



THE CARBON 14 CYCLE

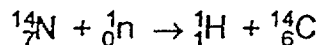
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Abstract

The purpose of this report is to assess the carbon 14 releases from a contaminated graphite incinerator and to compare these releases and their radiological impact with other ways in which carbon 14 is produced, both naturally and artificially in waste produced by the nuclear industry.

1. Recapitulation - Ways in which carbon 14 is formed

Carbon 14 has a half-life of 5730 years. It would have disappeared from the earth's atmospheric carbon dioxide a long time ago if it were not re-formed by the action of cosmic neutrons on the nitrogen in the atmosphere.



The carbon 14 formed is oxidized in the air. It is then converted into CO_2 and penetrates by photosynthesis into the metabolism of the plant and animal world. Assuming constant intensity of the cosmic radiation, the proportion in the carbon dioxide in the air, ${}^{14}\text{C}/\text{C}^{12}$, remains constant and equal to:

$$1.2 \times 10^{-12}$$

In plants assimilating CO_2 , the ratio of the concentrations in all their organic substances equals 1.2×10^{-12} while the plants are alive. The same is true for animals and humans feeding on plants.

After death, injection of carbon dioxide into the plants ceases and the ratio ${}^{14}\text{C}/\text{C}^{12}$ decreases following the formula:

$${}^{14}\text{C}/\text{C}^{12} \rightarrow 1.2 \times 10^{-12} \times e^{\frac{-0.69 t}{5730}}$$

where t = time in years.

Measurement of radioactive carbon 14, which emits a β particle with an energy of 0.156 MeV, and comparison with the C^{12} concentration allows the age of the organism (plant, animal, human) to be estimated, provided it lies between 1000 and 50 000 years.

Carbon 14 is produced continuously in the atmosphere at a rate of 27 000 curies or 10^{15} Bq per year by the action of cosmic neutrons.

The following three main modes of production of carbon 14 can be identified in nuclear reactions.

Reaction	Capture cross section in barns (10^{-24} cm^2)	Abundance of isotope in %
$^{14}\text{N} (n, p) ^{14}\text{C}$	1.8	99.63 N^{14} /Nitrogen
$^{13}\text{C} (n, \gamma) ^{14}\text{C}$	0.9×10^{-3}	1.1 C^{13} /Carbon
$^{17}\text{O} (n, \alpha) ^{14}\text{C}$	0.235	0.04 O^{17} /Oxygen

1 g of carbon 14 represents an activity of 4.6 curies.

2. Carbon 14 releases into the atmosphere

The nuclear tests carried out in the atmosphere since 1945 and particularly in the period from 1954 to 1962 were an important source of releases.

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has made an inventory of the 423 atmospheric explosions that produced a large quantity of radionuclides in the atmosphere (see Table 1).

Country	Period	Number	Power (Mt)
USA	1945-1962	193	138,6
USSR	1949-1962	142	357,5
UK	1952-1962	21	16,7
France	1960-1974	45	11,9
China	1964-1980	22	20,7

Table 1 - Main radionuclides due to nuclear tests

Radionuclide	Half-life	Activity
^3H	12.3 years	$2,4 \cdot 10^{20}$ Bq
^{14}C	5730 years	$2,3 \cdot 10^{17}$ Bq
^{54}Mn	313 days	$5,2 \cdot 10^{18}$ Bq
^{55}Fe	2.7 years	$2,0 \cdot 10^{18}$ Bq
^{85}Kr	10.7 years	$1,6 \cdot 10^{17}$ Bq
^{90}Sr	28.6 years	$6,0 \cdot 10^{17}$ Bq
^{89}Sr	50.5 days	$9,0 \cdot 10^{19}$ Bq
^{106}Ru	368 days	$1,2 \cdot 10^{19}$ Bq
^{131}I	8 days	$7,0 \cdot 10^{20}$ Bq
^{137}Cs	30.2 years	$9,6 \cdot 10^{17}$ Bq
^{140}Ba	12.8 days	$7,2 \cdot 10^{20}$ Bq
^{144}Ce	284 years	$3,0 \cdot 10^{19}$ Bq
^{238}Pu	87.7 years	$3,3 \cdot 10^{14}$ Bq
^{239}Pu	24 100 years	$7,8 \cdot 10^{15}$ Bq
^{240}Pu	6570 years	$5,2 \cdot 10^{15}$ Bq
^{241}Pu	14.4 years	$1,7 \cdot 10^{17}$ Bq
^{241}Am	433 years	$5,5 \cdot 10^{15}$ Bq

Carbon 14 is produced by the intense neutron fluxes on the nitrogen in the atmosphere and the cumulative production of carbon 14 by atmospheric explosions is estimated at 6×10^6 curies, i.e. 220 years of natural production.

Assuming that 90 % of the carbon 14 is projected to high altitude following the explosion, UNSCEAR has calculated that the integrated dose over the average human lifetime due to exposure to carbon 14 fallout would be in the order of 0.3 mSv, i.e. 30 mRem.

