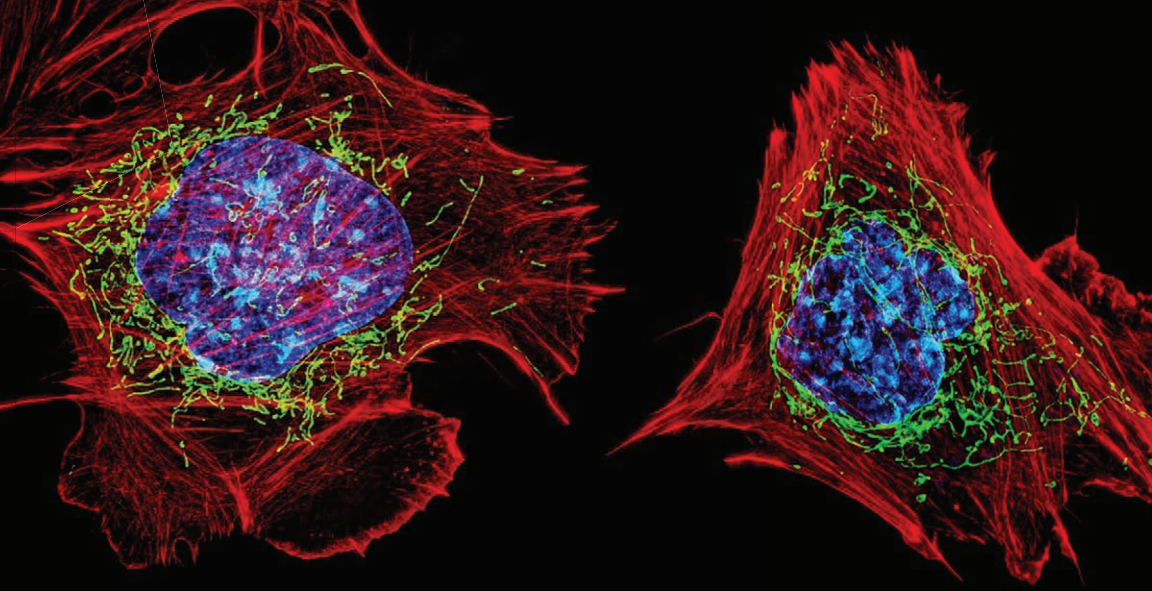
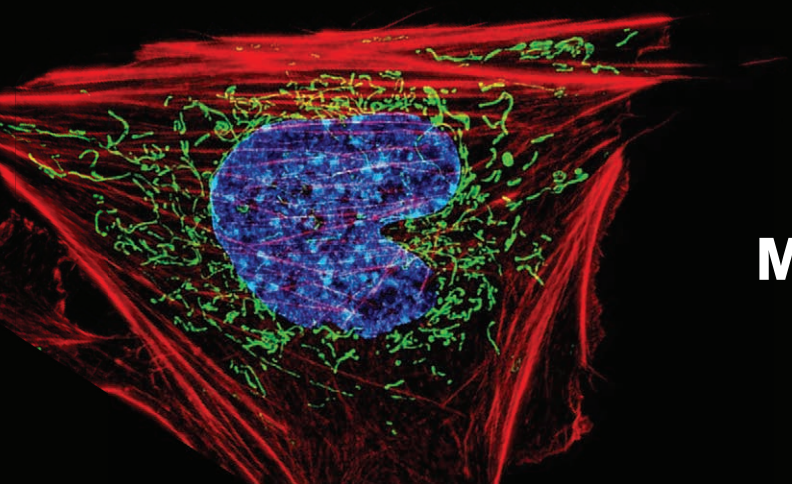


# Exploring Tritium Dangers

## Health and Ecosystem Risks of Internally Incorporated Radionuclides



**Arjun Makhijani, Ph.D.**

"I find this book extremely important and timely, especially considering the present plans for the large-scale release of radioactive, tritium-contaminated water from the damaged Fukushima-Daiichi Nuclear Power Plant. Efforts to trivialize the potential effects of tritium in the environment ignore organic-binding, the uptake, trophic transfer and bio-accumulation by marine organisms, and effects on the cells within individuals who eat contaminated seafood products, including on nuclear DNA, mitochondrial DNA, RNA and signaling proteins. Ignoring such effects threatens the health of ecosystems already affected by a variety of anthropogenic stressors and all who depend on them."

**Dr. Robert Richmond**
*Research Professor and Director,*
*Kewalo Marine Laboratory, University of Hawaii at Manoa*

"Rigorously independent and critical analyses in the field of ionising radiation and health, that draw upon and update evidence-based conclusions, like this one, are vital. More tritium is emitted by nuclear power plants and nuclear weapons production than any other radioactive pollutant.

For decades, it has been claimed that tritium is a minor hazard, dispersing widely in water, that its beta radiation is not penetrating and relatively weak. With harder to assess exposures and risks, such internal emitters have often not been adequately addressed by radiation protection standards. Similarly, tritium's different forms, distributed throughout organisms and affecting diverse cellular components, have been sidelined. This is particularly important to redress given our collective greatest responsibility is to protect the most vulnerable - pregnant women and young children, and the growing evidence that there is no threshold for risk for most if not all the long-term effects of radiation exposure."

**Tilman Ruff, MB, BS (Hons.)**
*Co-President, International Physicians for the Prevention of Nuclear War*

"This book is a must-read for anyone concerned about the impact of tritium on the environment. Written in an easy-to-read style, it challenges conventional wisdom and provides a comprehensive overview of the large variety of effects of tritium beyond the commonly understood link to cancer. Highly

recommended for anyone looking to understand the need for higher standards in radiation protection."

**Dr. Ferenc Dalnoki-Veress**
*Scientist-in-Residence and Adjunct Professor,*
*James Martin Center for Nonproliferation Studies,*
*Middlebury Institute of International Studies at Monterey*

# Exploring Tritium Dangers

Health and Ecosystem Risks of
Internally Incorporated Radionuclides

Arjun Makhijani, Ph.D.

Title: Exploring Tritium Dangers: Health and Ecosystem Risks of Internally Incorporated Radionuclides

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Cover image: "The cells shown here are fibroblasts, one of the most common cells in mammalian connective tissue. These particular cells were taken from a mouse. DNA within the nucleus (blue), mitochondria (green), and cellular skeleton (red) are clearly visible. This image was featured in an exhibit called Life: Magnified from June 3, 2014, to January 21, 2015, at Washington Dulles International Airport's Gateway Gallery."

Cover image credit: D. Burnette, J. Lippincott-Schwartz/NICHD downloaded from Wikimedia Commons at https://upload.wikimedia.org/wikipedia/commons/5/56/Cells_with_nuclei_in_blue%2C_mitochondria_in_green%2C_and_the_actin_cytoskeleton_in_red_%28119124186316%29.jpg



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*Dedicated to*

Kay Drey

*whose "favorite" radioisotope is tritium*
*and who plays "happy birthday" on the violin*

# CONTENTS

# Preface

The roots of this work are complex, but the most important by far is when Kay Drey and I found that we had independently arrived at the same conclusion: that tritium was a radionuclide that should be taken very seriously. However, in practice it had been treated rather cavalierly by the radiation establishment as a radiation danger even though it is radioactive water, makes food radioactive, crosses the placenta, and is a ubiquitous pollutant from both nuclear power and nuclear weapons. Kay has most generously funded this work.

Some of its technical roots go back to the early 1980s when Bob Alvarez introduced me to Anthony Guarisco. A veteran of the 1946 Bikini atom bomb tests, he had a pile of documents from the archive of Stafford Warren, who was Dean of the medical school of the University of California at Los Angeles when he died in 1982. Dr. Warren had been the chief of radiological safety at Bikini in 1946 during Operation Crossroads, the first nuclear bomb tests after World War II. Atomic veterans had been told by the Defense Nuclear Agency that their internal doses were negligibly small. The documents indicated otherwise. Radiological conditions were poor. Safety personnel complained about the "hairy-chested" approach of some naval officers who had a "disdain for the unseen hazard" of radiation.[1] The report that a colleague and I did was presented to the House Committee on Veterans Affairs by none other than Karl Morgan, one of the founders of the discipline of "health physics." It was my first major indication that internal doses were not being considered with the rigor they deserved. By "internal dose" is meant the energy from ionizing radiation

[1] As quoted in Makhijani and Albright 1983, p. 2.

deposited in the body or some part of it by radionuclides once they are inside the body.

A major joint project of the International Physicians for the Prevention of Nuclear War and the Institute for Energy and Environmental Research (IEER) in the 1990s concluded that every nuclear weapon state had first of all harmed its own people without informed consent. More evidence mounted of significant differences between external and internal radiation that were clearly not being taken into account given that the main focus was cancer risk for adults – a major problem, but not the only one.

In 1999, my then-colleague at IEER, Lisa Ledwidge, and I initiated a letter to the National Academies signed by over 100 scientists, academics, activists, physicians, and community leaders on the topic. We specifically urged the committee studying the health risks of "low-level" radiation to consider, among other things, the problem of radionuclides that can cross the placenta. While the committee updated cancer risks to men, women, and children, it did not seriously consider the question of radionuclides that cross the placenta. The issue of radioactive water and radioactive food that affects the embryo and fetus remains woefully and shockingly sidelined, as I wrote to yet another committee of the National Academies on the same topic, in January 2022.[2]

In the mid-2000's I learned that Argonne National Laboratory's RESRAD program to assess the risks of residual radioactivity contamination after remediation of polluted sites only calculated doses for "Reference Man" – officially defined as a 20 to 30-year-old "Caucasian male."[3] That made me determined to look into the problem of how risks were being calculated more deeply. That was also the point at which I realized that the "physics" in "health physics" was that the

[2] Makhijani 2022
[3] Makhijani 2009

body was simplified and represented as a bag of water – since we are mostly water. Models are simplified representations. Still, the body is highly organized system; that does not enter centrally into the model, even when modified by a concept called "relative biological effectiveness." I also realized that the approach to assessing chemical risks was quite different so that it was not possible in any systematic way to assess the combined risks of radiation and chemicals, though tens of thousands of toxic chemicals are in common use. In 2005, "the Environmental Working Group (EWG) in collaboration with Commonweal, researchers at two major laboratories found an average of 200 industrial chemicals and pollutants in umbilical cord blood from 10 babies…."[4]

The inability to systematically consider the combined effects of chemicals, not to speak of chemicals and radiation together, means that environmental protection is oriented not to protecting health but to controlling pollution. Limiting pollution from any particular toxicant is important, even critical. It is central to the regulation of industries. Yet, we experience pollutants in the environment together from the moment of conception – and indeed before, when ova and sperm are formed but still exist separately. I sometimes ask people (generally someone young in an audience) how old they are. And they tell their age, calculated, of course, from their date of birth. Yet, the ova from which we are made were created when our mothers were in their mothers' wombs, so a part of us is as old as our mothers.

These are vast problems and it has taken some time to sort out what a small non-profit like IEER, dedicated to the goal of bringing "scientific excellence to public policy issues in order to promote the democratization of science and a safer, healthier environment"

[4] EWG 2005.

should do. I decided we should focus on tritium. A ubiquitous pollutant, it exemplifies the types of risks that many other critical pollutants inflict on people and other living beings. For instance, much of the harm tritium does comes from the excess reactive oxygen species it creates in cells by ionizing water. This is also the source of much of the risk from heavy metals. Thus, understanding tritium also points the way to better understanding not only of other pollutants, but of how one might combine the risk estimates to better set environmental standards and protect public health. It also points to the complexity of the problem, how much we still need to learn, and the need for protective standards, given these gaps.

Parts of this monograph are taken from or adapted from my memorandum on low-level radiation research sent in January 2022 to a committee of the National Research Council.[5]

In order to illustrate that routine tritium pollution does actually rain down on people and ecosystems, IEER commissioned Matthias Rau, an atmospheric modeling expert in Germany, to model one year of routine tritiated water vapor emissions from the Braidwood nuclear power plant in Illinois. Braidwood achieved some notoriety after the discovery of tritium leaks and the fact that those leaks had migrated offsite and contaminated private water wells. His entire modeling results are in Appendix B.

I would like to thank Annie Makhijani, who is a Project Scientist at IEER; she did much of the research for this book, digging up documents in English and French and then proofreading this manuscript.

This book is dedicated to Kay Drey on the cover – an unusual way to acknowledge someone. But it is for a very good reason. She long ago concluded that tritium was a critical radionuclide to evaluate

[5] Makhijani 2022

for its health and ecological impacts; like the dangers at Bikini Atoll in 1946, it also has often been treated with disdain. This book shows that she has been prescient. Kay was also very generous and funded this entire research project. I am grateful to her on my own behalf and on behalf of everyone who might benefit from the analysis and exploration in this book. It is called "Exploring Tritium Dangers" because it has literally been a voyage of exploration into a variety of areas of radiation risk, especially some that have been relatively neglected.

First of all, there is tritium itself. Secondly, the risks during pregnancy and especially during the first trimester have, shockingly, received far less attention than they deserve, given the ubiquity of tritium as an anthropogenic pollutant from both nuclear weapons and nuclear power. And then there is its impact on the organelles in the cytoplasm of cells, once it gets into the body as radioactive water or bound up in the molecules of the food we eat. Specifically, tritium emits beta particles that create excess reactive oxygen species in cells, damaging mitochondria and mitochondrial DNA, which form the core of the energy system of all multi-cellular animals, plants, and fungi and also many single-celled ones that have nuclei. Oxidation and reduction – and hence the presence of reactive oxygen species – are a normal part of metabolism. For one thing, it is the way food is processed into energy. Reactive oxygen species are normally present. Here we are concerned with *excess* reactive oxygen species. By way of analogy, we need oxygen for our normal metabolic processes. But ozone pollution, on top of the normal oxidative processes, creates the damage. That's why we call it "pollution."

The word "exploring" in the title is also used because there is still so much to be learned beyond cancer and the conventional dose and risk assessments done in that context; they are of course critical and important in their own right. But this book is focused on other,

mainly non-cancer risks – the ones that need considerable exploration both from a scientific point of view and for protecting health and the environment.

*Arjun Makhijani*
*July 2022*

# Exploring Tritium Dangers

Health and Ecosystem Risks Of
Internally Incorporated Radionuclides

# 1

# Why Tritium?

Tritium is a radioactive isotope of hydrogen, which is the most abundant element by far in the universe – being nearly three-fourths of its observable mass. It is the simplest element, consisting of a single proton in the nucleus and an electron in orbit around it. Almost all the hydrogen on Earth (99.985 percent) is of this form. Essentially all the rest, 0.015 percent) is deuterium, which has, in addition to the proton, a neutron in the nucleus. Like ordinary hydrogen, it is not radioactive. Tritium is the other isotope of hydrogen, present naturally only in very minute amounts, being created by the interaction of cosmic rays with the atmosphere. The natural total inventory of tritium is less than 10 kilograms;[6] for comparison, the mass of the atmosphere is more than five million trillion kilograms.

So why worry about tritium? First, it is highly radioactive. The radiation it emits – a beta particle – poses dangers, like other radioactive materials, to people and other living beings. Tritium is ordinarily a gas, like hydrogen, or it exists in water, have replaced one or both atoms or $H_2O$, denoted respectively as HTO or $T_2O$. In either of these forms it is called "tritiated water," which is of course, radioactive. Tritium is unique in this – it makes water, the stuff of life, most of the mass of living beings, radioactive. And hence it makes food radioactive. On close examination then, there are a host of reasons to pay careful attention to the dangers of tritium:

[6] ANL 2007, pdf p. 56.

1. Tritium is the most common radioactive pollutant routinely discharged in the largest quantities as measured by radioactivity. For instance, the total radioactivity of tritium in the discharges of polluted water from the Braidwood nuclear power plant in Illinois in 2019 was more than *75,000 times the discharges of all other fission and activation products*. The total radioactivity of routine gaseous releases of tritium in the form of water vapor from the Braidwood nuclear power plant in Illinois in 2019 were more than one hundred times that of all other gaseous fission products and activated radioactive gases.[7]
2. Like plutonium, man-made quantities of tritium far exceed those in nature.
3. Tritium and plutonium are both used in nuclear weapons.
4. Like plutonium, uranium, and other alpha emitters, tritium is far more dangerous inside the body than outside the body.
5. Tritiated water is radioactive, but it is chemically indistinguishable from ordinary (non-radioactive) water, which is the majority of the mass of animals and plants. Our cells are mostly water; as a result tritiated water once ingested pervades in our bodies – and does the same for plants and animals.
6. Tritium has a long enough half-life, 12.3 years, that it persists in the environment for decades (in diminishing amounts as it decays); yet its half-life is short enough that it is extremely radioactive.[8] For a given mass, it is,

[7] Braidwood 2019, Table 2A, pdf p. 26 and Table 1A, pdf p. 20. Braidwood is used as a specific example throughout this monograph.

[8] The radioactivity per unit mass of a radioactive material is inversely proportional to its half-life – the shorter the half-life, the more radioactive particles a given mass of the material will emit. Materials with very short half-lives, like iodine-131 (8 days), are intensely radioactive and dangerous, but they do not

for instance, about 150,000 times as radioactive, in terms of disintegrations per unit time, as plutonium-239. *One teaspoon of tritiated water (as HTO) would contaminate about 100 billion gallons of water to the U.S. drinking water limit; that is enough to supply about 1 million homes with water for a year.*

7. Tritium crosses the placenta with facility. The concentration ratio of tritium in the fetus to that in the mother is more than one, whether the mother has had an intake before she became pregnant or after that.[9]
8. The core of the energy system of all multicellular beings consists of mitochondria, which are in the cytoplasm. Tritium ionizes water in the cytoplasm, setting in motion processes that can profoundly disrupt mitochondrial DNA and hence the system that converts food to usable a form, ATP, that the body uses for all functions.
9. Tritium can, by affecting ova during the time of their formation in utero and during the time of the maturation during pregnancy, impact future generations. It can also do so via being incorporated in sperm and spermatocytes.
10. Tritium can exemplify the ways in which other internal emitters can have non-cancer impacts, including during the early period of pregnancy, when internal radiation can result in miscarriages and malformations. At least

persist for long. Iodine-131 essentially decays completely to xenon-131 in about three months. Tritium with a half-life of 12.3 years is not as radioactive, but it is still highly radioactive and takes about 130 years to decay by the same amount as iodine-131 does in three months. Specific activity is also inversely proportional to atomic weight. Tritium being the lightest radioactive material is also more radioactive per unit mass on that account.

[9] NRPB 2001, Table 1, p. 13. This is also true of a few other radionuclides, notably carbon-14. By contrast, the fetal to maternal ratio for plutonium is less than one except for intakes in the last trimester.

some of these impacts can occur even at low doses, notably some types of damage to the central nervous system formation.[10]

11. The focus on adults, especially Reference Man, has resulted in an under-appreciation of the dangers and risks of tritium precisely because tritiated water is chemically the same as ordinary water. The official advice upon exposure has been to reduce the impact by drinking beer or other beverages: "Although the average biological half-life [of tritiated water] is 10 days, it can be decreased by simply increasing fluid intake, especially diuretic liquids such as coffee, tea, beer, and wine."[11] Not wrong advice, of course; yet, it is noteworthy that there are no cautions for women who are pregnant. In fact the words "pregnancy," "embryo," and "fetus," do not appear in this official handbook for "safe handling" of tritium.
12. Routine tritium pollution affects the drinking water of large numbers of people though it is generally below the U.S. drinking water standard limit. But that limit was set without detailed assessment of what it would take to protect the embryo or the fetus especially in the first trimester.
13. Nuclear power plants and many nuclear weapons relative activities are mostly located in relatively rural areas, where people have private wells. Drinking water regulations that limit concentrations of pollutants only apply to public water systems. This is reasonable from a certain viewpoint. Regulating private wells would im-

[10] ICRP 49, 1986, pp. 20-21 and p. 31
[11] DOE 1994, p. 19.

pose significant costs on rural families in terms of testing for and reporting on a host of chemical, biological, and radioactive pollutants. Yet, it has also created a huge loophole. There is no requirement for industries like power plants or chemical plants to offer funds for monitoring or even alert the public to the potential of contamination, unless some accident or leak brings it to the fore, when it is already too late for many.

14. Tritium is illustrative of a whole host of situations in terms of scientific assessment gaps, health and ecosystem impacts, social impacts, and regulatory issues. Thus, studying tritium allows one to draw lessons for other pollutants, including several other radionuclides, in all these aspects and thus also for the protection of health and the environment.

