TECHNICAL NOTE
ORP/TAD-76-3

PUBLIC HEALTH CONSIDERATIONS OF CARBON-14 DISCHARGES FROM THE LIGHT-WATER-COOLED NUCLEAR POWER REACTOR INDUSTRY

THE UNITED STATES
ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF RADIATION PROGRAMS

JULY 1976

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July 1976

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PREFACE

The Office of Radiation Programs of the Environmental Protection Agency carries out a national program designed to evaluate public health impact from ionizing and nonionizing radiation, and to promote development of control necessary to protect the public health and the environment. This report of current findings on public health considerations of carbon-14 discharges from the light-water-cooled nuclear power reactor industry was prepared to provide information to Federal, State, and local agencies as well as the public. Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors.

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ACKNOWLEDGEMENT

The authors gratefully acknowledge the contribution of C. B. Nelson who developed the modified Machta world-wide carbon-14 code and assisted in the preparation of the description of this code presented in this report.

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1. INTRODUCTION

Carbon-14 (C-14) is produced in nuclear power reactors by a variety of nuclear interactions. Any discharges of carbon-14 to the atmosphere can be considered as contributions of a permanent contaminant to the world-wide environment because of the long life of this radionuclide. Magno et al., (1) and other authors (2,3,4,5,6,7) have recently published papers estimating the discharges of carbon-14 from the light-water-cooled reactor (LWR) power industry and the impact of these discharges on the environment and public health. It has been estimated that the 100 year population dose commitment per gigawatt-year is a factor of 30 greater than for krypton-85 (1).

The potential environmental impact of carbon-14 was not considered during the early development of the LWR. Some of the influencing factors include:

a. Carbon-14 is not a fission product of significance nor is it a significant activation product in structural and shielding materials.

b. The capture cross-section of carbon-13 is low (0.9 mb) and the radioactive half-life of carbon-14 is relatively long (5,730 years (8)). Thus carbon-14 activity in nuclear reactors is produced in small quantities.

c. The major production of carbon-14 in the fuel results from the (n, p) reaction with nitrogen-14 which is present as an impurity in the fuel.

d. The major production of carbon-14 in the coolant results from the (n, alpha) reaction with oxygen-17 which is only present in its natural abundance of 0.037%.

For these reasons it appears likely that no comprehensive analysis was undertaken to determine the impact of carbon-14 from the nuclear power industry; however, environmental measurements had been made by Federal agencies in connection with atmospheric nuclear weapons testing (9).

This paper includes estimates of carbon-14 production in reactors and analyses of carbon-14 behavior in various waste treatment systems at both reactors and spent fuel reprocessing plants. Environmental transport models and carbon-14 dosimetric models are briefly reviewed to ascertain some probable errors in estimating the impact of carbon-14 on man and the environment. To the extent possible, this report provides an analysis of current findings and projects what work must be performed to provide answers to questions concerning the need for the control of nuclear power sources of this environmental contaminant.

2. CARBON-14 PRODUCTION IN LWR'S

Essentially all of the carbon-14 produced in LWR's results from the following two neutron induced reactions:

Reaction	Thermal Neutron Cross Section (2200 m/sec)	Natural Abundance of Target Element
nitrogen-14 (n, p) carbon-14	1.81 barns (1)	99.635% (10)
oxygen-17 (n, α) carbon-14	0.24 barns (1)	0.037% (10)

Other neutron induced reactions which produce carbon-14 in LWR's but at levels that are insignificant compared to the two above because of abundance and/or cross sections are:

Reaction	Thermal Neutron Cross Section (2200 m/sec)	Natural Abundance of Target Element
carbon-13 (n, γ) carbon-14	9E-4 barns*(1)	1.108% (10)
nitrogen-15 (n, d) carbon-14	2.4E-7 barns (11)	0.365% (10)
oxygen-16 (n, He-3) carbon-14		99.759% (10)

The production of carbon-14 by fission is negligible (U-235 fission yield is 1.7E-6 (12)) by comparison to that produced by the two major neutron induced reactions. The reactions of neutrons with nitrogen-14 and oxygen-17 will be taken in this report as giving adequate estimates of the total production of carbon-14 in the fuel and in the coolant at both BWR's and PWR's.

The activation equation used to calculate the production of carbon-14 in LWR's is as follows:

$$A = N\phi\sigma\,(1-e^{-\lambda t}) \times (1\ Ci/3.7E{+}10\ dis/sec)$$

where A = produced C-14 activity in curies
N = target atoms
ϕ = thermal neutron flux
σ = thermal neutron cross section
λ = decay constant for C-14 in years^{-1}
t = irradiation time in years

For λ = 1.2E-4 year^{-1} and t = 3 years, the activation equation is $A = (9.81E{-}15)\ N\phi\sigma$.

* 9E-4 = 9×10^{-4}

2.1 Carbon-14 Production in the Fuel

The activation calculation is based on the target atoms in a fuel loading of 33.5 MTHM and a three year exposure period. The 33.5 MTHM fuel loading produces a nominal electric power output of 1 GWe-yr for a burnup of 33,000 MW_t-days/MTHM and a thermal efficiency of 33%. Carbon-14 is calculated to be produced at the rate of 4 Ci/GWe-yr in the fuel by the (n, α) reaction on O-17 and 18 Ci/GWe-yr by the (n, p) reaction on N-14. The data base used to calculate these production rates in the fuel are discussed in the following paragraphs.

N (target atoms)

For 33.5 MT of uranium, there is 38 MT UO_2 containing 4.5E+6 grams of oxygen. Using a natural abundance of 0.037% for O-17, there is 1.6E+3 grams or 5.9E+25 atoms of O^{17} in the fuel loading [N(O-17) = 5.9E+25].

Nitrogen is present in the fuel interstices as an impurity and thus can vary significantly. For the purposes of this estimate, twenty ppm (by weight) is the assumed amount of nitrogen present as an impurity in the fuel (7). Therefore in a 33.5 MTHM fuel loading, there is 7.6E+2 grams nitrogen and 3.26E+25 atoms of N-14 in the fuel [N(N-14) = 3.26E+25].

ϕ (average thermal neutron flux)

An average thermal neutron flux of 5E+13 n/cm^2-sec for the fuel is assumed in this report (13,14).

σ (thermal neutron cross section)

For estimates of the production rate of carbon-14 in the fuel, consideration of the thermal neutron cross section at 2200 m/sec alone would result in high production rates since the cross sections for both reactions of interest varies as 1/v. Therefore, the previously presented cross sections at 2200 m/sec were multiplied by 0.6 to correct for the energy dependence of the thermal neutron cross section and the spectrum of thermal neutrons (13,14) [σ (N-14) = 1.1E-24 cm^2 and σ (O-17) = 1.4E-25 cm^2].

2.2 Carbon-14 Production in the BWR Coolant

The volume of coolant in the BWR/6 is 1,872 cubic feet at 1062 psia and 540°F (15). The specific volume of water at these temperature and pressure conditions is 0.0215 cubic feet per pound (16). Therefore, there is 39.5 MT of water in the BWR/6 core. The thermal power level for the BWR/6 is 3579 MWt which is equivalent to a nominal electric power output of 1.18 GWe for a thermal efficiency of 33%. A one year exposure period is used to calculate 8.9 Ci/GWe-yr in the BWR coolant by the (n, α) reaction on O-17 and 0.26 Ci/GWe-yr by the (n, p) reaction on N-14. The data base used to calculate these production rates in the BWR coolant are discussed in the following paragraphs.

N (target atoms)

For 39.5 MT of water, there is 4.6E+26 atoms of O-17 [N(O-17) = 4.6E+26]. No data could be found on the concentration of nitrogen dissolved in the coolant. However, since nitrogen impurities in the fuel proved to be the greatest source of carbon-14 production there, it is concluded that the production of carbon-14 by the reactions of neutrons on nitrogen-14 should be examined. One ppm (by weight) of nitrogen as an impurity in the water is assumed (17). One ppm of nitrogen in the coolant equates to 39.5 grams of nitrogen and 1.69E+24 atoms of nitrogen-14 in the BWR coolant [N(N-14) = 1.69E+24].

ϕ (average thermal neutron flux)

The simplifying assumption is made that the average thermal neutron flux in the fuel and coolant are equal. Therefore, an average thermal neutron flux of 5E+13 n/cm^2-sec in the coolant is assumed in this report.

σ (thermal neutron cross section)

The cross sections used to calculate the carbon-14 production rate in the fuel are used [σ (N-14) = 1.1E-24 cm^2 and σ (O-17) = 1.4E-25 cm^2].

2.3 Carbon-14 Production in the PWR Coolant

The volume of coolant in the core of the PWR is calculated to be 685 cubic feet using the effective cross section flow area of

the core (54.8 ft^2) and the core height of active fuel (12.5 ft) (18). The average temperature in the core coolant is 588°F and the operating pressure is 2235 psig. The specific volume of water at these conditions is 0.0226 cubic feet per pound (16). Therefore, there is 13.7 MT of water in the PWR core. The thermal power level for the chosen PWR (18) is 3473 MWt which is equivalent to a nominal electric power output of 1.146 GWe for a thermal efficiency of 33%. A one year exposure period is used to calculate 3.2 Ci/GWe-yr in the PWR coolant by the (n, α) reaction on O-17 and 0.09 Ci/GWe-yr by the (n, p) reaction on N-14. The data base for the average thermal neutron flux in the PWR coolant and the thermal neutron cross sections are identical to those used for the BWR coolant calculation. The data base for the target atoms in the coolant is discussed in the following paragraph.

N (target atoms)

For 13.7 MT of water in the PWR coolant, there is 1.6E+26 atoms of oxygen-17 [N(O-17) = 1.6E+26]. One ppm (by weight) of nitrogen as an impurity in the PWR core coolant is assumed (19). One ppm of nitrogen in the PWR coolant equates to 13.7 grams of nitrogen and 5.87E+23 atoms of nitrogen in the coolant [N(N-14) = 5.87E+23].

2.4 Total Carbon-14 Production in a LWR

The production rates of carbon-14 are summarized in Table 1 where it can be seen that they agree within a factor of two of those reported by Bonka et al. (3), Hayes et al. (12), ERDA (20), and Kelly et al. (7). Nitrogen impurities in the fuel is shown to be the greatest source of production of carbon-14 in the fuel. Reactions on oxygen-17 is shown to be the greatest source of production of carbon-14 in the coolant. Production rates for the BWR and PWR coolant as presented by ERDA (20) and Kelly et al. (7) are derived from measured discharges (21,2) and are about two times higher than the theoretical estimates presented in this report. It is also noted that the BWR coolant production rate is about a factor of 3 higher than that for the PWR coolant. The higher production rate for the BWR is probably due to the higher volume of water in the core where the (n, α) reaction on O-17 can take place.

TABLE 1

PRODUCTION OF CARBON-14 IN LIGHT WATER REACTORS

		Carbon-14 Production Rate (Ci/GWe-yr)				
		This Report	Bonka et al. (3)	Hayes et al. (12)*	ERDA-1535 (20)	Kelly et al. (7)*
BWR Fuel	O-17	4	8.4	10.9		2.7
	N-14	18	12.9	21.2		10.9
	Total	22	21.3	32.1	20**	13.6
BWR Coolant	O-17	8.9	9.9	11.5		
	N-14	.26	1.3	--		
	Total	9.2	11.2	11.5	16	16
BWR Sum (Fuel Plus Coolant)		31	32.5	43.6	36	29.6
PWR Fuel	O-17	4	7.1	4.0		2.7
	N-14	18	12.2	7.6		10.9
	Total	22	19.3	11.6	17**	13.6
PWR Coolant	O-17	3.2	9.8	3.3		
	N-14	.09	1.3	0.1		
	Total	3.3	11.1	3.4	6	6
PWR Sum (Fuel Plus Coolant)		25	30.4	15	23	19.6

* The production rates presented by Hayes et al. (12) and Kelly et al. (7) for 1000 MWt were multiplied by 3.03 (33% thermal efficiency to roughly present the values on a per GWe-yr basis for comparison purposes.