In power reactors, the materials subjected to neutron flux contain nitrogen as an impurity and also carbon and oxygen as major constituents of structures, fuel or the moderator.

For each type of reactor, it is possible to draw up an approximate inventory of the production of carbon 14, in either solid or gaseous form.

Table 2 gives the annual production of carbon 14 in both forms for the main types of reactor assuming operation of each reactor for 7000 hours a year.

Table 2 Carbon 14 production by different reactors

Type of reactor	Power in MWe	Total releases in curies/year	Gaseous effluents		Solid waste	
			Reactor	Reprocessing	Reactor	Reprocessing
PWR	900	50	4	14	18	14
	1300	70	5	20	25	20
Magnox or Gas-cooled	200 (1)	77	10	15	50 (1)	2
			10	30	72 (2)	3
AGR	660	110	8	5	72	25
PHWR	600	150	110	22		19
CANDU						
BWR	1000	75	8	16	35	16

(1) Example: Chinon A2: 1700 tonnes of graphite - 20 years operation
Pile activity = $50 \times 20 = 1000$ curies

(2) Example: SL A: 2400 tonnes of graphite - 18 years of operation
Pile activity = $72 \times 18 = 1296$ curies

Specific activity of graphite = 2×10^4 Bq/g

As an example, in PWRs carbon 14 is produced mainly in solid form by interaction between neutrons and the nitrogen in the stainless steel used to make the reactor internals, the nitrogen content ranging from 0.04 to 0.08 %.

Gaseous carbon 14 is produced mainly by activation of nitrogen entering the primary coolant and is released in the gaseous effluents. The average PWR release rate is about 5 to 10 curies per year of gaseous carbon 14.

For graphite reactors, the main carbon 14 production route is through irradiation of the moderator and, out of the 50 curies per year produced by a 200 to 250 MWe gas-cooled graphite-moderated reactor, 60 % is produced by interaction with the nitrogen impurities in the graphite and 40 % by interaction with the carbon 13 contained in the graphite pile.