This last reason has significant implications for the protection of health and the environment. Understanding tritium can potentially enable an understanding of synergistic interactions with non-radioactive pollutants. While the goal of actually addressing synergistic effects is too large for this monograph, the exploration in it points to directions for one way to systematically approach this difficult topic: by examining those pollutants that act by creating excess reactive oxygen species inside living cells.

# 2

# Physical and Radiological Features of Tritium

## a. Physical aspects

Tritium is a radioactive form of hydrogen with one proton and two neutrons in its nucleus, resulting in a total atomic mass of three. Figure II-1 shows the three isotopes of hydrogen.

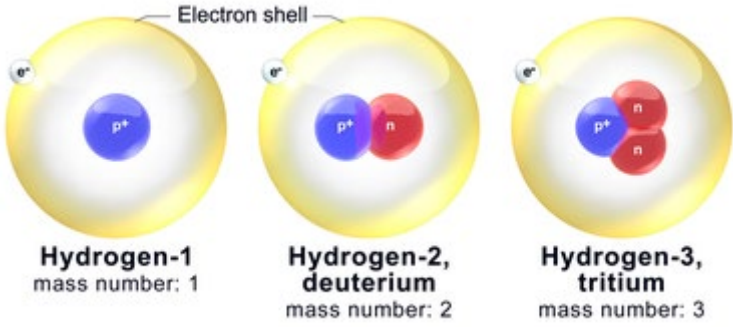



Figure II-1: Schematic of the three isotopes of hydrogen. Ordinary hydrogen H-1 or simply H, deuterium, H-2 or D, and tritium H-3 or T. Only tritium is radioactive Source: Bruce Blass, Wikimedia Commons, at https://commons.wikimedia.org/w/index.php?search=tritium+atom&title=Special:MediaSearch&go=Go&type=image

As a radioactive gas, tritium is essentially like hydrogen but with about three times the atomic mass. It is usually denoted by the symbol T. When its likeness to hydrogen is to be stressed, it is designated by the symbol H-3 – "H" for hydrogen, and "3" for its atomic mass (in round numbers)[12], also written

[12] The total number or protons and neutrons in the nucleus is called the "mass number" and is very close to the actual mass of a particular isotope.

as $^3H$. A nucleus of ordinary hydrogen consists of just one proton; it has no neutrons. While the mass number of T is 3 compared to 1 for hydrogen, tritium is still very light. As HT its molecular mass is just 4, compared to 32 for oxygen as $O_2$, for instance. As a small, light molecule tritium gas (HT or $T_2$), like $H_2$, diffuses readily through all but the most highly engineered containment vessels and mixes freely with the other forms of hydrogen.

Tritium is radioactive – that is it has an unstable nucleus that decays by emitting a beta particle. The emission of a beta particle transforms tritium into a new (and stable) element helium-3, which is a non-radioactive gas:

H-3 → He-3 + beta particle (an electron) + antineutrino

The time it takes for half of a given amount of tritium to decay into helium-3 is about 12.3 years. At the end of that period, half of the original tritium is still left. The other half has become helium-3 gas, which is inert. The harm that tritium does comes from the beta particles if and when they interact with living matter.

On average, these beta particles have an energy of 5,700 electron-volts[13] (5.7 kilo-electron-volts, keV for short). A typical chemical bond in a biological molecule is on the order of a few electron-volts. This means that a single beta particle can, on average, create on the order of a thousand ionizations. Ionization of water is especially important in the context of tritium entering the cytoplasm of cells. The ionization energy of water is about 12.6 electron-volts.[14] At that rate, the average beta particle energy of 5.7 keV would produce about 450 ionizations of water. The maximum beta particle energy, 18.6 keV, would result in over 1,470 ionizations of water molecules. Of course, the ionizations from a decay would not be confined to a single type of molecule (though water is the most abundant molecule the cytoplasm).

[13] ANL 2007, pdf p. 56.
[14] NIST 2001

The radioactivity of a substance is usually measured in the number of disintegrations per second or "becquerels" – so named after the scientist Henri Becquerel (abbreviated as "Bq"). The radioactivity per unit mass is called "specific activity." One gram of tritium undergoes about 360 trillion disintegrations per second, in other words its specific activity is 360 trillion Bq per gram or 360 TBq/g). Specific activity is also measured in a unit that honors Marie Curie – one curie corresponds to 37 billion disintegrations (Bq) per second, corresponding to the radioactivity of a gram of radium-226, which she discovered. In this measure, tritium has a specific activity of about 9,800 curies per gram. A curie is a very large amount of radioactivity. The amount of contamination of something is usually measured in trillionths of a curie, known as a "picocurie," billionths of a curie, known as a "nanocurie," or a millionth of a curie, called a microcurie.

Since it is chemically identical to hydrogen it combines with oxygen and forms water, known as "tritiated water," by replacing one or both atoms of non-radioactive hydrogen in water. Tritiated water is generally designated as HTO or $T_2O$, depending on whether it has one or two atoms of tritium in the water molecule respectively. Since tritium is radioactive, so is tritiated water. Since tritium is essentially chemically identical to hydrogen, tritiated water behaves and moves like water, interacting with living beings as ordinary non-radioactive water would but posing risks of harm to health due to its radioactivity. There are slight differences between the behavior of tritiated water and ordinary water since tritiated water has a higher molecular mass. But tritiated water moves in the environment like non-radioactive water.

## b. Tritium sources other than reactors

Tritium is produced naturally and artificially.

In nature, cosmic rays interact with the atmosphere to produce neutrons. Neutrons bombarding a nitrogen nucleus result in an ordinary carbon nucleus (C-12) and tritium (H-3):

N-14 + neutron → C-12 + H-3

Tritium is made naturally at a relatively steady rate; it also decaying at a steady rate. As a result there is an equilibrium amount of natural tritium – the balance between production and decay – in the environment. The French government's radiation research and safety institute, IRSN, estimates the annual production between 0.15 and 0.2 kilograms, which implies an equilibrium amount of about 3 kilograms.[15]

Most tritium in the environment is man-made, however. Atmospheric nuclear explosions (including the bombings of Hiroshima and Nagasaki) from 1945 to 1963, created an estimated 780 kilograms of tritium, of which 650 kilograms was in the Northern hemisphere and the rest in the Southern hemisphere.[16] The 1963 Partial Test Ban Treaty, which prohibited all nuclear tests except those underground, was ratified by the United States, the Soviet Union, and Britain. However, tritium continued to be created by the atmospheric tests of France, which ended in 1974, and of China, which ended in 1980; French testing in Polynesia would have created a proportionately greater impact in the Southern hemisphere per unit of explosive power.[17] About 20 kilograms – still much greater than the natural inventory remained in the environment in 2020, with 90 percent in the oceans, one percent in the atmosphere and the rest in water on the various continents.[18] Of course, the continuation of nuclear testing underground has left a legacy of subterranean radioactive pollution whose impacts are yet ill-understood. The Comprehensive Test Ban

[15] IRSN 2010, p. 4. Eisenbud 1987 gives an inventory of 2.65 kilograms (as cited in Zerriffi 1996, p. 3); this is close to the IRSN number, which is used in this book. Argonne National Laboratory gives a larger number for the equilibrium amount – 7.3 kilograms without citing a source or an estimated annual production by natural sources. ANL 2007, pdf p. 56.
[16] IRSN 2010, p. 4
[17] IRSN 2010, p. 4. Underground testing by the signatories of the Nuclear Non-Proliferation Treaty continued till 1996, after which India, Pakistan, and North Korea conducted underground tests.
[18] IRSN 2010, p. 4 gives an inventory of 40 kilograms for the year 2007.

Treaty, which bans all nuclear explosions, has not yet been ratified by enough countries, including some nuclear weapon states, to come into force.[19]

According to an Argonne National Laboratory compendium of fact sheets, published in 2005, the United States had made a cumulative 225 kilograms of tritium – about 70 times the entire natural inventory, for its weapons program; 75 kilograms remained, though the date is not provided.[20]

The inventory cited by Argonne appears to be from public sources dating back to around 1990.[21] U.S. tritium production was primarily for its nuclear weapons program. Tritium for weapons was also incidentally produced in reactors in the nuclear weapons complex, but these were shut by the early 1990s, though much of the tritium remains in the environment. Military tritium production has been done at the civilian Watts Bar 1 reactor in Tennessee since 2003. Based on the annual decay rate of a 225 kilogram tritium inventory, the replacement amount would be almost 13 kilograms a year; based on a 75 kilogram end-of-the-Cold-War inventory, the annual replacement amount would be about four kilograms. Other nuclear weapon states have also made tritium for weapons, of course. A global inventory on the order of 200 kilograms is a workable hypothesis of the purpose of this book.

The main tritium inventories other than from nuclear power plants and nuclear weapons related reactors can be summarized as follows:

- The equilibrium natural inventory is about 3 kilograms, that is roughly 30 million curies.

[19] In 2022, IEER prepared a series of short papers on the health and environmental impacts of nuclear weapons testing for the International Campaign to Abolish Nuclear Weapons. Some were authored by Arjun Makhijani; others were authored by Dr. Tilman Ruff, Co-President of International Physicians for the Prevention of Nuclear War. They can be downloaded at https://ieer.org/resource/disarmamentpeace/articles-on-the-health-and-environmental-impacts-of-nuclear-weapons-testing-at-the-major-test-sites/

[20] ANL 2007, pdf p. 56.

[21] Zerriffi 1996, p. 3 cites the same numbers with references to sources dating to 1987, 1992, and 1993. m

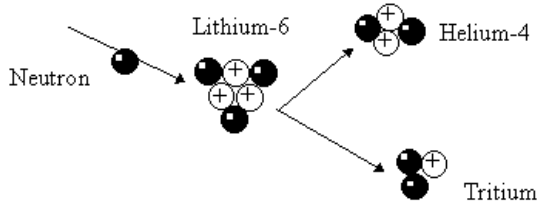
- The tritium remaining from atmospheric testing is about 20 kilograms – or roughly 200 million curies, 90 percent of which is in the oceans, one percent is in the atmosphere, and the rest is in continental water bodies (surface and groundwater). There is also a significant inventory from underground testing which has not been estimated here.
- It is difficult to estimate the inventory of tritium in the nuclear weapons establishments of various states, but based on publicly available information from the 1990s, the inventory is likely to be much larger than the natural and weapons testing amounts combined. The exact amount is not material to the purposes of this paper, except that some of this tritium may leak into the environment.

### c. Tritium production in reactors

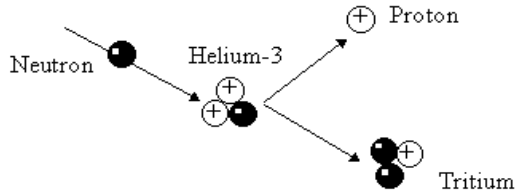
Tritium can be made in nuclear reactors or in accelerators. In a reactor, the basic technique of tritium production usually involves putting lithium-6 in a target rod and bombarding it with neutrons. In the United States, tritium for military purposes has been produced in a civilian reactor belonging to the Tennessee Valley Authority since 2003. Accelerator production of tritium, a new use of accelerator technology unproved on a large scale, would bombard helium-3 with neutrons. The two basic nuclear processes are shown in Figure II-2.

Figure II–2: Tritium Production Processes[22]

**Reactor Process:**
A neutron strikes a Lithium-6 nucleus making a Tritium nucleus and a Helium-4 nucleus.




**Accelerator Process:**
A neutron strikes a Helium-3 nucleus making a Tritium nucleus and a proton.




Source: Zerriffi 1996

Nuclear reactors, whether for weapons or power (or both), also produce tritium both as a fission product and as an activation product in the coolant water. Heavy water cooled and moderated reactors have created much larger amounts of tritium compared to reactors that use "light" (or ordinary) water for cooling and moderation. The most prominent example of a heavy water power reactor design is the CANDU reactor, used in Canada, India and a few other countries. The abundant production of tritium in reactors that use heavy water is because the nucleus of deuterium needs to capture just one neutron to be transmuted to tritium. The CANDU-produced tritium is separated for commercial purposes.

[22] Reprinted from DOE Factsheet, "What is Tritium?"

The quantity of tritium produced in light water reactors is not uniform; it depends on the type and size of the reactor. In pressurized water reactors (PWRs) most of the tritium that is released to the environment is produced by the interaction of neutrons with boron and lithium. The boron is added to the primary cooling water to control the rate of the nuclear reactions in the fuel and the lithium is added to control corrosion. This is not an issue with boiling water reactors (BWRs), in which neither boron nor lithium is added to the primary water. Primary cooling water is the water that removes the heat generated by fission reactions in the fuel in the reactor. In BWRs the water boils in the reactor vessel itself. In PWRs, the water in the reactor vessel ("primary water") does not boil. The high pressure primary water is used to boil water by transferring heat to a water in a secondary loop in a device called a steam generator. The condensing loop, where the steam is condensed back to water, is the tertiary cooling loop. Routine tritium discharges and emissions from the reactor are mainly associated with the primary water of the reactor.[23]

Tritium is also produced in the fuel rods of both PWRs and BWRs from ternary fission (fission in which there are three fission fragments). Only a small fraction of this leaks into the primary cooling water along with some other fission products through very small cracks and holes that form in a small number of the fuel rods. The PWR's cooling water is constantly taken out for chemical treatment, for volume control, and to reduce the radioactivity. Then most of it is sent back into the reactor vessel. The chemical treatment is mainly to reduce the amount of boron as the reactivity of the fuel decreases with time. Some of the fission products that leak into the primary water of the reactor are removed by passing the water through ion exchange resin filters; however, this does not affect tritiated water which, being chemically identical to water, just passes right through the filters. The part of the cooling water which is not returned to the reactor vessel is put in holding

[23] Heavy water reactors produce the most tritium per unit of water – about 1.9 grams per year in a 900 megawatt-electrical (MWe) – or almost 19,000 curies per year. IRSN 2010, p. 4

tanks. It is periodically released after further treatment and dilution to bring the tritium concentration to a level deemed "safe" by the nuclear industry and the Nuclear Regulatory Commission (NRC). Fresh water is mixed in with the balance of the primary water to make up for the water that is withdrawn into the holding tanks.

Overall, the annual production of tritium in a PWR is variable. In French PWRs, the 900 megawatt model produces 0.03 grams (or about 300 curies per year); the 1,300 MW model produces 900 curies per year.[24] However, for U.S. reactors, Argonne National Laboratory estimates the annual production in a typical reactor to be 2 grams[25] or about 20,000 curies.

There is also tritium in the spent fuel pools of reactors where the used fuel is stored. Spent fuel is transferred from the reactor to the storage pool through a canal so the water in the reactor and in the pool mix during that period. PWR spent fuel pools also contain boron.[26] Some neutrons are present due to the spontaneous fission of plutonium-240 and uranium-238 in the spent fuel. As a result, tritium is also produced in the spent fuel pools themselves. Routine emissions of tritium from reactors in recent times are mainly from spent fuel pools.[27]

The amount of tritium created in the pool evidently depends on the total amount of spent fuel stored in it. It also depends on the "burnup" of the fuel – that is the amount of energy that has been extracted from it. The higher the burn-up the more the concentration of plutonium-240 in the spent fuel. Pu-240, which undergoes spontaneous fission, is a source of neutrons, and therefore of tritium in PWR spent fuel pools that have boron added to them for criticality control – that is to prevent accidental chain reactions. Tritium concentration in spent fuel pool water varies over the time of a refueling cycle and also depends on how much spent fuel is in the pool. The concentration

[24] IRSN 2010, p. 4
[25] ANL 2007, pdf p. 56
[26] BWR spent fuel pools are not borated. NRC 2006, p. 5
[27] Sejkora 2006 and Sandike 2014

in PWR spent fuel pool varies; typical values are between 20 microcuries per liter and 50 microcuries per liter.[28] Some pool water evaporates (since the spent fuel is hot, so is the pool water) and is emitted into the atmosphere; like other moisture in the air, it comes down as rain, to be evaporated again or wind up in groundwater. A comparison of pool water concentration to the U.S. drinking water standard provides some indication of what is being emitted. 20 microcuries per liter is one thousand times the U.S. drinking water standard.

Figure II-3 shows airborne emissions of tritium (generally in the form of tritiated water vapor) in the atmosphere from both reactors and spent fuel pools for pressurized water reactors. However, emissions can and do exceed these amounts (see below).

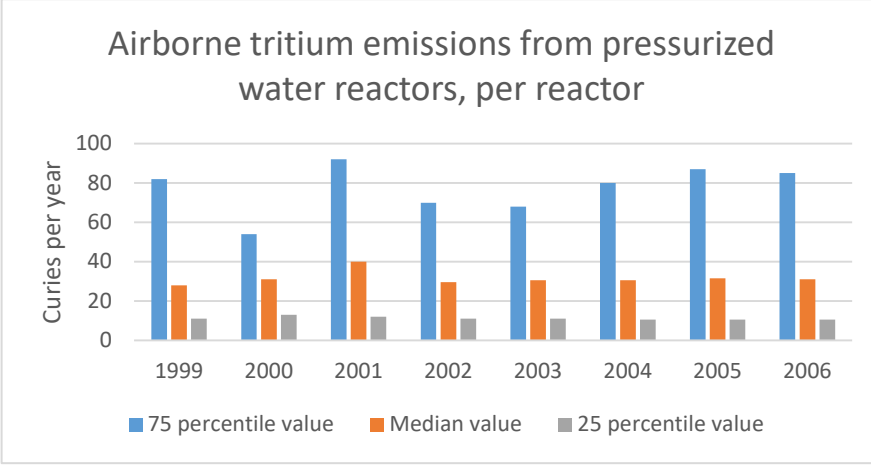



Figure II-3: Air borne tritium emissions from U.S. pressurized water reactors
Source: Jones circa 2007

## d. Secondary sources of tritium

Tritium also is found in a number of other places where it is not made. For instance, tritium is used in exit signs for illumination of the signs without a

[28] Sandike 2014, slide 10.

source of electricity. It is used in illuminated watch dials. When these are discarded into landfills, for instance, the landfills become contaminated. And tritium can then enter the environment from these landfills, including into water bodies.

Tritium is present at nuclear weapons production sites. Radioactive waste landfills at these sites contain large amounts of tritium, which routinely leaks into groundwater as well as surface water. Evaporation from contaminated surface water bodies, such as those at Savannah River Site, in South Carolina causes tritiated water to enter the atmosphere and rainout in the environs. Similarly these contaminated ponds and lakes are also sources of groundwater contamination.[29]

[29] Makhijani and Boyd 2004. Chapter III provides details about offsite tritium contamination (of the Savannah River and of groundwater) due to weapons production and dumping of contaminated materials on site.

# 3

# Tritium Discharges to the Environment and Concentrations of Tritium in the Environment

Prior to nuclear bomb testing, the tritium content of rainfall was about 16 picocuries per liter. Rainfall became contaminated with tritium after the start of atmospheric testing of nuclear weapons. The peak concentration, in 1963, was about 1,000 times greater – about 16,000 picocuries per liter.[30] The decay of tritium and the dilution of rainfall in surface waters, including the oceans has caused the concentration to fall back over the decades. The tritium content of rainfall due to natural and nuclear weapons testing factors is now back to approximately pre-testing days.

Tritium in surface waters due to natural and testing causes is on the order of 30 picocuries per liter (~1 Bq/liter) but can be higher up to ~110 picocuries per liter (~4 Bq/L). The concentrations vary by location but evaporation and mixing with ordinary, non-radioactive water tend to reduce the concentration over time (along with decay) unless there is a source of tritium.

Figure III-1 shows the variation in tritium concentrations in precipitation (rain and snow) from in 1955 to 1990. The box at left shows the concentration before the nuclear weapons era began (0.1 to 0.6 Bq/L or 3 to 16 picocuries per liter); the right box shows the concentration in 2008: 1 to 4 Bq/L (27 to 108 picocuries per liter). These concentrations would have declined by about half by 2020 due to the decay of tritium (half-life 12.3 years).

