclear power industry on environmental tritium levels indicates that future tritium releases from reactors and spent fuel processing plants may surpass the quantity of tritium remaining from nuclear weapons tests.

The Bureau of Radiological Health's environmental surveillance activities provide a continuing source of data on tritium concentration in rivers on which nuclear facilities are located. There is, however, a need for more specific source-oriented information on tritium, and its potential for becoming a public health problem. This paper is intended as a general information guide on tritium for persons engaged in Federal, State, county, and local public health activities. The various sources and mechanisms for release of tritium and its pathway from nuclear facilities to the environment are described. This information is essential in assessing the requirement for environmental surveillance and in evaluating, from a public health viewpoint, the upward trend of tritium levels in the environment.

Radiological Characteristics of Tritium

Because of its relatively short biological half-time (12 days) combined with a relatively low disintegration energy, tritium is one of the least hazardous radionuclides produced in nuclear reactors. A single ingestion of tritiated water having an activity of 1 microcurie will produce a total dose to the body tissues of 0.21 millirems (mrem). Continuous ingestion of

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water having a specific activity of 1 microcurie of tritium per milliliter will produce a dose rate of 170 rem per year to body tissues. From this value the annual average discharge concentration limit for tritium in water can be calculated to be 3×10^{-3} microcuries per milliliter. This quantity is the concentration limit for tritium in radioactive waste discharges to the environment contained in the Code of Federal Regulations (*1*).

Tritium decays by the emission of a beta particle and an anti-neutrino to form stable helium-3. The half-life for this process is 4,500 days or approximately 12 years (*2*). The beta particle and the neutrino carry varying fractions of the energy liberated by the disintegration. When there are many tritium atoms present, this process produces a continuous spectrum of beta particle energies.

The maximum energy of the beta emitted by tritium is 18.6 kilo-electron volts (Kev) and the average energy is 5.6 Kev. These energies are considerably lower than those of most other beta emitters which are usually about 100 times greater.

Sources of Environmental Tritium

Naturally occurring tritium. Tritium is one of the three isotopes of hydrogen. Normal hydrogen consists of approximately 99.9 percent protium (hydrogen-1) and 0.015 percent of deuterium (hydrogen-2), although variations in isotopic concentration can exist in natural sources. Unlike protium and deuterium, tritium (hydrogen-3) is unstable because of radioactive decay and occurs naturally only in trace amounts.

Naturally occurring tritium is formed principally by the interaction of cosmic radiation with oxygen and nitrogen atoms in the upper atmosphere. Tritium produced by cosmic-ray interactions prior to 1952 contributed to environmental concentrations ranging between 16 to 35 picocuries of tritium per liter of water or 5 to 10 tritium units (*3*) where 1 tritium unit is equivalent to 1 atom of tritium per 10^{18} atoms of normal hydrogen. Because of a constant rate of production, the tritium from natural processes will accumulate until an equilibrium condition is reached. This condition is achieved when the rate of removal of tritium from the environment by radioactive decay equals the rate of tritium production. Since the earth is several billion years old, this equilibrium rate has existed for some time and the amount of tritium present from natural sources is essentially constant.

Nuclear weapons tests. Past tests of nuclear weapons in the atmosphere are the greatest single source of tritium present in the environment today. Tests by both the United States and the Union of Soviet Socialist Republics, before the Atmospheric Test Ban Treaty in 1962, greatly increased the amount of tritium in the environment. Thermonuclear (fusion) weapons produce tritium from neutron interactions with lithium and deuterium with a production rate of 6.7 megacuries of tritium per megaton of fusion yield (*4*). This source contributed about 1,700 megacuries of tritium to the environment compared with an equilibrium value from natural sources equivalent to approximately 69 megacuries.

A large fraction of the tritium produced by these detonations was injected into the stratosphere. This part of the atmosphere does not mix rapidly with the lower portion of the atmosphere; therefore, the tritium is introduced into the troposphere over many years. Tritium in the lower atmosphere is rapidly removed by precipitation. Tritium removed from the atmosphere by precipitation will accumulate in the surface layers of the oceans and inland waters. Rainfall that follows periods of nuclear testing has several hundred times the tritium content normally present from natural tritium. The estimated time for these processes to remove half of the tritiated water from the lower atmosphere is 35 to 40 days (*3*).

Tritium From Nuclear Reactors

Tritium may be produced in nuclear reactors by the following five mechanisms: (*a*) fissioning of uranium, (*b*) neutron capture reactions with boron and lithium added to the reactor coolant, (*c*) neutron capture reactions with boron in control rods, (*d*) activation of deuterium (hydrogen-2) in water, and (*e*) high energy neutron capture reactions with structural materials.

The relative magnitude of tritium production by these sources is influenced by the reactor type, operating history, design characteristics, and materials of construction. The amount of tritium and manner in which it is released to the environment will also be affected by these parameters.

During the processing of spent reactor fuel, tritium, which has been produced in the fuel rods, is released to the environment and may be a significant source of environmental tritium contamination. A detailed discussion of this source, however, is not within the scope of this report.

Fission-product tritium. During the fission process the uranium nucleus usually splits into two more or less equal fragments plus several neutrons. About once in every 10,000 fissions, however, the nucleus is split into three portions (ternary fission) one of which may be a tritium nucleus. It requires 3.1×10^{16} fissions per second to produce a power level of 1 megawatt of thermal energy (Mwt).

Most nuclear powerplants generate electricity at a rate of approximately one-third of the thermal energy production rate. Therefore, approximately 10^{17} fissions per second are required to produce 1 megawatt of electrical energy (Mwe). This production rate corresponds to the production of about 10^{13} atoms of tritium per second or 50 millicuries of tritium per day for each megawatt of electrical energy generated. This tritium normally remains in the fuel unless it diffuses through the cladding material or a leak occurs.

A second source of fission-product tritium in nuclear reactors is due to traces of uranium on the outer side of fuel elements which remain from the fuel fabrication process. This "tramp" uranium may be only a few micrograms per square inch of fuel surface, but because of the large surface area provided by the many fuel rods in a reactor it can produce detectable fission-product concentrations in the coolant.

The reactor fuel is usually uranium oxide and is contained in tubes made of stainless steel or an alloy of zirconium (zircaloy). The primary function of the cladding material is to prevent the escape of fission products from the fuel element. These fission products would otherwise leak from the fuel and contaminate the primary coolant. Sometimes, because of a lack of uniformity in manufacturing, damage during shipment or handling, or as a result of unequal corrosion rates or temperature gradients, the cladding develops pinhole failures or defects through which fission products such as tritium may escape.

All new water-cooled power reactors being built in the United States are using zirconium-clad fuels; however, several older reactors use stainless steel clad fuel. Stainless steel is being replaced by zircaloy cladding because of the increased corrosion resistance and more favorable nuclear properties.

Experience has shown that the fraction of tritium escaping from fuel which is clad with zircaloy-2 is significantly less than that escaping from stainless steel clad fuels. This loss of tritium may be due to fewer defects in zirconium cladding or the ability of zirconium to combine chemically with hydrogen, thus limiting diffusion through the cladding and, consequently, limiting the release of tritium to the coolant.

Tritium produced by activation. Tritium is produced in water reactors by the activation of the naturally occurring deuterium present in the cooling water. Because of the small amount of deuterium in water and the low probability of its capturing a neutron to form tritium, this source of tritium in light water reactors is insignificant. In heavy water reactors which are cooled or moderated with deuterium oxide (D_2O), however, activation is the major source of tritium. Heavy water reactors for power production are not used in the United States at the present time but may be developed at some time in the future. There are, however, several heavy-water plutonium production reactors and small heavy-water moderated research reactors in operation.

A comparison of the tritium concentrations in the primary coolant of light and heavy water reactors is shown in the following table:

Reactor type	*Tritium concentration (microcurie per milliliter)*
Heavy water	7200
Pressurized water reactor	1.5
Do	.14
Do	.044
Boiling water reactor	.015

SOURCE: reference 5.

Tritium is also the product of neutron capture reactions in boron-10, boron-11, lithium-7 and lithium-6. Lithium-7 is usually formed by the $^{10}B(n,\alpha)^{7}Li$ reaction when boron is used in nuclear reactors for reactivity control.

In the past, lithium-6 was used in the coolant for pH control or in coolant purification demineralizers. This use of lithium-6 resulted in the production of elevated tritium concentrations in the primary coolant.

The use of lithium-6 has been abandoned for this reason and replaced by ammonia or lithium-7. Tritium may also be formed by the $^{14}N(n,T)^{12}C$ reaction with ammonia in the coolant or nitrogen in the containment atmosphere. The magnitude of these sources has not been fully assessed.

In boiling water reactors (BWR) boron is used for control in the form of boron carbide either as cruciform control rods or as absorber plates (curtains). Weaver and Harward (*6*) describe the differences in design between this reactor and a pressurized reactor. The rods are movable and function to control the power level of the reactor. Boron absorber curtains are generally used during the first fuel cycle to compensate for the lack of buildup of neutron absorbing fission products (poisons) and are then removed.

Both the absorber plates and the fuel rods are clad in similar fashion so that tritium which is formed from the boron must escape through the cladding by the same mechanism as the fission-product tritium.

Pressurized water reactors (PWR) use boron for reactivity control in the form of boric acid dissolved in the primary coolant. This chemical shim is used to obtain a more uniform power distribution and to compensate for long term changes in fission-product poison buildup, uranium burnup, and changes in neutron flux levels. The initial concentration of boron ranges from several hundred to several thousand ppm and decreases during the operating life of the fuel. Table 1 shows estimated tritium production rates in a typical 1,000 Mwt light water reactor for various irradiation times (7).

The production of tritium can also occur in high temperature gas-cooled reactors by ternary fission and by activation of helium-3 found in trace amounts in the helium coolant. The following table shows the estimated tritium production rates in a 120 Mwt air-cooled reactor (*5*).

Source	*Production rate (curies per year)*
Ternary fission	605
^{3}He activation in coolant	91
Total	696

SOURCE: reference 5.

Tritium release mechanisms. The tritium concentration in the primary coolant system is strongly dependent upon the coolant makeup rate or residence time of the coolant in the reactor. The effect of the average residence time of the coolant in the reactor system on coolant tritium concentrations is shown for boiling water reactors in table 2 and for pressurized water reactors in table 3. In both tables, the contribution of fission-product tritium is shown as a function of the fraction of the tritium which passes through the fuel cladding. The tritium produced by activation of the boron encased in the control rods of a boiling water reactor (BWR) has been assumed to remain within the rod and does not contribute to tritium levels in the coolant (7). The tritium level from the use of boron in a PWR varies with the boron concentration in the coolant and consequently shows a considerable range as shown in table 3.

Table 4 shows tritium levels in the primary coolant of several operating light water reactors. These measurements were obtained by the staff of the Northeastern Radiological Health Lab-

Table 1. Tritium production rates in a 1,000 Mwt light water nuclear reactor [1]

Irradiation time	Tritium produced (curies)		
	Activation of deuterium in coolant	Ternary fission	Boron and lithium reactions [2]
1 day	0.0003	13	50
1 week	.002	90	400
1 month	.008	400	1,600
1 year	.09	5,000	20,000

[1] Reference 7.

[2] These estimates represent upper limit production rates assuming boron control rods, boron chemical shim, and lithium-6 purification resin.

Table 2. Calculated tritium concentrations in the coolant of a 1,000 Mwt boiling water reactor [1]

Residence time of coolant	Coolant tritium concentrations (µCi per liter)				
	From deuterium activation	From ternary fission [2]			
		100 percent	10 percent	1 percent	0.1 percent
1 day	0.002	70	7	0.7	0.07
1 week	.01	500	50	5	.5
1 month	.04	2,000	200	20	2
1 year	.5	30,000	3,000	300	30

[1] Adapted from table II, reference 7.

[2] As a function of percent tritium leakage through cladding.

Table 3. Calculated tritium concentrations in the coolant of a 1,000 Mwt pressurized water reactor [1]

Residence time of coolant	Coolant tritium concentrations (µCi per liter)					
	From deuterium activation	From ternary fission [2]				From boron in coolant [3]
		100 percent	10 percent	1 percent	0.1 percent	
1 day	0.002	70	7	0.7	0.07	3-300
1 week	.01	500	50	5	.5	200-2,000
1 month	.04	2,000	200	20	2	800-8,000
1 year	.5	30,000	3,000	300	30	10,000-100,000

[1] Adapted from table II, reference 7.

[2] As a function of percent tritium leakage through cladding.

[3] Varies with boron concentration which decreases with time since last refueling.

oratory of the Bureau of Radiological Health. If the data for the three boiling water reactors are compared with the calculated tritium concentrations shown in table 2 for a residence time of 1 month, it can be estimated that approximately 1.0 percent of the fission-product tritium diffused through the stainless steel cladding, whereas only about 0.1 percent diffused through the zirconium cladding.

A similar comparison of the primary coolant tritium activities in PWR plants shown in table 5 with the calculated values in table 3 indicates that boron in the coolant appears to be the predominant source of tritium in the primary water. Based on a residence time of approximately 1 month, the only other source which could produce the observed levels would have to be leakage of almost all the fission-product tritium from the fuel. There is no apparent basis for assuming a higher tritium leak rate from the fuel of a pressurized water reactor than that estimated for a boiling water reactor.

Discharges From Nuclear Powerplants

Tritium which has entered the coolant of a nuclear reactor can reach the environment by several pathways. The number of pathways available and the relative amount of tritium discharged by each route depend upon the reactor design and reactor type.

Boiling water reactors. In a boiling water reactor the primary coolant flows through the reactor core, is converted to dry steam and then is fed to the turbine generator to produce power. The coolant is returned to the reactor through a condenser which removes the excess heat still remaining in the steam. The condenser cooling water and the reactor coolant are in close proximity in the condenser. This interface is maintained under a vacuum so that any leakage would be into the reactor and not to the environment.

Air ejectors on the condenser remove the oxygen and hydrogen produced by the dissociation of water in the reactor, air which has leaked into the condenser, and residual water vapor. The air and noncondensable gases are discharged to the plant stack through the off-gas system. Tritium in the form of elemental hydrogen gas may be discharged through the air ejector but a major part of the tritiated water vapor will be removed by condensation. Because of the long radiological half-life of tritium, its concentration in the gaseous stack effluent is not reduced by the 30-minute delay of waste gases in the system.

Pressurized water reactors. A pressurized water reactor consists of two coolant systems. The primary system conducts superheated water through the reactor core to steam generators and returns it to the reactor. The steam generator consists of rows of U-shaped tubes through which the heated primary coolant passes. Surrounding the tubes is the secondary coolant.

Because of a pressure difference between the primary and secondary system (2,000 pounds per square inch in the primary as opposed to about 1,000 pounds per square inch in the secondary) it is possible to convert the secondary coolant into steam. The secondary system then follows a path similar to the primary coolant of a BWR going through turbines and the condenser.

Table 4. Tritium in operating power reactors [1]

Reactor and type	Power level (Mwt)	Type of fuel cladding	Boron used in coolant	Tritium concentration μCi per liter		
				Primary coolant	Secondary coolant	Discharge canal water
A. Pressurized water reactor	600	Stainless steel	Yes	1,560	0.41	0.024
B. Pressurized water reactor	585	do	No [2]	2-44	(3)	.007
C. Pressurized water reactor	80	do	No [4]	145	(3)	(3)
D. Pressurized water reactor	50	Aluminum	No	116	.45	.019
E. Boiling water reactor	256	Stainless steel	No	8.8	(5)	.009
F. Boiling water reactor	240	do	No	13.3	(5)	.002
G. Boiling water reactor	700	Zirconium	No	[6] 1.6	(5)	[6] .0006

[1] Data from Northeastern Radiological Health Laboratory, Bureau of Radiological Health.

[2] Converted in late 1965 to a stainless steel fuel with full chemical shim (boron in coolant).

[3] Not reported.

[4] Boron is used for shutdown but not during routine operation.

[5] Not applicable.

[6] Data from Nuclear Engineering Laboratory, Bureau of Radiological Health.

NOTE: J. C. Drobinski, Jr., and E. J. Troianello of the Bureau's Northeastern Radiological Health Laboratory analyzed the tritium samples.

Table 5. Liquid radioactive waste discharges from operating power reactors [1]

Reactor and type [2]	Total annual discharge (curies)		Condenser cooling water available for dilution (gallons per minute)	Fraction of AEC discharge limit concentrations (percent) [3]	
	Gross activity less tritium	Tritium		Gross activity [4]	Tritium [5]
A. Pressurized water reactor	0.01	1,300	14,000	0.001-0.13	0.16
B. Pressurized water reactor	11.1	500	260,000	.03-4.7	.03
E. Boiling water reactor	.01	10	28,000	.0002-.05	.006
F. Boiling water reactor	1.3	20	100,000	.3-1.2	.0035
G. Boiling water reactor	4.1	5-10	167,000	.3-3.8	.001

[1] Reference 8.

[2] Corresponds to reactor designations in table 4.

[3] Reference 1.

[4] Applicable limit for continuous discharge for mixtures for radionuclides averaged over 12 months was 10^{-6}μCi per ml.

[5] Applicable limit is 3×10^{-3}μCi per ml.

