

# Tritium, carbon-14 and krypton-85

Jan Willem Storm van Leeuwen

Independent consultant  
member of the Nuclear Consulting Group

August 2019  
storm@ceedata.nl

## Note

In this document the references are coded by Q-numbers (e.g. Q6). Each reference has a unique number in this coding system, which is consistently used throughout all publications by the author. In the list at the back of the document the references are sorted by Q-number. The resulting sequence is not necessarily the same order in which the references appear in the text.

# Contents

1	Introduction
2	Tritium $^3\text{H}$ <ul style="list-style-type: none"><li>Hydrogen isotopes</li><li>Anthropogenic tritium production</li><li>Chemical properties</li><li>DNA incorporation</li><li>Health hazards</li><li>Removal and disposal of tritium</li></ul>
3	Carbon-14 <ul style="list-style-type: none"><li>Carbon isotopes</li><li>Anthropogenic carbon-14 production</li><li>Chemical properties</li><li>DNA incorporation</li><li>Suess effect</li><li>Health hazards</li><li>Removal and disposal of carbon-14</li></ul>
4	Krypton-85 <ul style="list-style-type: none"><li>Anthropogenic krypton-85 production</li><li>Chemical properties</li><li>Biological properties</li><li>Health hazards</li><li>Removal and disposal of krypton-85</li></ul>
	References

## TABLES

Table 1	Neutron reactions producing tritium and precursors
Table 2	Calculated production and discharge rates of tritium
Table 3	Neutron reactions producing carbon-14 and precursors
Table 4	Calculated production and discharge rates of carbon-14
Table 5	Calculated production and discharge rates of krypton-85

## FIGURES

Figure 1	Pathways of tritium and carbon-14 into human metabolism
----------	---

## 1 Introduction

The three radionuclides tritium ( $^3\text{H}$ ), carbon-14 ( $^{14}\text{C}$ ) and krypton-85 ( $^{85}\text{Kr}$ ) are routinely released into the human environment by nominally operating nuclear power plants. According to the classical dose-risk paradigm these discharges would have negligible public health effects and so were and still are permitted. This assumption turns out to be untenable based on the evidence of large epidemiological studies, such as [KiKK 2007] Q392 and [Geocap 2012] Q494. In addition the classical dose-risk paradigm does not include the evidence of non-targeted and delayed radiation effects, discussed in report m11 *Health effects of radioactivity*.

Of special importance are the radionuclides tritium and carbon-14,  $^{14}\text{C}$ . These radionuclides are biochemically indistinguishable from their non-radioactive isotopes, normal hydrogen H, respectively normal carbon (mainly  $^{12}\text{C}$ ). Carbon and hydrogen are two of the six primary building blocks (C, H, O, N, S, P) of proteins and DNA. A complicating factor is that both radionuclides are always discharged simultaneously and are incorporated in biomolecules when ingested or inhaled.

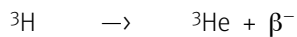
Krypton-85, a major fission product, is discharged routinely in large quantities by nuclear reactors and reprocessing plants. Although krypton is a noble gas there is evidence that it is biologically active inside the body.

## 2 Tritium $^3\text{H}$

### Hydrogen isotopes

Tritium is the radioactive isotope of hydrogen, symbol:  $^3\text{H}$ , H-3 or T, with a half-life of 12.32 years (values of 12.2 - 12.5 years are also mentioned) and a specific activity of 358 TBq/g. Chemically, tritium is indistinguishable from ordinary hydrogen H, or the other isotope deuterium ( $^2\text{H}$ , H-2 or D). Ordinary hydrogen and deuterium are stable isotopes, of which H is the most abundant: H 99.985% and D 0.015%.

In nature tritium is formed in the upper atmosphere, due to cosmic radiation. Global inventory of natural tritium is estimated at about 2.6 EBq (1 EBq = 1 exabecquerel =  $10^{18}$  disintegrations per second), equivalent with 7.3 kilograms [NCRP-62 1995] Q251. The cosmogenic production rate is estimated at about 0.15 EBq/yr. In other studies global tritium inventories of 2-7 kilograms are reported [Dworschak 1993] Q80, of which 1% in the atmosphere and 99% in the oceans as tritiated water HTO. Tritium decays to helium-3 with emission of low-energy beta radiation (electrons):



### Anthropogenic tritium production

A hydrogen bomb needs minimal 3 grams of tritium, US warheads contain an average of 5 grams, neutron bombs may contain 10-30 grams [Zerriffi 1996] Q257. Due to the decay of tritium, each warhead has to be refurbished after a number of years. Even with a constant number of warheads, tritium production has to be continued.

Thermonuclear explosions in the fifties and sixties of last century added large amounts of tritium to the atmosphere, peaking at about 115 EBq in 1963. It will decay to about 3 EBq in the year 2030.

The artificial emission of tritium by nuclear installations surpasses the natural generation by cosmic radiation [Dworschak 1993] Q80.

