# Pathways of radioactive contamination

Jan Willem Storm van Leeuwen Independent consultant

member of the Nuclear Consulting Group

July 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

- Releases of radioactivity into the human environment Natural radioactivity Human-made radioactivity Authorised routine releases Unplanned discharges Violent nuclear accidents Insidiuous disasters Risk factors
- 2 Dispersion of radionuclides Dispersion by air Dispersion via water Dispersion patterns
- Health hazard assessment
  Variables and parameters
  Timescale
  Radioactivity and half-life
  Biased standards
  Examples: technetium and ruthenium
- 4 Radionuclides in the food chain Bioaccumulation Observable effects in the environment Tritium and carbon-14 Tritium Incorporation of tritium in DNA Health hazards Incorporation of carbon-14 in DNA Uncertainties

References

### FIGURES

Figure 1	Difference in tree rings in pine logs from Chernobyl
Figure 2	Pathways of tritium and carbon-14 into the human metabolism

# 1 Releases of radioactivity into the human environment

# Natural radioactivity

In the front end processes of the nuclear chain, including extracting uranium from its ore, only naturally occurring radioactivity is involved: uranium and its decay daughters. In the front end of the nuclear chain uranium is separated from its radioactive decay daughters. The latter end up in the waste streams and are chemically mobile. Inevitably a part of this waste stream of mobilised natural radioactivity is discharged into the biosphere.

No principal difference exists between natural and human-made radioactivity; radioactive materials occurring in nature emit alpha, beta and gamma radioaction, and so do the radioactive materials generated in nuclear reactors. Consequently it is a misconception to think natural radioactivity is harmless to humans. Radioactive materials generated in a reactor consist of dozens of kinds of radionuclides which do not occur in nature, for reason of their relatively short half-lifes.

# Human-made radioactivity

The radioactivity of nuclear fuel increases in the reactor a billionfold by the fission process, due to the generation of fission products, actinides and activation products. For each radioactivity unit entering the reactor, a billion units will be generated in the reactor.

In view of the enormous amounts of radioactivity involved in the reactor operation and in the downstream processes, even the escape of a tiny fraction could involve large amounts of radioactivity with serious public health consequences. One 1 GWe reactor generates each year some 1000 nuclear bomb equivalents. If just 0.1% of the radioactivity content would be discharged into the human environment, that would still mean one nuclear bomb equivalent a year for each nuclear power plant. In practice the released fraction might be higher than 0.1%; nearly 400 nuclear power plants are operating globally.

There are many pathways along which radioactive materials from the nuclear energy system can enter the human environment. Broadly three categories of discharges of radioactive materials into the biosphere can be discerned: authorised routine releases, unauthorised releases and large-scale accidents.

# Authorised routine releases

The nominally operating nuclear process chain discharges routinely radioactive substances into the environment, in the front end processes, during the reactor operation as well in the back end processes. A part of these operational releases is hardly avoidable from a technical point of view, but another part is accepted for economic reasons (see report m34 *Conflict of interests, flexibility of regulations*. These discharges are authorised by regulations, based on models, which may be flexible under economic pressure. This chapter briefly addresses the authorised routine discharges from a number of processes of the nuclear chain:

- uranium mining
- other front end processes
- nominal reactor operation
- interim storage of spent fuel
- reprocessing.

# Unplanned discharges

No technical system is perfect. Inevitably radioactive substances will leak out of the technical installations and equipment of the nuclear energy system and disperse into the environment. Leakages can be curtailed by application of high quality standards of the technical systems and a stringent inspection regime, but cannot be eliminated. Due to ageing, corrosion and wear the quality of materials and structures will inevitably decline over time. Consequently the frequency and magnitude of spills and discharges tend to increase over time.