An important difference between boiling and pressurized water reactors for consideration of tritium releases is that in a PWR, leakage between the primary and secondary coolants must occur before the secondary system will be contaminated by tritium or other fission products. As shown in table 4, the tritium concentration in the secondary coolant of pressurized water reactors is only a small fraction of the primary coolant concentration. Thus, a much smaller amount of the tritium will be discharged in the form of elemental hydrogen through the condenser air ejector of a PWR as compared with a BWR.

Release of gaseous tritium. The amount of tritium discharged from both boiling and pressurized water reactors in a gaseous form is only about 1 percent of the total tritium discharge (*7*). Most of the tritium is released as tritiated water together with the liquid radioactive waste. In both types of reactors, a small part of the primary coolant water is continually drawn off, depressurized, cooled, and purified to remove fission and activated corrosion products.

The coolant stream is purified by filtration and demineralization which remove insoluble and soluble radionuclides. Gaseous activity, mainly noble gases and coolant activation products, is removed during coolant depressurization and then transferred to the gaseous radioactive waste processing system. Traces of tritiated water vapor and gaseous tritium are released from the coolant during this process.

Release of tritium in liquid wastes. Because tritium is in the form of tritiated water, it is not removed from the coolant by either filtration or ion-exchange. The purified water containing tritium may be transferred back to the reactor coolant system, stored for future use, or transferred to the liquid radioactive waste processing system.

The primary coolant is not continuously discharged to the waste disposal system. Leakage of the coolant from reactor coolant pumps and other components, however, can occur. This waste will be collected by the plant drainage system which drains to the liquid holdup tanks of the waste processing system.

The largest discharge of primary coolant to the waste treatment facility usually occurs after the reactor has been shut down for fuel reloading or maintenance. During fuel loading, water is added to the reactor cavity to provide radiation shielding and heat removal for the spent fuel elements as they are transferred from the reactor to the spent fuel storage pit.

The refueling water can mix freely with the reactor coolant and before startup this excess water must be drained from the reactor vessel. This excess water represents a considerable volume of the liquid processed by the radioactive waste system. It also contains considerable boron and, therefore, leads to additional tritium production after startup of the reactor. Liquid displaced by expansion of the reactor coolant as the plant power level increases during startup is also carried over to the waste processing system.

If the radioactivity level is sufficiently low, as determined by radiochemical analysis, the purified coolant may be discharged to the environment with the condenser cooling water. Liquid radioactive wastes consisting of purified reactor coolant, laundry wastes, and leakage from pumps and valves are stored in holding tanks before treatment. Treatment processes may include filtration, demineralization, or evaporation.

Although these processes are effective in reducing the concentration of other radionuclides by several orders of magnitude, generally they have no effect on tritium removal. The tritium is discharged to the condenser cooling water at almost the same concentration as in the reactor coolant except for a small amount of dilution by nontritiated wastes. The effluent concentrations are much lower, however, because of the large amount of dilution provided by the condenser cooling water.

Table 5 shows annual liquid radioactive waste discharges for several operating nuclear reactors. Tritium may comprise between 50 and almost 100 percent of the total amount of radioactive material discharged as liquid waste. Tritium discharge concentrations, however, are usually much less than 1 percent of the discharge limit as specified by the Code of Federal Regulations (*1*).

Tritium Detection

Tritium is undetectable by conventional methods of gross radioactivity analysis. Because tritium is usually in the form of tritiated water, it is usually evaporated during the procedures used to prepare water samples for radioactivity analysis. In addition, because of the low energy of the beta particle emitted during the decay of tritium, it is not detected by conventional gross beta counting techniques or in-line plant monitors.

As a result of these limitations, special analytical techniques such as liquid scintillation counting must be used to measure tritium.

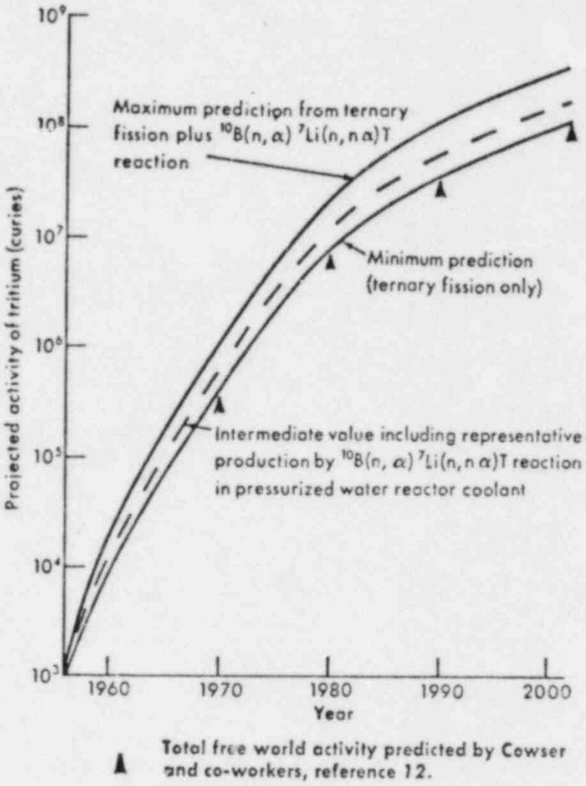
Limits for the discharge of unidentified mixtures of radionuclides cannot be correctly applied to tritium and due to the lesser biological hazard resulting from tritium, would be unnecessarily restrictive. The analytical techniques used to measure the activity of these mixtures usually include gross beta analysis, and since this technique is insensitive to tritium it would not be detected. Tritium may be separated from most other radionuclides by distillation of tritiated water. Tritium analysis can be made on the distillate using liquid scintillation counting. Conventional gross beta counting techniques can be used for the residue. The discharge limit can be calculated by ratioing the measured concentrations to the appropriate discharge limits ($3\times10^{-3}\mu$Ci per ml. for tritium and $3\times10^{-8}\mu$Ci per ml. for unidentified activity measured by gross beta analysis) and summing them. The limit $3\times10^{-8}\mu$Ci per ml. may be raised to $10^{-7}\mu$Ci per ml. if ^{129}I, ^{226}Ra, and ^{228}Ra are known to be absent. If the sum of the ratios is less than 1.0, the mixture may be released to the environment. If greater than one, further analysis or dilution is necessary before the liquid can be released.

Tritium Surveillance by BRH

Since 1964, the Radiation Surveillance Branch of the Bureau of Radiological Health has been operating a tritium sampling network which collects weekly samples of water for tritium analysis from 10 locations throughout the United States. Eight stations are downstream from nuclear facilities, and the other two serve to establish baseline levels. Samples from this network are analyzed monthly by the Bureau's Southeastern Radiological Health Laboratory.

Results of the analysis of samples from the tritium sampling network are reported periodically in *Radiological Health Data and Reports* (*9*, *10*). A special project to study tritium in surface waters of the western States is currently being conducted by the Southwestern Radiological Health Laboratory.

Projected cumulative activity of tritium produced in power reactors in the United States



Environmental tritium concentrations reported by the tritium sampling network for 1964–66 ranged between 2×10^{-6} and 2×10^{-5} microcuries per milliliter (2–20 nCi per liter) (*9*, *10*). Population exposure rates from continuous ingestion of this water would range from 0.33 to 3.3 mrem per year. The maximum concentration reported would contribute less than 3 percent of the normal average population exposure rate of 125 mrem per year from all natural sources of radioactivity (*3*).

The results obtained from this sampling network indicated, with one exception, that waters downstream from nuclear facilities did not show any significantly higher tritium concentrations than streams which did not have operating nuclear reactors on them (*9*). The one exception was a plantsite with several operating heavy-water moderated reactors on it. Releases of tritium in the liquid wastes from this plant did elevate tritium concentrations in the adjacent river above background levels. Tritium concentrations, however, amounted to less than 1 percent of the Atomic Energy Commission's discharge limit of $3 \times 10^{-3} \mu$Ci per ml.

A study by the nuclear engineering department of the University of Cincinnati, under a contract with the Bureau of Radiological Health, is developing projections of future radionuclide inventories based upon predicted growth trends in the nuclear power industry (*11*). The estimated tritium activity produced by increasing utilization of nuclear energy for electric power production forecast from this study is shown in the chart together with estimates made by the Oak Ridge National Laboratory (*12*). Based upon these projected levels, the total tritium produced from nuclear power facilities in the year 2000 will be more than 10^8 curies or approximately 5 percent of the maximum tritium activity present in 1963. At present, tritium releases from operating power reactors are only a small fraction of the discharge concentrations permitted by the Code of Federal Regulations and do not constitute a danger to health. The Public Health Service will continue to monitor the environment in the vicinity of nuclear reactors and fuel reprocessing plants and will evaluate any buildup of tritium in terms of a future hazard to health.

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TRITIUM PRODUCTION, RELEASES AND POPULATION DOSES AT NUCLEAR POWER REACTORS*

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ABSTRACT

Tritium is produced in light-water-cooled reactors as a product of ternary fission and by nuclear reactions with the coolant and with neutron-absorbing materials used for reactor control. Pressurized water reactors (PWRs) have greater amounts of tritium produced in or released into the coolant than boiling water reactors (BWRs). Consequently, tritium releases to the environment from PWRs [29 GBq/MW(e)-y (0.78 Ci/MW(e)-y)] are about 6½ times greater than from BWRs [4.4 GBq/MW(e)-y (0.12 Ci/MW(e)-y)]. Most of the tritium released from PWRs appears in the liquid effluent (about 85%), whereas 75% of the tritium released from BWRs is as airborne effluents. Radiation doses from these tritium releases are small; the average site collective (population) dose in 1981 was less than 0.002 person-sieverts per year (0.2 person-rem/ year). The total collective dose from all tritium releases was 0.08 person-sieverts (8 person-rem).

I. TRITIUM PRODUCTION IN LIGHT-WATER REACTORS

A. Fission

Tritium was identified as a product of the ternary fission of uranium-235 in 1959[1] and subsequently has been identified as a product of the fission of most fissile materials. A nominal fission yield of 1 triton per 10,000 ^{235}U fissions[2] would produce approximately 14 terabecquerels (380 curies) of tritium per metric ton of fuel having a burnup of 30,000 megawatt(t)-days. Fissions occurring in plutonium-239 and uranium-238 raise the total production to about 19 TBq (515 curies) per metric ton of fuel. This results in a total annual production of approximately 600 TBq (16,000 curies) per year for a 3,000 MW(t) [1,000 MW(e)] reactor operating for 300 full-power equivalent days.

The amount of ternary fission-product tritium which is released to the environment depends upon the fraction which escapes from the fuel rod into the reactor coolant. This fraction is affected by the materials used for the fuel rod cladding and its integrity. The zirconium alloy (Zircaloy) fuel rod cladding used in most current light-water reactors exhibits greater retention of tritium than the stainless steel cladding used in early reactors. Stainless steel is highly permeable to tritium. Early boiling water reactors (BWRs) with stainless steel cladding had normalized tritium releases between 0.02 to 0.03 TBq per electrical megawatt-year. (0.5 to 0.8 Ci/MW(e)-y)[3]; whereas a zirconium-clad BWR had tritium releases of 0.0015 to 0.0026 TBq/MW(e)-y (0.04 to 0.07 Ci/MW(e)-y)[3]. This improved retention appears to be due to the formation on the surface of the zirconium cladding of a hydrogen- impermeable oxide layer.[4,5] Approximately half of the total tritium inventory in the fuel may be bound to the cladding[6].

B. Boron Reactions

Boron is a neutron absorber and is used in several forms in LWRs for control.. In General Electric boiling water reactors and Combustion Engineering pressurized water reactors boron carbide (B_4C), is used in moveable control rods. (Most PWRs use AgInCd control rods which are not a source of tritium.) Boron is also used in the form of either a borosilicate glass (Westinghouse) or B_4C (Babcock & Wilcox and Combustion Engineering plants) in fixed "burnable" absorbers to compensate for fuel depletion and fission-product "poison" buildup. Pressurized water reactors also use a soluble boron

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* The views expressed in this work are the personal views of the authors and do not necessarily represent the official position of the Nuclear Regulatory Commission, the Pacific Northwest Laboratory or the Department of Energy.

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compound in the primary coolant to compensate for fuel depletion and, by changing the concentration through removal or addition of boron, to make changes in power levels.

The primary boron reactions which yield tritium are:

$^{10}B\ (n,\alpha)\ ^{7}Li\ (n,\ n\alpha)\ ^{3}H$,

$^{10}B\ (n,\ 2\alpha)\ ^{3}H$, and

$^{11}B(n,\ n2\alpha)^{3}H$.

The ^{10}B reaction yielding ^{7}Li is the primary neutron absorbing reaction having a thermal neutron absorption cross-section of approximately 3.8 E-25 m^2 (3,800 barns). The other two reactions have small cross-sections (less than 1E-30 m^2 or 0.1 barns.) The ^{11}B reaction has a threshold of 14 MeV. It is not an important source of tritium due to the small number of high-energy neutrons present in light-water reactors as well as its small reaction cross-section.

The amount of tritium produced by reactions with boron (B_4C) in the moveable control rods of a 1,000 MW(e) boiling water reactor is relatively large, about 115 TBq (3,100 Ci).[5] Due to differences in the use of the control rods (differences in the time the rods are kept in the reactor core) Combustion Engineering PWRs produce less tritium in the B_4C control rods ~ 33 TBq/y (~ 900 Ci/y).[5] About 37 TBq (1,000 curies) of tritium are produced per year in the fixed absorbers and 11-18.5 TBq (300-500 Ci) of ^{3}H are produced by the boron in the coolant of PWRs[2,3,5,6,8,9].

All of the tritium produced by reactions with soluble boron is produced directly in the coolant. Only about 0.2% of the tritium generated in the B_4C control rods appears to be released to the coolant.[5,7]. Releases from fixed absorbers using B_4C are also less than 1%. Borosilicate glass absorber plates appear to have less retention and releases have been estimated to be between 10 and 50% of the production [5,9].

C. Lithium Reactions

Lithium salts are added to the coolant of pressurized-water reactors to control acidity (pH). In operation the lithium concentration in the coolant varies between 0.2 and 2 parts per million. Natural lithium has an isotopic composition of 7.4% ^{6}Li and 92.6% ^{7}Li. Tritium is produced by neutron capture reactions with both isotopes:

$^{6}Li\ (n,\ \alpha)\ ^{3}H$ and

$^{7}Li\ (n,\ n\alpha)^{3}H$.

The ^{6}Li reaction has a considerably larger neutron absorption cross-section for tritium production than ^{7}Li. The ^{6}Li thermal neutron cross-section is approximately 9.5E-26 m^2 (950 barns).[10] The ^{7}Li reaction has a 3-MeV threshold and a cross-section of about 8.6E-30 m^2 (0.086 barns).

Commercial light-water reactors have employed lithium salts which are enriched to 99.9% ^{7}Li in order to reduce tritium production. The lithium used for pH control yields approximately 0.3 TBq/MW(e)-y (0.01 Ci/MW(e)-y) in an equilibrium fuel cycle.[6]

D. Deuterium Activation

Deuterium (^{2}H or D) occurs naturally, comprising approximately 0.015 per cent of natural hydrogen. Tritium can be formed by neutron activation of deuterium by the reaction:

$$^{2}H\ (n,\ \gamma)^{3}H$$

with a thermal neutron cross-section of 4.6 E-32 m^2 (0.46 millibarns).[10] Although this reaction is the major source of tritium in heavy-water moderated reactors, the low natural abundance of deuterium and its small activation cross-section make this a minor source in light-water reactors with an estimated production of 0.2 TBq (slightly less than 10 curies) per year in a 1,000 MW(e) reactor.[10]

E. Summary of Tritium Production

Estimates of the tritium production rates in 1,000-MW(e) light-water-cooled nuclear power reactors are summarized in Table 1. As indicated in the table, a boiling water reactor is estimated to produce slightly (10%) more tritium than an equivalent pressurized water reactor. However, due to production of tritium from boron in the coolant, pressurized water reactors are estimated to have more tritium available for releases to the environment.

II. TRITIUM RELEASES TO THE ENVIRONMENT

The amount of tritium released to the environment from commercial light-water reactors is highly variable as the data in Table 2 indicate. Typically, pressurized water reactors had tritium releases between 0.01 to 0.055 TBq per electrical megawatt-year (0.3 to 1.5 Ci/MW(e)-y). Boiling water reactors had lower releases 0.74 GBq to 0.013 TBq per MW(e)-year (0.02 to 0.34 Ci/MW(e)-y). This variation reflects not only differences in reactor type, design and materials of construction; but also differences in power generating history and waste management actices at individual sites. The higher ritium release from pressurized water eactors [average: 0.029 TBq/MW(e)-y (0.78 Ci/MW(e)-y)] compared to boiling water reactors [average: 4.4 GBq/MW(e)-y (0.12 Ci/MW(e)-y)] reflects the greater tritium production in and release into the coolant of PWRs. The higher tritium releases through stainless steel clad fuel is evident from the normalized (Ci/MW(e)-y) data for the LaCrosse BWR and the Haddem Neck and San Onofre PWR plants.