Table 1

Some neutron reactions producing tritium or precursors of tritium

$^6\text{Li}$	(n, $\alpha$ )	T
$^7\text{Li}$	(n,n $\alpha$ )	T
$^{10}\text{B}$	(n,2 $\alpha$ )	T
$^2\text{H}$	(n, $\gamma$ )	T
$^3\text{He}$	(n,p)	T
$^9\text{Be}$	(n, $\alpha$ )	$^6\text{Li}$
$^{10}\text{B}$	(n, $\alpha$ )	$^7\text{Li}$
$^{12}\text{C}$	(n, $\alpha$ )	$^9\text{Be}$
$^{14}\text{N}$	(n,t)	$^{12}\text{C}$
ternary fission		

Nuclear reactors generate large amounts of tritium. Estimates for LWR's vary from 743-1018 TBq/GWe.a tritium [IAEA-203 1981] Q74, [NEA 1980] Q75, [Dworschak 1993] Q80, [NRC 1996] Q16. Most tritium arises from ternary fission in the fuel, the rest from various neutron reactions on light elements present as impurities or components in fuel, coolant and cladding. Chemicals are added to the cooling water, to control the reactivity and the chemistry of the water, in particular boric acid  $\text{H}_3\text{BO}_3$  and lithiumhydroxide  $\text{LiOH}$ .

Heavy-water moderated reactors (HWR), like CANDU, produce a hundred times more tritium than a LWR (NCRP-62 1995 [Q251]), see Table 2, mainly due to the D (n,γ) T reaction in the heavy water moderator. It is not clear which fraction of this is annually released at the reactor site. Eventually, all tritium in the moderator will end up in the environment.

Production of tritium in gas-cooled reactors (Magnox, AGR) is about the same as in LWR's [NEA 1980] Q75.

Table 2

Calculated production and discharge rates of tritium in LWR's and HWR's in TBq/GW(e).a

Sources: [NCRP-62 1995] Q251, [NEA 1980] Q75

source	PWR NCRP-62	PWR NEA	BWR NCRP-62	BWR NEA	HWR NCRP-62	HWR NEA
fuel	555-925	750	555-925	750	555	750
coolant	30.7	40	2.3	0	$2.2 \cdot 10^{14}$	$9.0 \cdot 10^{14}$
total		790		750	$2.26 \cdot 10^{14}$	$9.08 \cdot 10^{14}$
discharged *	1.3 + 29.6	4 + 33	0.7 + 1.6	2 + 5	185 + 1850	600 + 150

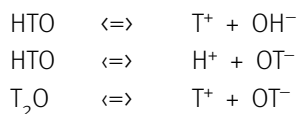
- Discharges at the reactor site. First number gaseous effluents, second liquid effluents

According to [NCRP-62 1995] Q251, most tritium, about 900 TBq/GWe.a, remains in the spent fuel elements. About 85-90% of the tritium retained the spent fuel elements is discharged into the air and sea at reprocessing plants, when the fuel is dissolved and reprocessed. About 10-15% remains fixed in the Zircalloy cladding, as zirconiumtrihydride.

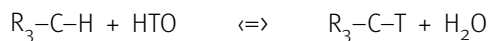
If the spent fuel is not reprocessed, this tritium remains in the elements. However, during the long interim storage period of the spent fuel (tens of years), before it is conditioned for final disposal, a part of the remaining tritium slowly diffuses through the Zircalloy cladding and will be discharged as yet.

### Chemical properties

Hydrogen atoms and so tritium atoms, are very mobile in the aquatic system and as a consequence in the biochemical system. In tritiated water quickly three chemical equilibria are established:



Exchange reactions with biochemical molecules may occur via various mechanisms, via ions or otherwise, schematically:



When the tritium atom decays to helium-3, a carboniumion may be formed, chemically a very reactive species:



Tritium is discharged into the environment as tritiated water HTO and relatively small amounts of hydrogen gas HT or T<sub>2</sub>. Tritium atoms, like ordinary hydrogen atoms, are very mobile in the aquatic system and as a consequence in the biochemical system. They are readily incorporated into biomolecules, which may enter the food chain. Direct ingestion via drinking water, prepared from river water, is another pathway.

Hydrogen atoms bound to the N, O or S atoms in biomolecules of a living cell are easily exchanged with hydrogen atoms from the water molecules in the cell. In a living cell this exchange process goes on continuously. When tritiated water HTO enters a cell, normal hydrogen atoms in the biomolecules are exchanged with tritium atoms. By this process biomolecules with built-in radioactive hydrogen atoms are formed. In this way tritium enters the food chain, for example in milk and vegetables. In the body organically bound tritium (OBT) is slowly formed by metabolic reactions. Once it is formed it stays in the body for much longer periods (20 to 30 times longer) than HTO [Fairlie 2008] Q443.

Tritium can enter the body via food as OBT and via drinking water containing tritiated water HTO. The biological half-life of OBT in the human body is much longer than of HTO. Normal hydrogen atoms in the DNA molecules are exchanged for tritium via OBT and HTO. The exposure of DNA to the radiation from tritium comes from within the DNA molecules themselves and from biomolecules and HTO adjacent to the DNA molecules. Despite the short-range of the  $\beta$ -radiation, its effects can be significant. See also [NCRP-62 1995] Q251, [NCRP-63 1979] Q253, [Fairlie 2007] Q373, [AGIR 2007] Q444.