** Fuel and cladding production rates from ERDA-1535 (20) were added and identified as a fuel production rate in this table.

3. SOURCES AND TREATMENT OF CARBON-14 IN LWR'S

3.1 Gaseous Source Terms for BWR's

Carbon-14 may be discharged from all of the following gaseous release pathways:

1. Main Condenser Air Ejector Off-Gas
2. Turbine Gland Seal
3. Containment Building (Including Drywell) Purge
4. Turbine Building Ventilation
5. Radwaste Building Ventilation

Where possible, measured concentrations of C-14 in exhaust streams have been used to calculate the annual discharge of C-14 from a nominal 1250 MWe BWR. Where measured concentrations are lacking, the C-14 average reactor coolant concentration determined at Oyster Creek is used in conjunction with standard assumptions (22,23,24).

Preliminary data from the Oyster Creek study (22) have indicated an average C-14 release rate of 0.182 μCi/sec from the air ejector. When extrapolated on an annual basis (80% capacity factor) for a 1250 MWe BWR, a total of 9.0 Ci/yr of C-14 is estimated to be released. A measurement of the turbine gland seal condenser exhaust showed the release rate to be <3E-4 μCi/sec (22). This is equivalent to an annual release of <0.015 Ci/yr for a 1250 MWe BWR at 80% capacity factor.

For the remaining gaseous release pathways, the average C-14 reactor coolant concentration measured at Oyster Creek 4.0E-6 μCi/ml (22), is used in conjunction with standard reactor coolant leak rate assumptions of 500 lb/hr, 1700 lb/hr, and 1000 gpd for the containment, turbine building, and radwaste facility, respectively (23). The leakage to the radwaste facility is at 1% reactor coolant activity while the others are at reactor coolant activity. All of the C-14 in these leakages are assumed to escape to the building atmosphere with subsequent release to the environment. A sample calculation for the containment purge is as follows: The assumed leak rate into the drywell and containment building is 500 lb/hr which equates to 2000 gallons per day using the approximate specific volume of 6 lb/gallon for water. The containment purge source term is therefore 2000 gal/day x 3785 ml/gal x 4.0E-6 μCi/ml x 365 day/year x 1E-6 Ci/μCi $\overset{\text{WE}}{\Lambda}$ 0.0088 Ci/yr for a 1250 MWe plant at 80% capacity (1000 MWe) or 0.0088 Ci/GWe-yr. Table 2 lists the gaseous discharge pathways and estimated annual discharge rate.

The Oyster Creek data (22) indicates that C-14 is released both as CO_2 and as other chemical species. At both the air ejector and turbine gland seal condenser, there was about twice the release rate of ^{14}C as

CO_2 as compared to other chemical species. The data of Kunz et al. (21) for BWR's showed that 95% of the C-14 activity was in the CO_2 fraction.

3.2 Liquid Source Terms for BWR's

Liquid discharge pathways for C-14 may include:

1. Clean (High Purity) Wastes
2. Dirty (Low Purity) Wastes
3. Chemical Wastes
4. Detergent (Laundry) Wastes

Except for detergent wastes, the average measured C-14 reactor coolant concentration at Oyster Creek (22) was used in conjunction with standard assumptions for flow rates and fractions of reactor coolant activity (23,24) to estimate annual releases of C-14 from a nominal 1250 MWe reactor at 80% capacity. Actual data for the concentration of C-14 in laundry waste at Oyster Creek (0.150 pCi/ml) (22) and a flow rate of 450 gallons per day (23) was used to obtain the detergent waste estimate. Table 2 illustrates the magnitude of each of these release pathways. The total liquid C-14 source term, 0.044 Ci/yr, is <1% of the gaseous source term at BWR's.

3.3 Gaseous Source Terms for PWR's

A nominal 1250 MWe PWR of Westinghouse design with a 80% capacity factor is considered. Gaseous discharge pathways may be broken down as follows:

1. Gaseous Waste Disposal System
2. Condenser Air Ejector Off-Gas
3. Turbine Gland Seal Exhaust
4. Fuel Handling Building Ventilation
5. Containment Purge
6. Auxiliary Building
7. Turbine Building

Gaseous waste disposal system gases evolve chiefly from the gas stripper-evaporator package in the boron recycle system. Measurements of C-14 in the gas decay tanks at Haddam Neck (Connecticut Yankee), Yankee Rowe, and Ginna (2,25,26) have shown an average concentration of about 5E-4 μCi/cc. Estimates of the flow of these gases have ranged from 0.1 to 1.0 scfm (23,27). Operating data at San Onofre 1, a 430 MWe PWR, showed about 30,000 ft^3 treated in a three month period (October 11,

TABLE 2

BOILING WATER REACTOR CARBON-14 SOURCE TERMS

(Nominal 1250 MWe at 80% Capacity Factor)

	Annual C-14 Discharge Rate (Ci/yr) or (Ci/GWe-yr)
GASEOUS SOURCE TERMS	
Main Condenser Air Ejector Off-gas	9.0
Turbine Gland Seal	< .015
Containment Purge	0.0088
Turbine Building Ventilation	0.030
Radwaste Facility Ventilation	0.000044
	9.0
LIQUID SOURCE TERMS	
Clean Wastes	0.029
Dirty (Low Purity) Wastes	0.013
Chemical Wastes	0.0023
Detergent Wastes	0.000075
	0.044
Grand Total:	9

1972 to January 10, 1973), which would correspond to about 350,000 ft^3/yr (0.66 scfm) for a 1250 MWe plant. A flow of 0.66 scfm along with the measured concentration of 5E-4 μCi/cc is used to calculate a release rate of 3.8 Ci/yr from the gaseous waste disposal system for 1250 MWe PWR with an 80% capacity factor. Measurements at Haddam Neck revealed that virtually all of the C-14 was in a non-CO_2 form (26). At Ginna, almost 90% was in the CH_4 or C_2H_6 form and only about 5% in the CO_2 form (2).

Condenser air ejector effluent was measured for C-14 content at Haddam Neck and Yankee Rowe and found to contain an average of 3.6E-7 μCi/cc. Over 85% of the C-14 was in a species other than the CO_2 form (26). Assuming an air ejector flow of 25 scfm for a 1250 MWe PWR at 3.6E-7 μCi/cc and an 80% capacity factor, a source term of 0.11 Ci/yr is estimated.

To calculate the release from the steam generator blowdown vent, the measured C-14 concentration in steam generator water at Haddam Neck (1.6E-7 μCi/ml) was used along with the other conditions associated with this PWR: 5 gpm total blowdown flow rate with 35% flashing to steam (23,26). Also, 35% of the C-14 in the blowdown was assumed to exit via the blowdown tank vent. A release of 4.5E-4 Ci/yr of C-14 is estimated for a 1250 MWe PWR with an 80% capacity factor.

No measurement of the turbine gland seal was available. However, by assuming that all of the C-14 in the 50 gpd primary to secondary leakage was transferred to the steam phase and that 0.1% of the main steam flow was routed to the turbine gland seal, a conservative C-14 annual release claculation can be made. The average of the primary coolant C-14 concentrations observed at Haddam Neck and Yankee Rowe, 1.67×10^{-5} μCi/ml, was used (25,26) in this calculation to arrive at the estimate of 9.2E-7 Ci/yr of C-14 via the turbine gland seal.

Using the measured C-14 concentration of 9E-9 μCi/cc in the Haddam Neck fuel handling building along with its ventilation rate of 70 m^3/min all year (26), a release of 0.69 Ci/yr is estimated via this pathway for a nominal 1250 MWe PWR.

Two approaches were used to obtain estimates for the annual discharge of C-14 from the containment. Using the average measured concentration (during operation) of 1.5E-6 μCi/cc for C-14 in containment air at Ginna, Haddam Neck and Yankee Rowe (2,25,26) and assuming a containment volume of one million cubic feet with four purges annually, a discharge of 0.52 Ci/yr of C-14 is calculated for a 1250 MWe PWR. Alternatively, using the "standard" leak rate of 40 gpd (23) of primary coolant into the containment and the average primary coolant C-14 concentration observed at Haddam Neck and Yankee Rowe (1.67E-15 μCi/ml) (25,26) an annual C-14

release of 7.4E-4 Ci/yr may be calculated. The former value of 0.52 Ci/yr will be used, however, as it is based on a direct measurement of the containment air. Over 90% of the containment C-14 at Ginna was in the CH_4 and C_2H_6 forms.

By assuming the "standard" leak rate of primary coolant to the auxiliary building, 20 gpd cold and 1 gpd hot (23), and a primary coolant C-14 concentration of 1.67E-5 μCi/ml (25,26), an annual auxiliary building C-14 discharge of 8.0E-4 Ci/yr of C-14 is calculated for a nominal 1250 MWe PWR at 80% capacity. At Haddam Neck, a concentration of <6.0E-9 μCi/cc was measured for the auxiliary building (26). This direct measurement is not sensitive enough to serve as a base for a second calculation of the auxiliary building discharge rate, so the value estimated using leak rate assumptions is employed in this report.

Using the standard assumption of 1700 lb/hr leakage into the turbine building (23) in conjunction with the estimated secondary coolant concentration of ∿1.6E-7 μCi/ml at Haddam Neck with 50 gpd primary to secondary leakage (26) and a 80% capacity factor, this second method yields an annual C-14 discharge estimate of 8.7E-6 Ci/yr for a 1250 MWe PWR. Another method for estimating the turbine building C-14 discharge is to assume that of the total C-14 entering the secondary system, only a fraction is escaping via turbine building ventilation exhaust. This fraction is the ratio of the standard leak rate of secondary coolant to the turbine building, 1700 lb/hr (23), to the total steam flow rate, 1.4E+7 lb/hr. Using a 50 gpd primary to secondary leak rate at a primary coolant concentration of 1.67E-5 μCi/ml (25,26), an annual C-14 discharge of 1.1E-7 Ci/yr is calculated for the turbine building.

A third method of calculation is to use a measured concentration and a ventilation rate to estimate the turbine building discharge rate. At Haddam Neck, a concentration of <8E-9 μCi/cc was measured for the turbine building (26). This direct measurement is not sensitive enough to serve as a base for a comparison calculation of the turbine building discharge rate, so the value estimated using leak rate assumption is employed in this report.

As in the case with BWRs, C-14 may appear associated with different chemical species. C-14 found in the gas decay tanks at Ginna was in the CH_4 and C_2H_6 forms (2). At Haddam Neck, virtually all of the decay tank gas was in the non-CO_2 form, whereas 80% of the containment C-14 and over 85% of the air ejector C-14 at Haddam Neck was in the

non-CO_2 form (26). Over 90% of the containment C-14 at Ginna was associated with CH_4 and C_2H_6 (2). Table 3 illustrates the PWR gaseous C-14 release pathways and the estimated magnitude of each.

3.4 Liquid Source Terms for PWR's

Four principal liquid waste streams may be identified at a Westinghouse PWR:

1. Boron Recycle System
2. Liquid Waste Disposal System
3. Steam Generator Blowdown
4. Turbine Drains

Using a flow rate of 1 gpm (23) through the boron recycle system, assumed to be at a primary coolant C-14 concentration of 1.67E-5 μCi/ml (25,26), and assuming that only 10% of the boron recycle liquids are discharged, a release of 2.7E-3 Ci/yr of C-14 may be calculated (at 80% capacity factor).

The release of C-14 from the liquid waste disposal system may be estimated using the "standard" assumptions for liquid waste stream flow rates and fractions of primary coolant activity (23), and the measured average primary coolant C-14 concentration of 1.67E-5 μCi/ml (25,26). A release of 1.9E-3 Ci/yr of C-14 is estimated from the liquid waste disposal system.