The mode of tritium release to the environment as well as the magnitude of activity released is highly variable from site to site and differs between pressurized water and boiling water reactors. The total tritium release to the environment from U.S. BWRs in 1981 was approximately 44 TBq (1,200 curies). Of this total, approximately 10 TBq (280 Ci) were released as liquid effluents and 34 TBq (920 Ci) were released as airborne effluents. Pressurized water reactors released approximately 768 TBq (20,710 Ci) of tritium; 690 TBq (18,600 Ci) as liquid effluents and 78 TBq (2,110 Ci) as discharges to the atmosphere. The activity of tritium per unit energy production released as airborne emissions from the two reactor types is similar (about 3.7 GBq/MW(e)-y (~0.1 Ci/MW(e)-y). However, the airborne releases comprise 80% of the tritium released at a typical BWR site but only about 15% of the tritium released from pressurized water reactors, the remaining 85% being released with liquid effluents.

The release mode for tritium can be modified to accommodate local environmental conditions of the site such as the availability of dilution water for liquid effluents. Thus, it is possible to have BWRs releasing 90% of the tritium via liquid effluents (Oyster Creek) and PWRs releasing over 60% of the tritium via airborne effluents (Rancho Seco).

III. RADIATION DOSES FROM TRITIUM RELEASES TO THE ENVIRONMENT

The models used for the dose calculations have been described elsewhere.[11,12] The total population doses from releases of radionuclides in 1981 have also been published[13]; however, the doses from tritium were not evaluated separately in that report.

A. Methods

The specific activity approach is used to estimate the long-term concentrations of tritium in vegetation, and animal food products. The tritium concentrations in water and in the atmosphere are estimated using established

Table 1. Tritium Production in Light-Water-Cooled Nuclear Power Reactors

[1,000 MW(e) plant operated for 300 full-power equivalent days]

Reactor type and Tritium Source	Amount of Tritium Produced		Amount Released to Reactor Coolant		
	TBq	Ci	TBq	Ci	% of production
Boiling Water Reactor (BWR)					
ternary fission	600	16,000	3.7	100	0.6
encapsulated boron	115	3,100	0.4	~ 10	0.3
deuterium activation	0.4	< 10	0.4	< 10	100
	715	19,100	4.4	120	0.6
Pressurized Water Reactor (PWR)					
ternary fission	600	16,000	3.7	100	0.6
encapsulated boron	37	~ 1,000	11	300	30
boron/lithium in coolant	11-18	300-500	11-18	300-500	100
deuterium activation	0.4	< 10	0.4	< 10	100
	650	17,400	30	800	5

Table 2. Reported Tritium Releases to the Environment in 1981 from Nuclear Power Reactors

1. Reactor Type and Site (number of units)	2. Energy Generation [MW(e)-h](a)	3.	4.	5.	6.	7.	8.	
		TRITIUM RELEASES						
		Activity (curies)(a)			Activity per Electrical Megawatt (Ci/MW(e)-y)(b)			
I. BOILING WATER REACTORS		Liquid	Airborne	Total	Liquid	Airborne	Total	
1. Duane Arnold (1)*	2.2E+06	0.0E+00*	NR*	--*	0.0E+00*	--	--	
2. Big Rock Point (1)*	4.7E+05	3.1E+00*	<1.0E+01*	<1.3E+01*	5.8E-02*	<1.9E-01	<2.4E-01*	
3. Browns Ferry (3)	1.8E+07	2.4E+02	3.8E+01	6.2E+01	1.2E-02	1.8E-02	3.0E-02	
4. Brunswick (2)	5.8E+06	2.3E+01	1.8E+01	4.0E+01	3.4E-02	3.0E-02	6.0E-02	
5. Cooper (1)	3.8E+06	<8.4E+00	4.5E+00	<1.3E+01	1.9E-02	1.0E-02	2.9E-02	
6. Dresden 2-3 (2)	8.6E+06	6.0E+00	3.2E+02	3.2E+02	6.1E-03	3.2E-01	3.3E-01	
7. James Fitzpatrick (1)	4.8E+06	4.1E+00	6.6E+00	1.1E+01	7.5E-03	1.2E-02	2.0E-02	
8. Edwin I. Hatch (2)	7.2E+06	2.1E+01	9.6E+00	3.0E+01	2.5E-02	1.2E-02	3.7E-02	
9. LaCrosse (1)*	2.4E+05*	7.7E+01*	2.3E+01*	1.0E+02	2.8E+00*	2.5E-01*	3.7E+00*	
10. Millstone-1 (1)	2.5E+06	2.6E+00	9.5E+01	9.7E+01	9.1E-03	3.3E-01	3.4E-01	
11. Monticello (1)*	3.3E+06*	4.2E-03*	1.1E+02*	1.1E+02	1.1E-05*	3.0E-01*	3.0E-00*	
12. Nine Mile Point (1)	3.3E+06	5.0E+00	6.3E+01	6.8E+01	1.3E-02	1.7E-01	1.8E-01	
13. Oyster Creek (1)	2.6E+06	2.7E+01	3.2E+00	3.0E+01	2.7E-02	3.2E-03	3.0E-02	
14. Peach Bottom 2-3 (2)	9.8E+06	3.7E+01	2.8E+01	6.5E+01	3.3E-03	2.6E-02	5.9E-02	
15. Pilgrim (1)	3.4E+06	3.4E+01	7.6E+01	1.1E+02	8.7E-02	1.9E-01	2.8E-01	
16. Quad Cities (2)	9.5E+06	1.2E+01	8.6E+01	9.8E+01	1.1E-02	7.9E-02	9.0E-02	
17. Vermont Yankee (1)	3.6E+06	3.7E-01	2.0E+01	2.0E+01	9.1E-04	4.8E-02	4.9E-02	
Mean ± S.E.	[Ci/reactor-y]	10. ± 0.4	38. ± 2.4	48. ± 2.4	0.022 ± 0.002	0.098 ± 0.009	C.12 ± 0.01	[Ci/MW(e)-y]
	[TBq/reactor-y]	0.38 ± 0.02	1.4 ± 0.1	1.8 ± 0.1	0.81 ± 0.06	3.6 ± 0.3	4.4 ± 0.4	[GBq/MW(e)-y]
II. PRESSURIZED WATER REACTORS								
1. Arkansas Nuclear One (2)	9.2E+06	6.9E+02	1.3E+02	8.1E+02	6.5E-01	1.2E-01	7.7E-01	
2. Beaver Valley (1)	4.7E+06	1.4E+02	9.5E-02	1.4E+02	2.6E-01	1.8E-04	2.6E-01	
3. Calvert Cliffs (2)	1.2E+07	1.0E+03	5.8E+00	1.0E+03	7.6E-01	4.4E-03	7.7E-01	
4. Donald C. Cook (2)	1.3E+07	9.2E+02	5.5E+00	9.2E+02	6.1E-01	3.6E-03	6.1E-01	
5. Crystal River (1)	4.0E+06	2.7E+02	1.6E+01	2.9E+02	5.9E-01	3.4E-02	6.3E-01	
6. Davis-Besse (1)	4.4E+06	1.6E+02	8.6E+00	1.7E+02	3.2E-01	1.7E-02	3.3E-01	
7. Joseph Farley (2)	5.5E+06	8.0E+02	1.9E+02	9.9E+02	1.4E+00	3.0E-01	1.6E+00	
8. Fort Calhoun (1)	2.2E+06	2.4E+02	8.7E+01	3.2E+02	1.0E+00	3.3E-01	1.3E+00	
9. Robert E. Ginna (1)	3.3E+06	2.4E+02	7.0E+01	3.1E+02	6.3E-01	1.8E-01	8.2E-01	
10. Haddem Neck (1)	4.1E+06	5.3E+03*	8.6E+01*	5.4E+03*	1.1E+01*	1.9E-01*	1.1E+01*	
11. Indian Point (3)	6.1E+06	8.8E+02	7.9E+00	8.9E+02	1.3E+00	1.1E-02	1.3E+00	
12. Kewaunee (1)	3.8E+06	2.5E+02	3.8E+00	2.6E+02	5.8E-01	8.8E-03	5.9E-01	
13. Maine Yankee (1)	5.2E+06	2.2E+02	4.5E+00	2.2E+02	3.7E-01	7.7E-03	3.8E-01	
14. McGuire (1)	1.9E+04*	6.2E+00*	6.5E-02*	6.3E+01*	2.9E+00*	3.0E-02*	2.9E+00*	
15. Millstone-2 (1)	6.1E+06	3.7E+02	1.4E+02	5.1E+02	5.3E-01	2.0E-01	7.4E-01	
16. North Anna (2)	1.0E+07	1.3E+03	3.1E+01	1.3E+03	1.1E+00	2.7E-02	1.1E+00	
17. Oconee (3)	1.4E+07	5.1E+02	5.8E+01	5.6E+02	3.7E-01	3.7E-02	3.6E-01	
18. Palisades (1)	3.5E+06	2.8E+01	6.4E+00	2.8E+02	7.0E-01	1.6E-02	7.2E-01	
19. Point Beach (2)	6.3E+06	6.5E+02	4.8E+02	1.1E+03	9.0E-01	6.6E-01	1.6E-01	
20. Prairie Island (2)	6.9E+06	5.6E+02	7.4E+01	6.4E+02	7.1E-01	9.0E-02	8.0E-01	
21. Rancho Seco (1)	2.6E+06	8.4E+01	1.4E+02	2.2E+02	2.8E-01	4.7E-01	7.5E-01	
22. H. B. Robinson 2 (1)	3.5E+06	1.9E+02	1.1E+01	2.0E+02	4.7E-01	2.6E-02	4.9E-01	
23. Salem (2)	7.3E+06	1.3E+03	3.9E+00	1.3E+03	1.5E+00	4.0E-03	1.5E+00	
24. San Onofre (1)	7.8E+05	3.0E+02*	1.2E+01*	3.1E+01*	3.3E+00*	1.4E-01*	3.5E+00*	
25. Sequoyah (1)	2.5E+06	7.6E+01	9.2E-01	7.7E+01	2.6E-01	3.2E-03	2.7E-01	
26. St. Lucie (1)	5.0E+06	3.2E+02	3.7E+02	7.0E+02	5.8E-01	6.6E-01	1.2E+00	
27. Surry (2)	7.5E+06	5.3E+02	6.3E+01	5.9E+02	6.2E-01	7.2E-02	6.9E-00	
28. Three Mile Island (2)*	-	7.2E+01*	6.5E+01*	7.3E+01*	-	-	-	
29. Trojan (1)	6.4E+06	1.0E+02	4.0E+01	1.4E+02	1.4E-01	5.5E-02	2.0E-01	
30. Turkey Point 3-4 (2)	5.4E+06	2.0E+02	6.9E-01	2.0E+02	3.2E-01	1.1E-03	3.2E-01	
31. Yankee (1)	8.8E+05	1.0E+02	3.1E+00	1.0E+02	1.0E+00	3.1E-02	1.0E+00	
32. Zion (2)	1.1E+07	8.7E+02	NR	-	6.7E-01	-	-	
Mean ± S.E.	[Ci/reactor-yr]	302 ± 4.	47. ± 2.	350 ± 4.	C.65 ± 0.01	0.12 ± 0.01	0.78 ± 0.02	[Ci/MW(e)-y]
	[TBq/reactor-yr]	11 ± 0.1	1.7 ± 0.07	13 ± 6.2	24 ± 0.4	4.6 ± 0.3	29. ± 0.6	[GBq/MW(e)-y]
III. HIGH-TEMPERATURE GAS -COOLED REACTOR								
Fort St. Vrain (1)	7.6E+05	2.2E+02	1.4E+01	2.3E+02	2.5E-01	1.6E-02	2.7E-01	

* not included in averages because of atypical design (LaCrosse, Monticello, Big Rock Point, Haddem Neck, San Onofre) non-operation (Three Mile Island), or low power production (McGuire) but included in total-release.

(a) Energy generation and activity release data is taken from J. Tichler and C. Benkovitz, "Radioactive Materials Released from Nuclear Power Plants - Annual report" NRC Report NUREG/CR-2907 Vol. 2 (June 1984) for 1981 releases.

(b) Calculated from the reported released activity (columns 3-5) divided by the energy generation (column 2) converted to MW(e)-y.

dispersion models. Tritium concentrations in vegetation and animal food products are determined from the calculated tritium specific activity (S.A.) in atmospheric moisture. The specific activity of atmospheric moisture is calculated as:

$$(S.A.)_{air} = [X/Q\ (r,\theta]Q/H$$

where H is the absolute humidity (taken to be 0.008 kg water/m^3 air) and $[X/Q(r,\theta)]Q$ gives the atmospheric tritium concentration (activity/ m^3) at point r,θ.

The specific activity in vegetation is estimated to be one-half (0.5) of the tritium specific activity in atmospheric moisture. Recently this assumption has been challenged by Murphy[14] who indicated that a value of 0.8 might be more appropriate. Dinner et al.[15] show that this value is dependent upon the type of vegetation; the factor of 0.5 being the mean for vegetation, but a factor of 0.8 would be more appropriate for leafy vegetables. Using the factor of 0.5 and an assumed average vegetation composition of 75% water, the transfer factor from air to vegetation is given by:

$$TF_{air\text{-}to\text{-}plant} = \frac{0.5(0.75\text{kg water/kg plant})}{(0.008\text{kg water/cu. meter air})}$$

$$= 47\ \frac{Bq/kg}{Bq/m^3} = 47\ \frac{m^3}{kg}.$$

Long-term transfer of tritium into meat and milk is calculated using tritium transfer factors which relate the concentration in the food product to the activity ingested by the animal. These factors are:

TF_{milk} = 0.01 Bq/L per Bq/day (=day/L)

TF_{meat} = 0.012 Bq/kg per Bq/day (=day/kg).

Doses are calculated for the average individual (adult) inhaling 8×10^3 m^3 of air per year, drinking 110 L of milk and 370 L of water and consuming 190 kg of produce, 95 kg of meat and poultry, 6.9 kg of fish and 1.0 kg of seafood per year. The whole body dose factors are based upon a quality factor (Q.F.) of 1.7 and are:

2.8×10^{-11} Sv/Bq (1.04×10^{-7} mrem/pCi) for ingestion and

4.3×10^{-11} Sv/Bq (1.58×10^{-7} mrem/pCi) for inhalation.

The population is assumed to be comprised solely of adults but the population distributions and sizes for the individual sites are used.[13]

B. Results

The calculated collective (population) radiation doses resulting from tritium releases from nuclear power reactors are shown in Table 3. The principal parameters that determine this dose are: the activity and mode of tritium releases to the environment, the amount of dilution available from atmospheric dispersion and water bodies receiving liquid effluent discharges, and the size and distribution of the population in the vicinity of the reactor site.

The total collective dose from tritium to the population residing within 80 kilometers (50 miles) of nuclear power reactors is small, 0.083 person-sieverts (8.3 person-rem). Most (93%) of this total is contributed by pressurized water reactors. The highest site collective dose of 0.048 person-sieverts (4.8 person-rem) may be high due to a possible underestimation of the available dilution water flow. This value accounts for almost 60% of the U.S. total collective dose. If this single value were neglected, the total dose at PWR sites would be 0.028 person-sieverts (2.8 person-rem) or 82% of the revised total of 0.034 person-sieverts (3.4 person-rem).

The highest site collective doses, 1.4 ± 0.2 (S.E.) person-millisieverts (0.14±0.02 person-rem) occurred at PWR sites located on lakes and impoundments. Over half (~ 55%) of this dose was received from drinking water ingestion. The smallest doses occurred at boiling water reactor sites also located on lakes or impoundments, (4.7±0.2)E-02 person-millisieverts or (4.7±0.2)E-03 person-rem. However, there was no reported drinking pathway at these sites, the doses being primarily from airborne emissions.