If a tritium atom decays during its stay in a DNA molecule it transforms into a stable helium-3 atom, a noble gas, while emitting a beta-particle. The helium atom does not form any chemical bonding and is ejected from the DNA molecule. The emitted beta particle causes additional ionisations and broken bonds in the vicinity of the decay event. Obviously such an event may cause a mutation or lesion in the DNA molecule. Studies in the 50's, 60's and 70's of the 20th century concluded that deleterious effects of tritiated water from nuclear installations were unlikely at its (then present) levels. This is not to imply that tritium at higher concentrations would be not carcinogenic or mutagenic [NCRP-63 1979] Q253. Apparently no fundamental, biochemical reaction mechanisms initiated by tritium decay (interaction of  $\beta$  radiation with chemical bonds, transmutation and recoil effects) in living organisms have been thoroughly experimentally investigated. Experimental research in mice and rats of biological effects of tritium is reported, but extrapolation to human health effects is difficult. Little direct evidence of tritium effects in humans exists, according to [Straume 1991] Q258.

The radiotoxicological classification of tritium is based on theoretical computations, starting from the relatively weak beta radiation of this nuclide [NCRP-63 1979] Q253. Indications are found that under certain circumstances the biological activity of tritium is higher than predicted [AGIR 2007] Q444, [Straume 1991] Q258, [NCRP-89 1987] Q260. These findings may point to non-targeted and delayed affects.

### **DNA incorporation**

Observations from experiments with mice (1966-1973) suggest that the most hazardous time for mammals exposed to tritium as thymidine, and possibly other DNA precursors as well, would be in utero or as neonatals. No effects following exposure of adult mice were demonstrable in these experiments [NCRP-62 1995] Q251.

### **Health hazards**

Studies in the 50's, 60's and 70's of the 20th century conclude that deleterious effects of tritiated water from nuclear installations appear unlikely at the (then) present levels. This is not to imply that tritium in sufficient amounts would be not carcinogenic or mutagenic [NCRP-63 1979] Q253.

Apparently no fundamental, biochemical reaction mechanisms initiated by tritium decay (interaction of  $\beta$  radiation with chemical bonds, transmutation and recoil effects) in living organisms are thoroughly experimentally investigated. Experimental research in mice and rats of biological effects of tritium is reported, but extrapolation to human health effects is difficult. Few if any evidence of tritium effects in humans exists [Straume 1991] Q258.

The long term effects of a significant higher tritium concentrations in the environment are not clearly understood. On global scale, the rise of the tritium concentrations may seem relatively harmless, but on regional scale relatively high concentrations in surface waters and prolonged exposure may occur. In Western Europe, with numerous nuclear power plants, rivers are used as resources for cooling water for the plants, but the same rivers provide the drinking water for millions of people. We found no empirical data on effects of chronic exposure to tritiated water HTO or tritiated organic material (OBT: organic bound tritium) in humans.

Health hazards of radioactivity are discussed in more detail in report m11 *Health effects of radioactivity*.