In principle many leaks are preventable but the occurrence of leaks in general is unavoidable and uncontrollable. Apart from the accidental spills from any nuclear facility, which are in fact unavoidable, there are releases from unnoticed and practically 'invisible' sources. A part of these uncontrolled and unauthorised discharges occur occasionally and may go on for long periods before detection or recognition, other occur frequently. This report briefly addresses unplanned releases related to:

- depleted uranium
- reprocessed uranium
- plutonium
- discarded nuclear-related components and materials (orphan sources)
- leaks, 'minor' accidents
- interim storage of spent fuel
- interim storage and shallow burial of other radioactive waste
- reprocessing plants
- decommissioning and dismantling of nuclear facilities.

# Violent nuclear accidents

In addition to the allowed and unplanned radioactive discharges large-scale accidents are possible, involving dispersion of massive amounts of radioactivity within a short time period over vast inhabitated areas, by explosions and related events. Potential sources of Chernobyl-like accidents are:

- nuclear reactors
- interim storage facilities of spent fuel
- reprocessing plants.

The potential sources and mechanisms of large-scale nuclear accidents are adressed in report **m22** *Severe nuclear accidents* and the consequences of the Chernobyl and Fukushima disasters are addressed in reports **m02** *Chernobyl disaster* and **m08** *Fukushima disaster*.

# Insidious disasters

Severe accidents can go on slowly and insidiously, when spent fuel or reprocessing waste gets dispersed during prolonged periods, without meltdown and/or explosions. The amounts of dispersed radioactivity could be massive and the contaminated area could be vast, albeit not of global scale. The affected areas could become inhabitable because of heavy and irreversible contamination of soil, groundwater, rivers, lakes and/or coastal seas.

# **Risk factors**

Accidental and inadvertent releases of radioactivity into the environment, including large-scale accidents, can be caused by various mechanisms, such as:

- ageing, wear, corrosion
- accidents
- natural disasters
- human failings
- illegal trade, criminality
- transport accidents
- terrorism and war actions
- economic pressure.

Reports **m21** *Nuclear safety* and **m38** *Nuclear power and the Second Law* address the factors which follow from the basic laws of nature and greatly increase chances of nuclear accidents, not only the frequency but also the severity of the accidents. Terrorism and war actions will briefly addressed in report **m23** *Nuclear terrorism*. Economic pressure will be addressed in reports **m34** *Conflict of interests, flexibility of regulations* and **m05** *Downplaying and denial of health effects*.

# 2 Dispersion of radionuclides

# Dispersion by air

The physical and chemical properties of the radionuclides determine their mobility when released from spent fuel in case of a meltdown. Nearly all elements of the Periodic System are represented in used nuclear fuel, and each element has his specific chemical and physical properties. A radioactive isotope of an element is often called a radionuclide. The chemical and physical properties of a radioactive isotope and a stable (non-radioactive) isotope of the same element are virtually identical.

The physical and chemical conditions during a severe nuclear accident are extreme: high temperatures, high radiation levels, presence of air, water and construction materials, high pressures resulting from explosions. At these conditions numerous chemical compounds are formed of the released radionuclides by reaction with air, water and many other compounds and elements.

At the high temperatures during the meltdown and explosions, many compounds evaporate and escape in gaseous form. At some distance from the destructed reactor, often high in the air, these gases condense to minute solid or liquid particles, which can be transported over many thousands of kilometers by the wind.

For dispersion by air the volatility of the radionuclides and their compounds is an important parameter. The more volatile, the larger the fraction which can escape from the molten fuel and the longer the distance travelled by air from the desintegrated reactor.

Regarding their volatility [UNSCEAR 2008] Q551 distinguishes four categories of radionuclides:

- inert gases
- volatile elements
- elements with intermediate volatility
- refractory elements.

Refractory elements released from hot desintegrating nuclear fuel form in reactions with air and water compounds with high melting and boiling points, which generally do not evaporate at the temperature of a reactor meltdown. Due to the fierce conditions, often with steam and hydrogen explosions, the refractory radionuclides are dispersed into the air as aerosols, which can be transported over long distances too. This phenomenon may be compared with the transport of volcanic ash over thousands of kilometers. See maps in reports **mo2** *Chernobyl disaster* and **mo8** *Fukushima disaster*.