IV. SUMMARY AND NEED FOR ADDITIONAL TRITIUM CONTROLS

The collective dose from tritium released to the environment totalled 0.083 person-sieverts (8.3 person-rem) in 1981. The average collective dose was 4.3E-04 person-sieverts (0.043 person-rem) at BWR sites and 2.6E-03 person-sieverts (0.26 person-rem) at PWR sites. The Nuclear Regulatory Commission has a cost-effectiveness criterion of $1,000 per man-rem reduction ($$10^5$ per person-sievert reduced) for requiring additional radioactive effluent treatment at light-water reactors.[16] Using that criterion, no treatment to retain tritium would be indicated as there are no processes which could reduce tritium releases and cost less than $300 per site. Existing tritium concentration processes (electrolysis, distillation, hydrogen distillation, etc.) would cost between $30-$50 per cubic meter[6] to process the 3,000 m^3 (100,000 ft^3) of coolant or liquid wastes at large power reactors. The

Table 3. Calculated Collective (Population) Whole-body Doses from Tritium Releases from Nuclear Power Reactors in 1981

1. Reactor Type and Site	2. Collective Dose (person-rem)	3.	4.	5. Percentage of Total Tritium Dose from:	6.	7.	8. Percentage of Total Dose Contributed by Tritium
	Liquid Effluents	Airborne Effluents	Total	Drinking Water	Aquatic Foods	Airborne Releases	
I. BOILING WATER REACTORS							
1. Duane Arnold	-	-	-	-	-	-	-
2. Big Rock Point*	3.8E-05*	1.2E-03*	1.25E-03*	0.16*	2.5 *	9.7 *	0.03 *
3. Browns Ferry	6.8E-04	1.0E-02	1.1E-02	5.3	1.0	94.	0.34
4. Brunswick	6.8E-05	1.5E-03	1.5E-03	0	0.4	~100.	0.01
5. Cooper	8.4E-08	3.5E-04	3.5E-04	0	0.02	~100.	2.2
6. Dresden 2-3	0.0E+00	2.8E-01	2.8E-01	0	0	~100.	0.08
7. James Fitzpatrick	3.9E-04	9.3E-04	1.3E-03	29.	0.2	70.	0.02
8. Edwin Hatch	1.2E-04	1.1E-03	1.2E-03	0	10.	90.	0.05
9. La Crosse*	3.4E-04*	4.5E-03*	4.6E-03*	0 *	7 *	93. *	0.085*
10. Millstone-1	4.2E-07	3.8E-02	3.8E-02	0	< 0.01	~100.	96.
11. Monticello*	1.2E-05*	4.9E-02*	4.9E-02*	0.14*	< 0.01	~100.	~100. *
12. Nine Mile Point	2.5E-03	8.9E-03	1.1E-02	22.	~ 0.08	78.	0.18
13. Oyster Creek	2.0E-04	2.0E-03	2.2E-03	0	9.	91.	0.015
14. Peach Bottom 2-3	9.7E-02	4.5E-02	1.4E-01	68.	< 0.01	32.	5.2
15. Pilgrim	1.5E-05	3.1E-02	3.1E-02	0	< 0.05	~100.	31.
16. Quad Cities	4.2E-03	2.0E-02	2.4E-02	18.	0.14	92.	31.
17. Vermont Yankee	1.8E-05	2.2E-02	2.2E-02	0	0.01	~100.	0.036
BWR Total (person-rem)	0.10	0.46	0.56	-	-	-	-
(person-mSv)	1.0	4.6	5.6	-	-	-	-
BWR Average (±S.E.)[a] (person-rem)	(8.1±2.0)E-03	(3.5±0.6)E-02	(4.3±0.6)E-02	10.	1.5	88.	12.8
(person-mSv)	(8.1±2.0)E-02	(3.5±0.6)E-01	(4.3±0.6)E-01	-	-	-	
II. PRESSURIZED WATER REACTORS							
1. Arkansas Nuclear One (2)	3.9E-04	1.2E-02	1.2E-02	0.	3.2	97.	0.3
2. Beaver Valley (1)	1.5E-02	1.5E-04	1.6E-04	99.	0.1	0.9	92
3. Calvert Cliffs (2)	3.7E-02	3.4E-03	4.0E-02	0.	92.	8.5	6.2
4. Donald C. Cook (2)	9.3E-02	9.4E-04	9.3E-02	98.	0.6	1.0	19.
5. Crystal River (1)	4.2E-01	8.9E-04	4.2E-01	0.	~100.	0.2	2.1
6. Davis-Besse (1)	8.4E-03	2.2E-03	1.1E-02	77.	3.0	20.	1.4
7. Joseph Farley (2)	2.0E-03	3.4E-02	3.6E-02	0.	5.6	94.	0.9
8. Fort-Calhoun (1)	4.8E+00	1.5E-02	4.8E+00	~100	< 0.1	0.3	79.
9. Robert E. Ginna (1)	7.5E-02	1.7E-02	9.2E-02	81.	0.3	18.	60.
10. Haddem Neck (1)	7.0E-03*	1.4E-01*	1.4E-01*	0. *	4.8*	95. *	42. *
11. Indian Point (3)	4.9E-04	3.9E-02	3.9E-02	0.	1.2	99.	0.45
12. Kewaunee (1)	3.1E-01	1.0E-03	3.1E-01	99.	0.9	0.3	17.
13. Maine Yankee (1)	1.5E-04	2.0E-03	2.1E-03	0.	7.1	93.	25.
14. McGuire (1)	7.3E-02*	2.8E-05*	7.3E-02*	~99. *	~ 1.0*	0.04*	~40. *
15. Millstone-2 (1)	6.3E-05	5.5E-02	5.5E-02	0.	0.1	~100.	90.
16. North Anna (2)	1.4E-02	5.9E-03	2.0E-02	0.	70.	30.	0.38
17. Oconee (3)	1.0E+00	1.5E-02	1.0E+00	98.	0.3	1.5	6.6
18. Palisades (1)	2.2E-02	1.1E-03	2.3E-02	87.	8.7	4.3	14.4
19. Point Beach (2)	2.7E-02	9.7E-02	1.2E-01	22.	< 0.1	78.	60.
20. Prairie Island (2)	3.1E-03	3.4E-02	3.7E-02	~ 0.	8.4	92.	89.
21. Rancho Seco (1)	6.9E-04	5.9E-02	6.0E-02	~ 0.	1.1	~100.	2.7
22. H.B. Robinson-2 (1)	1.4E-02	2.4E-03	1.6E-02	0.	89.	11.	0.9
23. Salem (2)	5.3E-03	2.5E-03	7.9E-03	0.	68.	32.	0.9
24. San Onofre (1)	1.0E-02*	-	-	0. *	-	-	1.2 *
25. Sequoyah (1)	4.0E-02	5.2E-04	4.0E-02	99.	0.1	1.3	5.9
26. St. Lucie (1)	6.2E-05	7.6E-02	7.6E-02	0.	< 0.1	~100.	17.
27. Surry (2)	4.5E-03	3.5E-02	4.0E-02	0.	11.	89.	2.1
28. Three Mile Island (2)	2.1E-03*	1.0E-01*	1.0E-01	2.0*	~ 0.1*	98. *	37. *
29. Trojan (1)	6.6E-05	1.8E-02	1.8E-02	< 0.1	0.3	~100.	7.8
30. Turkey Point 3-4 (2)	1.7E-04	1.2E-04	2.9E-04	0.	59.	41.	0.39
31. Yankee (1)	5.2E-04	1.8E-03	7.0E-03	~ 0.	22.	78.	1.9
32. Zion (2)	6.3E-05	2.1E-02	2.1E-02	0.3	< 0.1	~100.	6.2
PWR Total (person-rem)	7.0	0.79	7.8	-	-	-	-
(person-mSv)	70	7.9	78	-	-	-	-
PWR Average (±S.E.)[a] (person-rem)	0.25 ± 0.003	(2.0 ± 0.09)E-02	0.26 ± 0.03	30.	20.	50.	19.6
(person-mSv)	2.5 ± 0.03	0.20 ± 0.01	2.6 ± 0.3				
Grand Total (person-rem)	7.1	1.2	8.3	-	-	-	19.4
(person-mSv)	71	12.	83	-	-	-	

*Not included in averages because of atypical design (LaCrosse, Monticello, Big Rock Point, Haddem Neck, San Onofre), non-operation (Three Mile Island), or low Power production (McGuire), but included in total collective dose.

[a] Arithmetic mean and standard error of the collective tritium doses at individual sites. The standard error is calculated from the variations about the mean and does not include the uncertainties which might be inherent in the environmental transport, metabolic and dosimetric models.

resultant cost of \$90,000 - \$150,000 per site per year is clearly not commensurate with the small collective doses that result from these tritium releases. According to one study[8], even existing controls on tritium production such as the use of zirconium cladding, enriched ^{7}Li, and zirconium tritium "getters" would exceed the NRC's cost-effectiveness criterion (based upon their use only for tritium control). Reduction in liquid effluent releases by evaporation and release as airborne effluents would produce only small dose reductions and would be ranked low in terms of cost-effectiveness. The use of enriched ^{7}Li is most cost-effective (about \$5,000 per person-rem reduced)[8] and is currently used.

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ENVIRONMENTAL TRITIUM CONTAMINATION FROM INCREASING UTILIZATION OF NUCLEAR ENERGY SOURCES

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Abstract

ENVIRONMENTAL TRITIUM CONTAMINATION FROM INCREASING UTILIZATION OF NUCLEAR ENERGY SOURCES. Tritium is produced in nuclear reactors by ternary fission and by neutron capture reactions in ^{10}B, ^{11}B, ^{6}Li, ^{7}Li, ^{14}N and ^{2}H. The expanding use of nuclear reactors for power production will contribute additional amounts of tritium to the existing background levels due to atmospheric testing of nuclear devices and natural production by cosmic-ray interactions in the upper atmosphere.

Production processes, both in the environment and in nuclear and thermonuclear energy sources are reviewed. US power reactor experience indicates that the primary source of tritium production in boiling light-water reactors is ternary fission, while neutron capture reactions in ^{10}B and ^{7}Li are the principal sources in pressurized-water reactors employing chemical shim. Tritium from deuterium activation in heavy-water moderated reactors far exceeds the production of tritium by ternary fission. Environmental surveillance data show no apparent increase of tritium concentrations in water sources in the vicinity of light-water reactors. Due to tritium retention by the fuel cladding, a major portion of the tritium produced by ternary fission will be released during processing of the spent reactor fuel. Estimated releases from fuel reprocessing plants are compared with discharge data from operating plants.

Forecasts of future tritium production from nuclear reactors are provided up until the year 2000. These estimates are compared with existing equilibrium levels of naturally-occurring tritium and with the decreasing inventory of tritium remaining from atmospheric nuclear testing. The implications of these projections are evaluated from a public health viewpoint. A discussion of the possible impact of new thermonuclear energy sources on environmental tritium concentrations is also included.

1.0 INTRODUCTION

Tritium is a radionuclide on which attention is currently being focused because of its radiological characteristics and its appreciable production by an expanding nuclear power industry. This industry results in tritium releases to the environment as a consequence of normal operation of both nuclear power reactors and fuel reprocessing plants. The purpose of this paper is to discuss the production of tritium by several sources and to examine the environmental consequences of this production relative to the nuclear power industry.

Although tritium is generally considered to be one of the least hazardous radioisotopes (1), its long radioactive half-life (approximately 12 years) means that tritium discharged into the environment will accumulate over a relatively long period of time. Since tritium closely follows the reactions of ordinary hydrogen, it assimilates readily into water and into biological media.

The importance of tritium as an environmental contaminant is related to the development of nuclear power. Figure 1 shows current predictions of the growth of nuclear electricity in the United States and compares it with total generation both in the United States and the world (2). In the United States, the Atomic Energy Commission has estimated that 120-170,000 megawatts of electricity will be generated by nuclear reactors by the year 1980 (3). This estimate will represent about 25% of the total electrical power generated in that year. By the year 2000 the total installed nuclear capacity in the U. S. may be as high as 1,000,000 megawatts (4).

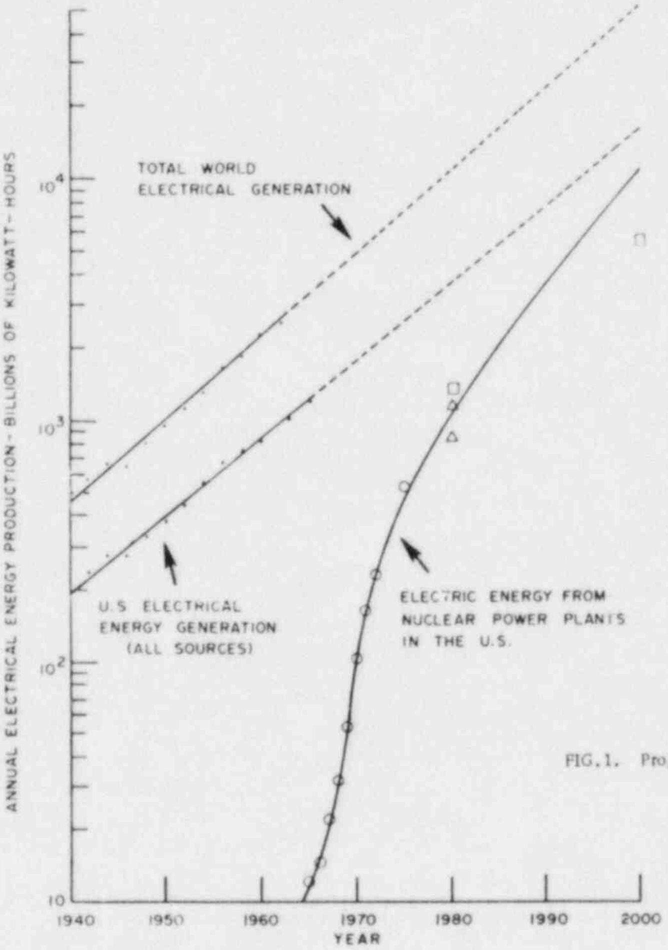

FIG.1. Projected energy requirements [2].

2.0 TRITIUM PRODUCTION IN NUCLEAR REACTORS

Tritium may be produced in nuclear reactors by several mechanisms some of which are shown schematically in Figure 2. These sources include:

1. Ternary fission having a triton as one of the fission fragments;
2. Neutron capture reactions with coolant additives such as boron, lithium and ammonia;

3. Activation of naturally occurring deuterium in light- and heavy-water moderated reactors;
4. Neutron capture reactions with poison material used in control rods and plates.

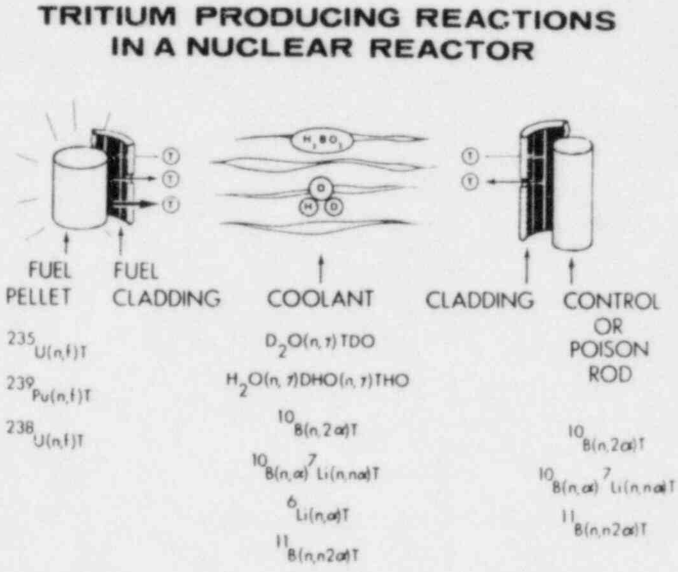

FIG.2. Schematic diagram of tritium production processes.

Other reactions with nitrogen, helium-3 or structural material may also produce tritium but the contribution to the total production rate from these reactions appears to be slight (5, 6). The relative importance of a given reaction mechanism for tritium production will depend upon the reactor type, design characteristics, operating history, and materials of construction. The manner in which tritium is released to the environment will also be a function of these parameters.

2.1 Fission Product Tritium

The discovery of tritium as a fission product was initially reported by Albenesius (7), and has since been verified by several investigators (8, 9). Measured tritium fission yields from thermal neutron induced fission of enriched or natural uranium range from 0.3 to 1.0 tritium atoms per 10^4 fissions (7-9). Theoretical fission yields have been calculated for other materials and for ^{235}U by Dudey whose results are shown in Table I (10). Measurements of the tritium released in fuel reprocessing indicate that the theoretical estimate of 1.3×10^{-4} is the most accurate (11).

Dudey has also estimated the tritium content of various reactor fuels as shown in Table II. Based upon these values we have calculated the tritium production rates shown in the last column of Table II. Since fuel burnups of 30,000 and 40,000 MW(th)-day/tonne have been forecast for replacement fuel loadings in proposed reactors and advanced reactors (3), we have extrapolated production rates for these burnups. These values are also contained in Table II.

TABLE I

ESTIMATED YIELD OF TRITIUM FROM FISSION (10)

Material	Neutron Energy (MeV)	Atoms ^{3}H Produced per Fission
^{235}U	Thermal	1.3×10^{-4}
	1.0	1.2×10^{-4}
	3.0	8.2×10^{-5}
^{238}U	Thermal	2.6×10^{-4}
	2.5	1.4×10^{-4}
^{239}Pu	Thermal	2.3×10^{-4}
	1.0	2.5×10^{-4}

Light-water nuclear reactors produce about 16.4 millicuries of tritium daily per megawatt of thermal energy. Fast reactors should produce more fission-product tritium than thermal reactors since the fission yield of plutonium-239 is higher than uranium-235 (2.5×10^{-4} and 1.3×10^{-4} atoms per fission, respectively) (10). These estimates yield tritium production rates of 18.7 Ci/yr-MW(e) for thermal reactors and 26.2 Ci/yr-MW(e) for plutonium-fueled fast reactors.

2.2 Deuterium Activation

Tritium can be formed by neutron activation of deuterium which represents about 0.015% of the atoms of hydrogen in nature (12). The thermal cross-section for the $^{2}H(n,\gamma)^{3}H$ reaction is 0.5 millibarn (13). Deuterium activation has been estimated to produce tritium in a 3295 MW(th) boiling-water reactor at a rate of about 0.15 μCi/sec or 4.45×10^{-3} Ci/MW(e)-yr. By comparison, the tritium production rate from fission was calculated to be 540 μCi/sec (14).

Because of the retention of fission-product tritium in the fuel element, the importance of tritium from deuterium activation as an environmental contaminant will be higher than its relative production rate would indicate. If only 0.1% of the fission-product tritium escaped from the fuel then deuterium activation could contribute over 27% of the activity discharged to the environment from a boiling-water reactor. In a pressurized-water reactor other sources contribute considerably greater amounts of tritium, thus deuterium activation is negligible by comparison.