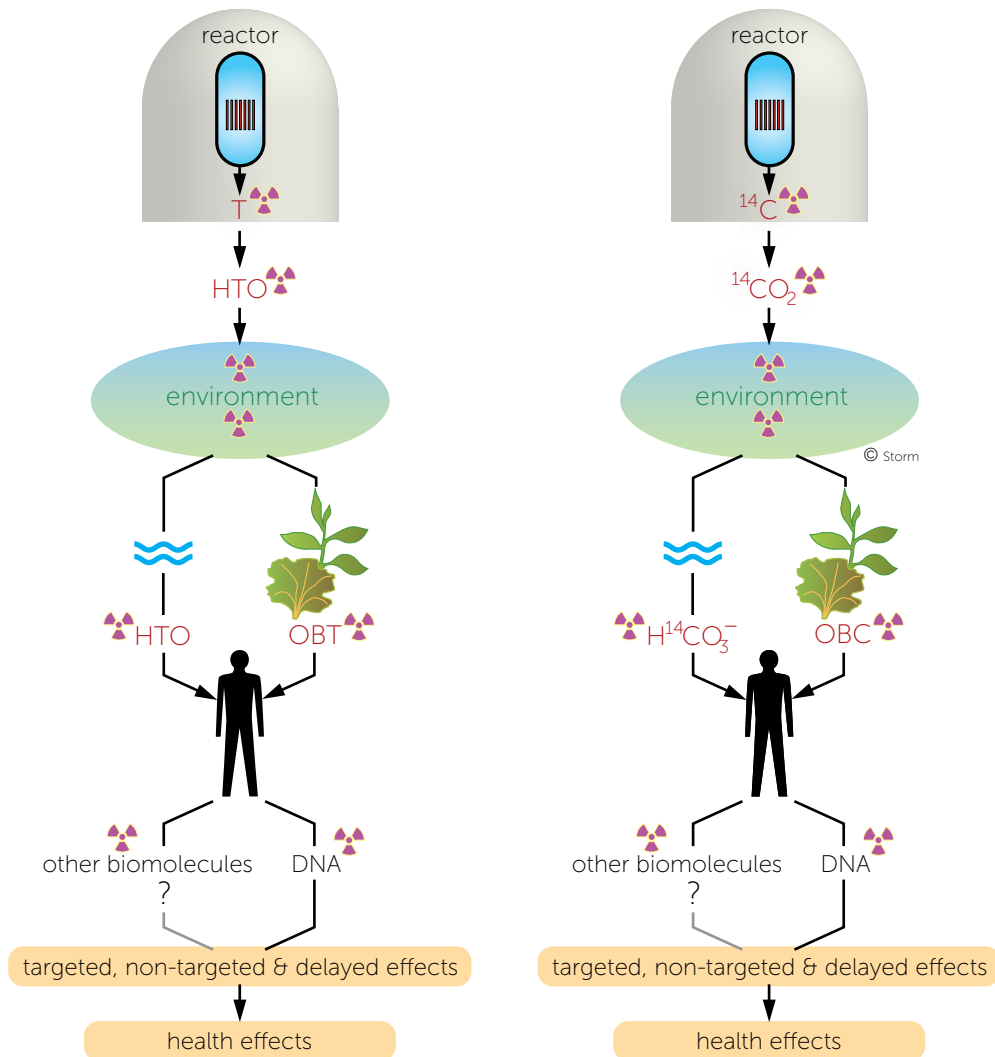


Figure 1

Pathways of radioactive hydrogen (tritium) and carbon-14 into the human metabolism. Both radionuclides are routinely released into the environment by operating nuclear power plants. The pathways are similar. It is generally assumed that damage to DNA molecules cause adverse health effects. Cell damage is not limited to the cells directly hit by radiation, due to the bystander effect. See report m11 *Health effects of radioactivity*.

### Removal and disposal of tritium

Tritium is released mainly as tritiated water HTO and partly as hydrogen gas HT or T<sub>2</sub>. Methods suitable for stripping tritium from large volumes wastes include fractional distillation of HTO-H<sub>2</sub>O, cryogenic distillation of HT-H<sub>2</sub> mixes (after electrolysing tritiated water), HT-HTO exchange and H<sub>2</sub>S-H<sub>2</sub>O exchange [NCRP-62

1995] Q251. After catalytic enrichment the tritium may be immobilized as zirconiumtride in steel containers [NEA 1980] Q75.

Practically, it is not feasible to extract tritium completely from tritiated water. Differences in physical properties of HT and HTO compared to H<sub>2</sub> and H<sub>2</sub>O are too small to allow an efficient separation. Moreover, all separation methods are very energy-intensive and require costly equipment.

Theoretically tritiated water can be immobilized in drying agents (e.g. silica gel, polymers, clays) or cement. The solid wastes can be packed in appropriate containers and disposed of in a permanent repository. If done, all the cooling water of the reactor were to be immobilized at every occasion the cooling water of the reactor has to be refreshed. Such a procedure would produce large numbers of containers with tritiated waste. In practice all tritiated water from nuclear power is being discharged into the environment.



### 3 Carbon-14

#### Carbon isotopes

Carbon has two stable isotopes, of which  $^{12}\text{C}$  or C-12, ordinary carbon, is the most abundant with 98,9%; the other stable isotope is  $^{13}\text{C}$  (1.1%). The element has an important radioactive isotope,  $^{14}\text{C}$  or C-14, with a half-life of 5730 years (other values also are reported). The specific activity is 0.165 TBq/gram.

In nature  $^{14}\text{C}$  is formed in the upper atmosphere by reactions neutrons from cosmic radiation with carbon and nitrogen atoms, mainly by the  $^{14}\text{N}$  (n,p)  $^{14}\text{C}$  reaction. The global inventory of cosmogenic C-14 in the atmosphere is estimated at 0.14 EBq and the production rate at 0.0014 EBq/yr [NCRP-81 1993] Q256. According to [NRC 1996] Q16 the total global inventory is 9.25 EBq, of which about 0.15 EBq in the atmosphere. Carbon-14 decays with emission of beta rays to nitrogen-14, which is stable:



#### Anthropogenic carbon-14 production

Table 3

Some neutron reactions producing carbon-14 and precursors

$^{13}\text{C}$	(n, $\gamma$ )	$^{14}\text{C}$
$^{12}\text{C}$	(n, $\gamma$ )	$^{13}\text{C}$
$^{14}\text{N}$	(n,p)	$^{14}\text{C}$
$^{15}\text{N}$	(n,d)	$^{14}\text{C}$
$^{16}\text{O}$	(n, $\gamma$ )	$^{17}\text{O}$
$^{16}\text{O}$	(n, $^3\text{He}$ )	$^{14}\text{C}$
$^{17}\text{O}$	(n, $\alpha$ )	$^{14}\text{C}$
ternary fission		

Table 4

Calculated production and discharge rates of carbon-14 in LWR's and HWR's, in TBq/GW(e).a

Sources: [NCRP-81 1993] Q256, [NEA 1980] Q75

source	PWR NCRP-81	PWR NEA	BWR NCRP-81	BWR NEA	HWR NCRP-81	HWR NEA
fuel	0.74	0.4	0.74	0.6		1.0
coolant	0.37-0.52	0.3	0.26	0.4	24	20
core hardware *	1.1-1.6		1.9-2.7			
total	2.2-2.9	4 + 33	2.9-3.7			

\* Cladding and structural components of fuel elements

Thermonuclear explosions in the 1950s and 1960s added about 0.36 EBq C-14, so the total global inventory of the atmosphere at present is estimated at 0.50 EBq.