[30] IAEA on tritium at http://www-naweb.iaea.org/napc/ih/documents/global_cycle/vol%20II/cht_ii_05.pdf pages 63-64

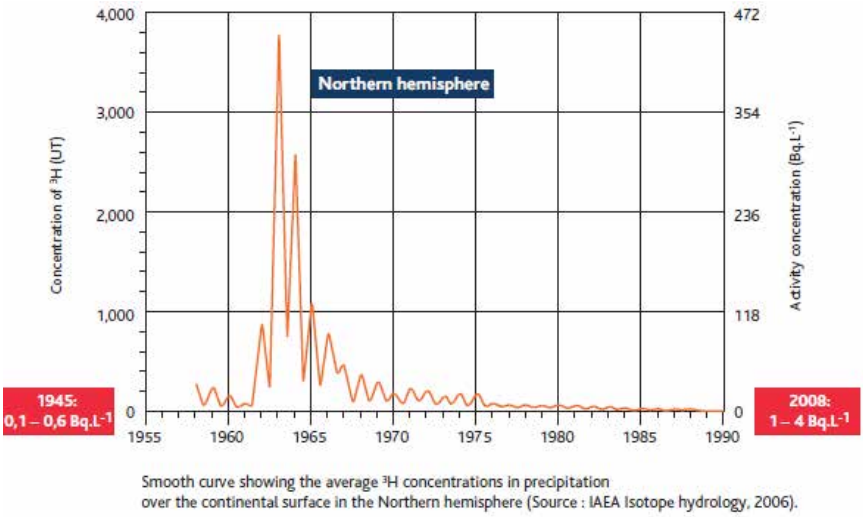



Figure III-1: Tritium concentrations in precipitation due to natural and bomb testing sources.
Source: IRSN 2010, p. 5, with permission. Copyright IRSN.

Figure III-1 shows tritium concentration in precipitation to be much less than 100 pCi/L from at least 1985, onward if not before. In contrast, nuclear power industry sources tend to cite the background as 100 to 300 pCi/liter. For instance, Ken Sejkora of Entergy used this range in his 2006 presentation.[31] Attributing a higher concentration to "background" would lead one to attribute tritium contamination in many areas to "background" even if it is largely or even mainly due to nuclear power plant emissions and discharges. Further, if one uses the actual typical background tritium concentration in precipitation, which is 20 to 30 pCi/L, the minimum detectable limit should be at about this level or below if the aim is to detect human impact on the environment. Specifically, that would enable reasonably accurate inferences about the contributions of nuclear power plant sources – i.e., reactors and spent fuel pools, low-level radioactive waste disposal facilities, etc. Typical detection limits used by nuclear power plant operators are much higher.

[31] Sejkora 2006, Slide 6.

Groundwater concentrations vary more, and may show the influence of bomb testing to a greater degree; they range from about that in surface water (20 to 30 picocuries per liter and sometimes even lower) to roughly 100 picocuries per liter. These are values for the mid-latitudes; concentrations are lower in the tropics.[32]

### a. Nuclear power plant emissions to the atmosphere and tritium contamination of rainfall and snow

The specific activity of tritium is very high – almost 10,000 curies per gram. This means that a single gam on tritium undergoes a very large number of disintegrations – almost 400 trillion – every second. As a result, a very small mass of tritium can contaminate a large amount of water. *For instance, one gram (the weight of about one-fifth of a teaspoon of water) of tritium in tritiated water (as HTO) will contaminate almost 500 billion liters of water up to the U.S. drinking water limit of 20,000 picocuries per liter.* The combination of two properties – tritiated water is chemically like ordinary water and tritium is highly radioactive – makes tritium a very pernicious pollutant that is difficult to contain and, once in the water, very difficult to remediate; in trace amounts, remediation is essentially impossible.

In addition to tritiated water, tritium can also be integrated into organic molecules and hence into body tissues. The tritium that replaces hydrogen in a carbon-hydrogen bond is difficult to remove and is, therefore, referred to as non-exchangeable organically bound tritium (OBT). Animal studies indicate that one to five percent of the tritiated water in mammals is incorporated into OBT in this way.

If tritiated water is used for irrigation, it can become organically bound in the plant's molecules. Direct intake of organically bound tritium through food makes it more likely that it will be incorporated as organically bound tritium in biomolecules in the human body compared to tritium ingested

[32] Marešova et al. 2017

by drinking tritiated water. However, it is important to remember that the term "organically bound tritium" is a catch-all for a heterogeneous group of compounds that can behave very differently in metabolic processes.

Both tritiated water and organically bound tritium can cross the placenta and irradiate developing fetuses *in utero*, thereby raising the risk of birth defects, miscarriages, and other problems. The forms of tritium discussed in this chapter are either tritiated water or OBT, unless otherwise specified.

***The most common form of tritium in the environment is as tritiated water in which a single atom of ordinary hydrogen has been replaced by tritium (HTO). In this book, the term "tritium" in the context of releases, contamination, etc. refers to tritiated water (HTO), unless otherwise specified.***

The main new source of routine tritium contamination of water after the end of atmospheric nuclear bomb testing is emissions to the atmosphere and discharges to surface water from nuclear power reactors (including spent fuel pools) and discharges from reprocessing plants.

Contamination of surface water bodies due to routine discharges from nuclear power plants can run to hundreds or even thousands of picocuries per liter. Rainfall can be much more contaminated. Its tritium levels in rainfall can range from close to background – that is a few tens of picocuries per liter to tens of thousands or even millions of picocuries per liter.[33] A 1985 study by Peterson and Baker estimated that about 780 curies of tritium per 1,000 megawatt electric (Mwe) from a PWR, operating at 82 percent capacity for the whole year, are released to the environment, of which 85 percent are waterborne effluents (663 curies) and the rest are airborne effluents (107 curies). The liquid effluents are discharged in batches in lakes, rivers, and oceans, often through underground pipes. Leaks can occur in such pipes and when they do, they contaminate the soil and groundwater. In boiling water reactors (BWRs) boron is

[33] Sejkora 2006

not added to the water and therefore tritium is not produced in boron-neutron reactions in the primary water.

The tritium in BWRs is mainly produced as a result of ternary fission in reactors. The Peterson and Baker study estimated that 120 curies per 1,000 Mwe are released per year to the environment, of which 75 percent is in gaseous form and the rest in liquid form. Leaks can also occur from BWRs that have pipes carrying primary water that are buried underground. At some plants tritium has leaked out of the cooling pool in which spent fuel is put after being unloaded from the reactor.

The estimates in the Baker study should be taken as indicative. In practice, the releases are much more variable from one reactor to the next. Figure III-2 shows tritium releases to the atmosphere from US pressurized water reactors (PWRs) and boiling water reactors (BWRs).

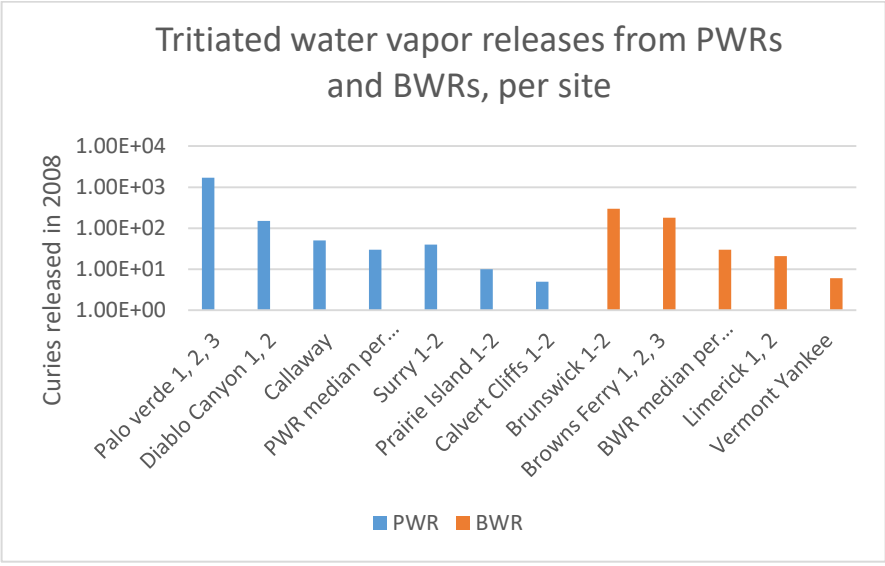



Figure III-2: PWR tritium releases to the atmosphere, 2008
Source: Adapted from data in NAS-NRC 2012, Figure 2.4

Figure III-2 allows the following conclusions regarding tritium emissions to the atmosphere from U.S. nuclear power plants, with the caveat that releases vary from year to year and the chart represents only one year of data (See Section b. below in this chapter):

1. On average, PWRs have much higher releases than BWRs. This is because much more tritium is made in PWRs in the course of reactor operation than in BWRs. However, there are some BWR sites where releases are higher than PWR sites at the low end of releases. At the lowest end, Hope Creek (not shown) released only a small fraction of a curie of tritiated water vapor in 2008.
2. The variation among PWRs is huge. The largest release from a single reactor, Palo Verde 1 (in Arizona), in 2008 was about 900 curies, compared to about 30 curies from Calvert Cliffs 1 (in Maryland). The differences stem from variations in reactor design and operating characteristics (like primary water chemistry).

Figure III-3 shows tritium release variations in 2012. The range is huge – from less than 10 curies to more than 1,000 curies (at left). The low end applies mainly but not exclusively to closed nuclear power plants.

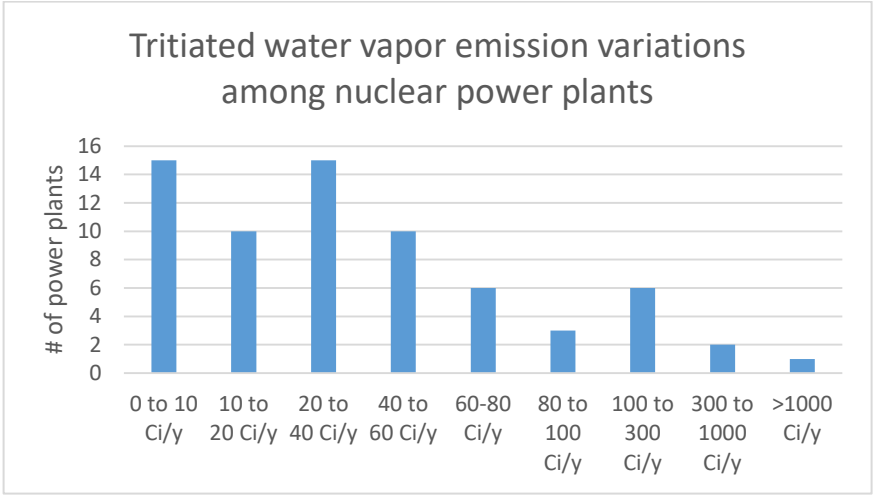



Figure III-3: Variation in tritium air emission per year from some US nuclear power plants
Data source: Sandike 2014, Slide 21.

Tritium emissions to the air generally affect the radioactivity in rain and snow and the tritium content of local water bodies. All of them can be and are often contaminated to levels far higher than would be present due to natural sources and bomb testing residuals.

In 2006, Ken Sejkora of Entergy Nuclear Northeast (Pilgrim Station), a company that owns nuclear power plants, presented an analysis of concentrations of tritium in rainfall due to emissions from a nuclear power plant of one curie per day continuous release. He estimated that concentration in rainwater falling on or around the site would depend on the rate and type of rainfall (as raindrops or as fine mist) and whether the winds were calm or not. Two scenarios of rainfall totaling 0.4 inches in a day (1 cm) resulted in estimates of tritium concentrations in water of 20,000 picocuries per liter (the drinking water limit) and as high as 36 million pCi/L – 1,800 times the drinking water limit – in case of rain in the form of a misty spray in calm atmospheric conditions.[34]

[34] Sejkora 2006

There is evidence of very high concentrations of tritium in precipitation in the form of a measurement at the Hope Creek nuclear power plant in New Jersey. The site has one boiling water reactor. In 2015, meltwater from an icicle was discovered to be contaminated with tritium at a level of 10 million picocuries per liter, or 500 times the maximum allowable annual average concentration under the drinking water standard:

> On February 18, 2015, PSEG identified some dripping from an icicle near the north end of the Hope Creek Turbine Building and collected a sample on February 19, 2015, of the dripping water as part of its on-going evaluation to identify the cause of the indication of tritium in ground water monitoring well BY. PSEG analyzed the sample and confirmed on March 3, 2015, that the dripping from the ice exhibited a tritium concentration of about 0.01 uCi/ml [10,000,000 picocuries per liter]. On March 4, 2015, PSEG informed the New Jersey Department of Environmental Protection, Bureau of Nuclear Engineering, in accordance with NEI 07-07, "Industry Ground Water Protection Initiative."[35]

The discovery was the result of an investigation of the cause of groundwater contamination with tritium. No cause other than rainwater and snow could be found:

> In late 2013, PSEG's groundwater monitoring program identified detectable tritium in a shallow onsite well (BY) near the corner of the Hope Creek Administration Building. The building is within the site restricted area and is not

[35] Dentel 2015

> accessible to members of the public. PSEG's evaluation, at that time, concluded that the detectable activity was associated with precipitation recapture of gaseous radioactive effluent releases since: 1) there were no sources of tritium near the area (e.g., pipes, tanks etc.); 2) there was no evidence of any leak or spill; and, 3) the concentrations of well sample results correlated well with gaseous radioactive effluent releases over the same time period.[36]

Such high concentrations in precipitation may indicate high episodic releases, raising questions about whether such releases are being adequately captured by monitoring. As noted above, the reported release to the atmosphere in 2008 was a small fraction of a curie. Thus, high year-to-year variations are also indicated if the source was the Hope Creek plant.

There is another nuclear power plant very near the Hope Creek plant – the Salem plant, which has two PWRs. The NRC document does not examine whether the highly contaminated rainfall came from Hope Creek emissions or Salem emissions or some combination of both.

In sum, there are two direct sources of offsite tritium contamination due to air emissions from a nuclear power plant site:

- Air emissions coming down as rainfall or snow and contaminating soil and water around the nuclear plants and on their sites.
- Air emissions contaminating groundwater on the site, which could then migrate offsite.

[36] Ibid. p. 19

The value of 10 million picocuries per liter in water from the melted icicle may be compared to the range of 20 million to more than 50 million picocuries per liter in spent fuel pool water cited above.

The NRC does not require monitoring of offsite private wells that are used for drinking or irrigation or both. Neither does it require monitoring of rainwater or snow, despite evidence of heavy contamination at least sometime. The Environmental Protection Agency (EPA) does some monitoring of radionuclides around the country, but does not focus on rainfall near nuclear plants. Further, private parties who own wells near the plants would generally not test them for radionuclides. EPA drinking water standards apply only to public water systems and thus do not cover private wells. The original intent was to spare private well owners the expense of monitoring. But this has created a loophole allowing contamination of wells due to emissions from nearby nuclear power plants.

Rainwater and melted snow percolate into the groundwater and contaminate it. Further, when releases are routine, soil pores will retain water and remain contaminated, especially in wet climates. Onsite groundwater contamination can migrate offsite. Tritium contamination from the Braidwood Illinois plant site migrated offsite to private wells, exceeding 1,000 picocuries per liter in one case.[37] In addition to rainfall and snowfall, tritium contamination of on-site groundwater can be due to leaks – as has been the case at several nuclear plants.

Most nuclear power plants in the United States have had tritium leaks at levels exceeding the drinking water limit of 20,000 pCi/L. According to the NRC:

> There are 61 nuclear power plant sites in the United States that are currently [2017] operating. Records indicate 43 of these sites at one time or another have had leaks or spills that involved tritium concentrations greater than or equal

[37] NRC 2005

> to 20,000 pCi/L. Six sites are currently reporting tritium, from a leak or spill, in excess of 20,000 pCi/L. Although many sites have had leaks or spills involving tritium, no site is currently detecting tritium in the offsite environment, or in drinking water, in excess of 20,000 pCi/L. Tritium rapidly disperses and dissipates in the environment, and as a result, tritium from leaks and spills is typically not detected outside the facility boundary.[38]

Sixteen of these plants have had peak groundwater tritium concentrations of more than 1 million picocuries per liter; concentrations have generally declined substantially since the dates of peak concentrations. They are shown in Table III-1.

Table III-1: US nuclear plants with peak groundwater concentrations in excess of 1 million picocuries per liter

| | Peak conc. PCi/L | Date of peak | 2017 conc. PCi/L |
|---|---|---|---|
| Browns Ferry | 7,520,000 | January 2015 | 3,493 |
| Brunswick | 19,000,000 | December 2010 | 280,943 |
| Callaway | 1,600,000 | July 2014 | 1,944 |
| Dresden | 10,312,000 | July 2004 | 251,000 |
| Duane Arnold | 2,150,000 | October 2012 | 2,700 |
| Grand Gulf | 2,240,000 | March 2014 | 3,200 |
| Hatch | 6,840,000 | September 2011 | 22,000 |
| Indian Point | 14,800,000 | February 2016 | 200,000 |
| Limerick | 3,950,000 | February 2009 | 369 |
| LaSalle | 1,230,000 | July 2010 | 11,000 |
| Millstone | 4,000,000 | November 2007 | 7,690 |

[38] NRC 2017

| | | | |
|---|---|---|---|
| Oyster Creek | 10,000,000 | 2009 | 2,250 |
| Palo Verde | 4,200,000 | March 1993 | not detectable |
| Quad Cities | 7,500,000 | 2008 | 15,700 |
| River Bend | 1,135,000 | February 2013 | 690,000 |
| Salem | 15,000,000 | April 2003 | 41,400 |

Source: NRC 2017

Note that this list has a mix of PWRs and BWRs; it consists of plants with one reactor on site and plants with more than one reactor on site; the high tritium concentrations are attributed to leaks and spills. The document listing these reactors does not mention the possible contribution of precipitation to groundwater contamination.

### b. Discharges to water bodies and concentrations in surface and drinking water

Nuclear reactors also routinely discharge tritium to water bodies – rivers, lakes, and oceans. The main source is when primary water, which is laced with tritium (and smaller quantities of other radionuclides) is discharged. Figure III-4 shows tritium releases in liquid effluents from several nuclear reactors of which the National Research Council began a study. No particular trend of increasing or decreasing releases in years that are decades apart is evident.

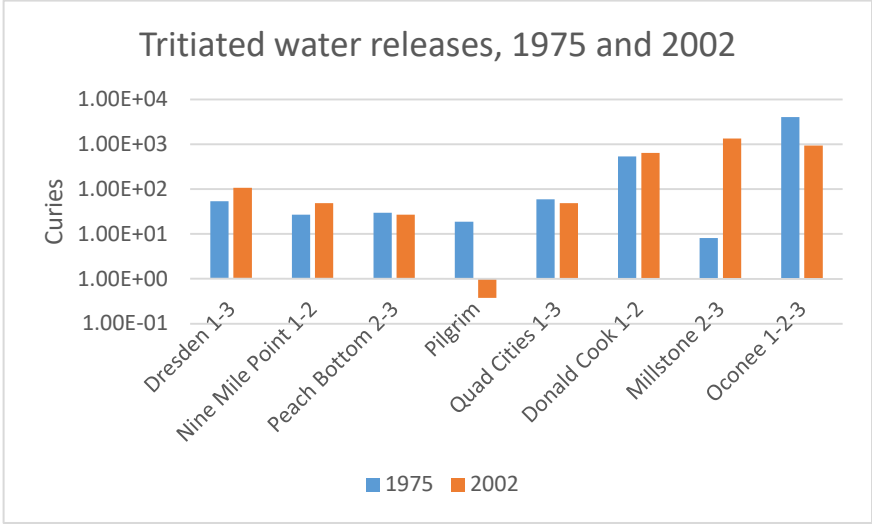



Figure III-4: Tritium releases in effluents of several reactors – BWRs at left and PWRs at right. Data read off from NAS-NRC 2012, Chapter 2. Figure 2.10, and converted to curies

Surface water becomes contaminated with tritium as a result of tritium releases. The concentrations vary widely, depending on the volume of water in the water body into which the discharge takes place, the nature of the water body, and the amount of the discharge. Data from environmental monitoring reports for the year 2016 of three nuclear power plants are shown in Table III-2 to illustrate the information available.

Table III-2: Surface and drinking water tritium concentrations at three nuclear power stations for 2016 (Note)

| | Year | Surface water | Drinking water |
|---|---|---|---|
| Braidwood | 2016 | 460 | 488 |
| Comanche peak | 2016 | 12,925 | Below detection limit |
| Oconee | 2016 | 4,285 | 317 |

Note: The measurements must be interpreted with caution due to possible high limits of detection and large measurement errors arising from that. Measurements below the limit of

detection are normally cited with the symbol "<" before the number. None of the values in Table 2 had that caution.
Sources: Exelon 2017, Vistra 2017, and Duke Energy 2017

There are also prior instances of drinking water contamination. Table III-3 shows four examples from 2006.