In heavy-water reactors the moderator and coolant can consist of 99.75% deuterium oxide. For this reason, deuterium activation represents a greater source of tritium than does fission. A comparison of the primary coolant tritium concentration for several types of reactors is shown in Table III. These data show that the tritium activity in heavy-water reactors is considerably greater than in light water reactors. Based upon the ratio of the deuterium content in light- and heavy-water, the tritium production rate in a heavy-water reactor would be about 30 Ci/MW(e)-yr.

TABLE II

ESTIMATED FISSION-PRODUCT TRITIUM CONTENT IN VARIOUS REACTOR FUEL MATERIALS (10)

Reactor Type	Fuel Material	Fuel Burnup Rate (MW(th)-day/Tonne)	Tritium Content (Ci/Tonne)	Tritium Production Rate* (mCi/MW(th)-day)
Light-Water	^{235}U	20,000	327	16.4
		30,000	490*	
Light-Water	^{239}U	20,000	540	27.0
		30,000	810*	
Light-Water	1/2 U- 1/2 Pu fuel	20,000	440	22.0
		40,000	880*	
LMFBR**	^{235}U	100,000	1510	15.1
LMFBR	^{239}Pu	100,000	3150	31.5
LMFBR	^{239}Pu-^{235}Pu core and blanket	40,000	1900	47.5

* Estimated by the Authors

** LMFBR - Liquid Metal Fast Breeder Reactor

TABLE III

MEASURED TRITIUM CONCENTRATIONS IN REACTOR PRIMARY COOLANT WATER (20)

Reactor Type	Tritium Concentration µCi/ml
Boiling Water	0.015
Pressurized Water	1.5
Heavy Water	7200

2.3 Tritium from Boron Reactions

Boron-10 is used in some nuclear reactor control elements due to its large neutron absorption cross-section of 3840 barns (13). Both natural boron which contains 19.7% boron-10 and boron which is enriched in this isotope may be used. Boron carbide (B_4C) is usually used in boiling-water reactors (BWR) and some BWR plants in operation may contain over 20 kilograms of boron-10 in this form. Additional boron may be used as a burnable poison in the initial cores of BWRs to offset the buildup of neutron absorbing fission-products. The amount of boron-10 present in this form could be about 24 kilograms in a 1000 MW(e) reactor. This boron, in the form of borated stainless steel curtains, is removed after the initial fuel cycle burnup when fission-product poisons are near equilibrium in the reactor core.

Pressurized-water reactors (PWR) do not routinely employ control rods having an appreciable amount of boron in them and usually use an alloy of silver, cadmium and indium for the neutron absorber. Boron may be present,

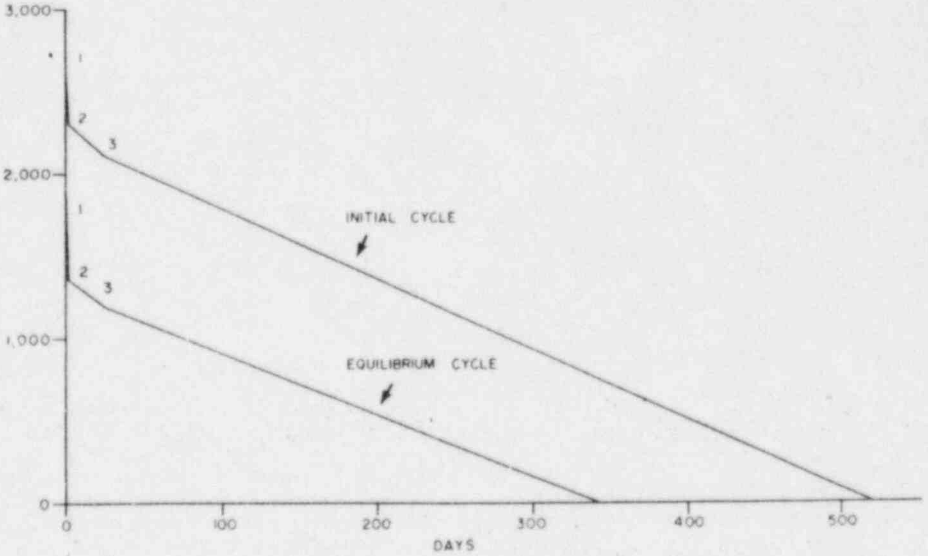

FIG.3. Variation of boron concentration over the fuel lifetime in a pressurized water reactor [16]. The regions indicated correspond to major changes in the removal rate.

TABLE IV

ESTIMATED TRITIUM PRODUCTION FROM BORON REACTIONS IN A 3391 MW(th) PRESSURIZED WATER REACTOR(17)

Source	Amount of ^{10}B (grams)	Reaction	Annual Tritium Production (Curies)	
			Initial Cycle	Equilibrium Cycle
Poison Plates	~ 3530	$^{10}B(n,2\alpha)^3H$	800	Not Used
		$^{10}B(n,\alpha)^7Li$ $^7Li(n,n\,\alpha)^3H$	1500	Not Used
Chemical Shim	~3400 (initial) ~3150 (equilibrium)	$^{10}B(n,2\alpha)^3H$	1100	780
		$^{10}B(n,\alpha)^7Li$ $^7Li(n,n\,\alpha)^3H$	not estimated (~2100)*	not estimated (~1450)*

* Estimated by authors from ratio of production in poison curtains.

however, in the form of boric acid dissolved in the coolant (chemical shim) or as poison plates in the initial core loading. The chemical shim is used to provide a more uniform power distribution throughout the reactor core and to compensate for long-term changes in reactivity such as the depletion of uranium or the accumulation of fission-product poisons (15). The boron concentration is not maintained constant but decreases over the operating life of the fuel as shown in Figure 3.

Tritium may be formed directly from boron-10 by the $^{10}B(n,2\alpha)^3H$ reaction or by the $^{10}B(n,\alpha)^7Li(n,n\ \alpha)^3H$ reactions. Estimates of the tritium production rate for a PWR employing chemical shim are shown in Table IV. Based upon the production of 2300 curies per year from the 3.4 Kg of boron-10 in the poison plates we estimate that the 20 Kg of boron-10 in a 700 MW(th) BWR would produce 13,000 curies of tritium annually, or about 21,500 curies of tritium per year by a 1000 MW(th) BWR plant. Smith has estimated a production rate of 20,000 curies of tritium per year for a comparable plant (18).

2.4 Tritium from Lithium

Natural lithium contains 7.42 percent of ^6Li which has a thermal cross-section of 675 barns for tritium production (13). One part per million of natural lithium as an impurity in the cladding and primary coolant of a small reactor could yield approximately 50 percent as much tritium as produced by fission (6). Lithium may be intentionally added to the coolant in the form of LiOH to control the acidity of the primary system. It may also be present from ion exchange resins in Li^+ form. The importance of lithium as a source of tritium is vividly depicted in Figure 4 which shows tritium discharges from the Shippingport Atomic Power Station in the United States. The tritium discharges were significantly reduced when resin containing only ^7Li was substituted for natural lithium (19).

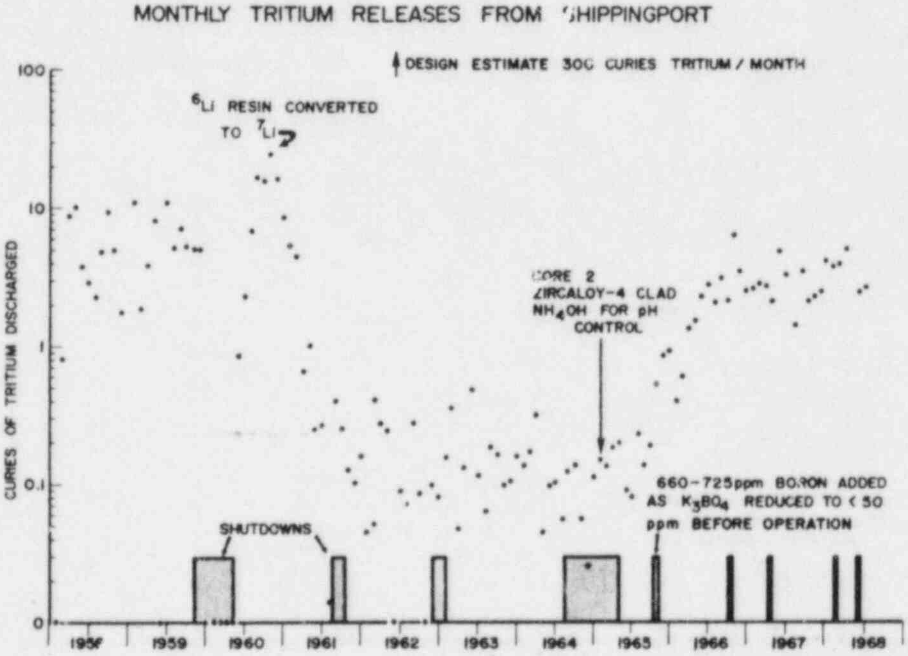

FIG.4. Tritium releases from a pressurized water reactor. The sharp decrease in the amount released after natural lithium was removed is evident [19].

Present reactor designs will employ 99.9% ^{7}Li in the ion exchange resins and LiOH in order to minimize tritium production (17).

3.0 TRITIUM RELEASES FROM NUCLEAR REACTORS

3.1 Retention of Fission-Product Tritium

Only a fraction of the fission-product tritium is released to the environment from a nuclear reactor. In order to reach the reactor coolant the tritium would have to diffuse out of the ceramic uranium dioxide (UO_2) fuel pellet and pass through the metallic fuel cladding as shown in Figure 2. For this reason, the bulk of the tritium is retained in the fuel elements and is not released until the elements reach the fuel reprocessing plant. Small amounts of tritium may also be produced from traces of uranium on the outer surfaces of the cladding.

Although the purpose of the metallic cladding around the fuel pellets is to contain fission products, the retention mechanisms for tritium are not well defined. These mechanisms may involve chemical reactions between tritium and the cladding material as well as physical diffusion. Three mechanisms have been proposed for the transmission of tritium through fuel cladding:

1. Direct penetration of tritons (6);
2. Effusion of tritium through minute holes or defects in the cladding material;
3. Diffusion of tritium atoms around grain boundaries and through intact cladding.

Releases of tritium by direct penetration of the cladding by tritons has been shown to be negligible (6), thus this mechanism is not as important as releases by effusion and diffusion processes.

The composition of the fuel cladding has been found to have an appreciable effect on the amount of tritium retained in the fuel (18, 20). Measurements of the tritium concentration in the coolant of boiling-water reactors indicate that the tritium release rate is approximately an order of magnitude greater in reactors employing stainless steel fuels than in those employing zirconium alloy (zircaloy) clad elements (18, 20). A limited amount of experience with zircaloy-clad fuels also indicates that there may be a three-fold difference in hydrogen retention between the two zirconium alloys, zircaloy-2 and zircaloy-4 (21).

3.2 Operating Experience

The majority of the tritium in the coolant, either from fission or boron and lithium reactions, is released to the environment with the liquid waste discharge. Table V shows amounts of tritium released from several pressurized-water (PWR) and boiling-water reactors (BWR) in the United States. The appreciable difference between the releases from PWR's and BWR's is believed to be due to the presence of boron in the primary coolant of the PWR (18, 20). Tritium release rates and tritium concentrations in the coolant of the boiling-water reactors indicate that boron present in cladded control rods or alloyed in poison curtains does not contribute appreciable amounts of tritium to the effluent (20).

Liquid wastes from reactors are discharged into the condenser cooling water which provides a large amount of dilution. For typical 1000 MW(e) PWR plants releasing 5000 Ci/yr of tritium the resultant concentrations in these discharges which reach the general public would be less than 0.2% of the radioactivity concentration guide (17). Reactors which use cooling towers

TABLE V

TRITIUM RELEASED TO THE ENVIRONMENT

FROM OPERATING POWER REACTORS

Reactor	Fuel Cladding	Power MW(e)(22)	Capacity Factor(22)	Tritium Release	
				Estimated Annual Curies (22)	Rate Ci/MW(e)-yr
<u>Boiling Water Reactors</u>					
Big Rock Point	Stainless steel	50	0.45	20	0.89
Humboldt Bay	Stainless steel, Zircaloy	52	0.80	20	0.50
Elk River	Stainless steel	24	0.70	10	0.60
Dresden - I	Zircaloy - 2	200	0.65	5-10	0.037-0.074
<u>Pressurized Water Reactors</u>					
Indian Point - I	Stainless steel	163	0.50	500	6.1
Yankee	Stainless steel	185	0.70	1300	10.0
Connecticut Yankee	Stainless steel	480	0.71[1]	1755[1]	5.1

[1] data from Connecticut Yankee monthly operating reports for 1968 (23).

TABLE VI

ESTIMATED TRITIUM RELEASES FROM U. S. COMMERCIAL NUCLEAR FUEL REPROCESSING PLANTS

Plant	Capacity (Tonnes/day)		Estimated Fuel Burnup (MWd/Tonne)	Tritium Content of Fuel (Ci/Tonne)	Design Annual Average Release Rate From Stack (Ci/day)	
	Design	Actual				
Nuclear Fuel Services, West Valley, New York	1	1	22,000	200	50 130**	(37)
Midwest Fuel* Recovery Plant Morris, Illinois	1.4	0.82	NS	200	432	(38)
Barnwell Nuclear* Fuel Plant, Aiken, South Carolina	6	5	35,000 40,000	400 500	1,640	(39)

* Under construction
** Released as liquid waste to storage lagoon
NS NOT SPECIFIED

have a smaller dilution capacity than those that use once-through condenser cooling. Under certain conditions extra care in discharging tritium may be required to insure that concentration limits are not exceeded.

Atmospheric release of tritium from light-water reactors with gaseous wastes has not been extensively studied but does not appear to be appreciable. The Bureau's Nuclear Engineering Laboratory has made some measurements at a BWR which indicate that atmospheric tritium releases may be less than 0.5 curies per year. By comparison, proposed BWR's are using a hypothetical estimate (based on considerable fuel cladding defects) of approximately 0.03 to 30 Ci/yr (24-27). Gaseous tritium releases from the Yankee pressurized-water reactor are less than 100 Ci/yr (28), or approximately 6% of the total tritium discharged from the plant.

In the United States, there has not been any development of large heavy-water reactors because of the availability of enriched uranium. Canada and several other countries, however, use this concept because natural uranium can be used without enrichment (29). Tritium concentrations in the primary coolant of heavy-water reactors could be about 10-20 mCi/g after long-term operation. Past experience at heavy-water reactors indicates that D_2O losses can approach several Kg/day although there is an important economic incentive to retain this material (30-35). Holmquist has stated that no heavy-water reactor can be assumed to have a lower D_2O leakage rate than 2-3%/yr or 6 Kg/day (36). The production reactors at the Savannah River Plant in the United States are estimated to release between 1,000 and 10,000 curies of tritium per month to the atmosphere from a D_2O leakage rate of 2-3%/yr (36). Based upon the leakage experienced at Canada's 200 MW(e) Douglas Point Nuclear Station (4 Kg/day) (34), a heavy-water reactor could release over 15,000 curies of tritium per year to the environment.

4.0 TRITIUM RELEASES FROM FUEL REPROCESSING PLANTS

The largest localized concentrations of tritium in the environment will be in the vicinity of fuel reprocessing plants where 68-99.9% of the fission-product tritium will be available for release when the fuel elements are processed. Table VI lists estimated tritium discharges for three commercial fuel reprocessing plants in the United States (37-39).

Several possibilities exist for the disposal of tritium from fuel reprocessing (40). These are as follows:

1. Disposal into ground water,
2. Dilution and disposal directly to surface water,
3. Distillation and release to the atmosphere, and
4. Concentration and storage with high level waste.

4.1 Ground Water Disposal

Disposal into ground water through seepage basins or wells has been pursued for several years at the major U. S. plutonium production sites. Disposal into ground water requires hydrological and geological studies to determine the direction and rate of ground water movement and extensive monitoring programs to assure that off-site drinking water supplies are not contaminated.

4.2 Surface Water Disposal

The Nuclear Fuel Services commercial reprocessing plant at West Valley, New York disposes of tritium by a combination of surface water releases and atmospheric releases. Release rates of tritium from this plant have been estimated to be 50 Ci/day up the stack and 130 Ci/day to storage lagoons which are eventually pumped into surface waters (37). These estimated releases are considerably higher than measured discharges as shown in Table VII.

TABLE VII

ANNUAL TRITIUM RELEASES IN LIQUID WASTE FROM

A COMMERCIAL FUEL REPROCESSING PLANT (41)

Data for 1967

Measured Tritium Releases to Storage Lagoon (Ci)	4200
Tonnes of Uranium Processed	136
Percent of Plant Capacity	37
Percent of Estimated Release of 130 Ci/day	9
Percent of Estimated Release Corrected for Capacity	25

Surface water disposal requires considerable dilution and therefore is practical only for sites on large rivers. Blomeke has estimated that a least 18 million gallons of water per day would be required per ton of fuel in order to dilute the released tritium to 10^{-3} μCi/ml or less (40). For this reason this technique has limited applicability for plants with capacities on the order of 5 MTU/day.