Large amounts of carbon-14 are generated in nuclear reactors, due to neutron-induced reactions of oxygen (in  $\text{UO}_2$  and  $\text{H}_2\text{O}$ ) and nitrogen and carbon which may be present as components of coolant, moderator, structural materials, fuel, or as impurities.

Calculated production rates in LWR's and HWR's (heavy-water moderated reactors, like CANDU) according to [NCRP-81 1993] Q256 and [NEA 1980] Q75 are summarized in Table 4. According to NRC 1996 [Q16] spent LWR fuel elements contain about 1.5 Ci  $^{14}\text{C}$  per Mg heavy metal (HM), or 0.056 TBq/Mg.

It is assumed that most  $^{14}\text{C}$  in cooling water is released at the reactor site and is discharged in the gaseous effluent, less than 1% in the liquid effluent. Few results of measurements of carbon-14 emissions are published [NEA 1980] Q75. In [OSPAR 2002] Q236 not a single measurement of C-14 emission is listed. In [NCRP-81 1993] Q256 one German publication (1982) is mentioned, which reported an annual release of 5.6 Ci/GW(e)y (0.21 TBq/GW(e).y) for PWRs and 13.5 for BWRs (0.50 TBq/GW(e).y). The experimental data from which the estimates are made are limited.

If the fuel is reprocessed, the  $^{14}\text{C}$  in the fuel will be emitted as  $^{14}\text{CO}_2$  at the reprocessing plant.

A PWR discharges its carbon-14 mainly as alkanes, over 80% methane and ethane  $^{14}\text{CH}_4$  and  $^{14}\text{C}_2\text{H}_6$ , a BWR in the form of carbon dioxide  $^{14}\text{CO}_2$  and a few percent of carbon monoxide  $^{14}\text{CO}$  and  $^{14}\text{CH}_4$  [NEA 1980] Q75. According to [NCRP-81 1993] Q256, a BWR discharges  $^{14}\text{C}$  in a chemical form other than  $\text{CO}_2$ .

In graphite moderated reactors, e.g. Magnox, AGR and RBMK, much more  $^{14}\text{C}$  is produced than in LWR's.

C-14 emission by reprocessing plants is mentioned as problem in [WNA-68 2003] Q248, but is not mentioned in a more recent version of October 2012.

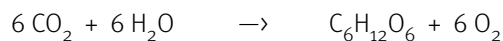
### Chemical properties

Carbon-14 atoms are chemically identical with stable carbon atoms (carbon-12 and about 1% carbon-13) and enters the biosphere mainly as  $^{14}\text{CO}_2$ . A part is emitted as hydrocarbons (methane, ethane) into the atmosphere, which are slowly oxidized to  $\text{CO}_2$ . Carbondioxide slightly dissolves in (rain) water and a chemical equilibrium is established:

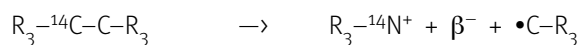


Via bicarbonate  $^{14}\text{C}$  can be bound in the bones of animals and humans.

By photosynthesis in plants,  $\text{CO}_2$  is bound into organic materials, which enter the food chain:



One or more C atoms may be  $^{14}\text{C}$ . In this way also tritium may be built in organic molecules and a combination also is possible. Near a nuclear power plant, both isotopes are present. When a  $^{14}\text{C}$  atom decays to a stable nitrogen atom several reactions are possible, e.g.:



Both fragments are chemically very reactive.

By the end of the second growing season, most food plants will be nearly at equilibrium with the atmospheric  $\text{CO}_2$ . The time delay for most foodstuff of plant origin to reach maximum concentration of  $^{14}\text{C}$  is not more than one year [NCRP-81 1993] Q256. Soft tissues in animals (and man) seem to lag behind the tropospheric  $^{14}\text{C}/^{12}\text{C}$  ratio by about one to two years. Since some tissues or body compartments have long turnover times, some approaching the life span, total body carbon in the adult subjected to a change in  $^{14}\text{C}/^{12}\text{C}$  intake ratio may not approach equilibrium for decades.

Fetal tissues will more nearly reflect the current  $^{14}\text{C}$  composition of the food available to the mother.

## DNA incorporation

Male sperm cells are produced from spermatogonia continuously in the adult male and consequently the  $^{14}\text{C}$  specific activity in the nuclear material must resemble that of the food with a lag time of one or two years. Female oocytes are laid down in the fetus before birth and thereafter remain dormant until they ripen just before being shed. It is uncertain how much of the adjacent tissue may be subject to turnover and renewal but, in any case, none of the DNA of the ovum, except the small fraction renewed by repair processes would contain  $^{14}\text{C}$  of the current specific activity. There will, therefore, be a delay of 16-40 years in the human female for expression of the genetic effects of a given  $^{14}\text{C}/^{12}\text{C}$  ratio. Calculation of the genetic effect to the current generation of an added increment of  $^{14}\text{C}$  to the atmosphere using a lag time of 1-2 years, therefore, is conservative according to [NCRP-81 1993] Q256.