A steam generator blowdown average concentration of 1.6E-7 μCi/ml and a blowdown rate of 5 gpm (total) with a 50 gpd primary to secondary leak was reported at Haddam Neck (26). If 65% of the blowdown liquid remains as a liquid waste after flashing (23), a release rate of 8.3E-4 Ci/yr of C-14 is calculated for the steam generator blowdown discharge pathway.

Data for the Haddam Neck plant showed a turbine drain (secondary system leakage) flow of 7570 gpd at a C-14 concentration of about 1.6E-7 μCi/ml, resulting in an annual C-14 discharge estimate of about 1.3E-5 Ci/yr (26).

Table 3 illustrates each of these liquid discharge pathways and the estimated C-14 source term for each. As was the case for BWRs, virtually all of the C-14 is discharged by gaseous pathways.

TABLE 3

PRESSURIZED WATER REACTOR CARBON-14 SOURCE TERMS
(Westinghouse Design, Nominal 1250 MWe at 80% Capacity Factor)

GASEOUS SOURCE TERMS	Annual C-14 Discharge Rate (Ci/yr) or (Ci/GWe-yr)
Gaseous Waste Disposal System	3.8
Condenser Air Ejector Off-gas	0.11
Steam Generator Blowdown Tank Vent	4.5E-4
Turbine Gland Seal	9.2E-7
Fuel Handling Building Ventilation	0.69
Containment Purge	0.52
Auxiliary Building Ventilation	8.0E-4
Turbine Building Ventilation	8.7E-6
	5.1
LIQUID SOURCE TERMS	
CVCS (Boron Recycle System)	.0027
Liquid Waste Disposal System	.0019
Steam Generator Blowdown	8.3E-4
Turbine Drains	1.3E-5
	5.4E-3

Grand Total: 5.1

3.5 Source Term Summary

It is evident that good direct measurements of C-14 in many of the discharge pathways are lacking. What measurements exist were obtained at light water reactors of early design. Therefore, great reliance has been placed on "standard" source term assumptions (23) as an aid in calculating C-14 source terms for reactors of the 1250 MWe power level. Furthermore, there is uncertainty as to the chemical species in which C-14 may occur, which has tremendous implications relative to any proposed treatment technologies.

C-14 production in coolant in light water reactors has been reported to be in the range of 9 to 16 Ci/GWe-yr for BWR's and 3 to 11 Ci/GWe-yr for PWR's as previously shown in Table 1. Estimates of C-14 in gaseous effluents have ranged from 6-11 Ci/GWe for PWRs and up to 8 Ci/yr for a 600 MWe BWR, most of which is produced by the reaction of neutrons with oxygen-17 in the coolant (1, 2, 21). These estimates are in reasonable agreement with the gaseous source terms derived in this report based on measurements: 9.0 Ci/GWe-yr for BWR's and 5.1 Ci/GWe-yr for PWR's. Also, the liquid C-14 source term is much smaller (< 1%) than the gaseous source term for each reactor type, based on the calculated source terms in this report.

3.6 Carbon-14 Control for BWR's and PWR's

At boiling water reactors, the condenser air ejector contributes about 98% of the total BWR C-14 source term (Table 2). The great majority of BWRs are proposing the use of recombiners and charcoal (at various temperatures) to control the release of noble gases from the air ejector. Under the operating conditions required to recombine elemental hydrogen and oxygen in the off-gas stream, hydrocarbons will be converted to carbon dioxide and water. Therefore, it is expected that C-14 will be primarily in the form of carbon dioxide at BWRs. Those BWRs that are not employing the recombiner-charcoal combination have typically proposed cryogenic distillation, which has CO_2 removal equipment incorporated as normal design practice. Potential processes for removal of CO_2 from the recombiner effluent (total volume flow: <30 scfm per 1,000 MWe) include caustic scrubbing, soda lime absorption, molecular sieves, and low temperature adsorption on silica gel or carbon. Considering only caustic scrubbing, with a nominal 90% CO_2 removal efficiency, total BWR releases of C-14 would be reduced by almost a factor of 10. BWR cryogenic distillation systems employ refrigerated carbon beds and liquid nitrogen-cooled "freezeout coils" to remove CO_2.

Approximately 75% of the C-14 from pressurized water reactor results from the gaseous waste disposal system. Many PWRs of Westinghouse design are planning to use the "cover gas recycle system," which results in no release (except for leakage) of radioactive waste system gases over the life of the plant. A few PWRs have chosen cryogenic charcoal adsorption, in which C-14 in the form of hydrocarbons will be converted to the CO_2 form. Again, caustic scrubbing could be introduced to remove C-14 as CO_2 with a nominal efficiency of 90%. Thus, PWR C-14 releases could be reduced from 5.1 Ci/yr to about 1.6 Ci/yr, a reduction of almost 70% in total C-14 release from a 1250 MWe PWR.

Cost estimates for CO_2 scrubbers may vary because of materials of construction and complexity of design and operation. Furthermore, the size of scrubbers required for PWRs is much smaller than employed by many non-nuclear industries. Indications are that a 100 cfm caustic scrubber (for BWR condenser air ejector off-gas after recombiner treatment) would result in an equipment cost of only a few thousand dollars, approximately $4,000, in 1970 dollars (28). Capital cost would therefore be in the $7,000-$8,000 range. Total annual costs (capitalization plus operating and maintenance) are estimated at about $4,000. Upgrading these estimates by the Marshall and Swift Cost Index to September 1974 (for chemical industries), the estimated capital cost of a caustic scrubber would be about $11,000 (29). Total annual costs in September 1974 dollars are estimated at about $6,000. Scrubbers for use in a PWR off-gas stream would be even less expensive as flow capacity would have to be only 10 scfm or less. However, costs for a PWR CO_2 caustic scrubber are taken as half those for a BWR, i.e., capital cost as $5,500, and total annual operating cost as $3,000.

It is possible that a solid adsorber, such as charcoal, or a solid absorber, such as an alkali metal alumino-silicate molecular sieve, would be even less expensive and perhaps more desirable in terms of handling and ultimate waste disposal. However, specific designs and cost information were not readily available.

On February 5, 1976, the U.S. Environmental Protection Agency awarded a contract to Science Applications, Inc. to assess carbon-14 control technology for LWR's. The contract which is expected to be completed within eight months after awarding should provide needed specific design and cost information. The contract report will address: (1) the capabilities of current LWR off-gas systems for control of C-14, (2) the cost of modifying current LWR off-gas cleanup systems, if possible, to control C-14, (3) the availability and cost of

specialized carbon-14 control systems for use at both LWR's and LWR fuel reprocessing plants, and (4) the potential capability for C-14 control and costs of off-gas treatment systems which have been proposed for use at LWR fuel reprocessing plants for control of other contaminants, such as krypton-85 and radioiodine. The information to be developed in these studies may point out certain dual-control capabilities for some off-gas treatment systems (i.e., such as control of both krypton-85 and carbon-14 by one system in a reprocessing plant) and thus alter other cost-effectiveness analyses.

4. CARBON-14 AT LWR FUEL REPROCESSING PLANTS

Spent fuel from LWR's can be reprocessed to recover valuable uranium and plutonium and to process the radioactive waste into forms more suitable for disposal than the spent fuel form. Current plans for reprocessing spent fuel include a cooling period of about 150 days to permit decay of the short-lived radionuclides, especially I-131. The control of radioactive iodine has proven to be most difficult but must be accomplished due to the extremely long half-life of I-129. Because of its volatility most of the iodine is released into the process off-gas systems at reprocessing plants where it constitutes one of the major contaminants, along with particulates, tritium, krypton-85 and NOx. Recently, Magno et al. (1) pointed out that there may be significant quantities of carbon-14 in the spent fuel and assumed that all of the carbon-14 would be discharged to the atmosphere. Thus, it appears that carbon-14 should be included as another major constituent, in terms of world-wide environmental and public health impact, of reprocessing plant off-gas releases.

Chapter two of this report presents a C-14 fuel production rate of 22 Ci in 33.5 MTHM or 0.66 Ci/MTHM. This production rate is comparable to the ORNL estimate of 0.464 Ci/MTHM as presented in reference (30) and the ERDA estimate of 0.4 Ci/MTHM (31). Using the production rate of 0.66 Ci/MTHM developed in this report, there would be 990 curies per year available for release at a typical LWR fuel reprocessing plant which has a throughput capacity of 1500 MTHM per year.

4.1 Behavior of Carbon in the Reprocessing Plant

The systems described here are those presented in the Barnwell Nuclear Fuel Plant separations facility final safety analysis report (32). The first process which would allow C-14 to escape is shearing. Here the fuel elements are cut into two- to five-inch segments. The segments fall by gravity into a dissolving basket and an 8 molar nitric acid dissolving solution. There is a slight air flow (250 scfm) from the shear to the dissolving solution to direct gases and particulates to the dissolver. In the dissolver the pieces are exposed to 90°C, 8 molar nitric acid (HNO_3). In this environment it is currently thought that 95-99% of the carbon present will escape to the off-gas system as CO or CO_2. The formation of monoxide or dioxide is expected because of the excess oxygen in solution from the UO_2 and HNO_3.

The off-gas process system at Barnwell, as described, is designed for the removal of particulates, radioiodine, and NOx only. The initial treatment of the dissolver off-gases is in the No. 1 iodine scrubber where the gases are scrubbed with a 0.4 molar solution of $Hg(NO_3)_2$ in 6-8 molar HNO_3 at 105°F. The gases then pass through the NOx absorber where they are scrubbed with water. The dissolver off-gases join the vessel off-gases and are scrubbed in the No. 2 iodine scrubber with 0.01 molar $Hg(NO_3)_2$ in 0.5 molar HNO_3 at 100°F. Final iodine adsorption is done using several silver zeolite beds and HEPA filters just prior to the gaseous stream being discharged to the stack. It appears that the described waste treatment system will have little effect on C-14 as CO_2 so a carbon-14 decontamination factor of 1 is assigned for a current LWR fuel reprocessing plant.

Another parameter which can be important in the air cleaning efficiency of various systems is the total quantity of contaminant present in the off-gas stream. Trace quantities of contaminants often prove difficult to collect or control. The quantity of carbon in the off-gas system of a reprocessing plant of the Barnwell design is developed as follows.

Assuming there are 990 Ci/yr released to the off-gas system, we find the C-14 flow rate is 0.138 Ci/hr, or about 0.031 gm/hr; assuming 7,200 operating hours per year. This rate must be compared to the natural abundance of CO_2 in the air. Since the standard atmosphere is 0.033% by volume carbon dioxide (33), 0.033% by volume of the air flowing in the dissolver off-gas (DOG) system is stable carbon dioxide. For comparison, these flow rates are presented in Table 4. Based on this estimate, it appears that sufficient carbon dioxide is present in the off-gas stream to permit reasonable assurance that it can be controlled. However, this factor should be recognized in the design of systems for the collection of carbon dioxide.

Because of the high molarity of the dissolving solution, the 1-5% of the C-14 remaining in the dissolver solution is likely to be incorporated into carbonic acid. From the dissolver it would proceed through the dissolver exchanger and would eventually go to the feed surge tank. From here it would pass through feed adjustment tanks, the centrifuge, the high activity feed tank, and eventually to high-level waste disposal. Since the behavior in the high activity process stream is difficult to predict, the above-mentioned path is considered the most likely but is certainly not the only potential path. However, there appears to be no set of conditions in which the C-14 would be released to the atmosphere following formation of carbonic acid.

In summary, it is estimated that at least 95 to 99% of the C-14 contained in the fuel will be released to the off-gas system and subsequently to the atmosphere since currently employed off-gas treatment systems will probably not remove CO_2 to any extent; while 1 to 5%, most likely closer to 1% than 5%, remaining in solution should remain in the high-level wastes. Therefore, a gaseous C-14 source term of nearly 990 Ci per year is estimated for a 1500 MT/yr plant using the Barnwell off-gas system design.