4.3 Atmospheric Disposal

The other two commercial fuel reprocessing plants shown in Table VI are under construction in the United States. Both plants will discharge only limited volumes of liquid waste thus the major means of environmental tritium release will be from atmospheric discharges (38-39). The estimated atmospheric tritium releases from these plants are shown in Table VI to be 432 and 1,640 Ci/day. The annual average off-site air concentrations for these plants are estimated to be less than 2% of the radiation concentration guide of 2×10^{-7} μCi/ml (38-39). Cowser, et.al., analyzed the dose that would be received from atmospheric releases from a plant reprocessing fuel equivalent to 100,000 MW(th) of reactor operation. They found that the annual dose received from released HTO would be about 7 mrem/yr (42).

4.4 High-Level Waste Storage

Retention of tritium in high-level waste storage tanks is dependent on the process used. Plants that use the Purex process produce between 10,000 and 100,000 gallons of liquid waste per ton of fuel (43). The Nuclear Fuel Services plant, which uses the Purex process, is estimated to retain about 10% of the tritium in long-term storage. The rest is disposed to surface water (about 65%) or the atmosphere (about 25%) principally because

of the large volume of liquid generated (37). Plants that use the fluoride volatility process, produce approximately 25-250 liters of liquid waste per ton of fuel which may have tritium concentrations of 1-10 Ci/l (43). Tank storage might be economically feasible for wastes of this activity.

5.0 TRITIUM ACCUMULATION IN BODIES OF WATER

5.1 General

All tritium, regardless of its mode of production or release, eventually deposits in the hydrosphere. Localized sources of environmental tritium such as reactors and fuel reprocessing plants may present certain public health considerations because of exposure resulting from accumulation in receiving waters. In general, these considerations are minor for streams which have a fast flushing time and sufficient dilution capability to keep concentrations low. This distinction, however, is not as apparent for facilities on lakes or impoundments where slow flushing times may result in a buildup of tritium.

In the United States some power reactors are located on a small lake or a stream which has been impounded to produce a lake large enough to provide sufficient cooling water. In some cases, several plants may be sited on a single large lake such as Lake Michigan. Tritium buildup for these two situations was examined in detail.

5.2 Tritium Buildup in Lakes

The tritium accumulated in a lake can be estimated by a model which assumes a constant outflow rate and a constant volume. The equation which governs this process is:

$$C(t) = \frac{R}{V\Lambda}\left[1 - e^{-\Lambda t}\right] + C_o e^{-\Lambda t} \qquad (1)$$

where,

$C(t)$ = the concentration ($\mu Ci/cm^3$) at time t,
R = the activity release rate (Ci/yr) into a lake,
r = the rate of water loss from a lake (cm^3/yr),
V = the volume of the lake (cm^3),
λ = the radioactive decay constant for tritium ($\lambda = 0.0561\ yr^{-1}$),
C_o = the initial activity present in the lake from natural and fallout sources, and
Λ = $\lambda + r/V$, the effective removal constant for both radioactive decay and the physical loss of water from the lake.

The average or mean residence time of water in the lake is defined by:

$$\tau_w = V/r \qquad (2)$$

while the mean residence time for tritium in the lake is given by:

$$\tau_T = 1/\Lambda \qquad (3)$$

5.3 Tritium Buildup in Small Lakes

An American power company is constructing a 700 MW(e) PWR nuclear power plant on a small lake created by impounding a natural creek, a site that is reasonably typical for the United States. The volume of the lake is approximately 1.34×10^9 cubic feet and the average discharge rate is 169 ft^3/sec. Annual discharges of tritium are estimated to be about 3820 Ci/yr. It was assumed that the plant would operate 40 years and that this would be equivalent to an infinite operating time, a conservative assumption.

Tritium buildup for three conditions was examined using the model represented in equation (1). The results of these calculations for these three conditions are shown in Table VIII. It is apparent that for realistic conditions (cases I and II) accumulated tritium concentrations would not exceed established concentration guidelines. Case III represents a hypothetical maximum estimate. All calculations were conservative because water loss by evaporation was omitted.

5.4 Tritium Accumulation in a Large Lake

Large lakes have a considerable dilution capacity for tritium released to them. However, this capacity may not be sufficient if several nuclear power plants are located on one lake. Lake Michigan, one of the Great Lakes in the United States is a good example of this situation. As shown in Figure 5 there will be seven nuclear power sites on the Lake by 1973 comprising some ten reactors, or about 7030 MW(e). The total U. S. generating capacity provided by nuclear power is scheduled to be 45,600 MW(e) by 1973; thus about 15% of this total will be located on Lake Michigan. Most of this capacity will be furnished by PWR's, a situation not typical of the national trend which tends to be an even distribution between BWR and PWR plants. The tritium release rate into Lake Michigan is assumed to be 5 Ci/yr-MW(e) because of this preponderence of PWR plants. This value was obtained using the average release shown in Table V and an average generating capacity of 70 percent of the design value.

Estimates of nuclear generating capacity on Lake Michigan are shown in Figure 6. The nuclear capacity in the United States in the year 2000 will be about 1,000,000 MW(e); about 18.75% of this capacity is assumed to be located on Lake Michigan.

The water retention time in Lake Michigan has been estimated by Rainey (44) and Stigall (45) to be 30.5 years and 75 years, respectively. The mean-life of tritium is 17.8 years, which is short enough to offset variations in these values, consequently the mean residence time for tritium in the Lake only varies between 11.2 and 14.5 years. The latter value was used in the projections of tritium concentrations for Lake Michigan that follow.

The activity of tritium in the Lake at time t, A(t), is given by:

$$A(t) = A(t-1)e^{-\Lambda} + \frac{RP(t)}{\Lambda} \quad (1-e^{-\Lambda}) \qquad (4)$$

where Λ is the removal constant, R is the tritium release rate (R = 5 Ci/MW(e)-yr), V is the lake volume, and P(t) is the anticipated power generation rate (MWe) estimated at midyear intervals. The first term accounts for the decay and removal of tritium produced during the previous year; the second term represents tritium produced during the year under consideration. The total tritium activity in the Lake in the year 2000 is estimated by this equation to be about seven megacuries.

TABLE VIII

TRITIUM ACCUMULATION IN A SMALL MAN-MADE LAKE FROM A SINGLE NUCLEAR PLANT

Case	Outflow Rate (ft^3/sec)	Volume (ft^3)	Mean Residence Time Water Days	Mean Residence Time Tritium Days	Estimated Equilibrium Tritium Concentration (μCi/ml)
I. Annual Average	169	1.34×10^9	92	90	7.1×10^{-5}
II. Low Flow	21	1.34×10^9	740	664	1.8×10^{-4}
III. No Outflow	0	1.34×10^9	∞	6510	1.8×10^{-3}

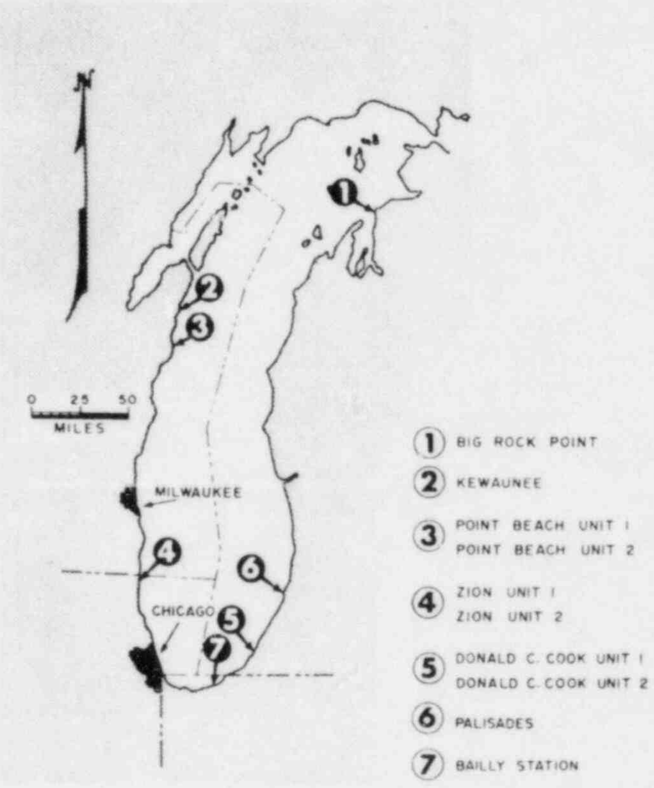

FIG.5. Nuclear power reactors at present planned for operation on Lake Michigan.

Tritium buildup in the Lake was calculated using equation (4) to account for a varying production rate due to increases in nuclear generating capacity. The concentration was calculated from:

$$C(t) = A(t)/kV \qquad (5)$$

where $C(t)$ is the average tritium concentration in the Lake, k is the fraction of the Lake volume available for dilution, and V is the total volume of the Lake.

The resulting tritium accumulation in Lake Michigan was determined by equation (5) for two conditions: 1) mixing with the total lake volume of 4781 km^3, and 2) mixing with 36.5 percent of the lake volume (~1780 km^3), a value estimated by Stigall from fallout data on ^{90}Sr as the effective dilution volume. The results of this calculation are shown in Figure 7. These data show that although the estimated tritium activity accumulated in the Lake by year 2000 is over seven megacuries, the concentration in the Lake would be less than 0.2% of the radioactivity concentration limit of 3×10^{-3} μCi/ml used by the United States Atomic Energy Commission (46).

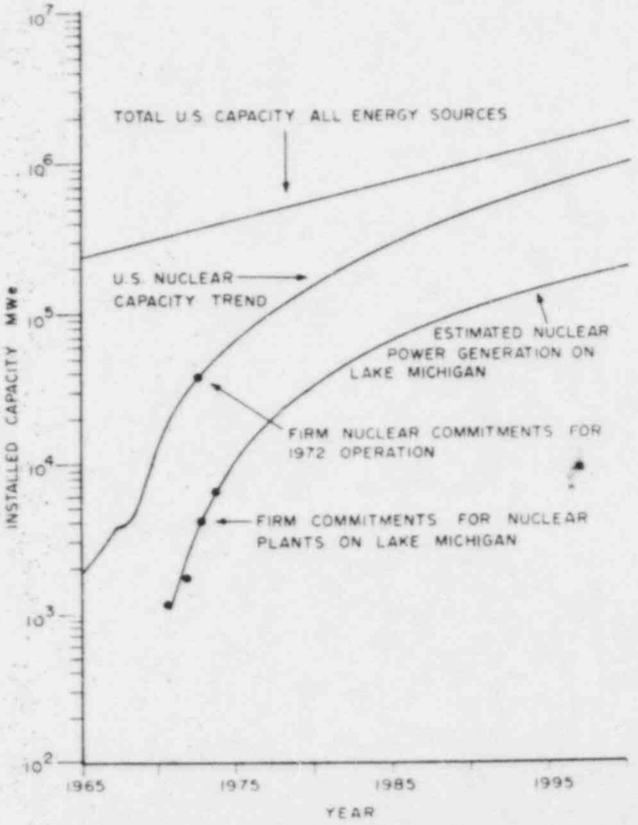

FIG.6. Projected power generating capacity from nuclear reactors discharging into Lake Michigan in comparison with total US power projections [4].

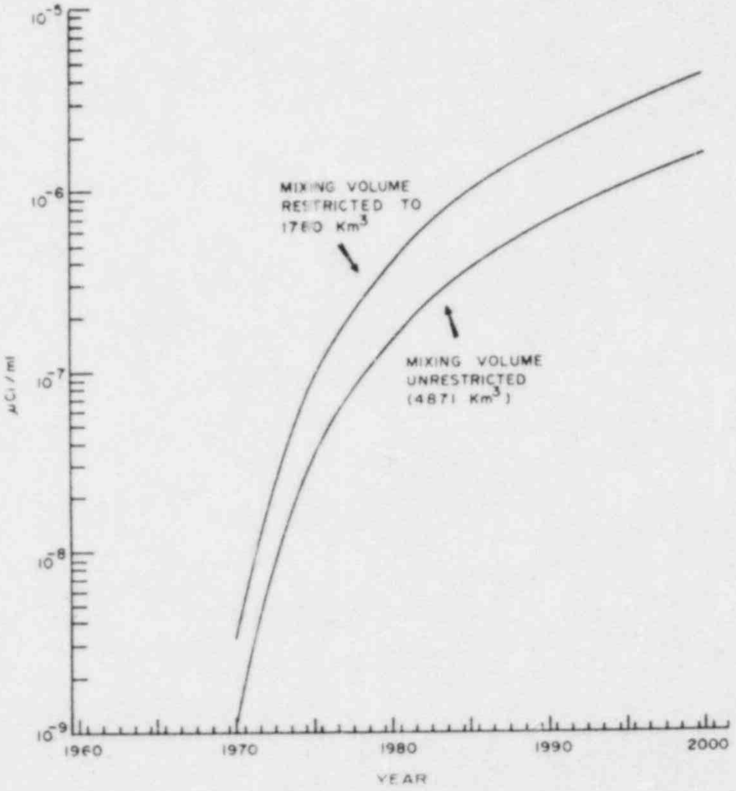

FIG.7. Estimates of future tritium concentration in Lake Michigan from nuclear reactor operation.
These values are in addition to tritium from fall-out and natural sources.

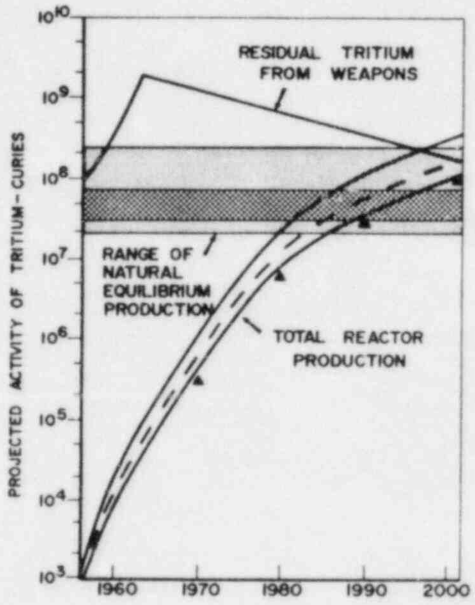

FIG.8. Comparison of tritium activity from: (a) natural production (wide range = all estimates; narrow range = most probable estimate); (b) residual weapons fall-out; (c) US reactor production; and (d) world-wide reactor production (▲) estimated in Ref.[56].

6.0 PROJECTED ENVIRONMENTAL TRITIUM LEVELS

6.1 General

The buildup of tritium in the environment from the nuclear power industry will be superimposed on tritium already present from natural production processes and from atmospheric testing of thermonuclear weapons. The relative contribution by the power industry in the United States is compared to these other sources in Figure 8. At present the nuclear power industry production is small compared to the other two sources and will not reach natural production levels until about 1985. The major source of tritium in the environment is from past nuclear weapons tests. This will continue to be the case up until about 1995 at which point reactor production will become the predominant source if the present trend continues and there are no further thermonuclear explosions.

6.2 Tritium Production in Nature

The production of tritium in nature is caused by cosmic-ray interactions with nitrogen and oxygen in the upper portion of the atmosphere. The principal reactions which produce tritium are high energy ($E > 100$ MeV) proton spallation reactions and $^{14}N(n,t)^{12}C$ and $^{16}O(n,t)^{14}N$ reactions with secondary neutrons from cosmic radiation (47-52).

Nir, et.al., reviewed all previous calculations of tritium production and estimated that the average production rate is 0.19 ± 0.09 ^3H atoms/cm^2-sec and that the total decay rate is 0.5 ± 0.3 ^3H atoms/cm^2-sec (50). This latter value corresponds to a total equilibrium activity inventory of approximately 69 megacuries of tritium. Estimates of the rate of production vary considerably, ranging from 0.09 - 2.0 ^3H atoms/cm^2-sec. This variation corresponds to equilibrium levels of 12 and 275 megacuries, respectively, as shown in Figure 8. Other natural processes such as spontaneous fission in thorium and uranium and reactions with boron, lithium, deuterium and helium-3 also produce tritium but the production rate is insignificant compared to the oxygen and nitrogen reactions (51-52).

6.3 Tritium from Nuclear Testing

Tritium is formed from fission in atomic weapons at a rate of 0.7 curies per kiloton of TNT explosive yield. Small amounts of tritium may also be formed by neutron interactions with nitrogen and oxygen (53). The largest source of tritium in the environment at the present time is from the detonation of thermonuclear (fusion) weapons in which tritium was produced by $^2H(n,\gamma)^3H$ and $^6Li(n,\alpha)^3H$ reactions with lithium deuteride in the devices. The yield of these reactions is estimated to be 6.7 megacuries per megaton equivalent of TNT (54). Nuclear testing has contributed about 1700 megacuries of tritium to the environment (55). This contribution is considerably higher than the equilibrium value from natural production which is about 69 megacuries (see Figure 8).

6.4 Tritium from Nuclear Energy

Projections of the environmental inventory from nuclear energy sources are dependent upon forecasts of power consumption rates and developments in reactor technology as indicated by the curves in Figure 8. Cowser's estimate (▲) of worldwide reactor production (56) is lower than the total U. S. estimate (2) shown by the three curves in the Figure. The curves of U. S. production in Figure 8 represent the contribution from fission (lower curve), fission plus soluble boron in PWR reactors (middle curve), and fission plus total boron in all reactors (upper curve). Boron control rods and curtains should not represent an appreciable source of tritium in the environment since the fraction of tritium which escapes from encapsulated boron is small and these elements are generally disposed intact as high-level solid waste.