Among the fission products are inert gases, which escape immediately into the atmosphere after desintegration of the nuclear fuel containment, for example krypton-85 (Kr-85, halflife 10.72 years) and xenon-133.

Examples of volatile elements are different radioisotopes of the fission products tellurium, iodine and cesium. Isotopes of these elements are dispersed in gaseous form in case of fire and/or fuel meltdown - both conditions were present at Chernobyl and Fukushima – for they form at high temperatures gaseous reaction products with other elements, such as oxygen, hydrogen and the halogens, and/or are volatile in elementary form at elevated temperatures.

Examples of radionuclides with intermediate volatility are radioisotopes of the elements strontium, ruthenium and barium.

Examples of refractory radionuclides are radioisotopes of the elements zirconium, molybdenum and cerium and all isotopes of uranium, plutonium and the minor actinides. Refractory radionuclides can also be dispersed over long distances as aerosols, as shown by the dispersion maps after the disasters of Chernobyl anf Fukushima.

With the lapse of time the airborne radionuclides are deposited on the ground surface by rain, except of course the inert gases.

# Dispersion via water

Above categorisation of radionuclides according to their volatility is important only for releases into the air. For releases into water the solubility of the elements and their compounds are the determining property. Often little relationship exists between volatility and solubility in water of an element and its chemical compounds.

Dispersion via water becomes the main route after the temperature of the destroyed reactors and cooling pools has fallen to below the boiling point of water, and when the radionuclides which have been dispersed by air are deposited on the ground by rain.

Via rain runoff, groundwater flows and direct deposition rivers and lakes may become heavily contaminated by radionuclides. Except the inert gases, nearly all radionuclides released by the disaster at Chernobyl will end up in the soil, groundwater, rivers and lakes of the contaminated regions. At Fukushima a large part of the released radionuclides were discharged, and are sill being discharged, into the sea.

A separate categorisation is needed for the mobility of radionuclides in water. The mobility of radionuclides in groundwater is not alone set by their solubility in water, but also by the extent to which dissolved compounds are adsorbed by substances in the soil, such as clay, peat and humus.

The issue of radionuclides in soil and groundwater is further complicated by the question: what happens with the solid particles of the refractory radionuclides released in the accident? These particles contain, among other, actinides (uranium, plutonium and minor actinides) and therefore are highly hazardous. Refractory means: the involved compounds have high melting points. However, refractory is not the same as chemically inert. On the contrary the actinides are chemically quite reactive. So the refractory oxides of these elements will relatively easily dissolve in water by reaction with organic acids in the water. In this way the actinides may become mobile in water, at a high rate if their chemical compunds are relatively soluble, e.g. neptunium, other at a slow rate, if their compounds are less soluble.

# **Dispersion patterns**

One of the fission products, Cs-137, is easily detectable and is released in large amounts in case of a reactor meltdown, and for that reason Cs-137 is generally used as marker of the dispersion of fission products from a wrecked reactor. The dozens of radionuclides released from melted reactor cores and destroyed spent fuel cooling pools during a large accident are very different in their physical and chemical properties, so their dispersion patterns might be different from each other. Besides, most radionuclides are not detectable in airborne surveys and a substantial number of radionuclides is not detectable even at ground level by means of the commonly used radiation detectors, which only detect gamma rays. Nevertheless all those radionuclides are hazardous.

Therefore it may be very well possible that the pattern of the health hazards resulting from contamination with a mixture of radionuclides in a certain region is quite different than the pattern following from the Cs-137dispersion alone. A region with little Cs-137 contamination may be heavily contaminated with other, hardly detectable radionuclides, posing possibly serious health hazards to the unaware inhabitants.

Redispersion of radonuclides by aerosols occurs in contaminated areas as a consequence of wildfires and burning of biomass for cooking and heating.

# 3 Health hazard assessment

# Variables and parameters

In case of accidents with nuclear fuel the release of all kinds of radionuclides present in spent fuel can be expected: fission products, actinides and activation products. Variables determining the severity of a nuclear accident are, among other:

- quantities of released radionuclides
- characteristics of released radionuclides
- size of the contaminated area
- rate of emission, how much time is available for evacuation and countermeasures?
- number of affected people.