## Suess effect

Before the nuclear age, an equilibrium existed between production and decay of natural  $^{14}\text{C}$ , so the  $^{14}\text{C}/^{12}\text{C}$  ratio in the atmosphere (as  $\text{CO}_2$ ) was constant. Via photosynthesis carbon from the air (as  $\text{CO}_2$ ) is integrated in organic material. Generally, every living organism has the same  $^{14}\text{C}/^{12}\text{C}$  ratio in its biochemical molecules. After death, no new  $^{14}\text{C}$  atoms are bound in the organism. With time the  $^{14}\text{C}/^{12}\text{C}$  ratio decreases by decay of the  $^{14}\text{C}$  atoms. This is the base of the carbon-14 dating method.

Due to the anthropogenic increase of the  $^{14}\text{C}$  inventory of the atmosphere, one might expect the  $^{14}\text{C}/^{12}\text{C}$  ratio in living organisms would increase. However, this effect is offset by the increase of the  $\text{CO}_2$  content by burning fossil fuels, which are free of  $^{14}\text{C}$ . In effect, the  $^{14}\text{C}/^{12}\text{C}$  ratio in the atmosphere decreases by dilution of  $^{14}\text{C}$  by stable C; this phenomenon is called the Suess effect, after the investigator who described it for the first time (in 1955).

Carbon-14 is emitted by a nuclear power plant as  $^{14}\text{CO}_2$  and  $^{14}\text{CH}_4$  and some higher alkanes [AGIR 2007] Q444. In air the alkanes are slowly oxidized to  $^{14}\text{CO}_2$ . Carbon-14 atoms are chemically identical with stable carbon atoms and enters the biosphere mainly as  $^{14}\text{CO}_2$ . Carbon dioxide slightly dissolves in (rain) water and a chemical equilibrium is established in which bicarbonate ions are formed. Carbon-14 can be bound in the bones of animals and humans via bicarbonate ions in drinking water.

Via photosynthesis in plants,  $^{14}\text{CO}_2$  can be bound into biomolecules: the built-in C-14 atoms are called organically bound carbon, or OBC. Via the food chain OBC can enter the human body and become incorporated into DNA molecules. A combination with tritium is possible. Both isotopes are present in the vicinity of all nuclear power plants, because tritium and carbon-14 are discharged into the human environment.

## Health hazards

Damage to DNA molecules occurs in a similar manner as by tritium. A carbon-14 atom decays into a stable nitrogen-14 atom by emitting a beta particle. The chemical properties of nitrogen differ widely from carbon and the beta particle causes secondary ionisations and lesions.

Some DNA molecules in the human body may remain practically unchanged for decades, for example in egg cells, so the medical consequences of carbon-14 ingestion may have long incubation times.

Experiments proved beta-particles (electrons) with remarkably low energy to be able to destroy vital parts of DNA and RNA molecules [Collins 2003] Q270. Up until about the year 2000 the common opinion was that DNA and RNA molecules could be damaged only by electrons with an energy of more than 10 eV. Experiments proved that electrons with energies as low as 3 eV can break both strings of the DNA double helix. There is evidence of a lower energy limit well below 1 eV.

The DNA repair mechanism is able to repair a single lesion, but complex and multiple lesions may result in

misrepair, ie mutations may occur.

Sparse data are available to base on estimates of genetic hazards associated with  $^{14}\text{C}$ -labelled nucleic acid precursors, according to NCRP-63 1979 [Q253] and NCRP-89 1987 [Q260]. This is confirmed by NCRP-81 1993 [Q256], which mentions only a few studies with theoretical calculations, concerning the dispersion of  $^{14}\text{CO}_2$  in air. As with tritium, we could not find any evidence that fundamental, biochemical reaction mechanisms initiated by carbon-14 decay (beta radiation, transmutation and recoil effects) in living organisms are thoroughly experimentally investigated.

The radiotoxicological classification of carbon-14 is based on theoretical computations, starting from the relatively weak beta radiation of the nuclide, and not on empirical biological evidence.

Long term effects of a significant higher carbon-14 concentration in the environment are unknown. On global scale, the rise of the concentrations may seem insignificant, but on regional scale relatively high concentrations in air, water and plants may occur.

Health hazards of radioactivity are discussed in more detail in report m11 *Health effects of radioactivity*.

#### **Removal and disposal of carbon-14**

Removal of  $^{14}\text{C}$  from gaseous effluents requires catalytic oxidation of hydrocarbons and CO to  $\text{CO}_2$ . The  $\text{CO}_2$  may be removed from the off-gas with a solvent-absorption process. The  $\text{CO}_2$  so collected could be discharged into a slurry of calciumhydroxide  $\text{Ca}(\text{OH})_2$  and converted to calciumcarbonate  $\text{CaCO}_3$ .

Another technique is the double caustic scrubbing process: absorption in sodium hydroxide solution and then reaction with calcium hydroxide solution for precipitating the carbonate from the first solution as calciumcarbonate [NEA 1980] Q75.