4.2 Control Systems at LWR Fuel Reprocessing Plants

The off-gas treatment system at Barnwell (34) is expected to remove essentially no C-14. In light of the Magno et al. findings (1), it would appear prudent to investigate the costs and removal efficiencies of carbon removal systems which could be installed at reprocessing plants. Of particular interest here would be a system in which other contaminants, such as tritium and krypton, would also be collected. Two widely used methods of carbon dioxide removal involve passing of the gas through either a lime bed or a caustic scrubber. These methods are known to be effective but produce large quantities of solid waste compared to the amount of C-14 removed. Other methods of CO_2 removal which are used on a commercial scale include ethanolamine scrubbing, hot and cold alkali carbonate scrubbing, water scrubbing, and molecular sieve adsorption. Depending on the amount of CO evolved during dissolution, catalytic conversion to CO_2 or separate systems for CO removal may be necessary. The removal efficiencies of all gas treatment systems are highly dependent on temperature, pressure, humidity, flow rate, and the fraction of the gas stream which is to be treated. Further study is necessary as to the system most applicable for utilization at a reprocessing plant.

The cryogenic process, which takes advantage of the widely differing boiling point of krypton (Kr), xenon (Xe), and CO_2 (actually the "boiling" point for CO_2 is a triple point) at atmospheric pressure, is a possibility for removing CO_2 from the gaseous effluent stream. The carbon dioxide could be frozen out prior to the Kr-Xe removal since its triple point is some 30°C higher than either the Xe or Kr boiling points. Solid CO_2 could be removed, sublimed and bottled for storage. This process has been proposed for collection of krypton in the off-gas stream. To avoid explosive concentrations of oxygen in the cryogenic systems, the proposals included a sophisticated pretreatment of the off-gas stream in which all oxygen would be removed. The carbon in this system would be converted entirely to CO_2 and could be removed as discussed above.

A second process, selective absorption, is under development for control of krypton at reprocessing plants. In this process, the off-gas is passed through a solvent (fluorocarbon solvents have received the most attention for noble gas collection) where certain gases are absorbed. In further steps, the dissolved gases are then selectively stripped out of the solvent. This process should be analyzed for the collection of CO_2.

In summary, the need for research on C-14 in reprocessing facilities exists in the following areas: amounts of C-14 produced in the fuel, chemical forms of the C-14 as it is evolved from the dissolver, unexpected chemical behavior elsewhere in any system, partitioning between the off-gas and liquid process systems, possible process pathways and reactions in those systems, probable decontamination factors for various systems, and costs of collection systems. Carbon-14 control technology and costs for LWR fuel reprocessing facilities is currently being evaluated by Science Applications, Inc. under a contract awarded by the U.S. Environmental Protection Agency. This contract is briefly discussed in section 3.6 of this report.

TABLE 4

AIR AND CARBON DIOXIDE FLOW RATES IN THE OFF-GAS SYSTEM AT BARNWELL (34)

Scrubber	Design Flow Rate (gm/hr)	CO_2 Flow Rate (gm/hr)	% of CO_2 Flow Rate which is $^{14}CO_2$*
No. 1 Iodine	1.18E+6	592	1.72E-2
NOx	1.22E+6	615	1.66E-2
No. 2 Iodine	8.62E+6	4330	2.35E-3

* On a mass basis where the $^{14}CO_2$ flow rate is 0.1 gm/hr.

5. ENVIRONMENTAL TRANSPORT OF CARBON-14

5.1 Local Transport

Carbon-14 is injected into the troposphere from LWR fuel cycle facilities and resulting local air concentrations around the points of release can be estimated using a diffusion model which estimates downwind concentrations for given site meteorological conditions. The EPA AIREM computer code (35) can be used to estimate local air concentrations based on estimated source terms and measured meteorological conditions at sites of interest. Local transport and resulting carbon-14 concentrations in local media other than air have not as yet been examined in detail in the literature for LWR carbon-14 discharges. Local media such as food should be investigated to assess whether potential concentrations of C-14 provide significant pathways. The concentration of carbon-14 in vegetation can be obtained by using the U.S. Nuclear Regulatory Commission model (42) which assumes that the ratio of carbon-14 to the natural carbon in the vegetation is the same as the ratio of carbon-14 to natural carbon in the atmosphere surrounding the vegetation. The concentration of natural carbon in the atmosphere is taken to be 0.16 gm/m^3 and the fraction of the total plant mass that is natural carbon is assumed to be 0.11.

5.2 World-Wide Transport

Carbon-14 injected in the troposphere becomes part of the carbon cycle and is constantly moving from inorganic reservoirs (CO_2 in the atmosphere and dissolved in water) to living systems and back again. Man is also affecting the carbon cycle by increasing the concentration of carbon-12 in the active carbon cycle by burning fossil fuels. The effect of these injections can only be estimated since specific carbon-14 concentrations and CO_2 concentrations are not known to a high degree of precision and carbon undergoes environmental transport processes which are not yet quantitatively defined. Predictions of world-wide future and past carbon dioxide concentrations have been derived from many different box models. The following quote from Minze Stuiver (36) gives insight into the validity of these models. "Box models that describe reservoir properties are used to assess the transfer of radiocarbon between the various reservoirs. Various simplifying assumptions have to be made for a rigorous mathematical treatment of the model. Such studies occasionally give the impression that a precise calculation provides a screen for an imprecise assumption, and the models should be considered a crude approximation only of the gross features of carbon transfer in nature." Livingstone (37) elaborates

on the current state of knowledge about the carbon cycle by stating that the science of the biosphere is still primitive, despite its importance. No pools in the carbon cycle seem to be known with satisfactory accuracy and it would be highly desirable to have a much wider net of stations measuring the atmospheric carbon dioxide and many more data for the equilibrium carbon dioxide of surface ocean water. (37)

The world transport model for carbon-14 that is currently used by the U.S. Environmental Protection Agency is that developed by Machta (38,39). This model is a relatively simple multireservoir exchange model consisting of seven compartments: stratosphere, troposphere, mixed layer of the ocean, deep ocean, short-term land biosphere, long-term land biosphere, and marine biosphere. The exchange rates between and within reservoirs are assigned by Machta except for the troposphere-mixed ocean layer rates which are obtained from a trial-and-error least squares fit procedure using atomic bomb produced $^{14}CO_2$ as a tracer. The model also takes into consideration the increase in the levels of tropospheric carbon dioxide due to the combustion of fossil fuel containing no carbon-14 which reduces the specific activity of C-14 in the carbon cycle (the "Suess Effect") and thereby reduces the long-term environmental dose commitment from carbon-14. Machta assumes that the characteristics of neither the oceans nor the biosphere vary with time. Carbon-14 inputs to the troposphere in the Machta model were due to nuclear weapon testing and cosmic rays. Magno et al. (1) modified the Machta model to allow injections of carbon-14 into the troposphere from the nuclear power industry. The modified Machta model in the form of a computer code is listed and discussed in detail in Appendix 1.

Given injections of fossil fuel $^{12}CO_2$ since the beginning of the industrial era (1860) and inputs of nuclear fuel cycle $^{14}CO_2$ to the troposphere, the modified Machta model can be used to estimate the C-12 content and the C-14 content (cosmic C-14 and nuclear fuel cycle C-14) in the stratosphere, troposphere, mixed layer of the ocean, deep ocean, short-term land biosphere, long-term land biosphere, and the marine biosphere. The Machta model basically computes net changes of C-12 and C-14 concentrations in various reservoirs since 1860 and the estimated reservoir concentration is computed by adding initial reservoir concentrations to the net changes in the concentration due to man-made injections of C-12 and C-14 and reservoir exchange rates over time. This basic model is rather simple and consideration is being given to investigating other world transport models for carbon-14 utilizing the work of Keeling et al. (40,41) and others.

6. DOSIMETRY FOR CARBON-14 DIOXIDE

6.1 Critical Organ Method to Estimate Local Short-Term Dose Equivalent Rates

To estimate the potential short-term dose equivalent rate, local intakes of carbon-14 from dosimetrically significant pathways must be calculated and then multiplied by the appropriate dose equivalent conversion factor. Dose equivalent conversion factors per picocurie intake for the inhalation and ingestion pathways were computed using the equation:

$$DECF_{a(or\ w)}\ [\text{mrem/pCi inhaled (or ingested)}] = 7.38E\text{-}2 \cdot \bar{E}/M \cdot f_{a(or\ w)}\ T_{eff}$$

where, T_{eff} = effective half-life in days

f_a = fraction of the inhaled activity which deposits in the critical organ

f_w = fraction of the ingested activity which deposits in the critical organ

M = mass of the critical organ in grams

$\bar{E}$ = effective energy absorbed in the critical organ per disintegration in MeV/dis

$DECF_a$ = dose equivalent conversion factor for the inhalation pathway

$DECF_w$ = dose equivalent conversion factor for the ingestion pathway

The biological parameter data utilized in the computation as well as the computed DECF's are presented in Table 5. The biological parameter data was taken from ICRP Publication 2 (43) except for the value of $\bar{E}$ (MeV/dis) which was taken from MIRD Supplement Number 5/Pamphlet No. 7 (44). The ICRP has updated values for organ mass in ICRP Publication 23 (45), but the values for the other biological parameters needed to calculate the dose equivalent conversion factors have not been updated. It is additionally noted that these DECF's developed from existing ICRP recommendations may be conservative based on more recent dose models as discussed by Rohwer et al. (46).

The dose equivalent rate per unit air concentration for the inhalation pathway, $DECF_a$, was computed using an adult breathing rate of 8000 m^3/yr and the previously computed dose equivalent conversion factors per picocurie intake. The adult breathing rate was obtained by averaging the adult man and adult woman breathing rates as presented in ICRP Publication 23 (45). The resulting $DECF_a$ values are shown in Table 6.

The dose equivalent rate per unit air concentration for the submersion pathway $\dot{DECF}_S$ was computed using the equation

$$\dot{DECF}_S \frac{(mrem/yr)}{(\mu\ Ci/cc)} = 7.2 \times 10^9 \times \bar{E} \frac{(MeV)}{(dis)}$$

Using a value of 0.0493 MeV/dis for $\bar{E}$, the resulting $\dot{DECF}_S$ is shown in Table 6. Because C-14 decays by beta emission, the estimated annual dose equivalent rate from submersion will be the dose equivalent rate at the clothing or outer skin surface of the body. This dose equivalent rate will decrease with tissue depth and the radiosensitive tissues underneath the normal inert layer of skin will receive only a fraction of the skin surface dose equivalent.

Given an air concentration, the local dose equivalent rate via inhalation and submersion from discharges of C-14 can be obtained by simple multiplication with the conversion factors in Table 6. The air concentration as a function of downwind distance from the point of C-14 discharge is calculated by multiplying the C-14 discharge rate "Q" in curies/second times the meteorological dilution factor "χ/Q" in sec/m^3 at the downwind distance where the dose equivalent rate is computed. The annual average dilution factor for an elevated release is found by assuming a sector-averaged concentration in each sector with the plume following a Gaussian distribution in the vertical direction.

The local dose equivalent rate via ingestion can be calculated if the C-14 activity in the vegetation is known. The yearly pCi intake is calculated by multiplying the vegetation activity in pCi/gm by the human vegetation intake rate in gm/yr. The ingestion dose equivalent rate is then calculated by multiplying the C-14 activity intake rate in pCi/yr by the $DECF_W$ factor in Table 5. If the C-14 activity in vegetation is not known, then the activity can be estimated by making the simplifying assumption that the C-14 specific activity in local vegetation is equal to local atmospheric specific activity. The local atmospheric C-14 specific activity is obtained by multiplying the source term "Q" in pCi/sec by the meteorological dilution factor "χ/Q" in sec/m^3 (at the distance where the specific activity is to be calculated) and dividing by the concentration of carbon-12 in gm/m^3. The vegetation activity is then estimated by multiplying the vegetation specific activity in pCi C-14/gm C-12 by the fraction of the plant mass that is C-12. A fraction of 0.1 C-12 in plant mass is employed by the U.S. Nuclear Regulatory Commission (42).