Table III-3: Tritium levels in drinking water at drinking water plants near four nuclear plants, 2006

| Nuclear Plant | Range pico-curies per liter | Mean (pico-curies per liter) | Distance from plant | Comments |
|---|---|---|---|---|
| McGuire 1&2 (NC) | 697 to 2,290 | 1,460 | 3.3 miles | North Mecklenburg Water Treatment Facility |
| Oconee 1,2,&3 (SC) | 298 to 370 | 340 | 18.9 miles | Anderson Water Plant |
| Vogtle 1&2 (near Savannah River Site, a nuclear weapons plant) | 471 to 1,040 | 766 | 76 miles | Purrysburg Water Treatment plant (Note 1) |
| Watts Bar 1 (TN) | 394 to 817 | 606 | 24 miles | Public water sampling location |

Source: Makhijani and Makhijani 2009, Table 2, p. 6

Note 1: Contamination downstream of Vogtle is a mix of discharges from the Vogtle Plant and the U.S. Department of Energy's Savannah River Site. According to the Savannah River Site's website, about 64% of the total releases were from Vogtle and the rest from Savannah River Site for an unspecified year. Year- to-year variation is expected to be substantial because of variations in flow and variations in tritium releases from both Vogtle and Savannah River Site. The effect of tritium releases to the atmosphere is not explicitly factored in; however runoff arising from tritium-contaminated rainfall may affect the concentrations.

### c. Tritium in the soil

Soil can become contaminated with tritium due to leaks from power plants. It is also contaminated due to tritium in rainfall. Pores in the soil have water in them in varying amounts; the level of tritium in this pore water is related to the level of tritium in the atmosphere. When contaminated rain falls on the soil at tritium levels higher than before the rainfall, the concentration of tritium in the pore water will increase. After the rainfall ceases, there is a disequilibrium between the tritium in the moisture in the air above the soil and the tritium level in the pore water at and near the soil surface. In that case, evaporation from the soil will increase the tritium contamination of the air in the vicinity. Plants can absorb tritium from the moisture in the air and from the water in the soil. They convert some of that moisture into organically bound tritium.

### d. Monitoring of tritium releases

The NRC requires power plant operators to monitor releases of radionuclides, onsite and offsite. The onsite and offsite releases from a plant are reported every year in the Effluent Report and the Environmental Report respectively. In the Effluent Report, the plant operator is required to give quarterly data on the amount of tritium curies released from each reactor, including the concentration of tritium before the water is sent to the underground pipe, the frequency at which the releases occur, and how long the releases last. In some cases, only the total discharges from two reactors may be measured. This may make it difficult to detect problems or to infer their existence from the reported data. Further, the amount of tritium discharged from PWRs is highly variable, as noted above. Other radionuclides are also reported.

Actual measurement practices at nuclear power plants vary considerably. The *Offsite Dose Calculation Manual Guidance* reports (NUREG-1301 for PWRs and NUREG-1302 for BWRs) direct the plant operator to have a lower limit of detection (LLD) of 2,000 picocuries per liter; that limit can be

increased to 3,000 picocuries per liter if no drinking water pathway exists. Most plant operators have lower LLDs (in the few hundreds of picocuries); however, these lower limits are not required. As a result, some power plant operators simply report that tritium levels are below the lower limit of detection. In some cases detection limit is not even specified.

Further, tritium measurements are done quarterly, using composites of samples that are collected at various intervals, commonly monthly. This means that samples from the times tritium is discharged (many times each quarter) and the times that it is not, are put together and averaged to give a quarterly result. There are two main problems with this approach. There is generally no independent verification by the NRC of when the samples are actually taken. The NRC (and hence the public) depends on the reactor operator's word that they are taken at the time of contaminated water discharge, and not before or after the discharge. As a result, there is no independent assurance of the representativeness of the samples and, hence, of the accuracy of the data in providing estimates of total tritium releases. Since tritium discharges are sometimes made into water bodies that are used for drinking downstream of the reactor (as is the case with the Braidwood plant), this lack of independent verification of discharges is troubling, especially in the context of batch sampling.

If the time the samples are taken is not coordinated with plant discharges occurring over a period of time, the samples may not be representative of the discharges; in such cases, the estimates of total tritium discharges would be inaccurate. There is at present no independent way for communities near nuclear plants to verify the accuracy of the discharges measurements and release estimates. This has become more important in light of the controversies surrounding the failure to report known tritium leaks.

It is noteworthy that despite evidence that serious contamination with tritium occurs in rainfall, and that it can be at levels hundreds of times greater than the drinking water limit, the NRC does not require routine monitoring of rainfall, either on the plant premises or offsite. Specifically,

the NRC does not require that the public residing near nuclear plants who have private wells to be alerted when there is a possibility that contaminated rainfall may exceed the drinking water standard of 20,000 picocuries per liter. This gap in requirements exists despite the fact, noted above, that the nuclear industry itself has called attention to the problem of potentially very high tritium concentration levels in rainfall. Granted that the drinking water standard is an annual average and rainfall is episodic; yet the drinking water standard is a common reference concentration and should provide a threshold for informing the public of environmental contamination. Indeed, the threshold for public information should be much lower (see Chapter 7, for recommended revisions to drinking water standards, including for tritium).

# 4

# Tritium Pathways and Residence Times in the Body

## a. Pathways

Once it is in the environment, tritium can reach people by a variety of pathways. It can be emitted from the source to the atmosphere, to surface water, or to groundwater. From there it can directly affect humans who ingest it or otherwise come into contact with is via drinking water, rainfall, or snowfall. There are also a large number of pathways via the soil, uptake by vegetation, consumption of plant matter by people, consumption of plant matter by animals or fish, and so on, with the eventual endpoint being a human being who is affected by tritium contaminated food, water, air or soil. Figure IV-1 below, from a publication of the French radiation research and safety agency, IRSN, shows various pathways for tritium transport in the environment, particularly as it might relate to agricultural activities.

Tritiated water is rapidly incorporated and reaches into all parts of the human body in a brief time after it enters the body via drinking water, food, or absorption through the skin. Here is the description of the biodynamics of tritiated water from the Argonne National Laboratory fact sheet on tritium:

> Tritium can be taken into the body by drinking water, eating food, or breathing air. It can also be taken in through the skin. Nearly all (up to 99%) inhaled tritium oxide can be taken into the body from the lungs, and circulating blood then distributes it to all tissues. Ingested tritium oxide is also almost completely absorbed, moving quickly

from the gastrointestinal tract to the bloodstream. Within minutes it is found in varying concentrations in body fluids, organs, and other tissues. Skin absorption of airborne tritium oxide can also be a significant route of uptake, especially for exposure to high concentrations of tritiated water vapor, as could occur under conditions of high humidity during hot weather, because of the normal movement of water through the skin. For someone immersed in a cloud of airborne tritium oxide (HTO), the uptake by absorption through the skin would be about half that associated with inhalation. No matter how it is taken into the body, tritium is uniformly distributed through all biological fluids within one to two hours. Tritium is eliminated from the body with a biological half-life of 10 days, the same as for water.[39]

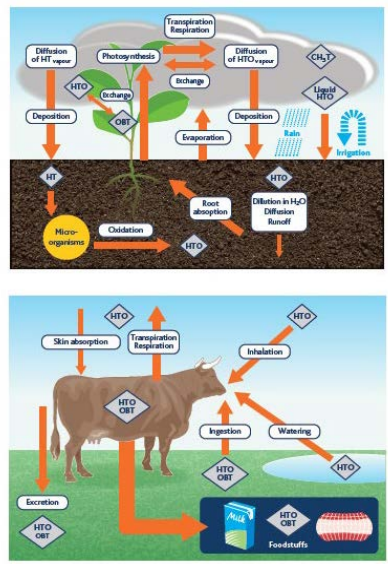



Figure IV-1: Illustrations of tritium pathways in agriculture. Source IRSN 2010, p. 9, with permission. Copyright IRSN.

[39] ANL 2007, p. 56; "tritium oxide" is tritiated water.

Note that tritium is distributed through all "biological fluids." In other words, the parts of the body, like bones, that have less water, will also have less tritium; those that have more water will have more tritium.

## b. Residence times

If there is a single intake of tritium, a biological half-life of 10 days means that almost 90% of the tritium will be excreted in about a month. But it will take almost two-and-a-half months to eliminate essentially all the rest – all of which goes right back into the environment.

However, if tritium is continuously present in the environment, as for instance in drinking water, it will build up in the body. A constant amount of 1 picocurie per liter in the drinking water and an intake of 1 liter per day will build up to about 18 picocuries per liter in the body. Intake rates vary by age. An adult male is usually considered to ingest 2 liters per day of water, as a reference amount (directly as water or as water contained in food and drink).

A part of the tritium in the environment and in the body becomes part of the hydrocarbon molecules of living beings – including all plants and animals. As noted, when it is part of organic molecules, tritium is called "organically bound tritium" (OBT). There are two kinds of OBT. A good description of OBT has been provided by the French safety agency, IRSN:[40]

> **Organically Bound Tritium (OBT):** this form, in which tritium is bound to organic matter, results from tritium being incorporated in various organic compounds during the synthesis process of living matter. Such organic compounds are distributed according to their specific chemical properties, which may explain the possible heterogeneous distribution of tritium among tissues. How stable tritium is within such compounds depends on the nature

[40] IRSN 2010, p. 3

of the bond between the tritium and the organic molecule and on the organic molecule's affinity with the different biological tissues. A distinction is made between:

**exchangeable tritium:** there is an exchangeable fraction when hydrogen atoms bound to oxygen, Sulphur or nitrogen are replaced by tritium and are readily accessible for new exchanges; this fraction of tritium bound in labile form to biomolecules is in equilibrium with the concentration of tritiated water in the intracellular environment;

**non-exchangeable tritium:** tritium is covalently bound to carbon. This is a permanent bond as long as the biomolecule itself is not transformed nor destroyed by an enzymatic reaction. The amount of time that tritium remains incorporated therefore depends on biomolecular turnover: fast in the case of molecules involved in the energy cycle, and slower in the case of structuring molecules or macromolecules such as DNA or energy reserve molecules.

These exchange mechanisms are common to all living organisms, plant and animal alike. The distribution between tritiated water, exchangeable and non-exchangeable tritium varies according to the respective intake of HTO or OBT, the nature of the organic bonds generating OBT and the metabolism of each individual species.[41]

Non-exchangeable OBT has a much longer half-life than exchangeable OBT. Further, the partition of tritium between tritiated water, exchangeable OBT and non-exchangeable OBT is dependent on the species.

[41] IRSN 2010, p. 3

### c. Organically Bound Tritium (OBT)

The issue of when, where, and how much tritium as water becomes organically bound and whether organically bound tritium bioconcentrates is a complex one. One principal route from water to OBT is via plants. When they take up tritiated water some of it becomes organically bound in the process of photosynthesis when plants make carbohydrates. From there the route can become more complex:

> Non-exchangeable OBT formed by photosynthesis appears initially in carbohydrates. Subsequently, metabolic reactions result in the incorporation of OBT in complex molecules such as polysaccharides, proteins, lipids and nucleic acids. The amount of OBT produced depends on a large number of environmental factors and plant parameters including light levels, oxygen and carbon dioxide concentrations, temperature, air circulation and water supply, all of which show considerable diurnal and seasonal variations. OBT concentrations are reduced by conversion back to HTO through maintenance respiration, but the process is slow. In the environment, OBT makes up a few percent of the total tritium activity in most plants but up to 90% in grains and hay crops, which have a high organic content.[42]

Phytoplankton bioconcentrate tritium. Two types of phytoplankton were exposed to tritiated water and then fed to mussels; both phytoplankton transformed HTO into OBT in their tissues but to different extents. The phytoplankton were also fed to mussels, in which tritium increased linearly with the amount fed to them.[43] The study's conclusions are as follows:

[42] Kim, Baglan, and Davis 2013, pdf p. 8. References within the quote are not included in the quote.
[43] Jaeschke and Bradshaw 2010

> Accumulation of organic tritium in the mussel tissues from tritiated-phytoplankton demonstrate an environmentally relevant transfer pathway of tritium even when water concentrations are reduced, adding weight to the assertion that OBT acts as a persistent organic pollutant. The persistence, potential for biomagnification and the increased toxicity of organic tritium increases the potential impact on the environment following a release of HTO. The study says that these issues should be taken into account in the legislation to adequately protect the environment and humans.[44]

Kim, Baglan, and Davis note that OBT concentration in plants and fruits, reflect total ("time-integrated") exposure to tritiated water.[45] In other words, plant tissues accumulate tritium as the plant takes up more and more tritiated water. Accidental discharges, reflecting non-equilibrium situations can result in significant bioaccumulation as has occurred in Wales due to accidental discharge of biopharmaceuticals; in that case "OBT concentrations in mussels and benthic fish were 1000 times higher than the HTO concentration in the water."[46] However, direct intake of tritiated water (in contrast to plants having a fraction of tritium as OBT) generally appears to results in roughly the same concentration of tritium in fish as in the water according to a 2020 study of contamination at the Savannah River Site in South Carolina.[47] The same study examined other radioactive and heavy metal contamination in addition to tritium and concluded that much was as yet not well understood:

[44] Jaeschke and Bradshaw 2010
[45] Kim, Baglan, and Davis 2013, pdf, p. 8.
[46] Kim, Baglan, and Davis 2013, pdf p. 11.
[47] Yu et al. 2020.

> This study reported fish tissue concentrations of $^{137}Cs$ and $^{3}H$, which have not been documented extensively in ecotoxicological studies. Our results suggested that industrial operations such as nuclear material production at SRS could have long-lasting impact on the aquatic ecosystem via the release of radionuclides and metals, and long-term monitoring of physiological effects and population level impact in biota exposed to these contaminants are recommended.[48]

The lack of ecotoxicological documentation indicates that while the concentrations may be documented, the ecological impact in terms of the toxicity, especially the combined toxicity, remains to be determined.

Over the long term, the tritiated water concentration, organically bound tritium, and seawater concentrations appear to become comparable, as indicated by research near the French reprocessing plant at La Hague. Background concentration of tritium in seawater, largely due to atmospheric testing of nuclear weapons is about 0.1 Bq per liter (2.7 picocuries per liter). The concentration in seawater near La Hague was more than a hundred times higher at 11 Bq/liter (almost 300 picocuries per liter). OBT concentrations in algae, fish, and shellfish built up to roughly the same level as the elevated levels near La Hague – that is roughly a hundred times higher than the background.[49]

A specific example where the importance of organically bound tritium relates to tritiated thymidine, which is an organic compound that can be incorporated into DNA. Experiments indicate that tritiated thymidine is "over 1000 times more efficient than HTO [tritiated water] on the basis of tritium concentration" in causing damage in "embryos up to the blastocyst stage."[50]

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[48] Yu at al. 2020
[49] ASN 2022, p. 62.
[50] ICRP 90 p. 14

Under the circumstances of exposure to such a tritiated DNA precursor, if even one-tenth of one percent of the tritium is in this form, its effect will be larger than tritiated water. This would not be the case for all forms of organically bound tritium, since thymidine is a DNA precursor. But it is one example of the type of damage that could be done by non-exchangeable organically bound tritium. The example and the research illustrate the large variations in response and the critical importance of considering the specific chemical forms of tritium when evaluating its impacts.

Tritiated water as well as organically bound tritium have greater average concentrations in fetal tissues than in maternal tissues (see Chapter 6 below). The health effects on the developing embryo/fetus itself (e.g., early miscarriages, malformations, and developmental effects) and on relevant organs at critical periods of fetal development are not well understood in human beings. Further, as Straume has noted "the incorporation of tritium into biomolecules of long-lived cells (e.g., neurons and oocytes) could result in large integrated doses over the lifetime of the cells."[51]

The impact of organically bound tritium needs much greater study in general; that is even more the case in regard to its impacts on the embryo and the fetus. Most of the discussion in the rest of this book does not depend on the specifics of the various types of organically bound tritium, their residence times in the body because there is little literature on the impacts on which this exploration is focused. For instance, the word "mitochondria," the focus of much of this book, does not even appear in the Canadian Nuclear Safety Commission's 2010 report on radiological safety and tritium.[52]

[51] Straume 1991 p. 5

[52] Canadian Nuclear Safety Commission 2010. This report acknowledges tritium risks, but only at very high doses, many orders of magnitude higher than usual exposures from nuclear power activities (p. viii). It is surprising that the report did not consider the impact on mitochondria and cellular metabolism before arriving at its conclusions even though this organelle is the core of the energy system of all multicellular plants, animals, and fungi.

# 5

# Harm From Internal Radiation

Health impact considerations of ionizing radiation have been largely focused on cancer risk. Within that framework, much of the attention has been devoted to the risk estimates derived from Hiroshima-Nagasaki survivor data. This discussion seeks, first of all, to broaden the range of risks considered to non-cancer risks and, within that, to accomplish three other things:

1. To review the differences in impact of internal radiation relative to external radiation, including with reference to the variation in impacts according to the endpoint under consideration.
2. To consider the impact of radionuclides that cross the placenta on the embryo and the fetus, including but not only the potential teratogenic impacts, especially in the first and second trimesters of pregnancy.
3. To focus on tritium as the radionuclide to illustrate the risks when the above two factors are combined, because tritium in the form of water is a pervasive pollutant in the nuclear age and also easily accesses every part of the human body, from the time of the formation of ova and semen to old age.

It is useful to review the science of health physics as it stands today in relation to considering external and internal radiation and the different types

of ionizing radiation. As a preliminary, a general note on radiation and biological damage is in order. Ionizing radiation, the kind this book is concerned with, splits molecules and creates ions – hence the name. As we are made of organic molecules, ionizing radiation can break up those molecules. Much of the study of radiation and cancer, for instance, involves the creation of breaks in the nuclear DNA caused by radiation. For instance, a photon or beta particle might create a "single strand break" in the DNA. These occur normally and naturally as well; the body has repair mechanisms and they are generally satisfactorily repaired. "Double strand breaks" occur when, as the term implies, both strands are broken – a situation in which repair is more complex – the DNA must find more than one location to reconnect, with a greater possibility of mis-repair, a mutation that can become the locus of a future cancer – or not, depending on future triggering events. Cell death may also result. Natural radiation causes such events, as does radiation put into the environment by human activities or medical radiation. Indeed, the objective of radiation therapy in cancer treatment is to kill the cancer cells; the risk is collateral damage to nearby healthy cells.

Overall, the best hypothesis for protection of human health in regard to cancer, based on numerous lines of evidence is that there is no threshold for cancer risk of radiation; for solid cancers the risk is linearly proportional to exposure.[53] That means that natural background radiation among the sources of cancer risk – along with all other risk factors. It should go without saying that the presence of natural risks should not be a license for pollution or infliction of human-caused risk.

The discussion in this chapter focuses on the impacts of internal radiation and how they are similar to or different from external radiation. That is because those differences are particularly important in the case of tritium (though not only tritium).

[53] BEIR VII 2006; EPA 2011.

## a. The model of the body as a bag of water

Understanding of the health risks and damage of ionizing radiation – the kind energetic enough to split apart molecules of matter – grew slowly at first after the discovery of X-rays by Wilhelm Röntgen (or Roentgen) in 1895. The medical community began to use X-rays widely and health damage appeared among its members very soon thereafter.[54] Exposure times were long and the results included symptoms of what we now know as high exposures, including loss of hair. Radium dial painting provided the next painful lessons. Women workers licked radium-laced paint brushes to shape their tips before application to dials to make them luminous. Necrosis of the jaw bone and bone cancer were the result. The practice was only banned after the women, suffering from painful osteonecrosis – death of bone tissue – and bone cancer filed a lawsuit against the United States Radium Corporation.

Understanding of radiation damage was accelerated after the start of the Manhattan Project, the massive U.S. World War II effort to make nuclear bombs. Lessons from the experience of X-ray exposure and radium dial painting were incorporated to set workplace exposure limits. A new discipline, called Health Physics, was born. As the term indicates, it was mostly created by physicists.

Basically, the approach was to treat the body like a bag of water. It was not an unreasonable starting point for the purpose of the Manhattan Project. After all, an adult's body is about 60 percent water; it is a larger percentage in utero and in newborns. Almost all the rest consist of a variety of hydrocarbons, like fats and proteins, made up of carbon, hydrogen, oxygen and a couple of dozen trace elements of which nitrogen and phosphorous are the most important by mass. Over 70 percent of the mass of the elements in the body consists of just two elements: hydrogen and oxygen, and another 10 percent is carbon. As an example of how the bag-of-water model is used,

[54] Sansare, Khanna, Kariodkar 2011.

experiments to understand the distribution of ionization in the body are carried out on "water phantoms," which are quite literally bags of water in the shape of a human body.