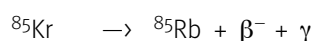
The  $\text{CaCO}_3$  can be immobilized using several agents, e.g. cement, for permanent disposal.

## 4 Krypton-85

Krypton is one of the six inert gases (noble gases), the other five being helium, neon, argon, xenon and radon. The atmospheric abundance is 1.14 ppm (volume).

There are six stable isotopes of krypton and a number of radioactive isotopes, of which krypton-85 ( $^{85}\text{Kr}$  or Kr-85) is the most important.

The half-life of  $^{85}\text{Kr}$  is 10.7 years, the specific activity 14.5 TBq/gram. It decays with emission of beta and gamma rays to stable rubidium-85.



Krypton-85 is present in the natural environment due to spontaneous and neutron-induced fissions of natural uranium and neutron capture by stable  $^{84}\text{Kr}$  of cosmogenic neutrons in the upper atmosphere. Calculated world equilibrium inventory is about 0.08 TBq in the upper three meters of total land and water surface and about 0.4 TBq in the atmosphere.

### Anthropogenic krypton-85 production

Nuclear weapons test (1945-1962) added about 0.19 EBq ( $1.9 \times 10^5$  TBq). The current world inventory of  $^{85}\text{Kr}$  is calculated at about 0.074 EBq [NCRP-44 1975] Q259.

In nuclear reactors large amounts of krypton-85 are produced, as one of the major fission products.

Table 5

Calculated production and discharge rates of Kr-85 in LWR's and HWR's, in TBq/GW(e).a

Sources: [NCRP-44 1975] Q259, [NEA 1980] Q75

source	PWR NCRP-44	PWR NEA	BWR NCRP-44	BWR NEA	HWR NCRP-44	HWR NEA
fuel	11000	14000	11000	14000	11000	14000
coolant	0	0	0	0	0	0
discharged *	<110	10	<110	10	<110	10

- Discharges at the reactor site in gaseous effluents

### Chemical properties

Krypton, like the other noble gases, is tasteless, colorless and chemically (almost) inert. A few fluor compounds of krypton are synthesized at cryogenic temperatures. The gas has a low solubility in water and a high solubility in nonpolar solvents. Noble gases can be bound in clathrates in which the atoms are physically entrapped in cages of organic molecules.

The decay product rubidium is an alkali metal and has similar chemical properties as potassium.

### Biological properties

Being chemically inert, krypton and the other noble gases are not usually involved in biological processes. They are, however, absorbed into the tissues of the body via inhalation and dissolution in body fluids and tissues. Xenon has been shown to combine with specific sites with certain protein molecules. Krypton is

characterized by low blood solubility, high lipid solubility and rapid diffusion in tissue. Exceptions to the the biologically inert characterization of inert gases have been noted by numerous studies. A comparatively high uptake of krypton by the adrenal gland has been reported. These phenomena are not understood [NCRP-44 1975] Q259.

### **Health hazards**

On global scale the genetic and overall carcinogenic effects from Kr-85 are calculated to be small as compared with other possible sources of deleterious effects.

The possible interaction of radiation from krypton-85 and solar ultraviolet (UV) should be mentioned. In order to understand better the implications of long-term <sup>85</sup>Kr releases to the atmosphere, epidemiological and laboratory studies should be undertaken to define the nature and degree of interaction, if any, of UV radiation with ionizing radiation in the induction of skin cancer [NCRP-44 1975] Q259.

### **Removal and disposal**

Absorption on charcoal or molecular sieves at ambient temperatures is only suitable for delaying the release of waste gases and not effective for control of <sup>85</sup>Kr.

Absorption on charcoal at liquid nitrogen temperatures is adaptable for recovery of krypton. Because the beds are cooled and heated alternatively, energy consumption for refrigeration is high. The system has several other severe disadvantages.

Cryogenic distillation is to be one of the more promising processes for krypton and xenon recovery. The gases can be stored in pressurized cylinders. This process also has a relatively high specific energy consumption. Selective absorption or extraction by fluorocarbon solvents has been tested.

On a laboratory scale other processes have been investigated: recovery by permselective membranes and clathrate precipitation from organic solvents [NCRP-44 1975] Q259, [NEA 1980] Q75.

In practice all krypton-85 is released into the atmosphere.