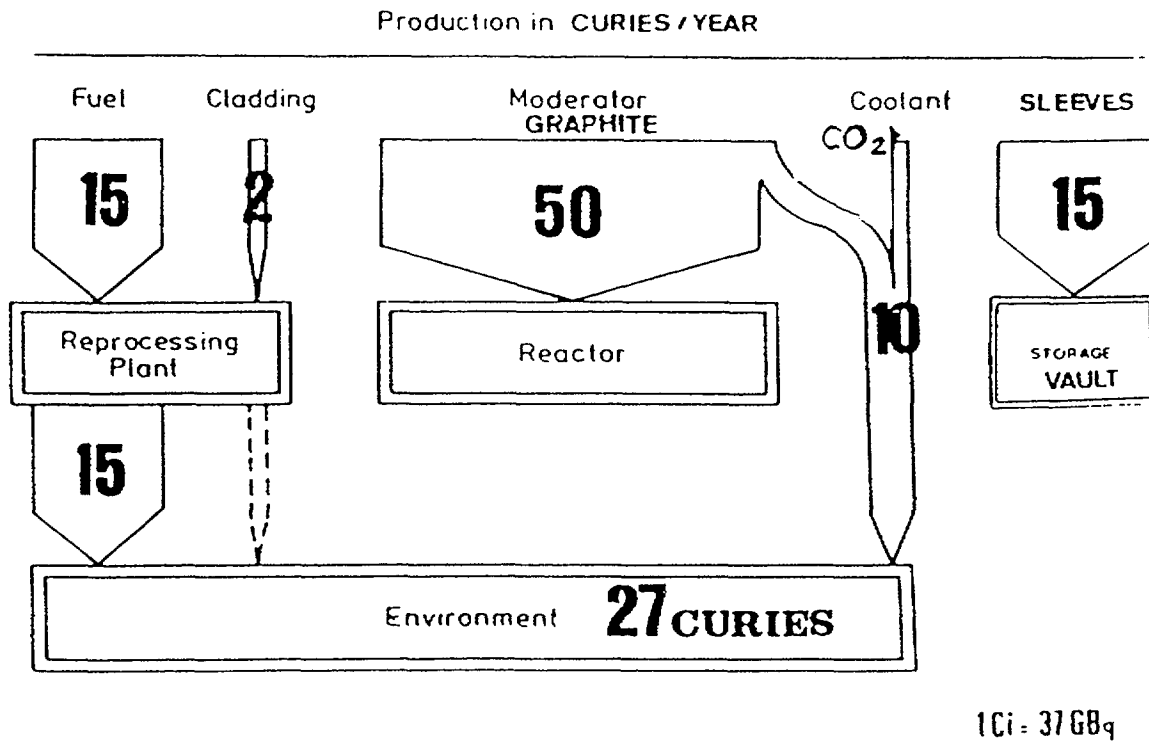


Fig 1 Schematic diagram of flows of ^{14}C produced in a 200 MWe Magnox reactor

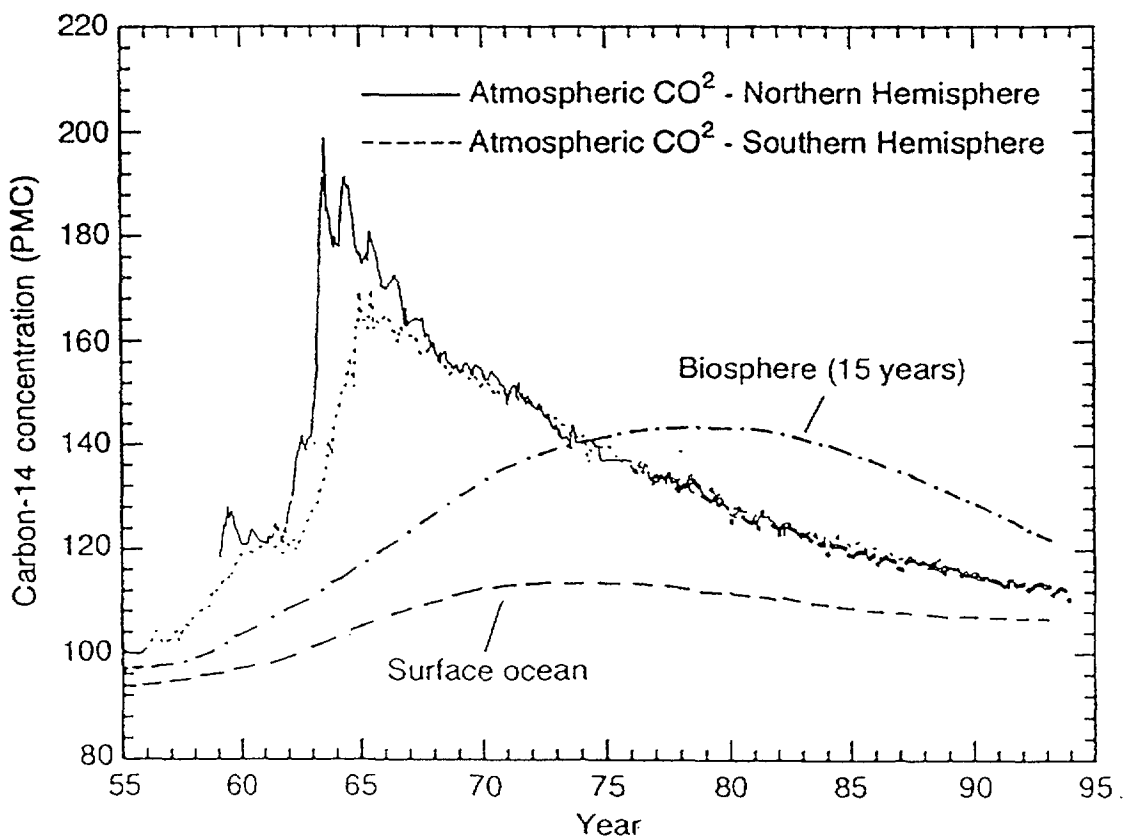


Fig. 2 Carbon-14 concentration in atmosphere; 1955 to 1994

A large portion of the carbon 14 gaseous releases from gas-cooled reactors comes from the purification of the CO₂ circuits used to cool the reactor and from the isotopic exchange between the moderator and the CO₂ circuit.

A gas-cooled reactor discharges into the environment about 27 curies of carbon 14 a year on average, i.e. 10¹² becquerels (see Figure 1).

3 Estimate of releases from an incinerator for contaminated and irradiated graphite

Fluidized bed incineration of contaminated graphite permits capturing the radionuclides contained in the graphite, except for the releases of tritium and carbon 14, which is converted into CO₂.

Since the average concentration of carbon 14 in the moderator graphite is 2 x 10⁴ Bq/g, the gaseous releases from an incinerator capable of processing 600 tonnes of graphite a year will be 12 x 10¹² Bq, i.e. 320 curies per year.

This corresponds to an increase of 1 % of the natural annual production; however, the impact must be measured over time, because of the long half-life of carbon 14 (5730 years).

Saturation can be considered to be reached after a time period equivalent to four half-lives, i.e. 20 000 years in the case of carbon 14.

The atmosphere's carbon 14 inventory is around 1.2 x 10¹⁷ becquerels, i.e. 3.1 x 10⁶ curies.

An incinerator operating for 50 years at a rate of 600 tonnes per year, i.e. incinerating a total of 30 000 tonnes of graphite, will produce an increase in the inventory of 5 x 10⁻³ times that from natural sources, which is small compared to the natural fluctuation of carbon 14 in the atmosphere (± 2%) in the last one hundred years.

The radiological impact calculated by several writers, including the IPSN, shows that, provided that precautions are taken during discharge (stack height, daytime or nighttime conditions), gaseous releases remain acceptable for the most exposed group according to ICRP (International Commission on Radiological Protection) criteria; in any case, it is much lower than the limit of 1 mSv/year established for the general public (ICRP 60) being, in fact, about 4 % of the allowable limit.

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