TABLE 5

CARBON-14 ADULT DOSE EQUIVALENT CONVERSION FACTORS PER UNIT INTAKE

Critical Organ	T_{eff} (days)	f_a	f_w	m(grams)	$\bar{E}$(MeV/dis)	$DECF_a$ (mrem/pCi inhaled)	$DECF_w$ (mrem/pCi ingested)
Total Body	10	0.75	1.0	7E + 4*	0.0493	3.9E - 7	5.2E - 7
Fat	12	0.38	0.5	1E + 4	0.0493	1.6E - 6	2.2E - 6
Bone	40	0.02	0.025	7E + 3	0.0493	4.2E - 7	5.2E - 7

* 7E + 4 equals 7×10^4

TABLE 6

CARBON-14 ADULT DOSE EQUIVALENT RATE CONVERSION FACTORS PER UNIT AIR CONCENTRATION

Critical Organ	Pathway	DECF $\left(\frac{\text{mrem/yr}}{\mu\text{Ci/cc}}\right)$
Total Body	Inhalation	3.1E + 9
Fat	Inhalation	1.4E + 10
Bone	Inhalation	3.3E + 9
Total Body	Submersion	3.6E + 8

6.2 Specific Activity Method to Estimate World-Wide Long-Term Dose Equivalent Rates

An intermediate result of the Machta model computation which was previously described is the specific activity of C-14 (pCi C-14/gm C-12) in the troposphere. Specific activities of C-14 are presented separately for nuclear fuel cycle injections of C-14 and for cosmic C-14. Therefore, the C-14 dose equivalent rates from the nuclear fuel cycle can be compared with background dose equivalent rates from cosmic C-14. In order to convert this specific activity to a dose equivalent rate in man, the conservative assumption is made that the specific activity of C-14 in the troposphere and in man are the same. The specific activity method assumes instantaneous equilibrium which is a conservative assumption thereby providing an upper limit to the estimated dose equivalent rate. Specific activity dose equivalent rate conversion factors were derived using the following equation:

$$\dot{DECF}\ (\text{mrem/yr per pCi C-14/gm C-12}) = 3.7E{+}10\ \text{dis/sec-Ci} \times 1\ \text{Ci}/1E{+}12\ \text{pCi} \times 0.0493\ \text{MeV/dis} \times 1.6E{-}6\ \text{ergs/MeV} \times 1\ \text{gm (tissue) rad}/100\ \text{ergs} \times M_C\ \text{gm C-12}/M_T\ \text{gm tissue} \times 3.15E{+}7\ \text{sec/yr} \times 1\ \text{rem/rad} \times 10^3\ \text{mrem/rem}$$

$$= 0.919\ M_C/M_T$$

where M_C and M_T is the mass of carbon and mass of tissue respectively for the organ or tissue that the DECF is being calculated. Values for M_C and M_T were obtained from ICRP Publication 23 (45) and calculated carbon-14 specific activity dose equivalent rate conversion factors for selected organs are presented in Table 7.

Carbon-14 specific activity dose equivalent rate conversion factors for body organs can also be calculated using "S" factors (average dose equivalent per unit accumulated activity) presented by Snyder et al. (47). The "S" factor is the dose equivalent to a target organ per unit integrated activity in the source organ which can be equated to the dose equivalent rate per equilibrium organ activity burden for steady state conditions. The specific activity dose equivalent rate conversion factors can be calculated using the equation:

$$\text{DECF (mrem/yr per pCi C-14/gm C-12)} = S\ (\text{rem}/\mu\text{Ci-day}) \times M_C\ (\text{gm C-12}) \times 1E{-}6\ \mu\text{Ci/pCi} \times 365\ \text{day/yr} \times 10^3\ \text{mrem/rem}$$

$$= 0.365\ SM_C$$

Carbon-14 "S" factors for 22 source organs and 24 target organs are presented in reference (47). When M_C values are obtained from ICRP Publication 23 (45), calculated DECF values are similar to those calculated using the previously described technique. The dose equivalent rate to the total endosteal cells per unit C-14 specific activity in cancellous bone, cortical bone, red marrow and yellow was calculated using the "S" factor technique and the DECF value is presented in Table 7. ICRP Publication 11 (48) discusses the importance of the dose equivalent rate to endosteal cells from low-energy beta-emitters such as C-14. The "S" factor method was utilized by ERDA (20) and values were calculated similar to those presented in Table 7 except for segments of the GI tract. The GI tract model in reference (20) includes the dose equivalent rate by the migrating contents in the segments of the GI tract whereas the factors in Table 7 do not include this contribution to the dose equivalent rate. Considering ^{14}C in tissue and in the migrating contents, DECF values of 0.14 and 0.16 mrem/yr per pCi C-14/gm C-12 for the stomach and lower large intestine respectively can be inferred from reference (20). Reference (20) should be reviewed for a discussion of the model employed to obtain the dose equivalent rate contribution from the migrating contents in the stomach and intestine of the GI tract.

The relative contributions of the ingestion and inhalation pathways to specific activity dose equivalent rate can be determined if the specific activity in the air and in the diet (food and water) are assumed equal. Assuming air is 0.033% CO_2, an adult breathing rate of 2.2×10^7 cc and that 75% of inhaled air is retained in the total body (f_a = .75), 2.92 gm/day of carbon is inhaled per day by standard man. Standard man intakes 300 gm/day of carbon from food and fluids (45). The relative contribution of the inhalation and ingestion pathways to the total C-14 dose equivalent rate for equilibrium conditions is assumed to be proportional to the intake of carbon for each pathway. Therefore, the ingestion pathway contributes 99% (300/302.9 x 100%) of the carbon-14 specific activity dose equivalent rate.

By assuming that the C-14 specific activity in air is the same in tissues of man, the dose equivalent rate to critical organs can be obtained by multiplying the C-14 tropospheric specific activity by the specific activity dose equivalent rate conversion factors presented in Table 7. The computed dose equivalent rate represents a measure of the annual potential impact of C-14 discharges on the average worldwide individual. Since carbon-14 is long-lived (half-life of 5,730 years) and therefore, represents a long-term potential source of exposure to a large number of people, the long-term impact on the population at large and on the individual must be assessed in addition to the annual

impact. The potential environmental impact in subsequent years is estimated by calculating the "environmental dose commitment" (49) which is the sum of all doses to individuals over the entire time period that the carbon-14 persists in the environment in a state available for interaction with humans. The population dose commitment in person-rems is usually expressed for a period of 100 years since it is difficult to predict the world population growth much beyond this time period. The total body individual and population dose commitment can be computed by using the specific activity method with the modified Machta Code as described in Appendix 1.

TABLE 7

CARBON-14 SPECIFIC ACTIVITY DOSE EQUIVALENT RATE CONVERSION FACTORS

Critical Organ	M_T (Weight of organ in grams)	M_C (Carbon in organ in grams)	$\dot{D}ECF$ $\frac{mrem/yr}{pCi\ C\text{-}14/gm\ C\text{-}12}$
Body fat	13500	10000	0.68
Kidneys (2)	310	40	0.12
Liver	1800	260	0.13
Lungs (2)	1000	100	0.09
Cortical Bone	4000	550	0.13
Trabecular Bone	1000	130	0.12
Red Marrow	1500	620	0.38
Yellow Marrow	1500	950	0.58
Total Endosteal Cells			0.33*
Lower Large Intestine	160	19	0.11
Stomach	150	18	0.11
Skin	2600	590	0.21
Testes (2)	35	3.1	0.08
Thyroid	20	2.1	0.10
Total Body	70000	16000	0.21

*This conversion factor represents contributions from C-14 in cancellous bone, cortical bone, red marrow and yellow marrow using reference (47).

7. CARBON-14 DOSE EQUIVALENT RATES AND HEALTH IMPACT

Estimated annual carbon-14 dose equivalent rates to individuals at the offsite location where maximum air concentrations occur at light-water-cooled reactors and fuel reprocessing plants are given in Table 8. The assumptions used for the calculation of the local dose equivalent rates are as follows:

(a) carbon-14 source terms
LWR fuel reprocessing facility - 990 Ci/yr
PWR - 5 Ci/yr
BWR - 9 Ci/yr

(b) maximum offsite atmospheric dispersion factor - "χ/Q"
fuel reprocessing facility - 5.0E-8
PWR - 2.5E-6
BWR with stack - 5.0E-8
BWR without stack - 2.5E-6

(c) concentration of carbon-12 in the troposphere = 0.174 gm C-12/m^3

(d) specific activity dose equivalent rate conversion factors are 0.21 and 0.08 mrem/yr per pCi C-14/gm C-12 for the total body and gonads respectively.

A specific activity model was employed for the local maximum individual dose equivalent rate calculation. This method assumes that the carbon-14 specific activity in the maximum individual is equal to carbon-14 specific activity in the air at the maximum point of offsite concentration. This methodology estimates a conservative upper bound to the maximum individual carbon-14 dose equivalent rate since the carbon-14 specific activity in man's diet will not be instantaneously equal to the specific activity in the air at the point of maximum offsite concentration. Any food or fluids that the maximum individual ingests that is uncontaminated or at a lower C-14 specific activity than that at the point of maximum offsite concentration will result in lower C-14 dose equivalent rates than those presented in Table 8. However, even using the conservative specific activity model, the local C-14 dose equivalent rates to the maximum individual presented in Table 8 are extremely low when compared to natural background dose equivalent rates. Using the linear, non-threshold dose effect relationships (50), less than one health effect due to carbon-14 exposure to the population within 50 miles of any of these facilities is expected during its entire operating life.

TABLE 8

MAXIMUM ANNUAL CARBON-14 DOSE EQUIVALENT RATES
TO INDIVIDUALS AT LWR FACILITIES IN mrem/yr

Organ	Fuel Reprocessing Facility	PWR	BWR (with stack)	BWR (without stack)
Total Body	1.9	0.48	1.7E-2	0.86
Gonads	0.72	0.18	6.6E-3	0.33

The health effects to the world's population from the U.S. LWR nuclear industry due to C-14 discharges present quite a different story. The potential committed health effects depends on the projected growth of both the world population and the U.S. nuclear industry. The potential health effects committed to the world's population for one year's LWR power production can be calculated using the following equation:

$$H(t) = \sum_i Q(t) \times D_i(t)/Q(t) \times J_i$$

where,

$H(t)$ = potential committed health effects for release of carbon-14 in calendar year t

$Q(t)$ = annual discharge of carbon-14 in curies

$D_i(t)/Q(t)$ = committed population dose in person-rem to organ i per curie release of carbon-14 in calendar year t

J_i = number of health effects per man-rem to organ i

The annual discharge of carbon-14 from the LWR nuclear power industry is equal to the LWR produced electrical power in GWe-yr times 30 Ci/GWe-yr. The production rate of C-14 of LWR's of about 30 Ci/GWe-yr is based on a production rate of 22 Ci/GWe-yr in the fuel and about 8 Ci/GWe-yr in the coolant of LWR's. For a LWR produced electrical power of 1 GWe-yr, 22 curies of C-14 would be discharged at a LWR fuel reprocessing plant and about 8 curies of C-14 would be discharged at a LWR with no control for carbon-14. Various scenarios have been presented for the growth of nuclear power in the U.S. For example, the Energy Research and Development Administration (ERDA) has presented a projection of nuclear capacity for the year 2000 as between 625,000 and 1,250,000 MWe. The Federal Energy Administration projects a range of 600,000 to 700,000 MWe for the nuclear energy capacity for the year 2000 (51). For the purposes of this report, two projections (cases 1 and 2) of power growth will be utilized to estimate a range of potential committed health effects due to C-14 discharges from the LWR power industry. Cases 1 and 2 are the moderate/low growth and high growth cases respectively of U.S. installed LWR capacity as presented in the ERDA update of WASH-1139(74) (52). Case 1 projections represent a slower growth rate of electricity with the need for new nuclear power plants being reduced. Case 2 projections

"reflects the Presidential objectives for 200 new nuclear power plants through 1985 and a continuation of a concerted nuclear effort in the longer-term coupled with continued high rates of growth in electric energy" (53). The installed LWR nuclear capacities for cases 1 and 2 for the years 1975 to 2000 are presented in Tables 12 and 13 which are worksheets showing a calculation of the world-wide potential health effects from carbon-14 for the two projected growth rates of the U.S. light-water-cooled nuclear power industry.