The bag-of-water model remains a useful way of understanding some aspects of radiation because the main way that ionizing radiation impacts the body is to break up the molecules of which it is composed – that is, to ionize them. This occurs when radiation particles[55] interact (or "collide") with those molecules and split them. Ionizations are directly caused by the energy of radioactive particles deposited in the body. "Radiation dose" is the measure of the amount of energy deposited in the body per unit weight of the body.[56] This ionizing energy deposited in a unit mass of the body provides a scientific measure of the number of ionizations to be expected. This, in turn, could serve as a proxy for risk of specific outcomes including a variety of diseases.

The matter is, of course, much more complex. One of the most important complexities that has long been recognized is that radiation that deposits its energy in many cells (known as "low linear-energy transfer", or "low-LET" radiation) creates different, and lesser, risks per unit of deposited energy than radiation that deposits its energy in a small, concentrated volume, known as "high linear energy transfer" or "high-LET" radiation. When

[55] Radioactive energy is carried by discrete particles. Electromagnetic radiation energy is carried in discrete packets called photons. Each photon carries a specific amount of energy corresponding to the frequency of the radiation. Ionizing photons are radiation of much higher frequency (and hence are much more energetic) than photons of visible light. This higher energy enables them to ionize molecules of matter. Other types of ionizing radiation include "beta particles," which are high energy photons, "alpha particles," which are high energy helium nuclei, proton beams if their energy is high enough, and neutrons (which ionize indirectly).

[56] A unit of radiation dose called a "rad" is the deposition of one erg of energy per gram of the body's (or organ's) weight; the same idea in metric units is called a "gray" (named after the British physicist Louis Harold Gray), in which one joule of energy is deposited in a kilogram (about 2.2 pounds) of the body. Both units measure the same thing, much as centimeters and meters both measure length, but different amounts of it. A gray is equal to 100 rad; it represents a very large amount of radiation for the body to absorb. Since the body is mostly water, deposition of energy in the body, notably in its soft tissues, is roughly equivalent to deposition of energy in the same mass of water.

cell biology is profoundly disrupted in a small region, as with high-LET radiation, the chances for normal repair processes to work are reduced and chances for damage, such as mutations, to persist are increased. This difference in the amount of biological damage per unit of energy deposited has been sought to be captured by a single parameter called "relative biological effectiveness." The word "effectiveness" refers to the *efficacy in causing damage*. To describe the amount of harm per unit of radiation dose, the amount of deposited energy per unit mass (rad or grays) is multiplied by the relative biological effectiveness factor for that type of radiation to get an empirical metric for radiation harm, measured in rem or sievert. A standardized relative biological effectiveness factor for regulatory purposes is called a "quality factor" or Q:

> Radiation harm in rem or sievert = (radiation dose in rad or grays) times relative biological effectiveness
>
> B (biological damage) rem (or Sv) = D (dose in rad or Gy) x Q

Unfortunately, the same word – "dose" – is applied to the metric of energy deposited per unit mass and to that number multiplied by the relative biological effectiveness factor to assess the damage done by the specific energy deposition under consideration. When assessing the issue of health impact, the term "biological damage" – represented by "B" in the above equation, is really a metric for biological damage or harm. As a metric for biological damage, with its own unit – sievert (or rem) as distinguished from the unit for deposited energy (gray (or rad), it deserves a specific word to describe it that is different from dose, which denotes the latter.[57] To be accurate, the

[57] I am indebted to Jessica Azulay for this insight. During one of IEER's technical training workshops several years ago, she asked why the term "dose" was used to refer to both deposited energy per unit mass, and also when that dose was multiplied by an empirical relative biological effectiveness factor that seeks to capture the relative harm of different types of radiation. She said that dose multiplied by relative biological effectiveness was meant to indicate biological radiation harm; therefore it was no

relative biological effectiveness factor should be specific to the type of biological damage that is under consideration – something that is almost never done in regulatory practice and, for the most part, in the scientific literature as well. In large measure that is because it is generally explicit or implicit that the risk under consideration is cancer risk, despite the fact that radiation exposure increases the risk for many other kinds of harm. Among other things, internal radiation exposure increases risks to pregnant women, the embryo and the fetus in a variety of ways, as discussed in this chapter.

Current standards generally assume that high energy gamma rays (such as the gamma rays emitted by cesium-137) and most beta particles (for example, the beta particles from strontium-90 decay) have a relative biological effectiveness factor of one — that is, the metric for harm is equal to the amount of energy deposited in the tissue and therefore equal to the dose. Alpha particles, on the other hand, which deposit all their energy in a smaller number of cells or even entirely in one cell, are assigned an RBE of 20. That is, the standards assume an alpha particle will do 20 times more biological damage than a gamma ray that deposits the same amount of energy in the body. Neutrons, which ionize indirectly, have different relative biological effectiveness factors, depending on the energy of the neutrons. These factors are usually applied in the context of cancer risk assessment.[58]

The biological damage caused by radiation evidently depends on the specific type of damage under consideration; this also means that the specific cells irradiated and the specific organ irradiated are also factors to be taken into account.

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longer a simple, physical measure. Using the same word "dose" for both was misleading and confusing. I agree. Dose multiplied by the relative biological effectiveness for a particular type of damage might be designated as "biological damage unit" or BDU for the specific harm at issue.

[58] In the case of the effectiveness of radiation in killing cells, a more precise metric is used (as this is important when radiation is used in cancer therapy to kill cancer cells). In that case, relative biological effectiveness is defined as the effectiveness of a particular kind of radiation to kill a cell relative to a 250 kilovolt photon. Waker 2012, slide 8. Waker notes that relative biological effectiveness factor depends on the endpoint – that is the specific biological impact -- being studied.

## b. Relative biological effectiveness

Examining different endpoints greatly expands the range of relative biological effectiveness factors that should be applied. Let us briefly consider why it is critical to apply relative biological effectiveness factors to the specific types of harm being evaluated using two examples:

- Sister chromatid exchange frequency; and
- The differences between the impacts of radiation on the embryo and fetus in early pregnancy versus assessing cancer risk.

Sister chromatid exchange is exchange of genetic material that occurs during mitotic cell reproduction. Excessive exchange can be harmful. Research by Hatsumi Nagasawa and John B. Little, published in the journal *Cancer Research*[59] compared sister chromatid exchange induced by low-LET radiation (high energy x-rays) with that induced by high-LET radiation (alpha particles, emitted in the process of plutonium-238 decay). They found that a rather high level of low-LET radiation – 1 to 2 grays (100 to 200 rads) produced the same level of genetic damage as indicated by sister chromatid exchange as just 0.31 milligray of high-LET radiation (plutonium-238 decay alpha particles), both being administered at high rates to Chinese hamster ovary cells. *This implies a relative biological effectiveness for the particular genetic endpoint of 3,200 to 6,400. Thus, the relative biological effectiveness of alpha radiation, based on these experimental results, would be 160 to 320 times greater than the factor of 20 applied in regulatory practice to assess cancer risk.* The typical factor used to compare the cell killing effectiveness of gamma rays with alpha radiation is 10. The factor for induction of sister chromatid exchange frequency indicated by this experiment is greater than that factor by 320 to 640 times.

[59] Nagasawa and Little, 1992

A central finding of the Nagasawa-Little paper was that 30 percent of the cells showed increased sister chromatid exchange frequency even though less than one percent of the cells were traversed by an alpha-particle. In other words, a very large part of the damage caused to the ovary cells was not only due to the high rate of linear energy transfer (high LET) in the directly affected cells. More than 30 "bystander" cells were impacted for every cell in which the alpha particle directly deposited energy. Very large relative biological effectiveness factors were also inferred by Khadim et al. in their experiment with hematopoietic clonal cells derived from stem cells that "survived the passage of one or more radiation tracks before the initiation of clonal proliferation" – that is, the experiment was for an endpoint of radiation that was not cell death. They observed "a high frequency of non-clonal aberrations in the clonal descendants compatible with α-emitters inducing lesions in stem cells that result in the transmission of chromosomal instability to their progeny." The effect was not observed when the cells were subject to x-rays.[60]

Some impacts of radiation exposure other than cancer are explored in the next chapter (Chapter 6); we consider here the basic biological difference between considering impacts on the embryo and fetus, especially early in the pregnancy, on the one hand and a fully developed adult on the other.

There is a central biological dynamic at the two times that is different. When an ovum is fertilized, creating a zygote, the stage is set for very rapid cell division. Enough mass must be created with enough complexity so that the blastocyst, as it is called, can implant itself on to the wall of the womb, usually between five and nine days after fertilization. There are no repair mechanisms. Damage to the blastocyst before and after implantation, or some problem with the zygote itself, is frequent – so frequent that a large fraction of fertilizations result in failed pregnancies either before implantation or in the weeks immediately following. The exact fraction is unknown

[60] Khadim et al. 1992

because such early miscarriages generally occur before a women realizes she is pregnant. On the order of a fifth of fertilizations fail to implant and even more result in an early miscarriage in the days after implantation.[61] Both genetic and environmental factors are involved; indeed, there is not a sharp dividing line between the two because ova and sperm can be and are damaged by environmental factors.

A basic biological-ecological way of looking at the problem is that for fully grown people, ionizing radiation impacts generally occur in the context of day-to-day routine homeostatic functioning – the body is in dynamic equilibrium with its environment. That is not at all the case for the embryo and the fetus that are developing and growing rapidly. For instance, the drastic negative impact of cell-killing in the earliest stages of pregnancy contrasts starkly with an alpha particle killing a cell in an adult most of whose cells are, with critical exceptions (such as oocytes), replaced in the space of days or months.

Thus, cell-killing, the lack of effective repair, the rapid creation of organs from stem cells are critical to understanding radiation (and other environmental) harm to the embryo and then fetus, especially in the phases of organ development (which for the brain continue after birth); quite the contrary is true for adults. In the latter case, dead cells are removed by normal metabolic processes, because cell death and replacement are part of the homeostatic functioning of grown people.

In contrast, embryonic and fetal stages are, by their nature, non-homeostatic. For adults, damaged cells that do not die are important, including for cancer risk, because it is those damaged surviving cells that become the loci for cancer, if there are subsequent triggering event(s).[62] Further, cancer risks for specific organs involve estimation of dose to the whole organ (integrated over time in the case of low dose-rates).[63] Teratogenic risks

[61] Danielson 2020
[62] See for example Section 2, EPA 2011.
[63] See EPA 2011 for instance.

during organogenesis are quite different. They involve non-stochastic effects – like cell killing or genetic damage – in rapidly dividing stem cells. In that context it is far more important to consider specific endpoints and specific cells; dose and damage are not averaged over the multitude of cells in a whole organ, which is still being formed.

It is important to note in this context that the concept of relative biological effectiveness is being applied to a non-stochastic effect, that is, a deterministic effect. The concept of relative biological effectiveness has generally been applied to adjust stochastic risk for a single end point – cancer – to radiation with different rates of linear energy transfer.[64] Generally two or more adverse events separated in time characterize cancer risk; repair mechanisms also play a role. Further, cancer risks for specific organs involve estimation of dose to the whole organ (integrated over time in the case of low dose-rates).[65] Teratogenic risks during organogenesis are quite different. They involve non-stochastic effects – like cell killing or genetic damage – in rapidly dividing stem cells. In that context it is far more important to consider specific endpoints and specific cells to estimate particular teratogenic risks.

In sum, a relative biological effectiveness factor or 2.2 for tritium compared to gamma radiation[66] may well be reasonable for adult cancer risk estimation, within the limitations of the concept. However, as the above discussion shows, specific factors must be calculated for endpoints other than cancer, especially in the context of pregnancy.

### c. Impact on mitochondria

Just as most health impact literature relating to ionizing radiation has focused on cancer risk, it has also focused on mutations in the nuclear DNA, which is in the form of the famous double helix. However, all multi-cellular

[64] See for example EPA 2011, Section 2.
[65] See EPA 2011 for instance.
[66] Canadian Nuclear Safety Commission 2010, p. ix

living creatures including plants, animals, and fungi as well as many others, contain organelles in their cytoplasm called mitochondria. Figure V-1 shows a schematic of a typical animal cell. The double-helix DNA is inside the nucleus; the cytoplasm, where radionuclides would first enter the cell, surrounds the nucleus. There are hundreds to thousands of mitochondria in each human cell (except red blood cells).

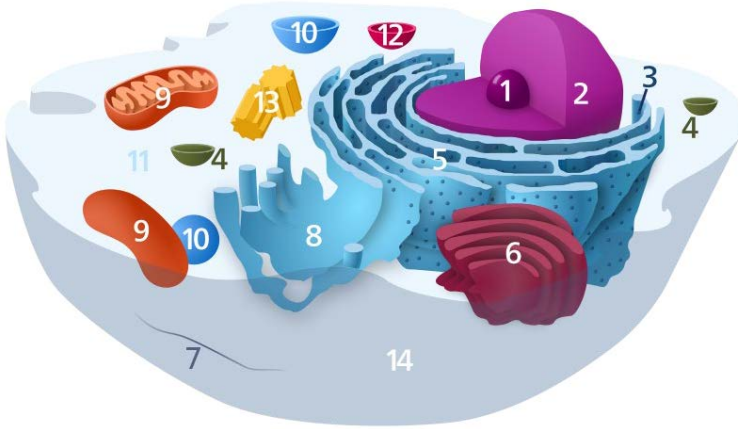


Figure V-1. Schematic of an animal cell. 1. Nucleolus; 2. Nucleus; 3. Ribosomes; 4. Vesicle; 5. Rough endoplasmic reticulum; 6. Golgi apparatus; 7. Cytoskeleton; 8. Smooth endoplasmic reticulum; 9. Mitochondrion; 10. Vacuole; 11. Cytosol; 12. Lysosome; 13. Centriole; 14. Cell membrane.
Source: Wikimedia commons at https://commons.wikimedia.org/wiki/File:Animal_Cell.svg

Mitochondria are the core of the body's energy system, converting a variety of organic molecules into ATP, the energy currency that enables us to function – that is, enables our hearts to beat, our lungs to expand and take in air and contract to exhale, our mouths to open and close so we can eat, or our fingers to move in order to type or to play the violin. They are also the communication system between the environment outside the cell and the nucleus. Each mitochondrion has its own DNA, abbreviated as mtDNA, which is circular in shape. (Nuclear DNA is often abbreviated nDNA when

both kinds of DNA are being discussed; we use this convention in the rest of this book.) Mitochondria, like other organelles in the cytoplasm, are bathed in the cytoplasmic fluid, called cytosol, which is primarily water. This is a critical fact in understanding the impact of ionizing radiation in cells, including its potential health impacts.

The cytoplasm occupies most of the volume of the cell and contains water that can be ionized by the beta particles of tritium. MtDNA is ten times more likely to suffer mutations due to ionizing radiation than nuclear DNA. Besides being the core of the body's energy system, mitochondria are also control aging and immune response. Thus damage to mtDNA can have profound systemic impacts on health – including, but not only, cancer. Yet the impact of radiation on mtDNA, on the body's energy system, on aging, and on the interactive damage from other pollutants is scarcely assessed in the literature on radiation risk. For instance, the BEIR VII report contains only a dozen or so mentions of mitochondria, with no examination of the systemic risks that mtDNA damage might create.

Further, mtDNA is an inheritance from the maternal side alone, unlike nuclear DNA, where the male and female each contribute 23 chromosomes. The metabolism of the entire body has its counterpart in the metabolic processes that take place in each cell. Basically, we get energy when organic molecules like carbohydrates are oxidized: we "burn" calories. Each cell has a balance of oxidants and anti-oxidants that is maintained as part of the finely tuned metabolic functioning of each cell and the whole body in healthy living beings. An excess of oxidants – chemical species that oxidize organic matter into simpler parts, and ultimately just carbon dioxide and water – can damage cells; a deficiency can cause a lack of energy to run the body. Figure V-2 shows a schematic of mitochondrial DNA, which is circular, and its regions.

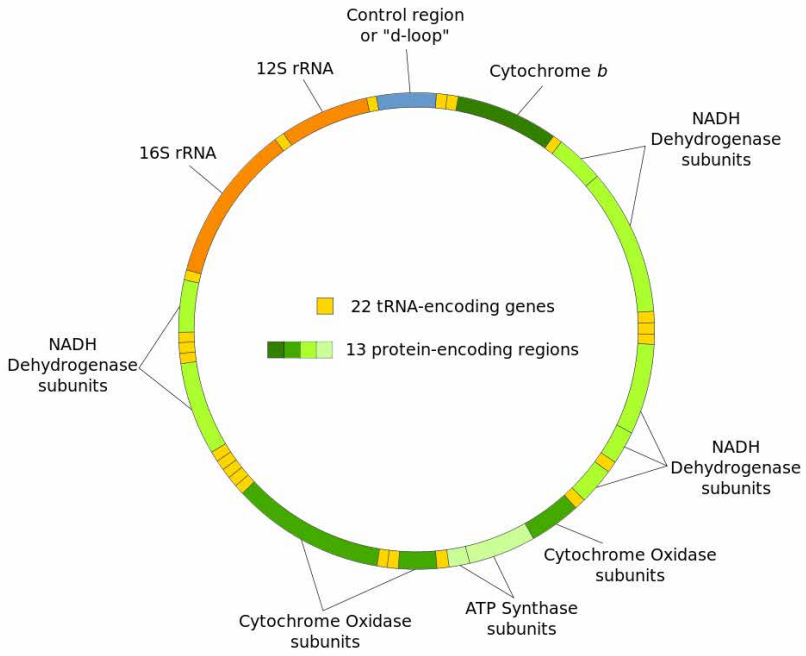


Figure V-2: Schematic diagram of mitochondrial DNA showing its regions

Source: Wikimedia at https://commons.wikimedia.org/wiki/File:Mitochondrial_DNA_en.svg

Commons Author: Derivative work, translation by Knopfkind, jalyout by jhc

One of the ways that ionizing radiation and many other pollutants damage living cells is by creating an excess of oxidants. There are many kinds of oxidants; those that contain oxygen are known as "reactive oxygen species" or ROS for short. Cell metabolism involves reducing and oxidizing chemical reactions; as a result, there are normally reactive oxygen species in cells, balanced by the presence of reducing species. It is *excess* reactive oxygen species that create damage, oxidizing in excess of restorative capacity. Excess reactive oxygen species are created in cells by pollutants. In the specific case of tritium, its beta particle emissions ionize molecules, including water, the "hydroxyl radical" (OH), which is the most reactive of reactive oxygen species, is among the ones created.

The 2006 National Academies report, BEIR VII, notes the potential for ROS to damage mitochondria:

> …oxidative damage cannot be considered a single entity, but is dependent on the chemical source of the oxidation. Mutants sensitive to hydrogen peroxide included an overrepresentation of mitochondrial respiratory functions, but those sensitive to diamide encompassed genes involved in vacuolar protein sorting. This makes it especially difficult to predict what kinds of damage would result from endogenous reactive oxidative species. Endogenous damage could present its own unique spectrum of genes required for resistance, different from each of the exogenous sources as well as from ionizing radiation.[67]

Pirini et al. have noted the special vulnerability of mtDNA among smokers:

> Mitochondrial DNA is extremely vulnerable. Defects in the respiratory chain can lead to an increased production of ROS, increasing in turn the probability of nuclear DNA damage and increases in mtDNA content, a compensatory mechanism for the damages to the respiratory chain and the lack of energy production. Moreover, genes belonging to oxidative phosphorylation and mitochondrial dysfunction pathways are differentially methylated among smokers.[68]

While Pirini et al. is about the damage caused by smoking, the mechanism discussed is the creation of excess reactive oxygen species, which is also the principal route for impact of ionizing radiation, including tritium beta particles.

[67] BEIR VII 2006, p. 42
[68] Pirini et al. 2015

Finally, mitochondria are the core of the energy system of all multicellular animals, plants and fungi. Thus ecological toxins, including radioactive materials, that cause mtDNA mutations create risks that could ripple through entire ecosystems.

## d. Considering tritium dangers and risks

Tritium outside the body should be considered differently from tritium inside the body (whether as tritiated water or organically bound tritium). Once tritium crosses into a cell, its beta particle energy will generally be deposited entirely within that cell. The average and maximum stopping distances of tritium beta particles in water are 0.42 microns and 5.2 microns, respectively.[69] This is somewhat smaller than the range of diameters of cell nuclei and, therefore, also smaller than cell diameters. As a result, each tritium decay will typically result in hundreds of ionizations, both in the nucleus and in the cytoplasm; the most energetic beta particles will create well over a thousand ionizations.[70]

As noted in the National Academies BEIR VII report, exposure to beta particles can create the highly reactive hydroxyl radical, creating oxidative stress in the cell. Hydroxyl radicals are produced when the ions created by collisions between beta particles and water molecules go on to collide with other water molecules. Essentially all of the energy of ionizing radiation emitted by tritium is in the form of beta particles.