Assessment of the health hazards caused by the large-scale dispersion of radionuclides into the environment as a result of spent fuel desintegration is an exceedingly complex matter, involving a number of parameters with regard to the released radionuclides, such as:

- quantities released from the cripled reactor(s) and/or cooling pool(s)
- halflifes
- dispersion mechanisms: fire, explosions, water inflow, etcetera
- volatility
- solubility in water
- chemical properties
- adsorption to substances in soil
- dispersion pattern
- exposure pathways: via air, water and food
- chemical form at moment of exposure
- exposure over time: during the first days or weeks and in the long run
- bioaccumulation in food
- migration into ground water, rivers, lakes and sea
- biomedical activity in the human body
- synergetic effects of different radionuclides in the human body

Some of these parameters are known chemical and physical properties of the various radionuclides, such as halflife, volatility and solubility, other parameters have to be measured, for example the dispersion pattern and the bioaccumulation in food. Some important parameters are poorly understood, for example biomedical activity and synergetic effects.

# Timescale

During the first weeks and months following a major accident involving destruction of a reactor and/or spent fuel pool, the inhabitants of the contamineated areas will be exposed to very high doses of short-lived radionuclides. After 20 halflifes an amount of a given radionuclide will have decayed to one millionth of the original amount, so contamination will be hardly demonstrable anymore. In case of, for example, iodine-131 this condition would be reached 160 days after the release. Evidently exposed people may contract large doses of short-lived radionuclides during the first days following the disaster. The detrimental health effects may become visible not until years after the exposure, long after the radionuclides which caused the health effects have decayed to stable atoms.

Several months after the disaster the radioactivity in a contaminated area are dominated by long-lived

fission products, such as <sup>137</sup>Cs and <sup>90</sup>Sr, and uranium, plutonium and minor actinides. As explained above, exposure to radionuclides then occurs chiefly via food and drinking water. Inhabitants of the contaminated areas will be exposed chronically to these radionuclides. The doses may be considered 'low', however defined, but the health hazards not always proportional to the level of radioactivity. Besides little, if anything, is known about chronic presence of human-made radionuclides inside of the human body.

# Radioactivity and half-life

The radioactivity of spent fuel at the moment of breakup of the core of an operating reactor is chiefly set by the fission products, especially the short-lived radionuclides. During the first few months after the fission process has stopped, the radioactivity of spent fuel decreases nearly a thousandfold, due to the decay of very short-lived fission products, for example xenon-133, tellurium-132 and iodine-131. After the first months the radioactivity decreases slowly and is chiefly set by cesium-137 and strontium-90. After some 300 years the radioactivity of spent fuel is set by the actinides, not to say that the fission products are unimportant after the crossover.

The rapid decay of some radionuclides to stable (non-radioactive) nuclides within the first few months after a reactor meltdown does not necessarily mean that the health hazards decrease proportionately. There are pathways possible by which people can be exposed to high doses of radionuclides even if the average contamination is low, for example via bioaccumulation in foodstuffs.

In addition, as a result of unpredictable criticality conditions uncontrolled fission can occur in the melted nuclear fuel, many times and also long after the initial meltdown occurred. Each criticality incident generates a fresh pulse of short-lived fission products.

# Biased standards

Often the exposure to artificial radiation is compared with the 'background radiation' in the involved region. A slight increase of the radiation from human-made sources above the 'background level' is disposed of in statements such as: 'harmless' or 'not significant'. This view raises some objections:

- Background radiation does not equal 'harmless level of radiation'.
- Due to the cumulating discharges of long-lived radionuclides present in the large areas involved, the background radiation rises with time. Consequently this way of presenting health hazards is strongly biased.
- Official exposure standards are based on a once contracted dose of radiation, and not on chronic exposure during years or even decades.
- Official exposure standards are based on gamma and X-ray radiation from external sources, not on the presence of radionuclides inside the human body, let alone the presence of a mix of alpha and beta emitting radionuclides.
- Due to the large numbers of incidences, the effects become statistically demonstrable.