An increase in the utilization of large heavy-water power reactors could substantially increase environmental tritium levels. It has been estimated that the tritium inventory would be doubled if 10% of the power production was from heavy-water reactors (57). Development of thermonuclear power sources could also increase tritium levels in the environment. The tritium production rate from a thermonuclear reactor has been estimated to be over 100,000 times greater than in a fission reactor (57); however, most of this material would be contained in the reactor for use as fuel material. Present information is insufficient to project future estimates of tritium from this source.

7.0 PUBLIC HEALTH SIGNIFICANCE

The principal form of tritium in reactors is tritiated water and conventional concentration processes such as ion exchange and evaporation do not remove appreciable amounts of tritium from liquid effluents. For this reason tritium may comprise between 50 and almost 100% of the total activity in reactor discharges to the environment (20). The Radioactivity Concentration Guide (RCG) for tritium, however, is much higher than for most common fission products. The RCG of tritium is 10^{-3} μCi/ml and because of its short biological half-life (12 days), its low disintegration energy, and uniform distribution in the body, its relative health significance is smaller than for other fission products such as ^{90}Sr and ^{137}Cs.

The estimated tritium release from a 1000 MW(e) pressurized-water reactor is approximately 7000 curies per year (average of Table V). Plants employing single-pass condenser systems to remove excess heat have circulation rates of about 1.5×10^{15} cm^3 per year and the resulting concentration would be about 5×10^{-6} $\mu Ci/ml$ or 0.5% of the RCG for exposure of the general population (10^{-3} $\mu Ci/ml$). The volume of water discharged from reactors employing cooling towers is considerably smaller and may be only 2000 gallons per minute. Under these circumstances the effluent concentration could approach 20% of the guideline value. Under certain circumstances it may be necessary to restrict discharges or to provide supplementary dilution for these plants.

The Public Health Service operates a tritium surveillance network in the United States to provide information on nationwide trends and tritium concentrations in water downstream from large nuclear installations. The results reported by this network are published periodically (59-62). Present levels are 0.2-10 nCi/l and the dose contribution from the maximum concentration has been estimated to be less than 1.6 mrem/year (62).

The buildup of tritium in small confined bodies of water may approach concentrations approximately 20% of the RCG which suggests that environmental surveillance would be indicated to monitor this accumulation. Our predictions for Lake Michigan show that the tritium concentrations in the Lake will be about 40 nCi/l, most of which will be contributed by the reactors on the Lake. Continuous consumption of this water for a fifty-year period would result in a total integrated dose of only 350 mrem, which is considerably less than that from natural radiation sources.

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DISCUSSION

J. TADMOR: I should like to ask whether the figures presenting the projection of tritium production in nuclear reactors up to the year 2000 include possible future fusion reactors.

A recent study made by F.L. Parker indicates that tritium waste from reactors of this type would present a major problem from a disposal point of view.

Another remark I would like to make concerns the possibility of tritium release into the atmosphere following the distillation of liquid waste. One of the problems encountered in this method of disposal is fog formation due to the large amounts of water distilled, and the consequent deposition of tritium in the neighbourhood of the release point. Have you any comment to make on this problem of fog formation?

B.J. MASON: In answer to your first question, I do not believe that the possibility of the development of fusion reactors has been taken into consideration. It would certainly not seem to be the case in the light of Mr. Parker's paper.

With regard to the fog question, I have no comment to make, but I have raised the matter of a 'rain-out' effect which I believe to be related to fog in that either process would result in fairly high levels of tritium in local environments. Apparently no consideration has been given to the possibility of such localized concentrations.

S.O.W. BERGSTRÖM: A tritium release of 15 000 Ci/yr from heavy-water reactors seems to be an upper limit. The Ågesta heat and power reactor has been operating for a long time with a tritium leakage of some 100 Ci/yr. This corresponds to 5-10 Ci/MW(e)/yr, a rate which it should be possible to reduce in the case of larger reactors. The dominating source of environmental tritium is therefore likely to be fractions other than those from deuterium activation in cooling water. In the power plants the higher water leakage rate from the light-water reactor results in tritium levels which may in fact be in excess of those which will be caused by the heavy-water type.

B.J. MASON: 15 000 Ci/yr was an estimate of what could result from the heavy-water reactor. The data used for the estimate are those available from one particular Canadian plant.

J.K. MIETTINEN: I understand that when calculating the radiation dose values for humans you used the biological half-life (12 days) given by the ICRP for tritiated water and a homogeneous distribution in the body.

In nature, in a lake containing 40 nCi T_2O/litre, part of this tritium will be present in plankton and fish, for example in the form of tritiated amino acids, and will be taken up as such by people consuming fish. It will then be built up into nucleic acids in the body and will certainly have a much longer biological half-life and give a genetic dose several orders of magnitude higher than if homogeneously distributed in the body water.

Have you calculated the contribution of this biologically bound tritium to the radiation doses received by human beings?

B.J. MASON: The authors have not made this calculation since data are not readily available. Evans, at the Savannah River plant, has indicated that the dose calculations based upon ICRP recommendations should be increased by a factor of 1.4 because of tissue-bound tritium. I agree with you that some effort must be made to take this tissue-bound tritium into consideration.

A. LAFONTAINE: In studying the problem of the radiological capacity of rivers and lakes in respect of tritium provision must be made for an adequate safety factor that allows for changes (i.e. an increase in strictness) in the maximum permissible concentrations recommended by the ICRP.

The biological cycle of tritium does in fact appear to indicate that it does not behave solely as an element of tritiated water, but that it can be concentrated in certain elements of the biological cycle, as has been indicated by Mr. Miettinen. Moreover, the distribution of tritium in the body is not as regular as was believed.

Account should also be taken of the possibility of tritium release partly in the form of molecules different from tritiated water, notably in fuel reprocessing plants.

B.J. MASON: All I can say is that I agree with you.

ERRATA

ENVIRONMENTAL TRITIUM CONTAMINATION FROM
INCREASING UTILIZATION OF NUCLEAR ENERGY SOURCES
H.T. Peterson, Jr., J.E. Martin, C.L. Weaver, E.D. Harward
SM-117/78

TABLE IV

ESTIMATED TRITIUM PRODUCTION FROM BORON REACTIONS IN A 3391 MW(th) PRESSURIZED WATER REACTOR(17)

Source	Amount of ^{10}B (grams)	Reaction	Annual Tritium Production (Curies)	
			Initial Cycle	Equilibrium Cycle
Poison Plates	~3530	$^{10}B(n,2\alpha)^3H$	800 (510)	Not Used
		$^{10}B(n,\alpha)^7Li$ $^7Li(n,n\,\alpha)^3H$	1500 (960)	Not Used
Chemical Shim	~3400 (initial) ~3150 (equilibrium)	$^{10}B(n,2\alpha)^3H$	1100 (346)	780 (480)
		$^{10}B(n,\alpha)^7Li$* $^7Li(n,n\,\alpha)^3H$*	8.8 ~~not estimated~~	8.8 ~~not estimated~~
		$^6Li(n,\alpha)^3H$**	8.8 ~~(~2100)*~~	8.8 ~~(~1450)*~~

~~* Estimated by authors from ratio of production in poison curtains.~~

* ^7Li concentration controlled to less than 2.2 ppm

** ^6Li impurity in 99.9% ^7Li

NOTE: Westinghouse has advised the authors that a design change is presently being made which will reduce tritium production to the values shown in parenthesis in the table. These values would also change the text in Sec. 2.3

Table VI

Plant	Capacity (Tonnes/day) Design	Actual
Nuclear Fuel Services, West Valley, New York	1	0.82

Table VII

Percent of Plant Capacity	45
Percent of Estimated Release Corrected for Capacity	20

Section 4.3 Atmospheric Disposal, last line

annual dose received from released HTO would be about 9 mrem/yr (42).

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Tritium in the Environment From Nuclear Powerplants

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TRITIUM, an isotope of hydrogen with an atomic mass number of three, is produced in nuclear reactors in substantial quantities. Although tritium is one of the least hazardous radioactive nuclides, its continued production and long half-life for radioactive decay may lead to increased levels in the environment. Because tritium is an isotope of hydrogen, it can be metabolized in the form of tritiated water and incorporated into body fluids and tissues. This source of population exposure requires that public health agencies be cognizant of the significance of tritium as an environmental contaminant.

The nuclear power industry has expanded rapidly during the past few years, and the U.S. Atomic Energy Commission predicts that this high rate of growth will continue during the next decade. Nuclear power reactors and fuel reprocessing plants release tritium to the environment under normal operating conditions.

Due to the stratospheric fallout from previous atmospheric testing of nuclear weapons, measurable levels of tritium are already present in the environment. An analysis of the effect of an expanding nuclear power industry on environmental tritium levels indicates that future tritium releases from reactors and spent fuel processing plants may surpass the quantity of tritium remaining from nuclear weapons tests.

The Bureau of Radiological Health's environmental surveillance activities provide a continuing source of data on tritium concentration in rivers on which nuclear facilities are located. There is, however, a need for more specific source-oriented information on tritium, and its potential for becoming a public health problem. This paper is intended as a general information guide on tritium for persons engaged in Federal, State, county, and local public health activities. The various sources and mechanisms for release of tritium and its pathway from nuclear facilities to the environment are described. This information is essential in assessing the requirement for environmental surveillance and in evaluating, from a public health viewpoint, the upward trend of tritium levels in the environment.

Radiological Characteristics of Tritium

Because of its relatively short biological half-time (12 days) combined with a relatively low disintegration energy, tritium is one of the least hazardous radionuclides produced in nuclear reactors. A single ingestion of tritiated water having an activity of 1 microcurie will produce a total dose to the body tissues of 0.21 millirems (mrem). Continuous ingestion of

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water having a specific activity of 1 microcurie of tritium per milliliter will produce a dose rate of 170 rem per year to body tissues. From this value the annual average discharge concentration limit for tritium in water can be calculated to be 3×10^{-3} microcuries per milliliter. This quantity is the concentration limit for tritium in radioactive waste discharges to the environment contained in the Code of Federal Regulations (*1*).

Tritium decays by the emission of a beta particle and an anti-neutrino to form stable helium-3. The half-life for this process is 4,500 days or approximately 12 years (*2*). The beta particle and the neutrino carry varying fractions of the energy liberated by the disintegration. When there are many tritium atoms present, this process produces a continuous spectrum of beta particle energies.

The maximum energy of the beta emitted by tritium is 18.6 kilo-electron volts (Kev) and the average energy is 5.6 Kev. These energies are considerably lower than those of most other beta emitters which are usually about 100 times greater.

Sources of Environmental Tritium

Naturally occurring tritium. Tritium is one of the three isotopes of hydrogen. Normal hydrogen consists of approximately 99.9 percent protium (hydrogen-1) and 0.015 percent of deuterium (hydrogen-2), although variations in isotopic concentration can exist in natural sources. Unlike protium and deuterium, tritium (hydrogen-3) is unstable because of radioactive decay and occurs naturally only in trace amounts.

Naturally occurring tritium is formed principally by the interaction of cosmic radiation with oxygen and nitrogen atoms in the upper atmosphere. Tritium produced by cosmic-ray interactions prior to 1952 contributed to environmental concentrations ranging between 16 to 35 picocuries of tritium per liter of water or 5 to 10 tritium units (*3*) where 1 tritium unit is equivalent to 1 atom of tritium per 10^{18} atoms of normal hydrogen. Because of a constant rate of production, the tritium from natural processes will accumulate until an equilibrium condition is reached. This condition is achieved when the rate of removal of tritium from the environment by radioactive decay equals the rate of tritium production. Since the earth is several billion years old, this equilibrium rate has existed for some time and the amount of tritium present from natural sources is essentially constant.

Nuclear weapons tests. Past tests of nuclear weapons in the atmosphere are the greatest single source of tritium present in the environment today. Tests by both the United States and the Union of Soviet Socialist Republics, before the Atmospheric Test Ban Treaty in 1962, greatly increased the amount of tritium in the environment. Thermonuclear (fusion) weapons produce tritium from neutron interactions with lithium and deuterium with a production rate of 6.7 megacuries of tritium per megaton of fusion yield (*4*). This source contributed about 1,700 megacuries of tritium to the environment compared with an equilibrium value from natural sources equivalent to approximately 69 megacuries.

A large fraction of the tritium produced by these detonations was injected into the stratosphere. This part of the atmosphere does not mix rapidly with the lower portion of the atmosphere; therefore, the tritium is introduced into the troposphere over many years. Tritium in the lower atmosphere is rapidly removed by precipitation. Tritium removed from the atmosphere by precipitation will accumulate in the surface layers of the oceans and inland waters. Rainfall that follows periods of nuclear testing has several hundred times the tritium content normally present from natural tritium. The estimated time for these processes to remove half of the tritiated water from the lower atmosphere is 35 to 40 days (*3*).

Tritium From Nuclear Reactors

Tritium may be produced in nuclear reactors by the following five mechanisms: (*a*) fissioning of uranium, (*b*) neutron capture reactions with boron and lithium added to the reactor coolant, (*c*) neutron capture reactions with boron in control rods, (*d*) activation of deuterium (hydrogen-2) in water, and (*e*) high energy neutron capture reactions with structural materials.

The relative magnitude of tritium production by these sources is influenced by the reactor type, operating history, design characteristics, and materials of construction. The amount of tritium and manner in which it is released to the environment will also be affected by these parameters.

During the processing of spent reactor fuel, tritium, which has been produced in the fuel rods, is released to the environment and may be a significant source of environmental tritium contamination. A detailed discussion of this source, however, is not within the scope of this report.

Fission-product tritium. During the fission process the uranium nucleus usually splits into two more or less equal fragments plus several neutrons. About once in every 10,000 fissions, however, the nucleus is split into three portions (ternary fission) one of which may be a tritium nucleus. It requires 3.1×10^{16} fissions per second to produce a power level of 1 megawatt of thermal energy (Mwt).

Most nuclear powerplants generate electricity at a rate of approximately one-third of the thermal energy production rate. Therefore, approximately 10^{17} fissions per second are required to produce 1 megawatt of electrical energy (Mwe). This production rate corresponds to the production of about 10^{13} atoms of tritium per second or 50 millicuries of tritium per day for each megawatt of electrical energy generated. This tritium normally remains in the fuel unless it diffuses through the cladding material or a leak occurs.

A second source of fission-product tritium in nuclear reactors is due to traces of uranium on the outer side of fuel elements which remain from the fuel fabrication process. This "tramp" uranium may be only a few micrograms per square inch of fuel surface, but because of the large surface area provided by the many fuel rods in a reactor it can produce detectable fission-product concentrations in the coolant.

The reactor fuel is usually uranium oxide and is contained in tubes made of stainless steel or an alloy of zirconium (zircaloy). The primary function of the cladding material is to prevent the escape of fission products from the fuel element. These fission products would otherwise leak from the fuel and contaminate the primary coolant. Sometimes, because of a lack of uniformity in manufacturing, damage during shipment or handling, or as a result of unequal corrosion rates or temperature gradients, the cladding develops pinhole failures or defects through which fission products such as tritium may escape.

All new water-cooled power reactors being built in the United States are using zirconium-clad fuels; however, several older reactors use stainless steel clad fuel. Stainless steel is being replaced by zircaloy cladding because of the increased corrosion resistance and more favorable nuclear properties.

Experience has shown that the fraction of tritium escaping from fuel which is clad with zircaloy-2 is significantly less than that escaping from stainless steel clad fuels. This loss of tritium may be due to fewer defects in zirconium cladding or the ability of zirconium to combine chemically with hydrogen, thus limiting diffusion through the cladding and, consequently, limiting the release of tritium to the coolant.

Tritium produced by activation. Tritium is produced in water reactors by the activation of the naturally occurring deuterium present in the cooling water. Because of the small amount of deuterium in water and the low probability of its capturing a neutron to form tritium, this source of tritium in light water reactors is insignificant. In heavy water reactors which are cooled or moderated with deuterium oxide (D_2O), however, activation is the major source of tritium. Heavy water reactors for power production are not used in the United States at the present time but may be developed at some time in the future. There are, however, several heavy-water plutonium production reactors and small heavy-water moderated research reactors in operation.

A comparison of the tritium concentrations in the primary coolant of light and heavy water reactors is shown in the following table:

Reactor type	*Tritium concentration (microcurie per milliliter)*
Heavy water	7200
Pressurized water reactor	1.5
Do	.14
Do	.044
Boiling water reactor	.015

SOURCE: reference 5.

Tritium is also the product of neutron capture reactions in boron-10, boron-11, lithium-7 and lithium-6. Lithium-7 is usually formed by the $^{10}B(n,\alpha)^{7}Li$ reaction when boron is used in nuclear reactors for reactivity control.

In the past, lithium-6 was used in the coolant for pH control or in coolant purification demineralizers. This use of lithium-6 resulted in the production of elevated tritium concentrations in the primary coolant.

The use of lithium-6 has been abandoned for this reason and replaced by ammonia or lithium-7. Tritium may also be formed by the $^{14}N(n,T)^{12}C$ reaction with ammonia in the coolant or nitrogen in the containment atmosphere. The magnitude of these sources has not been fully assessed.