The hydroxyl radical can create a very substantial amount of intracellular damage, including DNA damage:

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[69] NRC 2015, p. E-38

[70] For instance, the ionization energy of water is about 12.6 eV; at that rate, the average beta particle energy of 5.7 keV would produce about 450 ionizations of H2O molecules. Source: NIST 2001. The maximum beta particle energy would result in over 1,470 ionizations of water molecules. Of course, the ionizations from a decay would not be confined to a single type of molecule (though water is the most abundant). Ionizations of organic molecules which have a binding energy of a few electron volts would be even more abundant.

> The relatively long-lived (about $10^{-5}$ s) OH• radical is believed to be the most effective of the reactive species; as an oxidizing agent, it can extract a hydrogen atom from the deoxyribose component of DNA, creating a DNA radical. *Early experiments demonstrated that about 70% of the DNA damage can be prevented by the addition of OH• scavengers*….Because OH• is so highly reactive, it has been estimated that only the radicals formed within about 3 nm of DNA can react with it….[71]

The literature on cellular damage due to excess reactive oxygen species in cells is quite abundant, since excess ROS are created by a variety of pollutants in a variety of circumstances. Since that is also the main mechanism for damage caused by tritium, it is possible to tap into this larger literature to gain insights on tritium beta particle harm as well.

Excess ROS can attack mitochondrial DNA; in turn, mitochondrial dysfunction may cause genomic instability, possibly with heritable impacts.[72] It can also contribute to neurological diseases.[73] What would be the impact of such mitochondrial dysfunction produced by the excess reactive oxygen species created by ionizations in the cytoplasm in the embryo and fetus? Ruder et al. examine the impacts and risks of diet on pregnancy, mainly through assessing the oxidative impacts of diet – beneficial, when there are anti-oxidants, and stressful when there are not. Specifically, these impacts are hypothesized to include early failed pregnancies:

> Reproductive failure is a significant public health concern. Although relatively little is known about factors affecting fertility and early pregnancy loss, a growing

[71] BEIR VII 2006, p. 29. Italics added.
[72] Kim et al. 2006.
[73] Arun, Liu, and Donmez, 2016

body of literature suggests that environmental and lifestyle factors play an important role. There is sufficient evidence to hypothesize that diet, particularly its constituent antioxidants, and oxidative stress (OS) may influence the timing and maintenance of a viable pregnancy. We hypothesize that conditions lead to OS in the female affect time-to-pregnancy and early pregnancy loss.[74]

[74] Ruder et al. 2008

# 6

# Teratogenic Impacts on the Embryo and Fetus

We can now consider the question of the impacts and risks of radiation exposure, especially internal radiation exposure, to the embryo and the fetus as a topic in its own right.

## a. Transfer of radionuclides to the embryo and fetus

Internal exposure of the embryo and fetus occurs when radionuclides are transferred from the mother to the embryo and the fetus through the fluids that are exchanged between the two. Evidently, radionuclides can only reach the embryo and fetus if they are in the woman's body when she is pregnant. But if the biological half-life of the radionuclide is long enough, it will still be in in her body when she becomes pregnant, and be available for transfer to the embryo and fetus.

A 2001 report of the National Radiation Protection Board published the transfer ratios of various common radionuclides from the mother to the fetus.[75] For some radioactive materials, like plutonium, the ratio of fetal to maternal concentration depends on whether the intake was before or during pregnancy and also on when during the pregnancy the intake occurred. For others, the ratios are independent of time of intake. Table VI-1 shows some of the fetal to maternal radionuclide concentration ratios as published in the British report.

[75] NRPB 2001. The NRPB is a British radiation board.

Table VI-1 Fetal to maternal radionuclide concentration ratios

| Element | Intakes prior to Pregnancy | Intakes during Pregnancy |
|---|---|---|
| H in HTO | 1.6 | 1.6 |
| H in OBT | 1.6 | 1.6 |
| Organic carbon | 1.5 | 1.5 |
| Phosphorus | 0.5 | 10 |
| Sulphur | 1 | 2 |
| Potassium | 1 | 1 |
| Cobalt | 0.2 | 1 |
| Zinc | 2 | 2 |
| Technetium | 1 | 1 |
| Ruthenium | 0.01 | 0.2 |
| Cesium | 1 | 1 |
| Lead | 1 | 1 |
| Bismuth | 0.1 | 0.1 |
| Thorium (see note) | 0.03 | 0.1;0.3;1 |
| Uranium | 0.1 | 1 |
| Plutonium (see Note) | 0.03 | 0.1;0.3;1 |
| Americium | 0.01 | 0.1 |

Source: NRPB 2001, Table 1; only some of the radionuclides in that table are shown here
Note: The three ratios shown for thorium and plutonium are for intakes during the first, second, and third trimesters.

These facts about maternal-fetal radionuclide transfer establish a clear imperative to limit and, if possible, eliminate, man-made additions to the radionuclide burdens in ecosystems because once there, the long-lived ones will tend to become part of the food chain. The short-lived ones may also cause severe impacts, as for instance, when women who were pregnant at

the time of atmospheric nuclear weapons testing or in the immediate aftermath of the Chernobyl and Fukushima accidents were exposed to short-lived iodine isotopes. Other radionuclides, such as zinc-65, which has a half-life of about eight months, are also important in this context. Zinc is an essential mineral that plays many vital roles in the body, including in its immune system, in healing wounds, and in the process of cell division. Note that the fetal to maternal ratio of zinc is two, so that this radioactive zinc-65 would be even more concentrated in the fetus than carbon-14 or tritium. For instance, zinc-65 was an important contributor to radiation dose due to U.S. nuclear testing in the Marshall Islands. Fish, a staple of the Marshallese diet, was an exposure pathway, including for pregnant women.[76]

### b. The establishment view of teratogenic impacts

In the first part of the pregnancy, the potential harms and risks can be studied under the general rubric of "teratogenic impacts." In the matter of radiation risk, it is useful to start with the 1988 study of internal radiation exposure done by the U.S. National Academies in one of their periodic reports on radiation health effects. This report, commonly known as the BEIR IV report,[77] had a section in Chapter 8 entitled "Fetal Effects, Teratogenesis, and Neonatal Effects of In Utero Exposure." Two kinds of impacts were discussed: early miscarriages, including failures of implantation of the blastocyst; and harms caused during the period of organ formation, known as organogenesis.

In regard to "preimplantation loss" of the embryo, BEIR IV postulates that there is a threshold of 10 rad (0.1 gray) based on laboratory experiments on animals subjected to external radiation and "[t]heortical considerations" because a minimum number of cells must be killed before there is

[76] Franke 2002

[77] "BEIR" stands for "Biological Effects of Ionizing Radiation." The most recent report in this series was published in 2006, and is known as the BEIR VII report, since two others were published between BEIR IV and BEIR VII>

a preimplantation loss.[78] The report also arrives at a similar conclusion about teratogenic effects during organogenesis. A part of the reasoning was as follows:

> During the major organogenesis state, the *embryo appears to be sensitive to all the known teratogenic effects of radiation*….Windows of one to a few days are commonly observed during which a given developmental abnormality can be induced during the major organogenesis stage. Thresholds are expected theoretically and have been observed; single doses below about *10 rad of low-LET radiation* appear ineffective.[79]

The threshold observations are based on irradiation of animals with external radiation in experiments. The National Academies and the International Commission on Radiological Protection (ICRP) have recognized a teratogenic impact on the central nervous system as having no threshold. This conclusion is based on analysis of outcomes of women who were pregnant at the time of the atomic bombings of Hiroshima and Nagasaki in August 1945.

The ICRP issues numbered reports on a variety of topics relating to the impacts of radiation. ICRP 49 dealt with impacts of in utero radiation on the brain. The report included an analysis of women who were pregnant when the United States dropped atomic bombs on Hiroshima and Nagasaki in August 1945, and who survived those bombings. The epidemiologic analysis of the outcomes is described in ICRP 49 as follows:

> First, 30 of the 1 599 pregnancies included in the revised clinical sample terminated in a child with severe mental

[78] BEIR IV, p. 383.
[79] BEIR IV 1988, p. 383, italics added.

> retardation and, second, 18 of these, or 60%, had disproportionately small heads, that is, a head with a circumference more than two standard deviations below the mean observed among the 1 599. Of those pregnancies that terminated in a mentally retarded child…no fewer than 19 (and 17 of the 21 who received exposures of 0.01 Gy or more) were exposed in the 8th through the 15th week after fertilization. *This is many times the expectation based on the assumption of no effect of fetal age at exposure*. In this context, to reiterate, severe mental retardation implies an individual unable to form simple sentences, to solve simple problems in arithmetic, to care for himself or herself, or is (was) unmanageable or institutionalized.[80]

The term "severe mental retardation" is used here in the clinical sense provided by ICRP 49, as quoted above. ICRP 49 provides a dose-response estimate of 0.4 per gray – ranging from "one case per hundred individuals *exposed to less than 0.01 Gy* [1 rad] to approximately 40 cases per hundred at an exposure of 1 Gy [100 rad]."[81] Finally and importantly, while a substantial majority of the impacts observed occurred between the 8th and 15th weeks, an increase in "severe mental retardation" was also noted for exposures well beyond that time – up to 25 weeks after fertilization.[82]

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[80] ICRP 49, 1986, p. 20; italics added

[81] ICRP 49 1986, p. 31, italics added. The evidence of severe mental retardation at less than one rad and a linear dose-response curve leading to a conclusion that there is no threshold is consistent with other evidence for lack of thresholds. This includes the evidence for substantial increases in childhood leukemia and brain cancer at 1 rad presented to you on October 27, 2021 by Dr. Rebecca Smith-Bindman, Medical Perspectives, October 27, 2021, slide 11; hereafter Smith-Bindman 2021.

[82] ICRP 49, 1986, Table 1, p. 21. ICRP 49 found that "simplest statistical model consistent with the data appears to be a linear one without threshold." p. 31. The ICRP analysis also reported on a decline in IQ that was "consistent with the interpretation that there is a dose-related shift in IQ and that this could explain the increase in clinically classified cases of severe retardation. They do not exclude the possibility of two separate effects…. The statistical uncertainties in the data, and the known problems of obtaining a high consistency in intelligence testing, prevent quantitative statistical analysis of these data from refining these qualitative conclusions."

The facts and analysis in ICRP 49 make very clear that teratogenic impacts should have become a principal element of the scientific and public health discussion of the impacts of low-levels of exposure decades ago – dating back at least to ICRP 49. Certain conclusions are clear from BEIR IV and ICRP 49:

1. Radiation exposure can result in early failed pregnancies; they are hard to detect because women may not realize that they are pregnant from the very fact of an early failure.
2. Radiation exposure can produce a variety of teratogenic effects, especially during the most sensitive period of organogenesis.
3. There is no threshold for "severe mental retardation" caused by radiation exposure between the 8th and 25th week of pregnancy. A linear no-threshold for impact means that exposures during this period would be expected to produce the severe disabilities, with the number being proportional to the population dose to pregnant women in the relevant period of pregnancy.

### c. Reconsidering certain aspects of the establishment view

There are also issues relating to teratogenic impacts that need revisiting.

BEIR IV considered failed pregnancies only in the earliest post-fertilization period and teratogenic impacts only in the balance of the embryonic period up to 50 days of the pregnancy for all but the central nervous system. In the latter case it considers a window up to 19 weeks.[83] This is very important, but far too limited. Harm can occur leading to failed pregnancies can occur earlier and later than that window of 50 days.

[83] BEIR IV 1986, pp. 383-384.

For instance, Table 4 of ICRP 49 mentions possible neural tube defects ("dysraphic abnormalities") during the third and fourth weeks of pregnancy.[84] As the Centers for Disease Control and Prevention has explained, failure of neural tubes to close can lead to severe teratogenic effects, including spina bifida and anencephaly (the failure of the brain to develop at all, or develop incompletely). The CDC states that "Pregnancies affected by anencephaly often result in miscarriages, and the infants who are born alive die very soon after birth."[85] Thus a broader view is needed even for miscarriages. This is also true of teratogenic impacts more generally.

For instance a 1979 synthesis study, "based exclusively on the abundant but dispersed data in the literature,"[86] covered the period of central nervous system development starting at 15 days after conception through the entire pregnancy and the post-natal period, when the pre-frontal cortex continues to develop. Thus, the vulnerability period for central nervous system teratogenic impacts extends through the entire pregnancy as the Teratology Society also makes clear. According to the overview provided, a variety of risks extend throughout pregnancy, with different organ development risks being prominent in different periods. The Society summarizes possible adverse outcomes during pregnancy as follows:

> Teratogenic exposure during any period or phase of development can have dire consequences….In general, disruption of the earliest developmental stages (gametogenesis; fertilization, cleavage, and blastulation) results in the loss of the conceptus (that is, a miscarriage, often before the woman realizes she is pregnant). Disruption somewhat later during primary morphogenesis and organogenesis often results in major structural anomalies (a

[84] ICRP 49 1986, p. 30.
[85] Beth Burke et al., p. 6.
[86] Zamorano and Chuaqui 1979, pdf p. 1.

> "birth defect" for example, a neural tube defect, such as spina bifida; a ventral body wall defect, such as gastroschisis; a heart defect, such as the formation of a single outflow tract; a limb anomaly, such as phocomelia; or a facial cleft, such as cleft lip or palate). Disruption during the late embryonic and fetal period generally results in abnormal organ differentiation, growth, and function (for example, cognitive impairment, hearing loss, neonatal hypoglycemia, lung immaturity). Thus, the timing of a particular teratogenic exposure can result in drastically different outcomes.[87]

The postulated threshold of 10 rad (0.1 Gy) also needs re-examination since it is based on laboratory experiments on animals using external radiation, as regards both the laboratory setting and the radiation source.

First, research on wildlife compared to laboratory experiments conducted since then puts this conclusion into considerable doubt. Specifically, a Chernobyl study compared laboratory experiments on a single species with the impact of radiation on wildlife seen in the field over wide range of dose rates. It concluded that wildlife is eight times more sensitive to radiation than indicated by laboratory experiments as measured by the median value of hazardous dose rate "suggesting that organisms in their natural environmental [sic] were more sensitive to radiation."[88] This single study is suggestive rather than definitive. Still, the magnitude of the field and laboratory difference provides ample basis for caution in assuming that a threshold for most teratogenic effects, if it exists, is as high as 0.1 Gy (10 rad) of low-LET radiation.

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[87] Bleyl and Schoenwolf 2014

[88] Field radioecological studies are critical. For instance, wildlife in the Chernobyl exclusion zone appears to be much more sensitive to ionizing radiation than indicated by laboratory experiments – Gernier et al. 2013.

Second, a relative biological effectiveness factor that is specific as to the type of radiation and the harm being considered needs to be applied to internal radiation when relating it to the postulated threshold of 10 rad (0.1 gray) of external radiation exposure (see Chapter 5 above). BEIR IV applies a relative biological effectiveness factor of 10, based on in-vitro cell killing data, to external low-LET radiation data to conclude that there is a threshold of one rem for internal high-LET radiation. Thus, for internal alpha radiation, the threshold inferred by BEIR IV is one rem.

However, cell death is not the only relevant endpoint. As discussed in Chapter 5, experimental data indicate that the relative biological effectiveness factor for increased sister chromatid exchange frequency is in the thousands – that is, at least two orders of magnitude higher than for external radiation. This would indicate a threshold for damage of less than 10 millirem (0.1 millisievert). The Khadim et al. experiment indicates that there may be certain impacts, like non-clonal genetic aberrations that are unique to internal radiation because they were not in evidence when external radiation was applied.[89] Such phenomena indicate that for some endpoints there are qualitative differences between internal and external exposure. For those endpoints, relative biological effectiveness is not an applicable concept. Internal radiation appears to produce some genetic impacts that external radiation may not.

### d. Multigenerational impacts

The previous section relates to teratogenic impacts on the embryo and fetus. Beyond that, consideration of multigenerational impacts is also essential. That there are intergenerational risks is clearly indicated by the established fact of maternal to fetal radionuclide transfers.[90] Radionuclide body burdens acquired prior to pregnancy also impact the embryo and fetus due

[89] Khadim et al. 1992.
[90] ICRP 88 2002, NRPB 2001, Pacific Northwest National Laboratory 1996.

to radionuclide transfer from the mother, if the biological half-life of the radionuclides is long-enough.

There are a number of areas to of concern relating to the goal of protecting women and future generations:

- How long does each radionuclide transferred to the fetus stay in the child as it develops into an adult? What are impacts on subsequent generations of those body burdens? How does that vary between males and females?
- What is the impact on ova of internal radiation exposure during the oogenesis period of pregnancy and also due to exposure during the post-puberty maturation of ova? Specifically, what are the risks of germline mutations during oogenesis? What are the corresponding risks related to sperm, which are transient, as well as to primary spermatocytes?
- What are the risks of mutations in mitochondrial DNA during oogenesis and the related implications for the female child and succeeding generations? How many generations can mitochondrial DNA mutations be expected to last?
- What are the intergenerational risks of germline mutations in oocytes due to ionizing radiation, including internal exposure, producing teratogenic outcomes in the children of a woman who was exposed as a fetus?
- Can mitochondrial DNA damage acquired during pregnancy create general health vulnerabilities for subsequent generations, for instance, via compromised intra-cellular metabolism?

- What radionuclides are important for multigenerational impact, based on the following considerations?
  - Ubiquity of pollution – tritium is a prominent example in terms of the quantities routinely emitted and discharged from nuclear power plants, reprocessing plants, and other military and commercial facilities, whether as water, water vapor, organically bound, or other relevant chemical forms.
  - Specific radionuclide pollutants that impact some locations more than others a result of specific activities carried out there, such as uranium mining, uranium milling, nuclear weapons testing, and nuclear power plant accidents, with Chernobyl and Fukushima being the most prominent examples; however in the case of atmospheric testing and some nuclear accidents the damage can be and has been over vast areas, and even global;
  - Leaks and discharges from a variety of waste disposal and waste management activities as well as residual contamination of soils and waters at nuclear facilities. Uranium and radionuclides in the decay chain of uranium-238 are a common example.
- What pathways for fetal and multi-generational exposure have been understudied or even ignored?

A few examples related to nuclear weapons testing will illustrate some of the issues. Both pre-pregnancy exposure and intakes during pregnancy can result in fetal radionuclide burdens. Many women throughout the world, particularly in areas of high fallout during atmospheric weapons testing, had significant intakes of radionuclides. People who lived downwind from the Trinity test had direct inhalation doses. They also collected rainwater

from their roofs in barrels and used it for drinking and cooking. This was, of course, typical of many areas in the world during atmospheric testing. They were also affected by various pathways due to fallout depositing on laundry hung out to dry. Collection of drinking water and outdoor laundry drying are frequently mentioned by the affected public in matters of exposure; yet, they have often not been given their due.

Nor are the problems limited to the immediate vicinity of the test sites. For instance, the 1954 CASTLE test series in the Marshall Islands created significant cumulative fallout thousands of miles from the Enewetak test location.[91] The issue of zinc-65 exposure may be especially important, all the more since it has not been covered in much-used official scientific literature on fallout.[92] Moreover, as shows in Table VI-1 above, the fetal concentration of zinc-65 is twice as large as that in the mother. According to the Centers for Disease Control and Prevention, "Children need zinc to grow and develop. Zinc is a mineral that is important for immune function, wound healing, and the senses of smell and taste."[93] Radioactive zinc will behave identically in the body to the non-radioactive zinc that all people, and especially children need. Thus, it can impair all the functions that zinc is responsible for promoting. Specifically, impairment of immune system development can set the stage for a variety of health vulnerabilities later in life.

Finally, a large, though uncertain fraction of pregnancies, end in miscarriages. One estimate indicates that 40% to 50% of all pregnancies may be "occult" miscarriages – that is miscarriages that are not clinically recognized and often not even recognized by the women themselves. [94] A large part of the reason is that many of these occult miscarriages occur in the first few weeks after fertilization. The fraction of these that might be related to environmental causes or to combinations of environmental and genetic

[91] List 1955; see the map on p. 20 (pdf p. 26). Hot spots are indicated as far as Mexico City to the east of the Marshall Islands and Colombo, Sri Lanka to the west.
[92] Franke 2002
[93] CDC 2022
[94] Calculated from Rice 2018, Figure 3. The percentage varies according to the age of the woman.

causes is unknown. Given the ubiquity of environmental pollution, both radioactive and non-radioactive, an assessment of the environmental contribution to failed pregnancies should be a high priority along with other issues of reproductive justice.