# Examples: technetium and ruthenium

# Technetium-99

One of the hardly retainable radionuclides in spent fuel is technetium-99, one of the high-yield fission products. In the fission process  $^{99m}$ Tc is formed, which transforms in a matter of hours (half-life 6 hours) into  $^{99}$ Tc, which has a half-life of 0.21 million years and decays to radio-iodine  $^{1291}$  (half-life 16 million years). The most stable chemical form of technetium in water is the pertechnetate ion  $^{99}$ TcO<sub>4</sub><sup>-</sup> ion. Pertechnetate

compounds are highly soluble in water and it is difficult to extract pertechnetate ions from waste liquids. Removal of fission products is generally based on cation reactions, such as <sup>137</sup>Cs<sup>+</sup>, so an additional anionbased separation step is required to remove the negative pertechnetate ion.

The discharges of 99Tc by the European reprocessing plants have been very high in the past, probably the radionuclide was not retained at all. During the past decade the discharges gradually decreased, partially due to lower throughput of spent fuel, partially as a results of better retaining processes [OSPAR 2009]Q579. In the human body the pertechnetate ion accumulates in the thyroid gland as it mimics the iodide ion I<sup>--</sup>. Bioaccumulation of 99Tc occurs in sea organisms, especially seaweed [DECC 2009] Q582.

# Ruthenium-106

Ruthenium-106, <sup>106</sup>Ru, (half-life 1.0 years) is another hardly retainable radionuclide and also a major fission product. Ruthenium is a noble metal and is one of the platinum group metals, together with rhodium, palladium, osmium, iridium and platinum.

Ru-106 is a very hazardous compound because it is chemically toxic and also because of its radiotoxicity [Mun et al. 2004] Q578. All ruthenium compounds should be regarded as highly toxic and as carcinogenic. Compounds of ruthenium stain the skin very strongly. It seems that ingested ruthenium (e.g.a s aerosol) is retained strongly in bones. Ru-106 is one of the radionuclides involved in atmospheric testing of nuclear weapons. It is among the long-lived radionuclides that have produced and will continue to produce increased cancers risk for decades and centuries to come [Lenntech 2013] Q586.

Ruthenium tetraoxide, RuO<sub>4</sub>, is the most likely gaseous chemical form under the conditions prevailing in the containment of a nuclear reactor and during an accident involving spent fuel. The compound is highly toxic, volatile, highly reactive and explosive.

# Environmental effects of ruthenium

Very few data are available on ruthenium impact on plants and estimates of its uptake have deducted levels of 5 ppb or less, although algae appear to concentrate it. No negative environmental effects have been reported [Lenntech 2013] Q586. Obviously this observation does not mean that deleterious effects of radio-ruthenium in the biosphere and in the human body are absent: very likely such effects are not investigated.

# 4 Radionuclides in the food chain

Radionuclides dissolved in water unavoidably enter the food chain and drinking water. As long as the radionuclides persist in the soil and groundwater, may be for centuries, people will be chronically exposed to a mix of radionuclides.

The part of the report [UNSCEAR 2008] Q551 addressing the Chernobyl disaster mentions briefly what is called 'the main pathways of radionuclides in the terrestrial environment'. Other than statements that the radioactive content of the surface and ground waters have decreased in the years after 1986, discussions of possible health effects attributable to these pathways are missing in the report.

# Bioaccumulation

Nearly all released radionuclides will end up in water, soil and sea and will enter the food chain and drinking water, depending on the chemical behaviour of the various radionuclides. Some organisms tend to accumulate some elements. Sea weed for example accumulates iodine in its cells to relatively high concentrations, and consequently also radio-iodine. In view of its long halflife of 16 million years, contamination with I-129 may become a chronic problem in contaminated coastal seas, such as in Japan near Fukushima. This phenomenon is also present in the coastal seas near Sellafield (UK) and La Hague (France), where the reprocessing plants at those locations discharge large amounts of <sup>129</sup>I into the sea, originating from the reprocessed spent fuel.