The worksheets also present the 100-year world-wide population dose commitment per curie of C-14 discharged by calendar year. These conversion factors were obtained by utilizing the modified Machta world-wide carbon-14 transport computer code which is described earlier in this report. The 1970 world population was estimated as 3.56E+9 with an annual growth rate of 1.9 percent (49).

The conversion from population dose to potential health effects was performed using the following carbon-14 dose-risk conversion factors:

400 cancers(200 fatal and 200 non-fatal) per 10^6 person-rem to the whole body

200 genetic effects per 10^6 person-rem to the gonads

As is shown in the worksheets (Tables 12 and 13), it is estimated that between about 3900 and 5500 potential health effects will be committed to the world's population by year 2000 from carbon-14 discharges from the U.S. LWR nuclear industry. These health effects estimates assume that all C-14 generated in LWR power production is discharged to the atmosphere, that the 100-year population dose commitment adequately reflects the impact of carbon-14 in the atmosphere and that the linear dose-effect relationship is valid at these low dose rates.

8. SUMMARY

This review has presented C-14 source term estimates and the state of carbon-14 control technology as it is presently known. Production rates of C-14 in light-water reactors were estimated as 22 Ci/GWe-yr in the fuel and 9.2 and 3.3 Ci/GWe-yr respectively in the BWR and PWR coolant. Based on limited measurements at LWR's, gaseous C-14 emissions of 9.0 and 5.1 Ci/GWe-yr were estimated for the BWR and PWR. Caustic scrubbers were discussed as a potential C-14 control system for LWR's with estimated C-14 discharge reductions of almost a factor of 10 for BWR's and about three for the PWR. Based on production rates of C-14 in the fuel of light-water reactors and assuming that all the C-14 in the spent fuel would be discharged to the atmosphere, a gaseous C-14 source term of about 990 Ci per year was estimated for a 1500 MT/yr LWR fuel reprocessing plant. Carbon-14 undergoes environmental transport processes which are not yet quantitatively well defined; however, preliminary estimates indicate that the 100-year population total body dose commitment per gigawatt-year of electric power is more than an order of magnitude greater for carbon-14 than krypton-85. The evaluation of the proper point of application of the control technology for C-14 is not as evident as it is for Kr-85, where essentially 99% of the gaseous Kr-85 discharge originates from the fuel reprocessing plant. Preliminary data indicates that C-14 source terms from LWR reactors are significant in comparison to LWR fuel reprocessing C-14 source terms.

There are many areas in which more study is necessary to be able to more precisely estimate the impact of C-14 on man and the environment. Investigations are urgently needed for the following areas:

1. measurements of C-14 production rate and chemical form and mechanisms affecting production such as O-17 and N-14 present in the fuel and coolant,

2. further measurement of carbon-14 discharge rates at reactors and fuel reprocessing plants,

3. studies to determine the capabilities of current reactor and fuel reprocessing off-gas systems for the control of C-14 including measurements on the partitioning between the off-gas and liquid process systems, possible pathways and chemical reactions and probable decontamination factors for various systems,

4. studies to assess the availability and cost of specialized carbon-14 control systems for use at both reactors and fuel reprocessing plants,

5. an evaluation of currently used carbon-14 world-wide environmental transport models with special emphasis on a sensitivity analysis of model parameters, such as exchange rates, and inputs such as C-12 from the combustion of fossil fuels on calculated C-14 dose equivalent contributions from LWR fuel cycle C-14 discharges,

6. an assessment of potential C-14 concentrations in local pathways to determine the extent of the buildup of C-14 levels above natural background as a function of distance from nuclear facilities,

7. an evaluation of recent C-14 internal dose studies which may yield data more appropriate to calculating C-14 internal dose equivalent conversion factors than the data in existing ICRP recommendations,

8. an evaluation of the biological significance of enhanced C-14 specific activities in the biosphere.

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APPENDIX 1

WORLD-WIDE CARBON-14 TRANSPORT MODEL

The basic Machta model described in references (38) and (39) was modified by Magno et al. (1) to allow estimates of the dose equivalents from nuclear fuel cycle carbon-14 discharges to be assessed. The modified Machta computer code is listed in Table 9 and a definition of symbols used in the code is presented in Table 10. The computer program listed in Table 9 requires input data for the annual release of fossil fuel CO_2 to the troposphere expressed in 10^{16} grams carbon-12 for the years 1860 to 1968. Nuclear power fuel cycle power production data are input into the code along with the year that the power was produced. A conversion factor is utilized to convert the yearly power production in GWe to a monthly discharge of carbon-14 in KCi to the troposphere. Fossil fuel injections are projected for 1969 to 1979 using an assumed annual growth rate of four percent and are projected for 1980 to 2080 using an assumed annual growth rate of 3-1/2 percent. The carbon-12 and carbon-14 input data are utilized in the Machta equations described in reference (39) to compute net changes in world-wide compartments since 1860 for cosmic C-14, C-12 due to fossil fuel discharges, and nuclear power fuel cycle C-14. The seven world-wide compartments described by the Machta equations are the stratosphere, troposphere, mixed layer of the ocean, deep ocean, short-term land biosphere, long-term land biosphere, and marine biosphere.

Net changes of cosmic and nuclear power fuel cycle C-14 in the tropospheric compartment are added to the initial amounts present in 1860 to calculate the total amount of C-14 present as a function of time. These total amounts of C-14 are divided by the total amount of C-12 present to compute C-14 specific activities in the troposphere as a function of time. The specific activity (pCi C-14/gram C-12) in man is assumed to be the same as that calculated to be present in the troposphere at equilibrium conditions. Total body dose equivalent rates due to C-14 discharges from the nuclear power fuel cycle are then estimated for the world-wide individual and the population using a specific activity dose equivalent model.

Magno et al. (1) utilized the computer program listed in Table 9 to estimate tropospheric inventories of carbon-14 for the period 1980 to 2020 resulting from projected discharges from the nuclear industry. Table 11 is a listing of the input data that was utilized to calculate a carbon-14 100-year population dose commitment of 38 person-rem/Ci from releases in 1980.