In boiling water reactors (BWR) boron is used for control in the form of boron carbide either as cruciform control rods or as absorber plates (curtains). Weaver and Harward (*6*) describe the differences in design between this reactor and a pressurized reactor. The rods are movable and function to control the power level of the reactor. Boron absorber curtains are generally used during the first fuel cycle to compensate for the lack of buildup of neutron absorbing fission products (poisons) and are then removed.

Both the absorber plates and the fuel rods are clad in similar fashion so that tritium which is formed from the boron must escape through the cladding by the same mechanism as the fission-product tritium.

Pressurized water reactors (PWR) use boron for reactivity control in the form of boric acid dissolved in the primary coolant. This chemical shim is used to obtain a more uniform power distribution and to compensate for long term changes in fission-product poison buildup, uranium burnup, and changes in neutron flux levels. The initial concentration of boron ranges from several hundred to several thousand ppm and decreases during the operating life of the fuel. Table 1 shows estimated tritium production rates in a typical 1,000 Mwt light water reactor for various irradiation times (*7*).

The production of tritium can also occur in high temperature gas-cooled reactors by ternary fission and by activation of helium-3 found in trace amounts in the helium coolant. The following table shows the estimated tritium production rates in a 120 Mwt air-cooled reactor (*5*).

Source	*Production rate (curies per year)*
Ternary fission	605
^{3}He activation in coolant	91
Total	696

SOURCE: reference 5.

Tritium release mechanisms. The tritium concentration in the primary coolant system is strongly dependent upon the coolant makeup rate or residence time of the coolant in the reactor. The effect of the average residence time of the coolant in the reactor system on coolant tritium concentrations is shown for boiling water reactors in table 2 and for pressurized water reactors in table 3. In both tables, the contribution of fission-product tritium is shown as a function of the fraction of the tritium which passes through the fuel cladding. The tritium produced by activation of the boron encased in the control rods of a boiling water reactor (BWR) has been assumed to remain within the rod and does not contribute to tritium levels in the coolant (*7*). The tritium level from the use of boron in a PWR varies with the boron concentration in the coolant and consequently shows a considerable range as shown in table 3.

Table 4 shows tritium levels in the primary coolant of several operating light water reactors. These measurements were obtained by the staff of the Northeastern Radiological Health Lab-

Table 1. Tritium production rates in a 1,000 Mwt light water nuclear reactor [1]

Irradiation time	Tritium produced (curies)		
	Activation of deuterium in coolant	Ternary fission	Boron and lithium reactions [2]
1 day	0.0003	13	50
1 week	.002	90	400
1 month	.008	400	1,600
1 year	.09	5,000	20,000

[1] Reference 7.

[2] These estimates represent upper limit production rates assuming boron control rods, boron chemical shim, and lithium-6 purification resins.

Table 2. Calculated tritium concentrations in the coolant of a 1,000 Mwt boiling water reactor [1]

Residence time of coolant	Coolant tritium concentrations (μCi per liter)				
	From deuterium activation	From ternary fission [2]			
		100 percent	10 percent	1 percent	0.1 percent
1 day	0.002	70	7	0.7	0.07
1 week	.01	500	50	5	.5
1 month	.04	2,000	200	20	2
1 year	.5	30,000	3,000	300	30

[1] Adapted from table II, reference 7.

[2] As a function of percent tritium leakage through cladding.

Table 3. Calculated tritium concentrations in the coolant of a 1,000 Mwt pressurized water reactor [1]

Residence time of coolant	Coolant tritium concentrations (μCi per liter)					
	From deuterium activation	From ternary fission [2]				From boron in coolant [3]
		100 percent	10 percent	1 percent	0.1 percent	
1 day	0.002	70	7	0.7	0.07	3-300
1 week	.01	500	50	5	.5	200-2,000
1 month	.04	2,000	200	20	2	800-8,000
1 year	.5	30,000	3,000	300	30	10,000-100,000

[1] Adapted from table II, reference 7.

[2] As a function of percent tritium leakage through cladding.

[3] Varies with boron concentration which decreases with time since last refueling.

oratory of the Bureau of Radiological Health. If the data for the three boiling water reactors are compared with the calculated tritium concentrations shown in table 2 for a residence time of 1 month, it can be estimated that approximately 1.0 percent of the fission-product tritium diffused through the stainless steel cladding, whereas only about 0.1 percent diffused through the zirconium cladding.

A similar comparison of the primary coolant tritium activities in PWR plants shown in table 5 with the calculated values in table 3 indicates that boron in the coolant appears to be the predominant source of tritium in the primary water. Based on a residence time of approximately 1 month, the only other source which could produce the observed levels would have to be leakage of almost all the fission-product tritium from the fuel. There is no apparent basis for assuming a higher tritium leak rate from the fuel of a pressurized water reactor than that estimated for a boiling water reactor.

Discharges From Nuclear Powerplants

Tritium which has entered the coolant of a nuclear reactor can reach the environment by several pathways. The number of pathways available and the relative amount of tritium discharged by each route depend upon the reactor design and reactor type.

Boiling water reactors. In a boiling water reactor the primary coolant flows through the reactor core, is converted to dry steam and then is fed to the turbine generator to produce power. The coolant is returned to the reactor through a condenser which removes the excess heat still remaining in the steam. The condenser cooling water and the reactor coolant are in close proximity in the condenser. This interface is maintained under a vacuum so that any leakage would be into the reactor and not to the environment.

Air ejectors on the condenser remove the oxygen and hydrogen produced by the dissociation of water in the reactor, air which has leaked into the condenser, and residual water vapor. The air and noncondensable gases are discharged to the plant stack through the off-gas system. Tritium in the form of elemental hydrogen gas may be discharged through the air ejector but a major part of the tritiated water vapor will be removed by condensation. Because of the long radiological half-life of tritium, its concentration in the gaseous stack effluent is not reduced by the 30-minute delay of waste gases in the system.

Pressurized water reactors. A pressurized water reactor consists of two coolant systems. The primary system conducts superheated water through the reactor core to steam generators and returns it to the reactor. The steam generator consists of rows of U-shaped tubes through which the heated primary coolant passes. Surrounding the tubes is the secondary coolant.

Because of a pressure difference between the primary and secondary system (2,000 pounds per square inch in the primary as opposed to about 1,000 pounds per square inch in the secondary) it is possible to convert the secondary coolant into steam. The secondary system then follows a path similar to the primary coolant of a BWR going through turbines and the condenser.

An important difference between boiling and

Table 4. Tritium in operating power reactors [1]

Reactor and type	Power level (Mwt)	Type of fuel cladding	Boron used in coolant	Tritium concentration μCi per liter		
				Primary coolant	Secondary coolant	Discharge canal water
A. Pressurized water reactor	600	Stainless steel	Yes	1,560	0.41	0.024
B. Pressurized water reactor	585	do	No [2]	2–44	(3)	.007
C. Pressurized water reactor	80	do	No [4]	145	(3)	(3)
D. Pressurized water reactor	50	Aluminum	No	116	.45	.019
E. Boiling water reactor	256	Stainless steel	No	8.8	(5)	.009
F. Boiling water reactor	240	do	No	13.3	(5)	.002
G. Boiling water reactor	700	Zirconium	No	[6] 1.6	(5)	[6] .0006

[1] Data from Northeastern Radiological Health Laboratory, Bureau of Radiological Health.

[2] Converted in late 1965 to a stainless steel fuel with full chemical shim (boron in coolant).

[3] Not reported.

[4] Boron is used for shutdown but not during routine operation.

[5] Not applicable.

[6] Data from Nuclear Engineering Laboratory, Bureau of Radiological Health.

NOTE: J. C. Drobinski, Jr., and E. J. Troianello of the Bureau's Northeastern Radiological Health Laboratory analyzed the tritium samples.

Table 5. Liquid radioactive waste discharges from operating power reactors [1]

Reactor and type [2]	Total annual discharge (curies)		Condenser cooling water available for dilution (gallons per minute)	Fraction of AEC discharge limit concentrations (percent) [3]	
	Gross activity less tritium	Tritium		Gross activity [4]	Tritium [5]
A. Pressurized water reactor	0.01	1,300	14,000	0.001 –0.13	0.16
B. Pressurized water reactor	11.1	500	260,000	.03 –4.7	.03
E. Boiling water reactor	.01	10	28,000	.0002– .05	.006
F. Boiling water reactor	1.3	20	100,000	.3 –1.2	.0035
G. Boiling water reactor	4.1	5–10	167,000	.3 –3.8	.001

[1] Reference 8.

[2] Corresponds to reactor designations in table 4.

[3] Reference 1.

[4] Applicable limit for continuous discharge for mixtures for radionuclides averaged over 12 months was 10^{-6}μCi per ml.

[5] Applicable limit is 3×10^{-3}μCi per ml.

pressurized water reactors for consideration of tritium releases is that in a PWR, leakage between the primary and secondary coolants must occur before the secondary system will be contaminated by tritium or other fission products. As shown in table 4, the tritium concentration in the secondary coolant of pressurized water reactors is only a small fraction of the primary coolant concentration. Thus, a much smaller amount of the tritium will be discharged in the form of elemental hydrogen through the condenser air ejector of a PWR as compared with a BWR.

Release of gaseous tritium. The amount of tritium discharged from both boiling and pressurized water reactors in a gaseous form is only about 1 percent of the total tritium discharge (*7*). Most of the tritium is released as tritiated water together with the liquid radioactive waste. In both types of reactors, a small part of the primary coolant water is continually drawn off, depressurized, cooled, and purified to remove fission and activated corrosion products.

The coolant stream is purified by filtration and demineralization which remove insoluble and soluble radionuclides. Gaseous activity, mainly noble gases and coolant activation products, is removed during coolant depressurization and then transferred to the gaseous radioactive waste processing system. Traces of tritiated water vapor and gaseous tritium are released from the coolant during this process.

Release of tritium in liquid wastes. Because tritium is in the form of tritiated water, it is not removed from the coolant by either filtration or ion-exchange. The purified water containing tritium may be transferred back to the reactor coolant system, stored for future use, or transferred to the liquid radioactive waste processing system.

The primary coolant is not continuously discharged to the waste disposal system. Leakage of the coolant from reactor coolant pumps and other components, however, can occur. This waste will be collected by the plant drainage system which drains to the liquid holdup tanks of the waste processing system.

The largest discharge of primary coolant to the waste treatment facility usually occurs after the reactor has been shut down for fuel reloading or maintenance. During fuel loading, water is added to the reactor cavity to provide radiation shielding and heat removal for the spent fuel elements as they are transferred from the reactor to the spent fuel storage pit.

The refueling water can mix freely with the reactor coolant and before startup this excess water must be drained from the reactor vessel. This excess water represents a considerable volume of the liquid processed by the radioactive waste system. It also contains considerable boron and, therefore, leads to additional tritium production after startup of the reactor. Liquid displaced by expansion of the reactor coolant as the plant power level increases during startup is also carried over to the waste processing system.

If the radioactivity level is sufficiently low, as determined by radiochemical analysis, the purified coolant may be discharged to the environment with the condenser cooling water. Liquid radioactive wastes consisting of purified reactor coolant, laundry wastes, and leakage from pumps and valves are stored in holding tanks before treatment. Treatment processes may include filtration, demineralization, or evaporation.

Although these processes are effective in reducing the concentration of other radionuclides by several orders of magnitude, generally they have no effect on tritium removal. The tritium is discharged to the condenser cooling water at almost the same concentration as in the reactor coolant except for a small amount of dilution by nontritiated wastes. The effluent concentrations are much lower, however, because of the large amount of dilution provided by the condenser cooling water.

Table 5 shows annual liquid radioactive waste discharges for several operating nuclear reactors. Tritium may comprise between 50 and almost 100 percent of the total amount of radioactive material discharged as liquid waste. Tritium discharge concentrations, however, are usually much less than 1 percent of the discharge limit as specified by the Code of Federal Regulations (*1*).

Tritium Detection

Tritium is undetectable by conventional methods of gross radioactivity analysis. Because tritium is usually in the form of tritiated water, it is usually evaporated during the procedures used to prepare water samples for radioactivity analysis. In addition, because of the low energy of the beta particle emitted during the decay of tritium, it is not detected by conventional gross beta counting techniques or in-line plant monitors.

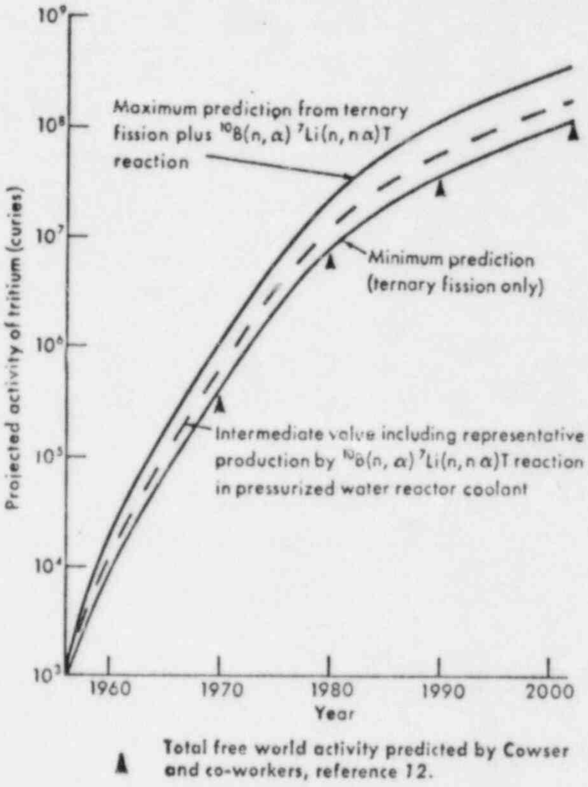
As a result of these limitations, special analytical techniques such as liquid scintillation counting must be used to measure tritium.

Limits for the discharge of unidentified mixtures of radionuclides cannot be correctly applied to tritium and due to the lesser biological hazard resulting from tritium, would be unnecessarily restrictive. The analytical techniques used to measure the activity of these mixtures usually include gross beta analysis, and since this technique is insensitive to tritium it would not be detected. Tritium may be separated from most other radionuclides by distillation of tritiated water. Tritium analysis can be made on the distillate using liquid scintillation counting. Conventional gross beta counting techniques can be used for the residue. The discharge limit can be calculated by ratioing the measured concentrations to the appropriate discharge limits ($3 \times 10^{-3} \mu$Ci per ml. for tritium and $3 \times 10^{-8} \mu$Ci per ml. for unidentified activity measured by gross beta analysis) and summing them. The limit $3 \times 10^{-8} \mu$Ci per ml. may be raised to $10^{-7} \mu$Ci per ml. if ^{129}I, ^{226}Ra, and ^{228}Ra are known to be absent. If the sum of the ratios is less than 1.0, the mixture may be released to the environment. If greater than one, further analysis or dilution is necessary before the liquid can be released.

Tritium Surveillance by BRH

Since 1964, the Radiation Surveillance Branch of the Bureau of Radiological Health has been operating a tritium sampling network which collects weekly samples of water for tritium analysis from 10 locations throughout the United States. Eight stations are downstream from nuclear facilities, and the other two serve to establish baseline levels. Samples from this network are analyzed monthly by the Bureau's Southeastern Radiological Health Laboratory.

Projected cumulative activity of tritium produced in power reactors in the United States



▲ Total free world activity predicted by Cowser and co-workers, reference 12.

Results of the analysis of samples from the tritium sampling network are reported periodically in *Radiological Health Data and Reports* (*9, 10*). A special project to study tritium in surface waters of the western States is currently being conducted by the Southwestern Radiological Health Laboratory.

Environmental tritium concentrations reported by the tritium sampling network for 1964–66 ranged between 2×10^{-6} and 2×10^{-5} microcuries per milliliter (2-20 nCi per liter) (*9, 10*). Population exposure rates from continuous ingestion of this water would range from 0.33 to 3.3 mrem per year. The maximum concentration reported would contribute less than 3 percent of the normal average population exposure rate of 125 mrem per year from all natural sources of radioactivity (*3*).

The results obtained from this sampling network indicated, with one exception, that waters downstream from nuclear facilities did not show any significantly higher tritium concentrations than streams which did not have operating nuclear reactors on them (*9*). The one exception was a plantsite with several operating heavy-water moderated reactors on it. Releases of tritium in the liquid wastes from this plant did elevate tritium concentrations in the adjacent river above background levels. Tritium concentrations, however, amounted to less than 1 percent of the Atomic Energy Commission's discharge limit of $3 \times 10^{-3} \mu$Ci per ml.

A study by the nuclear engineering department of the University of Cincinnati, under a contract with the Bureau of Radiological Health, is developing projections of future radionuclide inventories based upon predicted growth trends in the nuclear power industry (*11*). The estimated tritium activity produced by increasing utilization of nuclear energy for electric power production forecast from this study is shown in the chart together with estimates made by the Oak Ridge National Laboratory (*12*). Based upon these projected levels, the total tritium produced from nuclear power facilities in the year 2000 will be more than 10^8 curies or approximately 5 percent of the maximum tritium activity present in 1963. At present, tritium releases from operating power reactors are only a small fraction of the discharge concentrations permitted by the Code of Federal Regulations and do not constitute a danger to health. The Public Health Service will continue to monitor the environment in the vicinity of nuclear reactors and fuel reprocessing plants and will evaluate any buildup of tritium in terms of a future hazard to health.

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