Cesium-137 is accumulated in, among other, mushrooms, berries and game. In Scotland there are sheep farms which are still contaminated by Cs-137 from Chernobyl at higher than allowed levels.



#### Figure 1

Difference in width and color of tree rings in pine logs from Chernobyl. The year of the accident in 1986 is clearly visible from the change in color of the wood. This is Figure 3 from [Mousseau et al. 2011] Q615.

# Observable effects in the environment

The consequences of the radioactivity for the plant and animal life in the contaminated regions have been and are still being investigated by a number of scientists in the USC Chernobyl + Fukushima Research Initiative at the University of South Carolina. See [Mousseau 2014] Q610 for a list of the more than 60 completed studies and results and the goals for 2014-15.

One of the findings are the adverse effects on the growth of pine trees in the contaminated areas, see the striking photograph in Figure 1.

One may wonder what are the long-term effects of radioactive contamination for humans, in view of the pronounced effects for trees? It seems extremely improbable that chronic exposure to many different radionuclides, even at 'low' levels, would not have health effects.

# Tritium and carbon-14

Special examples of volatile radionuclides are tritium and carbon-14. Tritium, formed in large quantities in the nuclear fuel and in the cooling water, is chiefly released as radioactive water (tritriated water HTO). All cooling water from nuclear power plants s released into the environment.

Carbon-14 is mainly formed in the cooling water of a nuclear reactor and is released as radioactive carbon dioxide ( ${}^{14}CO_2$ ), methane ( ${}^{14}CH_4$ ) and some higher alkanes. In the atmosphere the alkanes are oxidized to carbon dioxide.

Both tritium and carbon-14 are routinely released into the human environment by nominally operating nuclear power plants. In reprocessing plants, in Europe La Hague in France and Sellafield in UK, all tririum and carbon-14 generated in the nuclear fuel are released into the air. 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*.

Carbon dioxide dissolves in rainwater and so a part of the released carbon-14 ends up in groundwater and enters the food chain.

Tritium and carbon-14 are biochemically indistinguishable from their non-radioactive isotopes, normal hydrogen H, respectively normal carbon (mainly C-12). 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, including DNA, when ingested (via food and drinking water) or inhaled. Despite the relatively low energy of the radioactive decay of tritium and carbon-14, their biological impact might be more drastical than recognized in the classic radiological models. For more details see report **m42** *Tritium, carbon-14 and krypton-85*.

# Tritium

Tritium (symbols T,  $^{3}$ H or H-3) 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.



### Figure 2

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.* 

Tritium can also enter the body via food as organic bound tritium (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 adjecent 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 bèta radiation with chemical bonds, transmutation and recoil effects) in living organisms have been thouroughly 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.

# Incorporation of tritium in DNA

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

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.

# Incorporation of carbon-14 in DNA

Male sperm cells are produced from spermatogonia continuously in the adult male and consequently the <sup>14</sup>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 <sup>14</sup>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 <sup>14</sup>C/<sup>12</sup>C ratio. Calculation of the genetic effect to the current generation of an added increment of <sup>14</sup>C to the atmosphere using a lag time of 1-2 years, therefore, is conservative according to [NCRP-81 1993] Q256.

# Uncertainties

The effects of a large nuclear accident are exceedingly serious, the more so because radioactive contamination is practically irreversible. The exposure of the residents in the affected areas to radioactive contamination is not limited to the period of the disaster, when the radionuclides are being dispersed into the environment. The gaseous radionuclides released during the accident rapidly disperse into the atmosphere and so are diluted to low, but still measurable concentrations around the globe, as is illustrated by the dispersion maps in report **mo2** *Chernobyl disaster*. In this way the background radiation and exposure to radioactivity increases, especially in the Northern Hemisphere.