TABLE 9

LISTING OF MODIFIED MACHTA WORLD-WIDE
CARBON-14 TRANSPORT COMPUTER CODE

```
      CARB14                                                                    1
      DIMENSION YI(2652),T(2652),T14(2652),S(2652),BS(2652),BS14(2652),         2
     1BL(2652),BL14(2652),ZM(2652),ZM14(2652),S14(2652),D(2652),D14(2652        3
     2),CI(2652),CT(2652),DCTCI(2652),IYR(2652),                                4
     3CT14(2652),DCT(2652)                                                      5
      DIMENSION BBS(2652),BBL(2652),BMB(2652),BM(2652),BM14(2652),TB4(26        6
     152),TBR(2652),ZMB4(2652),DB(2652)                                         7
      DIMENSION TDI(2652),SB(2652)                                              8
      DIMENSION TITLE(80)                                                       9
      DATA YI/2652*0./,T/2652*0./,T14/2652*0./,S/2652*0./,BS/2652*0./,          10
     *     BS14/2652*0./,BL/2652*0./,BL14/2652*0./,ZM/2652*0./,                 11
     *     ZM14/2652*0./,S14/2652*0./,D/2652*0./,D14/2652*0./,                  12
     *     CI/2652*0./,CT/2652*0./,DCTCI/2652*0./,IYR/2652*0/,                  13
     *     CT14/2652*0./,                                                       14
     *     DCT/2652*0./,                                                        15
     *     BBS/2652*0./,BBL/2652*0./,BMB/2652*0./,BM/2652*0./,                  16
     *     BM14/2652*0./,TB4/2652*0./,TBR/2652*0./,ZMB4/2652*0./,               17
     *     DB/2652*0./                                                          18
      DATA TDI/2652*0./,SB/2652*0./,LYR/0/                                      19
  502 FORMAT (80A1)                                                             20
  503 FORMAT (1H1,30X,80A1)                                                     21
      ISM=0                                                                     22
      BCC=10.                                                                   23
      IT=11                                                                     24
      READ 502,(TITLE(I),I=1,80)                                                25
      PRINT 503,(TITLE(I),I=1,80)                                               26
      READ 100,(YI(I),I=12,1080,12)                                             27
      READ 105,(YI(J),J=1092,1296,12)                                           28
      DO 2 I=12,1080,12                                                         29
    2 YI(I)=YI(I)*0.8                                                           30
      DO 3 J=1308,1452,12                                                       31
    3 YI(J)=YI(J-12)*1.04                                                       32
      DO 4 I=1464,2652,12                                                       33
    4 YI(I)=YI(I-12)*1.035                                                      34
      DO 8 J=12,2652,12                                                         35
      YI(J)=YI(J)/12.                                                           36
      K=J-11                                                                    37
      DO 8 I=K,J                                                                38
      YI(I)=YI(J)                                                               39
    8 CONTINUE                                                                  40
      DO 7 I=1,2652                                                             41
    7 YI(I)=YI(I)*1.1                                                           42
      READ  NUCLEAR GW(E) CAPACITY, AND CONVERT TO RELEASE RATE OF C-14          43
      READ(5,1210) NYR,CONV                                                     44
 1200 READ(5,1210) MYR,GEM                                                      45
 1210 FORMAT(1X,I4,F10.0)                                                       46
```

```
      IF (LYR.EQ.0) LYR=NYR-1                                          47
      I1=(LYR+1-1860)*12+1                                             48
      I2=(MYR+1-1860)*12                                               49
      DO 1215 I=I1,I2                                                  50
 1215 TDI(I)=GWEM*CONV/(12.*1000.)                                     51
      LYR=MYR                                                          52
      IF(MYR.LT.NYR) GO TO 1200                                        53
C *** C1 & C2 - FACTORS TO CONVERT GRAMS OF C TO PPM IN ATMOSPHERE      54
      C1=30./(0.85*0.521*12.)                                          55
      C2=30./(0.15*0.521*12.)                                          56
      LG=492                                                           57
      T0=50.68                                                         58
      SET T014 TO BE EQUIVALENT TO 6.13 PCI/GM PRIOR TO 1860           59
      T014=299.9                                                       60
      XLDM=1./(1600.*12.)                                              61
      F=0.25                                                           62
      PL=2.5/12.                                                       63
      PS=3.1/12.                                                       64
      PM=2./12.                                                        65
      ALINV=1.016                                                      66
      ALPH1=0.9643                                                     67
      ALPH2=0.984                                                      68
      XLST=0.5/12.                                                     69
      XLTS=0.15*0.5/(0.85*12.)                                         70
      CI(1)=C1*YI(1)                                                   71
      CT(1)=CI(1)                                                      72
  998 IYR(1)=1860                                                      73
      XLLL1=0.22/12.                                                   74
      XLLL2=0.10/12.                                                   75
      ZM0=T0*XLLL1/XLLL2                                               76
      ZM014=0.95*T014*XLLL1/XLLL2                                      77
      D0=61.*T0-ZM0                                                    78
      XLMD=XLDM*D0/ZM0                                                 79
      DO 30 I=2,2652                                                   80
  610 ZM2=0.                                                           81
  620 TB441=0.                                                         82
      TB43=0.                                                          83
      ADS14=0.                                                         84
      ZMB2=0.                                                          85
      T41=0.                                                           86
      T1441=0.                                                         87
      T3=0.                                                            88
      ZM143=0.                                                         89
      T143=0.                                                          90
      ZM142=0.                                                         91
      IF(I.LE.24) GO TO 555                                            92
      ZMB2=ZMB4(I-24)                                                  93
      ZM2=ZM(I-24)                                                     94
      ZM142=ZM14(I-24)                                                 95
  555 IF(I.LE.36) GO TO 566                                            96
      T3=T(I-36)                                                       97
      ZM143=ZM14(I-36)                                                 98
      T143=T14(I-36)                                                   99
      TB43=TB4(I-36)                                                   100
```

```
  600 IF(I.LE.LG) GO TO 777                                                  101
      TB441=TB4(I-LG)                                                        102
      T1441=T14(I-LG)                                                        103
      T41=T(I-LG)                                                            104
C  T  EQUATION 1, T(I)=C-12 IN TROPOSPHERE (10**16 GRAMS)                   105
  777 T(I)=T(I-1)*(1.-XLLL1  -XLTS)+   BCC*XLLL2*ZM(I-1)+XLST*S(I-1)         106
      T(I)=T(I)-F*(PL*(T(I-1)-T41)/T0+PS*(T(I-1)-T3)/T0)+YI(I)               107
C ZM  EQUATION 4, ZM=MAN-MADE C-12 IN OCEAN MIXED LAYER (10**16 GRAMS)      108
      ZM(I)=ZM(I-1)*(1.-XLMD-BCC*XLLL2)+XLLL1*T(I-1)+XLDM*D(I-1)             109
C  S  EQUATION 10, S= C-12 IN THE STRATOSPHERE (10**16 GRAMS)                110
      S(I)=S(I-1)*(1.-XLST)+XLTS*T(I-1)                                      111
      D(I)=D(I-1)*(1.-XLDM)+XLMD*ZM(I-1)                                     112
      ZM14(I)=ZM14(I-1)*(1.-ALINV*XLLL2  -XLMD) -BCC*XLLL2*ZM(I-1)*(ZM       113
     1014+ZM14(I-1))/(ZM0+ZM(I-1))+XLLL1  *T14(I-1)*ALPH2                    114
      ZM14(I)=ZM14(I)+XLDM*D14(I-1)-ALPH1*PM*((ZM014+ZM14(I-1))/(ZM0+ZM(     115
     1I-1))-(ZM014+ZM142)/(ZM0+ZM2))                                         116
      S14(I)=S14(I-1)*(1.-XLST)+XLTS*T14(I-1)+ADS14                          117
      D14(I)=D14(I-1)*(1.-XLDM)+XLMD*ZM14(I-1)                               118
      T14(I)=T14(I-1)*(1.-ALPH2*XLLL1  -XLTS)+XLLL2  *ZM14(I-1)*ALINV+XL     119
     1ST*S14(I-1)                                                            120
       T14(I)=T14(I)+BCC*XLLL2  *ZM(I-1)*(ZM014+ZM14(I-1))/(ZM0+ZM(I-1))     121
      T14(I)=T14(I)-ALPH1*PL*(T14(I-1)/(T0+T(I-1))-T1441/(T0+T41))           122
      T14(I)=T14(I)-ALPH1*PS*(T14(I-1)/(T0+T(I-1))-T143/(T0+T3))             123
      BS(I)=BS(I-1)+F*PS*((T(I-1)-T3)/T0)                                    124
      BL(I)=BL(I-1)+F*PL*((T(I-1)-T41)/T0)                                   125
      BS14(I)=BS14(I-1)+ALPH1*PS*(T14(I-1)/(T0+T(I-1))-T143/(T0+T3))         126
      BL14(I)=BL14(I-1)+ALPH1*PL*(T14(I-1)/(T0+T(I-1))-T1441/(T0+T41))       127
      BM14(I)=BM14(I-1)+ALPH1*PM*((ZM014+ZM14(I-1))/(ZM0+ZM(I-1))-(ZM014     128
     1+ZM142)/(ZM0+ZM2))                                                     129
      BM(I)=0.                                                               130
  888 CT(I)=C1*T(I)                                                          131
      DCT(I)=CT(I)-CT(I-1)                                                   132
      DCTCI(I)=DCT(I)/C1(I-1)                                                133
      CT14(I)=C1*T14(I)                                                      134
      CI(I)=C1*YI(I)                                                         135
      IYR(I)=(I-1)/12+1800                                                   136
      TBR(I)=-ALPH2*XLLL1  *TB4(I-1) +XLLL2  *ZMB4(I-1)*ALINV                137
      TBR(I)=TBR(I)+ BCC*XLLL2  *ZM(I-1)*ZMB4(I-1)/(ZM0+ZM(I-1))             138
      TBR(I)=TBR(I)-ALPH1*PL*(TB4(I-1)/(T0+T(I-1))-TB441/(T0+T41))           139
      TBR(I)=TBR(I)-ALPH1*PS*(TB4(I-1)/(T0+T(I-1))-TB43/(T0+T3))             140
      TB4(I)=(1.-XLTS)*TB4(I-1)+XLST*SB(I-1)+TBR(I)+TDI(I)                   141
      SB(I)=(1.-XLST)*SB(I-1)+XLTS*TB4(I-1)                                  142
      ZMB4(I)=ZMB4(I-1)*(1.-ALINV*XLLL2  -XLMD)- BCC*XLLL2  *ZM(I-1)*ZMB     143
     14(I-1)/(ZM0+ZM(I-1))+ALPH2*XLLL1  *TB4(I-1)                            144
      ZMB4(I)=ZMB4(I)+XLDM*DB(I-1)-ALPH1*PM*(ZMB4(I-1)/(ZM0+ZM(I-1))-ZMB     145
     12/(ZM0+ZM2))                                                           146
      DB(I)=DB(I-1)*(1.-XLDM)+XLMD*ZMB4(I-1)                                 147
      BBS(I)=BBS(I-1)+ALPH1*PS*(TB4(I-1)/(T0+T(I-1))-TB43/(T0+T3))           148
      BBL(I)=BBL(I-1)+ALPH1*PL*(TB4(I-1)/(T0+T(I-1))-TB441/(T0+T41))         149
      BMB(I)=BMB(I-1)+ALPH1*PM*(ZMB4(I-1)/(ZM0+ZM(I-1))-ZMB2/(ZM0+ZM2))      150
   30 CONTINUE                                                               151
```

```
      PRINT 250                                                          152
      DO 40 J=12,2652,12                                                 153
   46 PRINT 215,IYR(J),YI(J),CI(J),T(J),CI(J),T14(J),DCTCI(J),ZM(J),ZM14 154
     1(J),S(J),S14(J),DCI(J)                                             155
   40 CONTINUE                                                           156
      PRINT 255                                                          157
      DO 42 J=12,2652,12                                                 158
   47 PRINT 225,IYR(J),D(J),D14(J),BL(J),BL14(J),BS(J),BS14(J),BM(J),BM1 159
     14(J)                                                               160
   42 CONTINUE                                                           161
      PRINT 265                                                          162
      DO 44 I=12,2652,12                                                 163
   51 PRINT 275,IYR(I),To4(I),TBR(I),ZMB4(I),DB(I),BBS(I),BBL(I),BMB(I), 164
     1S=(I)                                                              165
   44 CONTINUE                                                           166
C     PREPARE SUMMARY OF NPWR C14 CALCULATIONS                           167
      PRINT 1220                                                         168
 1220 FORMAT(1H1,'YEAR',4X,'   ANNUAL   ',' CUMULATIVE ',                169
     *           ' NPWR C14  ',' NPWR C14  ','NATURAL C14 ',             170
     *           ' INDIVIDUAL ',' INTEGRATED ',' POPULATION ',           171
     *           ' POPULATION ',' POPULATION '/                          172
     *         9X,' INJECTION  ',' INJECTION  ',' ACTIVITY  ',           173
     *           'SP ACTIVITY ','SP ACTIVITY ',' DOSE RATE  ',           174
     *           ' INDIV DOSE ','10**9 PERS. ',' DOSE RATE  ',           175
     *           '    DOSE    '/                                         176
     *         9X,'    KCI     ','    KCI     ','    KCI     ',         177
     *           '   PCI/GM   ','   PCI/GM   ','   MREM/A   ',         178
     *           '    MREM    ',','           ','10**6 REM/A ',         179
     *           ' 10**6 REM  '/)                                        180
      SDOSE=0.                                                           181
      TTDI=0.                                                            182
      CDOSE=0.                                                           183
      DO 1230 I=1452,2652,12                                             184
      STDI=0.                                                            185
      SPTB=To4(I)/(T(I)+10)/10.                                          186
      CALCULATE SPECIFIC ACTIVITY IN PCI/GM.                             187
      SPT14=(T14(I)+1014)/(T(I)+10)/.9538                                188
      PDOSE=0.                                                           189
      DO 1225 J=1,12                                                     190
      K=I-12+J                                                           191
      STDI=STDI+TDI(K)                                                   192
      POP=4.30*EXP((K-1452)/(12.*53.))                                   193
      DOSE=TB4(K)/(T(K)+10)/732.                                         194
      SDOSE=SDOSE+DOSE                                                   195
 1225 PDOSE=PDOSE+POP*DOSE                                               196
      TTDI=TTDI+STDI                                                     197
      DOSE=DOSE*12.                                                      198
      CDOSE=CDOSE+PDOSE                                                  199
 1230 PRINT 1240, IYR(I),STDI,TTDI,TB4(I),SPTB,SPT14,DOSE,               200
     *           SDOSE,POP,PDOSE,CDOSE                                   201
      CALL EXIT                                                          202
```

```
1240 FORMAT(1X,I4,4A,10(1PG10.3,2X))                                          203
 100 FORMAT (1F6.0)                                                           204
 105 FORMAT (10F7.0)                                                          205
 215 FORMAT (1H ,I5,5X,11F9.3)                                                206
 225 FORMAT (1H ,I5,5X,8F9.3)                                                 207
 250 FORMAT (5H1YEAR,08A,9H     YI    ,9H     CI    ,9H      T    ,9H     CT   208
    1 ,9H    T14    ,9H   DCTCI   ,9H     ZM    ,9H     M14   ,9H      S    ,9H  209
    2  S14   9H    DCT    //)                                                 210
 255 FORMAT (5H1YEAR,07A,9H     D     ,9H     D14   ,9H     BL    ,9H    BL14  211
    1 ,9H    BS     ,9H    BS14   ,9H     BM    ,9H    BM14   //)            212
 265 FORMAT (5H1YEAR,07A,9H    TB     ,9H     TBR   ,9H    ZMB4   ,9H     DB   213
    1 ,9H   BBS     ,9H    BBL    ,9H     BMB   ,9H     SB    )              214
 275 FORMAT (1H ,I5,5X,8F9.3)                                                 215
 708 STOP                                                                     216
     END                                                                      217
```

TABLE 10

DEFINITION OF SYMBOLS USED IN THE
WORLD-WIDE CARBON-14 COMPUTER CODE

I = time in number of months since 1860

YI(I) = monthly release of fossil fuel CO_2 in the troposphere in 10^{16} grams carbon-12

NYR = end year for nuclear power discharges of carbon-14 to the troposphere

CONV = conversion factor used to convert nuclear power production in GW(e) to curies per year of carbon-14 discharges to the troposphere (units of Ci/GW(e)-yr.)