## References

- [Q16]  
NRC 1996  
Rasmussen N C (chair) et al.,  
*Nuclear Wastes. Technologies for separations and transmutation*,  
National Research Council, NRC  
Washington DC: National Academy Press, 1996.
- [Q74]  
IAEA-203 1981  
*Handling of Tritium-bearing wastes*,  
Technical Report Series No 203,  
International Atomic Energy Agency (IAEA), Vienna, 1981.
- [Q75]  
NEA 1980  
*Radiological significance and management of Tritium, Carbon-14, Krypton-85, Iodine-129, arising from the nuclear fuel cycle*,  
Report by the NEA Group of Experts  
Nuclear Energy Agency (NEA),  
OECD, Paris, April 1980.
- [Q80]  
Dworschak 1993  
Dworschak H,  
'Characteristics, origin, production, applications', in:  
F. Mannone (ed.),  
*Safety in Tritium Handling Technology*,  
EUR 15144 ECSC, EEC, EAEC, Brussels/Luxembourg, 1993,  
Dordrecht: Kluwer, 1993.
- Q236  
OSPAR 2002  
Liquid discharges from nuclear installations in 2000,  
Radioactive Substances Series  
OSPAR Commission 2002,  
ISBN 0 946956 91 X  
<p00158\_Liquid Discharges from Nuclear Installations, 2000.pdf>  
[www.ospar.org/v\\_publications/](http://www.ospar.org/v_publications/)  
retrieved 21 November 2013
- [Q248]  
WNA-68 2003 ???  
Energy subsidies and external costs,  
World Nuclear Association, Information Paper # 68  
[www.world-nuclear.org/info/inf68.html](http://www.world-nuclear.org/info/inf68.html)
- [Q251]  
NCRP-62 1995  
Tritium in the environment,  
National Council on radiation Protection and Measurements,  
NCRP Report 62,  
Washington DC, January 1995,  
(reprint of 1st edition 9 March 1979 and 2nd edition 15 May 1989)
- [Q253]  
NCRP-63 1979  
Tritium and other radionuclide labelled organic compounds incorporated in genetic material,  
National Council on radiation Protection and Measurements,  
NCRP Report 63,  
Washington DC, 30 March 1979.
- [Q256]  
NCRP-81 1993  
Carbon-14 in the environment,  
National Council on radiation Protection and Measurements,  
NCRP Report 81,  
Washington DC, 15 May 1985, first reprinting April 30 1993.
- [Q257]  
Zerrieff 1996  
Zerrieff H,  
*Tritium: the environment, health, budgetary and strategic effects of the Department of Energy (DOE) decision to produce tritium*,  
Institute for Energy and Environmental Research IEER,  
Takoma Park, Maryland, January 1996, revised March 1996.  
[www.ieer.org/reports/tritium.html](http://www.ieer.org/reports/tritium.html)  
(retrieved July 2013)
- [Q258]  
Straume 1991  
Straume T,  
*Health risks from exposure to tritium*,  
UCRL-LR-105088,  
Lawrence Livermore National Laboratory,  
University of California, Livermore, CA, February 22, 1991.
- [Q259]  
NCRP-44 1975  
*Krypton-85 in the atmosphere. Accumulation, biological significance and control technology*,  
National Council on radiation Protection and Measurements,  
NCRP Report 44,  
Washington DC, July 1, 1975
- [Q260]  
NCRP-89 1987  
Genetic effects from internally deposited radionuclides,  
National Council on radiation Protection and Measurements,  
NCRP Report 89,  
Washington DC, 15 August 1987.
- Q270  
Collins 2003  
Collins G P,  
Fatal attachments. Extreme low-energy electrons can wreck DNA,  
Scientific American, September 2003, pp14-15.

Q373

Fairlie 2007

Fairlie, I,

Tritium hazard report: pollution and radiation risk from  
Canadian nuclear facilities,

Greenpeace, June 2007,

file: tritium-hazard-report-pollu.pdf

[www.greenpeace.org/canada/en/documents-and-links/  
publications/](http://www.greenpeace.org/canada/en/documents-and-links/publications/)

Q392

KiKK 2007

Kaatsch P, Spix C, Schmiedel S, Schulze-Rath R,

Mergenthaler A & Blettner M,

Epidemiologische Studie zu Kinderkrebs in der  
Umgebung von Kernkraftwerken

(KiKK-Studie),

Vorhaben StSch 4334 (in German),

Im Auftrag des Bundesministeriums für Umwelt,

Naturschutz und Reaktorsicherheit und des Bundesamtes  
für Strahlenschutz, Germany, 2007,

4334\_KiKK\_Gesamt\_T.pdf

[www.bfs.de/de/bfs/druck/Ufoplan/](http://www.bfs.de/de/bfs/druck/Ufoplan/)

[Q443]

Fairlie 2008

Fairlie I,

The hazards of tritium – revisited,

Medicine, Conflict and Survival,

Vol 24:4. October 2008. pp 306 -319.

[Q444]

AGIR 2007

Review of Risks from Tritium,

Report of the independent Advisory Group on Ionising  
Radiation (AGIR),

Documents of the Health Protection Agency, Radiation,  
Chemical and Environmental Hazards, RCE-4,

Oxford. United Kingdom, November 2007,

filename: RCE\_Advice\_on\_tritium.pdf

[http://www.hpa.org.uk/web/HPAweb&HPAwebStandard/  
HPAweb\\_C/1197382220012.](http://www.hpa.org.uk/web/HPAweb&HPAwebStandard/HPAweb_C/1197382220012)

Q494

Geocap 2012

Sermage-Faure C, Laurier D, Goujon-Bellec S, Chartier M,

Guyot-Goubin A, Rudant J, Hémon D & Clavel J,

Childhood leukemia around French nuclear power plants  
– The Geocap study, 2002-2007,

International Journal of Cancer, doi: 10.1002/ijc.27425,

February 2012,

<http://onlinelibrary.wiley.com/doi/10.1002/ijc.27425/pdf>  
download 5 March 2012.