Within a relatively short period the bulk of the short-lived radionuclides, for example I-131 have decayed into stable nuclides. For above reasons it may be impossible to assess the doses radionuclides people are exposed to during the days and weeks immediately following the disaster. During this 'short' period heavy radioactive contamination could have happened, causing serious health damage which manifests itself after long incubation periods.

The non-volatile radionuclides are deposited on the ground and a number of them will enter the food chain. Via food and water the contamination with long-lived radionuclides will last nearly forever. The health effects of chronic exposure to low concentrations of a mix of radionuclides is not systematically investigated and not included in the exposure models used by the nuclear world. Statements that low concentration do not harm, or even would be beneficial (hormesis), are not based on empirical data. On the contrary, results from many studies, among other the KiKK and Geocap studies, prove that concentrations far below the official safe threshold values are harmful and can cause cancer deaths and other diseases.

### References

#### 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, Bethesda, MD, 15 May 1985, first reprinting April 30 1993.

#### 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.

#### Q260

NCRP-89 1987 Genetic effects from internally deposited radionuclides, NCRP Report 89, National Council on Radiation Protection and Measurements, Bethesda, MD, December 1, 1975.

#### Q373

Fairlie 2007 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/

### 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 Reaktorsicherkeit und des Bundesamtes für Strahlenschutz, Germany, 2007, 4334\_KiKK\_Gesamt\_T.pdf 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.

#### 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.

# Q551

UNSCEAR 2008 UNSCEAR 2008 Report. Sources and effects of ionizing radiation. ANNEX D. Health effects due to radiation from the Chernobyl accident, United Nations, April 2011. <11-80076\_Report\_2008\_Annex\_D.pdf> www.unscear.org/docs/reports/11-80076\_Report\_2008\_ Annex\_D.pdf retrieved 11 Sept 2013

#### Q578

Mun et al 2004 Mun C, Cantrel L & Madic C,

A Literature Review on Ruthenium Behaviour in Nuclear Power Plant Severe Accidents,

Institut de Radioprotection et de Sûreté Nucléaire (IRSN) paper not dated, probably 2003-2004 (youngest reference of 2003)

<A\_Literature\_Review\_on\_Ruthenium\_Behaviour\_in\_Nuclear\_ Power\_Plant\_Severe\_Accidents\_.pdf> http://hai-isrn.archives-ouvertes.fr/docs/oo/17/76/21/PDF/ retrieved 19 November 2013

#### Q579 OSPAR 2001

Liquid discharges from nuclear installations in 1999, OSPAR Commission, 2013, ISBN 0 946956 63 4 vpoo130\_text\_99.doc> www.ospar.org/v\_publications/ retrieved 21 November 2013

#### Q582 DECC 2009

UK Strategy for Radioactive Discharges, Department of Energy & Climate Change, July 2009, Crown copyright 2009. (0084424.pdf) http://www.scotland.gov.uk/Resource/Doc/280203/ http://decc.gov.uk/en/content/cms/what\_we\_do/uk\_supply/ energy\_mix/nuclear/issues/radioactivity/radioactivity.aspx

Q586 Lenntech 2013 Ruthenium – Ru <Ruthenium (Ru) – Chemical properties, Health and Evironmental effects.pdf> www.lenntech.com/periodic/elements/ru.htm retrieved 22 November 2013

Q610 Mousseau 2014 Mousseau T A, The USC Chernobyl + Fukushima Research Initiative, University of South Carolina <Mousseau\_-\_IPPNW\_Press\_Release\_-\_Chernobyl\_ Fukushima\_Feb\_2014.pdf> www.tschernobylkongres.de/fileadmin/user\_upload/ Arnoldshain\_Doku/

Q615 Mousseau et al. 2013 Mousseau TA, Welch SM, Chizhevsky I, Bondarenko O, Milinevsky G, Tedeschi DJ, Bonidoli-Alquati A & Møller AP, Tree rings reveal extent of exposure to ionizing radiation in Scots pine Pinus Sylvestris, DOI 10.1007/s00468-013-0891-Z published online 11 June 2013 ‹Mousseau-et-al-TREES-2013.pdf› www.academia.edu/3803036/