# 7

# Tritium Standards in Water

Remediation of research deficiencies and gaps, including those identified above, is going to take time. The potential for teratogenic damage, the established conclusion that there is no threshold for central nervous system teratogenic effects, and at least plausible arguments that there is no threshold for other teratogenic impacts point to the necessity and urgency of interim protective measures as research clarifies the specific harms and risk parameters for various endpoints, including the potential harms in the first trimester of pregnancies (two trimesters in the case of the central nervous system).

In particular, a tightening of drinking water standards is urgently needed, especially for tritium, which is the most common routine radiological pollutant that is routinely emitted and discharged in larger quantities from commercial nuclear facilities than any other pollutant. For the reasons discussed below, a tightening of the drinking water standard to 400 picocuries per liter (from the current 20,000 picocuries per liter[95]) would be much more protective; it is all the more needed in view of the long neglect of protection of pregnant women and the embryo and fetus.

There is already a substantial official history pointing to the need to tighten drinking water standards for tritium because tritium in drinking presents risks to very large populations. For instance, the specific public health criterion used by the official Ontario Drinking Water Advisory Council, was a lifetime fatal cancer risk of one in a million people for tritium-contaminated drinking water; tritium is emitted from the province's

[95] 40 CFR 141 (2013)

heavy water-moderated nuclear power plants. The recommended limit was 20 Bq/liter (540 picocuries/liter).[96] The Department of Energy remediation guideline for tritium in runoff water during the decommissioning of its Rocky Flats Plant in Colorado provides another example where the same risk criterion was used and the limit was set at 500 picocuries per liter, which is 40 times more stringent than the U.S. drinking water standard.[97] The concern in that case was the nearby drinking water reservoir that supplies the Denver area.

The Ontario and Colorado calculations were done using somewhat dated dose conversion factors to calculate the lifetime risk. The updated values in Federal Guidance Report 13 show that the tritium concentration limit corresponding to the same a one-in-a-million life time fatal cancer risk should be 400 picocuries/liter, whence the recommendation. The State of California has a drinking water guideline of 400 picocuries per liter (15 Bq/L) of tritium.

While tritium is the most common man-made pollutant, it is not the only one. The same criterion should also apply to other man-made radionuclides that cross the placenta. My calculations for drinking water limits at the one-in a million risk level based on Federal Guidance Report 13 are reproduced below in Table 1; they were presented to the New Mexico Water Quality Commission.

Table 1: Recommended drinking water standards for certain man-made radionuclides in pCi/L (or Bq/L)

| **Radionuclide** | **pCi/L** | **Bq/L** |
|---|---|---|
| Americium-241 | 0.19 | 0.007 |
| Cesium-137 | 0.64 | 0.024 |

[96] The Advisory Council's recommendation was rejected in favor of a much more lax standard (7,000 Bq/L) even though the Council noted that the province's power plants could meet the 20 Bq/L limit if they did not exceed their discharge limit – ODWAC 2009, p. 5.

[97] Canada uses metric units; Ontario's advisory level was 20 becquerels/liter (Bq/L), which is the rounded value corresponding to the one-in-a-million life time risk; it is equal to 540 picocuries per liter)

| | | |
|---|---|---|
| Plutonium239/240 | 0.15 | 0.006 |
| Strontium-90 | 0.35 | 0.013 |
| Tritium | 400 | 15 |

Source: Witness Statement of Arjun Makhijani to the New Mexico Water Quality Commission on behalf of Amigos Bravos, August 27, 2009, p. 8. The statement and the transcript of the oral testimony are at https://ieer.org/resource/press-releases/mexico-strengthens-limits-public/

The U.S. drinking water standard applies only to public water systems. As a result, private wells are exempt, sparing them the expense of monitoring and other compliance-related expenses. However, this also allows nuclear power and weapons plants to emit and discharge tritiated water that can adversely impact private water supply. It is important to consider interim restrictions on such emissions and discharges to protect drinking water sources not covered by the EPA regulations to the equivalent standard of 400 picocuries per liter (15 Bq/L). Further, in the US and elsewhere there are groundwater protection standards to ensure the continuing quality of aquifers irrespective of whether water is drawn by public or private users. This places a requirement on polluters not to pollute aquifers irrespective of actual or potential use.

# 8

# Concluding Reflections

There is a very wide range of possible harms from internal radiation, especially of radiation that deposits all its energy in a single cell, such as the beta particles emitted by tritium decay and alpha particles, such as those emitted by several plutonium isotopes, including plutonium-238 and plutonium-239, the three natural isotopes of uranium, U-234, U-235, and U-238, and the man-made uranium isotope, U-233

Three broad categories of harm especially deserve more attention; while research is done to improve knowledge, precautionary action and standards are needed to protect human health:

1. **Teratogenic impacts**, especially in the first trimester of pregnancy, including early failed pregnancies, central nervous system harms, and organ malformations. In this context, there is ample evidence to revisit the use of the concept of "relative biological effectiveness" of internal radiation compared to external gamma radiation. Non-cancer impacts, such as sister chromatid exchange frequency and bystander effects need to be researched, along with the variety of post-natal health impacts they may have.
2. **Mitochondrial impacts**, including mitochondrial DNA (mtDNA) mutations, caused when intra-cellular radiation results in the creation of excess reactive oxygen species (ROS) in the cytoplasm. There is a broad category of adverse health impacts that may be

associated with excess ROS. For instance, mtDNA mutations in ova may result in multigenerational harms, since mtDNA is inherited exclusively from the mother. As another example, disruption of mitochondrial functions could result in a broad array of adverse impacts because they compromise the functioning of intra-cellular metabolism. As discussed in this book, mitochondria are the core of the energy system of cells and, hence, of the body. Further, since all plants, fungi, and animals have the same mitochondrial metabolic system, radionuclides like tritium that disrupt their functioning could also cause a broad array of ecological harms. This aspect of research is especially important in relation to tritium because it is the most ubiquitous routine pollutant associated with both nuclear weapons and nuclear power.

3. **Potential synergistic impacts** of internal radiation and non-radioactive pollutants. Such impacts have been difficult to estimate for a variety of reasons, including the different metrics used to assess dose and the way those metrics translate into standards for health and environmental protection. For radiation the basic metric has been energy deposited per unit mass. In turn, that metric is used to set limits for radionuclides in the environment, including drinking water, or for remediation of contaminated sites. For instance, the limit for tritium in drinking water is 20,000 picocuries per liter; it is derived from dose – energy deposited per unit mass; it implies, a standard assumption about drinking water consumption. Other limits are set directly for dose. For instance,

dose from man-made beta-emitters other than strontium-90 and tritium, is set at 4 millirem per year to the whole body or any organ. The limit for drinking water contamination is derived from that dose limit.[98] This approach cannot be used for non-radioactive toxic materials, since their mechanism of toxicity is not energy deposition in the body; rather they interact chemically, and hence biologically, with the body in a variety of ways. Even the units are incommensurate. Rather than energy deposited, drinking water concentration limits for organic toxic chemicals and heavy metals are expressed in terms of mass per unit volume (usually milligrams per liter).[99] The radiation and chemical metrics are incommensurate so far as assessing harms to health and ecosystems is concerned. However, to the extent that the avenue of harm is via creation of excess ROS, whether by chemical toxicants or radionuclides, there is a clear avenue for the study and assessment of the combined adverse impacts of radiation and chemical exposures, including especially (but not only) internal radiation exposure.

This book has focused on tritium for the reasons described in the preface and Chapter 1. Other beta emitters can and should also be examined in the same manner. Among the most important include carbon-14, strontium-90, cesium-137, and radioisotopes of iodine. The disproportionate damage

[98] U.S. drinking water standards are specified at 40 CFR 141. Radionuclide standards are at 40 CFR 141.66.

[99] 40 CFR 141.50 for organic chemicals and 40 CFR 141.51 for heavy metals.

that alpha radiation causes has long been recognized but usually in the context of cancer risk is dealt with by applying a relative biological effectiveness factor of 20 – a value that is not applicable to a variety of other situations.

People who have suffered the impacts of radioactive pollution, notably, but not only, indigenous peoples, have long complained of teratogenic impacts during pregnancy and of multi-generational impacts. The widespread impacts due to uranium mining in many countries, including countries that have neither nuclear weapons or power, and on indigenous lands of countries that have one or both, are exemplary of the problem. This exploration of tritium and its impacts points to the need for a broader and much more serious consideration of the health harm from internal exposure to radionuclides. This should extend well beyond the usual consideration of nuclear DNA mutations to include mitochondrial DNA and excess reactive oxygen species impacts on cellular metabolism. Finally, given that the umbilical cords of newborns have been found to have over 200 chemicals in them, [100] the establishment of precautionary standards for chemicals and for chemicals and radiation combined is a critical and urgent matter for public and environmental health.

[100] EWG 2005

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# Appendix A:
# Radioactive rain

As noted in Chapter 3, nuclear power plants routinely discharge tritiated water and emit tritiated water vapor through their stacks. IEER commissioned a special modeling of tritium releases during one year from a single nuclear power plant, the Braidwood plant in Illinois. This plant was chosen for a few reasons:

- It is a pressurized water reactor, the most common type of power reactor in the world and in the United States.
- Tritium leaks at the plant led to serious controversies around the plant's health and environmental impacts.
- A physician and engineer, Dr. Sauer, who lived in the area and whose daughter contracted brain cancer compiled cancer data in the environs of the plant. While not an epidemiological study, the data are suggestive.
- Despite the controversies, no significant actions for requiring monitoring of the impact of radioactive precipitation on private wells, drinking water and locally produced food has been required.

We noted in Chapter 7 it was reasonable to not require owners of private wells to have their water tested for pollutants in the same manner as required for public drinking water systems. But this has created a very big loophole that allows the industrial neighbors, including nuclear power plants, of rural residents to emit pollutants like tritium that impact their

drinking water. This is an example of a serious environmental protection failure.

Even Milton Friedman, an eminent apostle of the free market and limited government, noted that the freedom of individuals should be limited in a variety of ways. He noted this in the context of determining the role of government. Among other things he noted that "one man's freedom to murder his neighbor must be sacrificed to preserve the freedom of the other man to live." In the same general context, he also opined that people should not be free to pollute the water flowing through their property because that action "in effect forc[es] others to exchange good water for bad" involuntarily. Government action of was needed when people are in situations where "it is not feasible for them, acting individually, to avoid the exchange or to enforce appropriate compensation."[101] This precisely describes the situation in which neighbors of NRC licensees (and many other industries) find themselves. It is therefore the responsibility of the EPA and the NRC, even in a minimalist interpretation of the appropriate role of government, to devise appropriate remedies, including preventing the industries from putting people in the position of exchanging good water for bad.

The rest of this Appendix was prepared by the consulting firm of Matthias Rau in Germany from modeling done for the year 2015 using data filed by Exelon, the owner of the Braidwood plant, with the Nuclear regulatory Commission.

[101] Friedman 1962 Chapter 2.

# Appendix B


# Braidwood Illinois Tritium Release

Client: Arjun Makhijani, Ph.D.
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Heilbronn, September 09, 2020

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# 1 Task

Subject of the investigation is the dispersion of tritium, its washing out with rain and the resulting ground contamination caused by the exhaust air of the nuclear power plant in Braidwood, Illinois.

The engineering office M. Rau was commissioned by the Institute for Energy and Environmental Research, Maryland, with this study.

# 2 Methodology

The dispersion calculations will be carried out with the German dispersion model ARTM [1] (Lagrangian dispersion model) used for licensing and regulatory purposes concerning long term releases caused by normal operation of nuclear installations.

The model calculates dispersion, dry and wet deposition of gaseous releases from nuclear installations. The results are two-dimensional fields of concentration and contamination and time series of concentration and contamination for given points.

As input, the model requires a meteorological time series and mean release values for the relevant period.

This study examines in special the gaseous release of tritium and the resulting contamination by precipitation in the area surrounding the Braidwood nuclear power plant.

Results are calculated on the basis of mean release rates for 2017 for the four quarters and the year as a whole, differentiated for continuous and discontinuous releases.

# 3 Model description and required data

## 3.1 Model description

The numerical calculation of ground contamination by wash out for tritium will be carried out with the model-package ARTM, an adoption of the well-known AUSTAL2000-model for modelling radionuclide transport, deposition and radionuclide doses. AUSTAL2000 is in compliance with the German Guidance TI Air. The core of AUSTAL2000 as well as ARTM is a Lagrangian particle model [1].

The Lagrangian particle model differs fundamentally from the majority of the established numerical modelling techniques which are founded on the computed solution of the advection-diffusion equation. This model tracks point-like particles representing a trace species on their path through the atmosphere. The particles travel with the mean wind and are additionally subjected to the influence of turbulence. The effect of the turbulence is modelled by adding an additional random velocity to the mean motion for the particle. This random velocity, which is derived from a Markov process, is a function of the turbulence intensity and is different for each particle. The

concentration distribution is determined by counting the particles in given sampling volumes and expressed as mean values over the volume and time intervals. The main advantages of the Lagrangian particle model are that the model concept largely reflects the natural phenomena involved in turbulent diffusion. It can be applied to any source geometry desired for any temporal behaviour of a spatially variable source. Required meteorological input information includes the fields of the mean wind components, the wind fluctuations and the diffusion coefficients which can be generated by meteorological pre-processors. For time-dependent calculations, the input parameters must be made available as a time series of fields. Furthermore, emission data will be required. The model output is a time sequence of the spatial distribution of the concentration of the emitted species and its transformation products. Based on the calculated time sequences mean values and momentary values for different time periods can be.

The ARTM-model has additional functions compared to AUSTAL2000 as e.g.

- Determination of the radiological dose caused by cloud radiation (gamma submersion),
- Calculation of ground contamination caused by wet deposition,
- Considering radioactive decay during dispersion.

## 3.2 Model options for the calculations

The following options have been used for the calculations:

### 3.2.1 Study Area

The calculations were done for a terrain (study area) of about 9,6 x 9,6 km² with a nested grid with fine resolution near the plant and a rough resolution in greater distances. The finest grid has an extension of 1.2 x 1.2 km² and a resolution of 10 x 10 m². This is followed by grids of 2.4 x 2.4 km² and 20 x 20 m², 4.8 x 4.8 km² and 40 x 40 m², and finally the coarsest grid of 9.6 x 9.6 km² and 80 x 80 m². Centre of the calculation area is the stack of the NPP Braidwood.

### 3.2.2 Role of Topography

The surrounding area of the NPP is nearly flat. Topography plays a minor role and is therefore not taken into account for the dispersion calculations.

### 3.2.3 Building Effects, Plume Rise and Effective Release Height

Larger buildings of the power plant can significantly influence the dispersion of tritium near the source. On the one hand, they can lead to a reduction in the effective release height and, on the other hand, they can reduce the concentration due to increased turbulence in the vicinity of the building and the resulting greater amount of mixing. These effects decrease with increasing distance from the buildings and source.

In the present study, the 3D wind field near the source is not explicitly modeled for the following reasons:

- The four locations of particular interest are already 500 m or 1 km away from the source (see chapter 4.2).
- The objective of the study is the contamination of the ground with tritum due to its washing out by rain. For the calculation of the washout rate an integration over the entire height of the propagation plume is carried out. Therefore the effective height of the release is less important than for the calculation of the concentration near the ground.

However, the influence of the buildings on the release height cannot be completely excluded. In addition, at higher wind speeds downwash can lead to a lower effective release height. Therefore, an effective release height of 33 m is assumed, which is half of the stack height. This is a rather conservative estimate for the present situation.

A possible increase of the effective source height by plume rise is not considered.

### 3.3 Emission data

Emission data are taken from:

*Braidwood Nuclear Power Station, Annual Radioactive Effluent Release Report for 2017, Unit 1, 2 and ISFSI (Docket Numbers 50-456, 50-457 and 72-73).*

With regard to emissions, a distinction must be made between continuous releases and releases in batch operation. Batch operation only occurs at certain times. The values of the gaseous tritium releases per quarter and the annual sum provided by the licensee with regard to the continuous releases and the releases in batch operation are given in Table 1.

**Table 1: Gaseous continuous and batch release of Tritium for 2017**

| | 1st Qtr | 2nd Qtr | 3rd Qtr | 4th Qtr | Year |
|---|---|---|---|---|---|
| Continuous release in Ci | 26,9 | 140 | 45,3 | 23,6 | 235,8 |
| Batch release in Ci | 1,86 | 3,56 | 3,91 | 37 | 46,33 |
| Total release in Ci | 28,8 | 144 | 49,2 | 60,6 | 282,6 |
| Portion Batch/Total in % | 6,5 | 2,5 | 8,0 | 61,1 | 16,4 |
| Relation Batch/Continuous in % | 6,9 | 2,5 | 8,6 | 156,8 | 19,7 |

In addition to the above data, the proportion of batch releases to total releases and the ratio of batch releases to continuous releases are given. The portion of batch releases to total releases during the first three quarters is between 2.5 % and 8 %. In the fourth quarter, however, their portion is 61.6% and more than 1.5 times higher than the continuous releases. The absolute level of releases is highest in the second quarter and thus 2.5 to 5 times higher than in the other

quarters. The proportion of releases in this quarter makes up about 50% of the total year's releases.

Releases in batch mode only occur at certain times, and it is not known when the individual periods occur or how long they continue. The data that the licensee provides for the batch operations are: The number of batch releases, their total duration and the durations of the shortest and the longest period.

The data are given in Table 2.

**Table 2: Some characteristic data of gaseous batch releases of Tritium for 2017**

| | 1st Qtr | 2nd Qtr | 3rd Qtr | 4th Qtr | Year |
|---|---|---|---|---|---|
| Total Number of Batch Releases | 52 | 62 | 49 | 70 | 233 |
| Total Time Period for Batch Releases in minutes | 8410 | 37800 | 13800 | 37400 | 97410 |
| Maximum Time Period for a Batch Release in minutes | 1510 | 12800 | 2300 | 2190 | 12800 |
| Average Time Period for a Batch Release in minutes | 162 | 610 | 282 | 534 | 418 |
| Minimum Time Period for a Batch Release in minutes | 19 | 22 | 23 | 22 | 19 |

The model used for the dispersion calculations expects as input beside a meteorological time series (see next chapter) a release rate (in Bq/s) averaged over the studied period. This approach corresponds to the situation prevalent in Germany where the gaseous releases of a nuclear plant are usually uniform. The associated regulations specify the conditions under which a release may be regarded as uniform. If there is no uniform release in an individual case, either a conservative result is estimated with the aid of factors or individual analyses are performed. In the case of discontinuous releases, it would in principle also be possible to apply the dispersion model with a time-dependent release series (corresponding to the time-dependent meteorological time series). For such an approach, however, the exact course of the release must be known.

In the case of the nuclear power plant in Braidwood, there is a continuous release, which can be assumed to be uniform and discontinuous releases in batch mode. Only the frequency, the total duration and the total release are known about the batch releases, but not when the individual releases occur and how long they continue in individual cases.

In this case, the following approach is applied in order to perform dispersion calculations and obtain reliable results:

- It is assumed that the batch release rate is the same for all batch releases (mean release rate in batch mode).
- This release rate results from the total emissions of all batch releases in relation to the total duration of all batch releases.
- Thus, a release rate due to continuous release and a release rate due to batch releases are obtained for the dispersion calculation.

With these prerequisites, the data specified in the last two lines of Table 3 results. The table contains also the relevant data for release rates: The total values of continuous releases, of releases in batch mode, of the quarterly hours and the duration of batch modes.

**Table 3: Continuous release rates and mean release rate during batch mode of gaseous Tritium for 2017**

| | 1st Qtr | 2nd Qtr | 3rd Qtr | 4th Qtr | Year |
|---|---|---|---|---|---|
| Continuous release in Ci | 26,9 | 140,0 | 45,3 | 23,6 | 235,8 |
| Batch release in Ci | 1,86 | 3,56 | 3,91 | 37,00 | 46,3 |
| Total time of period in hour | 2160 | 2184 | 2208 | 2208 | 8760 |
| Total time batch releases in hour | 140 | 630 | 230 | 623 | 1624 |
| Continuous release rate in nCi/s | 3459 | 17806 | 5699 | 2969 | 7477 |
| Mean release rate in batch mode in nCi/s | 3686 | 1570 | 4722 | 16488 | 7927 |

Because the batch releases cannot be allocated to a certain time period, the specified release rate is assumed for all periods of batch mode. On the basis of these data, the resulting tritium input with the rainwater can be calculated caused by the emissions of the power plant. The following must be noted:

- At which location at a certain time how many tritium is deposited with the rain depends not only on the release rate but also on the wind direction and rain intensity.
- The impact of tritium with rainwater due to the continuous releases is uniform at all times. In this sense, the calculated values represent the actual contamination by the continuous emissions.