MYR = year of interest for nuclear power production

GWEM = electrical capacity for year of interest

TDI (I) = carbon-14 discharged per month in KCi from the nuclear power fuel cycle to the troposphere

BCC = buffering factor which accounts for the changes in partial pressure of CO_2 in the mixed layer resulting from changes in the carbon content of the mixed layer; taken as 10

LG = lag time in months for return of material from the long-term biosphere to the troposphere

TO = total quantity of carbon as $^{12}CO_2$ in the troposphere in 1860; taken as 50.68×10^{16} grams carbon-12

TO14 = total quantity of cosmic C-14 atoms as $^{14}CO_2$ in the troposphere in 1860; taken as 299.9×10^{26} atoms of carbon-14 based on a tropospheric specific activity of 6.13 pCi C-14/gram C-12

XLDM = fractional transfer from the deep oceans to the mixed oceans per mohth

F = fraction of land biosphere whose growth is assumed to be CO_2 limited, taken as 0.25

PL = net primary production into the long-term land biosphere on a monthly basis in 10^{16} grams carbon

PS = net primary production into the short-term land biosphere on a monthly basis in 10^{16} grams carbon

PM = net primary production into the marine biosphere on a monthly basis in 10^{16} grams carbon

ALINV = 1/ALPH2

ALPH1 = fractionation factor for carbon transferring from air or water to the biosphere

ALPH2 = fractionation factor for carbon transferring from air to water

XLST = fractional transfer from the stratosphere to the troposphere per month

XLTS = fractional transfer from the troposphere to the stratosphere per month

XLLL1 = fractional transfer from the troposphere to the mixed oceans in one month

XLLL2 = fractional transfer from the mixed oceans to the troposphere in one month

ZMO = carbon content in the mixed oceans in 1860 in 10^{16} grams of carbon-12

ZMO14 = cosmic C-14 content in the mixed oceans in 1860 in 10^{26} atoms of carbon-14

XLMD = fractional transfer from the mixed oceans to the deep oceans per month

T(I) = net change since 1860 of the carbon content of the troposphere due to the carbon dioxide from the man-made combustion of fossil fuels in 10^{16} grams of carbon-12

ZM(I) = same for mixed oceans

S(I) = same for stratosphere

D(I) = same for deep oceans

BS(I) = same for short-term land biosphere

BL(I) = same for long-term land biosphere

BM(I) = same for marine biosphere

T14(I) = net change since 1860 of the cosmic carbon-14 content of the troposphere in 10^{26} atoms of carbon-14

ZM14(I) = same for mixed oceans

S14(I) = same for stratosphere

D14(I) = same for deep oceans

BS14(I) = same for short-term land biosphere

BL14(I) = same for long-term land biosphere

BM14(I) = same for marine biosphere

CT(I) = net change since 1860 of the ppm_v of carbon-12 in the troposphere due to injections of carbon dioxide from the man-made combustion of fossil fuels

DCT(I) = monthly change of CT (carbon-12 in ppm_v) in the troposphere due to injections of carbon dioxide from the man-made combustion of fossil fuels

CI(I) = monthly discharge of carbon-12 in ppm_v to the troposphere from the man-made combustion of fossil fuels

DCTCI(I)= DCT(I)/CT(I-I) = ratio of monthly change of C-12 in the troposphere to C-12 discharged on a monthly basis as CO_2

TBR(I) = net change per month of carbon-14 in the troposphere in KCi of carbon-14 resulting from injections of carbon-14 to the troposphere from the nuclear power plant fuel cycle

TB4(I) = cumulative total nuclear power plant fuel cycle carbon-14 content of the troposphere in KCi of carbon-14

SB(I) = same for stratosphere

ZMB4(I) = same for mixed oceans

DB(I) = same for deep oceans

BBS(I) = same for short-term land biosphere

BBL(I) = same for long-term land biosphere

BMB(I) = same for marine biosphere

SPTB = nuclear power plant fuel cycle carbon-14 specific activity in the troposphere in pCi C-14 per gram C-12

SPT14 = cosmic carbon-14 specific activity in the troposphere in pCi C-14 per gram C-12

STDI = carbon-14 discharged annually in KCi from the nuclear power fuel cycle to the troposphere

POP = world population in billions

DOSE = average world-wide individual total body dose equivalent rate in mrem per year due to carbon-14 discharges from the nuclear power fuel cycle

SDOSE = integrated world-wide individual total body dose equivalent in mrem due to carbon-14 discharges from the nuclear power fuel cycle

PDOSE = annual world-wide population total body dose equivalent rate in million person-rems per year due to carbon-14 discharges from the nuclear power fuel cycle

TTDI = cumulative carbon-14 injection to the troposphere in KCi from the nuclear power fuel cycle

CDOSE = cumulative world-wide population total body dose equivalent in million person-rems due to carbon-14 discharges from the nuclear power fuel cycle

TABLE 11

SAMPLE INPUT DATA FOR THE WORLD-WIDE
CARBON-14 TRANSPORT COMPUTER CODE

```
     RUN WITH 1600 YEAR OCEAN                                                218
0.010                                                                        219
0.010                                                                        220
0.011                                                                        221
0.011                                                                        222
0.012                                                                        223
0.012                                                                        224
0.013                                                                        225
0.014                                                                        226
0.014                                                                        227
0.015                                                                        228
0.016                                                                        229
0.016                                                                        230
0.017                                                                        231
0.018                                                                        232
0.019                                                                        233
0.020                                                                        234
0.021                                                                        235
0.022                                                                        236
0.023                                                                        237
0.024                                                                        238
0.025                                                                        239
0.026                                                                        240
0.027                                                                        241
0.029                                                                        242
0.030                                                                        243
0.031                                                                        244
0.032                                                                        245
0.033                                                                        246
0.034                                                                        247
0.036                                                                        248
0.035                                                                        249
0.037                                                                        250
0.038                                                                        251
0.040                                                                        252
0.042                                                                        253
0.044                                                                        254
0.047                                                                        255
0.049                                                                        256
0.051                                                                        257
0.054                                                                        258
0.061                                                                        259
```

```
0.063
0.065
0.067
0.069
0.071
0.073
0.076
0.078
0.081
0.090
0.091
0.092
0.094
0.095
0.097
0.098
0.100
0.101
0.103
0.108
0.109
0.110
0.110
0.111
0.112
0.113
0.113
0.114
0.115
0.105
0.108
0.111
0.114
0.117
0.120
0.123
0.126
0.129
0.132
0.127
0.132
0.136
0.143
0.149
0.155
0.161
0.167
0.174
0.180
 .148    .157    .160    .162    .168    .183    .195    .202    .213    .225
 .238    .233    .243    .260    .278    .285    .300    .307    .325
1980    1000.0
1980       1.
```

TABLE 12

CASE 1. WORLD-WIDE COMMITTED POTENTIAL HEALTH EFFECTS FROM CARBON-14 FOR THE U.S. LWR NUCLEAR POWER INDUSTRY

Year	LWR Installed Nuclear Capacity Cumulative Total	LWR Produced Power(1)	C-14 Annual Injection	C-14 100 Yr. ~~Population~~ Environmental Dose Commitment Conversion Factors		C-14 100 Yr. Population Dose Commitment	
				Total Body(2)	Gonads(3)	Total Body	Gonads
	(GWe)	(GWe-yr)	(KCi/yr)	(person-rem/Ci)		(10^6 person-rem)	
1975	37.1	25.6	.768	45.6	17.4	.035	.013
1976	43.5	30.0	.900	46.1	17.6	.041	.016
1977	50.5	34.8	1.04	46.6	17.7	.049	.018
1978	57.5	39.7	1.19	47.1	17.9	.056	.021
1979	65.5	45.2	1.36	47.6	18.1	.065	.025
1980	75.6	52.2	1.57	48.0	18.3	.075	.029
1981	92.4	63.8	1.91	48.5	18.5	.093	.035
1982	112.7	77.8	2.33	49.0	18.7	.11	.044
1983	133.8	92.3	2.77	49.5	18.9	.14	.052
1984	157.3	108.5	3.25	50.0	19.1	.16	.062
1985	179.3	123.7	3.71	50.4	19.2	.19	.071
1986	202.0	139.4	4.18	50.9	19.4	.21	.081
1987	225.6	155.7	4.67	51.4	19.6	.24	.092
1988	254.0	175.3	5.26	51.9	19.8	.27	.10
1989	283.9	195.9	5.88	52.4	20.0	.31	.12
1990	315.5	217.7	6.53	52.8	20.1	.34	.13
1991	348.7	240.6	7.22	53.3	20.3	.38	.15
1992	382.7	264.1	7.92	53.7	20.5	.43	.16
1993	416.6	287.5	8.63	54.2	20.7	.47	.18
1994	450.7	311.0	9.33	54.7	20.8	.51	.19
1995	483.8	333.8	10.0	55.1	21.0	.55	.21
1996	516.2	356.2	10.7	55.6	21.2	.59	.23
1997	547.6	377.8	11.3	56.0	21.3	.63	.24
1998	579.1	399.6	12.0	56.4	21.5	.68	.26
1999	609.7	420.7	12.6	56.8	21.6	.72	.27
2000	638.6	440.6	13.2	57.1	21.8	.75	.29
Total		5009.5	150.2			8.1	3.1

Committed Potential Health Effects = 8.1 E+6 (total body person-rem) x 400E-6 (cancers/total body person-rem) + 3.1 E+6 (gonadal person-rem) x 200E-6 (genetic effects/gonadal person-rem) = 3860

(1) assumed capacity factor is 69%
(2) 0.21 mrem/yr per pCi C-14/gm C-12 in the total body
(3) 0.08 mrem/yr per pCi C-14/gm C-12 in the gonads

TABLE 13

CASE 2. WORLD-WIDE COMMITTED POTENTIAL HEALTH EFFECTS FROM CARBON-14 FOR THE U.S. LWR NUCLEAR POWER INDUSTRY

Year	LWR Installed Nuclear Capcity Cumulative Total	LWR Produced Power(1)	C-14 Annual Injection	C-14 Cumulative Injection	C-14 100 Yr. Population Dose Commitment	
					Total Body(2)	Gonads(3)
	(GWe)	(GWe-yr)	(KCi/yr)	(KCi)	(10^6 person-rem)	
1975	41.8	28.8	.864	.864	.039	.015
1976	50.7	35.0	1.05	1.91	.048	.018
1977	57.7	39.8	1.19	3.11	.056	.021
1978	66.7	46.0	1.38	4.49	.065	.025
1979	76.1	52.5	1.57	6.06	.075	.028
1980	91.9	63.4	1.90	7.96	.091	.035
1981	117.6	81.1	2.43	10.4	.118	.045
1982	146.0	100.7	3.02	13.4	.148	.056
1983	172.2	118.8	3.56	17.0	.176	.067
1984	203.3	140.3	4.21	21.2	.211	.080
1985	234.3	161.7	4.85	26.0	.244	.093
1986	267.9	184.9	5.55	31.6	.282	.108
1987	304.4	210.0	6.30	37.9	.324	.123
1988	344.0	237.4	7.12	45.0	.370	.141
1989	386.4	266.6	8.00	53.0	.419	.160
1990	430.7	292.2	8.92	61.9	.471	.179
1991	478.3	330.0	9.90	71.8	.528	.201
1992	529.3	365.2	11.0	82.8	.591	.226
1993	582.5	401.9	12.1	94.8	.656	.250
1994	638.2	440.4	13.2	108.	.722	.275
1995	694.0	478.9	14.4	122.	.793	.302
1996	751.3	518.4	15.6	138.	.867	.331
1997	809.9	558.8	16.8	155.	.941	.358
1998	869.5	600.0	18.0	173.	1.02	.387
1999	929.8	641.6	19.2	192.	1.09	.415
2000	988.4	682.0	20.5	212.	1.17	.447
Total		7081.4			11.52	4.39

Committed Potential Health Effects = 11.5E+6 (total body person-rem) x 400E-6 (cancers/total body person-rem) +
4.4E-6 (gonadal person-rem) x 200E-6 (genetic effects/gonadal person-rem)
= 5481

(1) assumed capacity factor is 69%
(2) 0.21 mrem/yr per pCi C-14/gm C-12 in the total body
(3) 0.08 mrem/yr per pCi C-14/gm C-12 in the gonads