- The total impact of tritium with rainwater due to discontinuous releases is calculated too high, since the release rate is used for the entire period and therefore the total release applied for the calculation is higher than the actual one. The smaller the batch periods and the batch releases, the greater the overestimation.

## 3.4 Meteorological Data

The quality of dispersion calculations strongly depends on the quality of the dispersion model used and the meteorological input parameters. Therefore, the analysis of meteorological data representative for the site is very important for fixing the model options for the calculations. The required parameters are wind speed, wind direction, atmospheric stability, mixing layer heights, temperature and precipitation. These parameters should be available at the site or near the site in high temporal resolution (1-h-values) for a desired time period. The existence of a representative meteorological time series at the site is a precondition for the calculation of long-term (yearly averaged) as well as short-term (max. daily, max. hourly) pollution concentrations and deposition.

### 3.4.1 Meteorological data basis

The dispersion calculations at the Braidwood site (41.264°N, 88.218°W) are to be carried out with the ARTM model for the year 2015. The dispersion model requires a seasonal series with hourly values of wind speed, wind direction, precipitation rate and dispersion class according to Klug / Manier. The determination of the dispersion class according to [2] depends on the following input variables:

- date and time
- location coordinates for determining the times of sunrise and sunset
- roughness length z0 at site
- wind speed
- cloud coverage in eighths
- type of coverage, if applicable

No spatially representative measurement data for the model area are available from the immediate vicinity of the site. However, at the National Climatic Data Center at gis.ncdc.noaa.gov/maps/ncei/cdo/hourly hourly observation data of various measurement networks are available for download. Data from the following measuring stations in the Braidwood regional area were tested for their suitability and representativeness for the Braidwood site:

**Table 4: Measuring stations in the Braidwood regional area which were tested for their suitability and representativeness for the Braidwood site**

| Location | NCDC code | Coordinates | Distance Direction | Wind | Rain | Cloud Cover |
|---|---|---|---|---|---|---|
| Mors Muni-J.R. | 04867 | 88.419°W 41.425°N | ~25 km NW | X | - | (-) |
| Greater Kankakee Airport | 04880 | 87.846°W 41.121°N | ~35 km ESE | X | X | (-) |
| Lansing Municipal AP | 04879 | 87.532° W 41.540 N | ~65 km NE | X | X | (-) |
| Pontiac Municipal Airport | 04889 | 88.625° W 40.924° N | ~51 km SW | X | X | (-) |
| Joliet Regional Airport | 14834 | 88.167° W 41.500° N | ~26 km N | X | X | (-) |
| Lewis University Airport | 04831 | 88.085° W 41.604° N | ~39 km NNE | X | X | (-) |

There are no hourly precipitation rates at the Mors Muni site. The degree of coverage at all stations is indicated in only five classes of eighths (0, 1-2, 3-4, 5-7 and 8).

### 3.4.2 Preparation of NCDC data

The times of the NCDC data are related to UTC (coordinated universal time). For ARTM a time series was generated for the period Jan 01, 2015 00:00 to Dec 31, 2015 23:00 Chicago time (UTC - 6 hours). The time specifications in the compiled time series refer to the beginning of the hour intervals.

The averaging period of the measured wind data is unknown. However, the wind measurements are not available as hourly averages. All measurement data with a quality code of 0, 1, 4, 5 or 9 were accepted, all other data discarded. Calms and variable wind directions were coded according to the AKTERM definition [3].

From the precipitation data, measurements with a condition code 1 were initially rejected as faulty. The remaining precipitation data are available in mm, related to different time intervals. They were harmonized in successive hourly values (mm/h).

Data on cloud cover were only used for quality codes of 0, 1, 4, 5 or 9, otherwise marked as not defined. The five classes were converted to eighths according to the following table:

**Table 5: Assignment of the measured five cloud cover classes to the eight classes used for the simulations**

| measured (eighths) | compiled (eighths) |
|---|---|
| 0 | 0 |
| 1-2 | 2 |
| 3-4 | 4 |
| 5-7 | 6 |
| 8 | 8 |

The sunrise and sunset times according to [2] were calculated for each day from the location coordinates. Together with wind speed and degree of coverage, the hourly dispersion categories were determined according to Klug/Manier.

### 3.4.3 Spatial representativeness

Figure 1 shows the approximate spatial distribution of the wind roses of the six stations generated from the 2015 measurement data. All wind roses show large frequencies of wind directions from the third quadrant between south and west and a secondary maximum from northeastern directions. The northeast maximum is more pronounced the closer you get to Lake Michigan. It will therefore be associated with sea winds, which form during sunny weather conditions and can reach up to 100 km inland in the course of the day. The opposite nocturnal land breeze is usually less pronounced. It does not stand out from the frequent southwesterly winds.

Braidwood is about 75 km from Lake Michigan. Similar to the Mors Muni station, which is about at the same distance, the land-sea wind will play a much weaker role here. The more pronounced sea wind at the stations Lansing, Lewis and Joliet, as well as the isolated NE maximum at Greater Kankakee seem less characteristic for Braidwood.

Mors Muni, Pontiac, and Greater Kankakee all show a wide distribution of frequent winds from south to west. However, the frequencies within this quadrant are very irregularly distributed at Mors Muni and show several singularities in 10° sectors. It cannot be ruled out that local influences may be responsible for this.

Pontiac, on the other hand, shows a more even distribution of wind directions in the third quadrant than Mors Muni and a less pronounced secondary maximum from the northeast. Although the station is, except for Lansing, furthermost from Braidwood (50 km distance), the wind conditions measured there seem to be most representative for the conditions around Braidwood. The dispersion calculations are therefore carried out with the processed measurement data of Pontiac.

Figure 1 shows the compiled measurement data of Pontiac station for the year 2015 in the form of a wind rose and as frequency distributions over nine wind speed classes and six dispersion

classes respectively. A roughness length of 0.10 m and an anemometer height of 10 m were assumed for the compilation of the measurement data.

The annual average wind speed is 4.2 m/s. Therefore, the neutral dispersion class III/1 predominates with about 44% of the annual hours. Stable stratification conditions (classes I and II) are existent in almost ⅓ of the annual hours. Neutral to unstable conditions exist in ¼ of the annual hours.

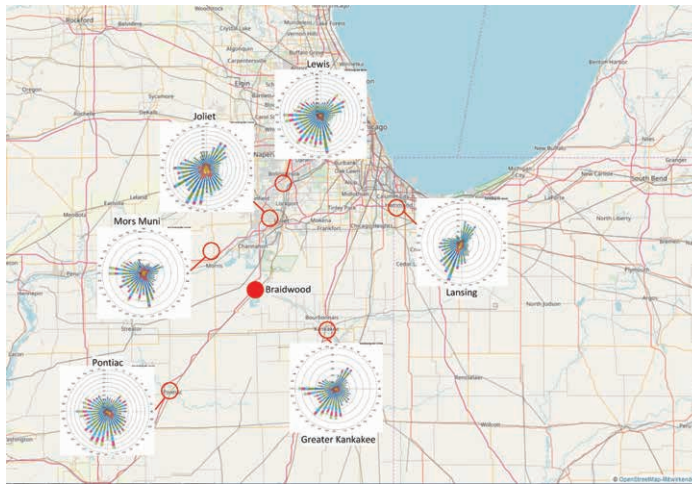


**Figure 1:** **approximate spatial distribution of the wind roses of the six stations generated from the 2015 measurement data.**

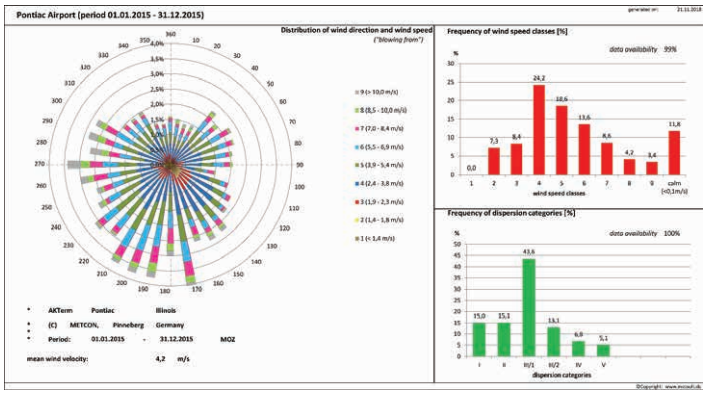


**Figure 2:** **compiled measurement data of Pontiac station for the year 2015 in the form of a wind rose and as frequency distributions.**

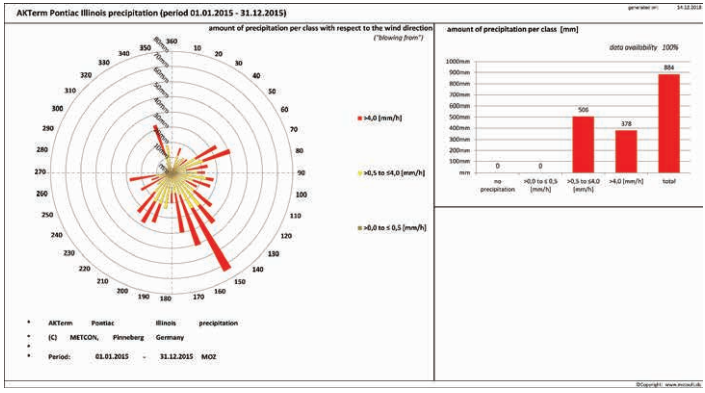


**Figure 3:** **Precipitation data of Pontiac station for the year 2015 in the form of a precipitation rose and a frequency distribution of precipitation intensity.**

# 4 Results from Dispersion Calculation

## 4.1 Mean Tritium Contamination in the Surrounding Area

The tritium contamination by precipitation due to the releases of the Braidwood nuclear power station in 2017 is shown. Results are presented for the four quarters as well as the total year and for the continuous release only as well as for the sum of continuous release and batch release.

In Figure 4 to Figure 43 of the appendix 1 the two-dimensional tritium contamination by precipitation due to the releases of the Braidwood power plant in 2017 is presented. The results are shown for each of the four calculation grids for the four quarters and the full year, for continuous release only, and for continuous release and batch release. This results in a total of 40 graphs. The legend is chosen in such a way that the entire value range of a calculation field is covered as far as possible. The values shown are the contamination in Ci/m².

Four assessment points (P1 to P4) are still marked in the graphics. These are located as follows in relation to the source:

- P1: 500 m southeast of the source: 135°
- P2: 1 km north of the source: 0°
- P3: 1 km north-northwest of the source: 330° and
- P4: 1 km north-northeast of the source: 30°.

When interpreting the results, the following should be noted:

- As far as continuous operation is concerned, the results should be considered as realistic.
- As far as the sum of continuous and batch release is concerned, there is an overestimation of the actual values as previously stated.
- At a single location, the actual impact is between zero (if the location for all actual batch releases was never affected due to wind direction and rain) and the calculated value (if the location for all actual batch releases was always affected due to wind direction and rain).

## 4.2 Daily Impact of Tritium at some Locations

Figure 44 to Figure 55 of the appendix 2 shows the maximum daily mean values to be expected for each month at the four assessment points. The daily average exposure is shown as a monthly time series for the sum of continuous and batch release.

The results are to be interpreted as follows:

- At a single location and on a specific day, the calculated values correspond approximately to the actual values if a batch release has occurred on that day and the assumed release rate

(mean value for all batch operations of the quarter) corresponds approximately to the real rate.

- However, the exact release rates of the individual batch operations are not known. The actual release rate of a batch mode can therefore be higher or lower than the assumed mean rate. The calculated values can therefore be correspondingly lower or higher than the calculated values.
- If no batch mode was actually performed on the corresponding day, the calculated values are too high by the portion of the assumed batch operation. The portion of batch mode operation compared to the portion of continuous emissions can be seen in Table 3-3. They are approximately the same in the first and third quarters. In the second quarter, the portion of batch mode is less than 10% of the portion of continuous operation. In the fourth quarter, the portion of batch mode is 5.5 times higher than that of continuous mode.

### 4.3 Tritium activity in rainwater

Until now, the contamination with tritium has been treated either as an integral value over a longer period of time or as daily values. A more detailed analysis also allows to obtain information about the activity of tritium in rainwater. The results of such an analysis depend on the regarded point and will be carried out in the following for the four already investigated locations.

From the results of the dispersion calculation, the hours can be determined for a given location during which a wet deposition with tritium occurred and consequently it rained at these locations. The criterium for this is a calculated wet deposition greater than 0.

A first result of such an analysis is the number of hours (hours of rain) per quarter and per point of interest as shown in Table 6. However, this analysis does not yet tell us how much precipitation has fallen and how much the tritium concentration in the air and subsequently in the precipitation is.

**Table 6: Number of rain hours at four assessment points (P1 to P4) per quarter and in total**

| | P1 | P2 | P3 | P4 |
|---|---|---|---|---|
| 1st Qtr | 5 | 13 | 6 | 11 |
| 2nd Qtr | 21 | 40 | 28 | 45 |
| 3rd Qtr | 16 | 27 | 32 | 29 |
| 4th Qtr | 2 | 40 | 52 | 27 |
| **Total** | **44** | **120** | **118** | **112** |

For each hour it is known how much precipitation was in that hour and how much tritium was deposited by wet deposition. Thus the activity of tritium in rainwater can be calculated. The following tables Table 7 to Table 1 show the results of such an analysis for each of the four points for the days on which at least one hour of precipitation occurred. In the tables are indicated: The date, the number of hours, the average tritium activity in the precipitation over these hours, the maximum tritium activity in these hours and the amount of precipitation.

**Table 7: Tritium activity in rainwater at assessment point P1 on days with precipitation**

Count: Number of hours with precipitation
Mean: Mean Tritium activity in nCi/l
Maximum: Maximum Tritium activity in nCi/l
p: Amount of precipitation in l/m²

| Date | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 1/3/2015 | 3 | 0.423 | 1.167 | 3.8 |
| 3/1/2015 | 2 | 1.558 | 2.117 | 3.1 |
| 4/19/2015 | 2 | 0.015 | 0.030 | 1.6 |
| 5/5/2015 | 1 | 0.002 | 0.002 | 1.3 |
| 5/8/2015 | 2 | 0.449 | 0.858 | 4.8 |
| 5/30/2015 | 4 | 0.425 | 1.291 | 6.1 |
| 6/7/2015 | 4 | 0.144 | 0.352 | 11.5 |
| 6/8/2015 | 1 | 1.125 | 1.125 | 2.8 |
| 6/11/2015 | 1 | 0.293 | 0.293 | 1 |
| 6/12/2015 | 1 | 0.004 | 0.004 | 2.8 |
| 6/13/2015 | 1 | 0.043 | 0.043 | 1.8 |
| 6/15/2015 | 2 | 0.033 | 0.062 | 5.6 |
| 6/25/2015 | 2 | 0.029 | 0.043 | 0.6 |
| 7/7/2015 | 1 | 0.036 | 0.036 | 1.5 |
| 7/9/2015 | 2 | 0.796 | 1.587 | 0.6 |
| 7/20/2015 | 1 | 0.091 | 0.091 | 7.4 |
| 7/29/2015 | 2 | 0.419 | 0.835 | 5.4 |
| 8/9/2015 | 3 | 1.319 | 3.692 | 8.2 |
| 8/18/2015 | 2 | 0.717 | 1.427 | 17 |
| 9/10/2015 | 1 | 0.002 | 0.002 | 9.1 |
| 9/11/2015 | 2 | 0.413 | 0.664 | 0.6 |
| 9/18/2015 | 2 | 0.144 | 0.222 | 8.4 |
| 11/22/2015 | 1 | 0.004 | 0.004 | 0.5 |
| 11/27/2015 | 1 | 0.022 | 0.022 | 1.5 |
| **Year** | **44** | **0.415** | **3.692** | **107** |

**Table 8: Tritium activity in rainwater at assessment point P2 on days with precipitation**

Count: Number of hours with precipitation
Mean: Mean Tritium activity in nCi/l
Maximum: Maximum Tritium activity in nCi/l
p: Amount of precipitation in l/m²

| Date | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 1/11/2015 | 1 | 0.002 | 0.002 | 0.3 |
| 1/31/2015 | 2 | 5.979 | 6.320 | 0.9 |
| 2/6/2015 | 1 | 1.975 | 1.975 | 0.5 |
| 2/7/2015 | 2 | 0.431 | 0.639 | 1.8 |
| 3/1/2015 | 2 | 0.599 | 0.949 | 3.3 |
| 3/3/2015 | 2 | 0.612 | 1.209 | 1.8 |
| 3/25/2015 | 1 | 0.090 | 0.090 | 2.3 |
| 3/29/2015 | 2 | 0.097 | 0.183 | 1.3 |
| 4/2/2015 | 1 | 0.030 | 0.030 | 1 |
| 4/9/2015 | 2 | 0.136 | 0.258 | 2.8 |
| 5/4/2015 | 2 | 0.103 | 0.161 | 3.8 |
| 5/5/2015 | 1 | 0.011 | 0.011 | 0.2 |
| 5/8/2015 | 3 | 0.938 | 2.576 | 0.9 |
| 5/10/2015 | 2 | 0.039 | 0.047 | 1.8 |
| 5/11/2015 | 2 | 0.974 | 1.915 | 0.6 |
| 5/14/2015 | 2 | 0.328 | 0.634 | 1.6 |
| 5/16/2015 | 2 | 0.432 | 0.810 | 0.6 |
| 5/26/2015 | 2 | 0.429 | 0.817 | 8.1 |
| 5/29/2015 | 2 | 0.142 | 0.278 | 5.1 |
| 6/7/2015 | 5 | 0.164 | 0.415 | 12 |
| 6/11/2015 | 2 | 0.070 | 0.076 | 0.8 |
| 6/12/2015 | 3 | 0.055 | 0.137 | 17.9 |
| 6/13/2015 | 2 | 0.055 | 0.069 | 0.6 |
| 6/15/2015 | 1 | 0.031 | 0.031 | 4.1 |
| 6/20/2015 | 2 | 0.015 | 0.016 | 3.3 |
| 6/24/2015 | 1 | 0.371 | 0.371 | 2.5 |
| 6/25/2015 | 3 | 0.979 | 2.886 | 3.8 |
| 7/6/2015 | 1 | 0.996 | 0.996 | 15.7 |
| 7/8/2015 | 3 | 0.796 | 2.194 | 5.9 |
| 7/11/2015 | 2 | 0.163 | 0.318 | 8.6 |
| 7/13/2015 | 1 | 0.036 | 0.036 | 5.3 |
| 7/16/2015 | 3 | 0.546 | 1.512 | 4.9 |
| 7/29/2015 | 2 | 0.071 | 0.098 | 2.1 |
| 8/9/2015 | 2 | 0.090 | 0.153 | 3.1 |
| 8/17/2015 | 3 | 0.701 | 1.709 | 8.2 |
| 8/18/2015 | 3 | 0.407 | 1.189 | 15.5 |

| Date | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 9/10/2015 | 1 | 0.007 | 0.007 | 0.8 |
| 9/11/2015 | 3 | 0.713 | 1.292 | 1.6 |
| 9/18/2015 | 3 | 0.173 | 0.474 | 6.2 |
| 10/21/2015 | 1 | 0.001 | 0.001 | 0.3 |
| 10/23/2015 | 2 | 1.006 | 1.091 | 2 |
| 10/31/2015 | 4 | 1.112 | 2.302 | 4.3 |
| 11/5/2015 | 2 | 0.930 | 1.838 | 0.6 |
| 11/11/2015 | 3 | 0.092 | 0.266 | 9.2 |
| 11/16/2015 | 1 | 0.001 | 0.001 | 0.5 |
| 11/17/2015 | 1 | 0.008 | 0.008 | 2 |
| 11/22/2015 | 1 | 1.853 | 1.853 | 0.5 |
| 11/25/2015 | 1 | 0.969 | 0.969 | 0.5 |
| 11/26/2015 | 3 | 1.091 | 1.800 | 2.3 |
| 11/27/2015 | 3 | 0.626 | 1.402 | 6.4 |
| 11/30/2015 | 1 | 0.064 | 0.064 | 0.3 |
| 12/1/2015 | 1 | 0.100 | 0.100 | 0.5 |
| 12/13/2015 | 4 | 0.495 | 1.772 | 2.9 |
| 12/14/2015 | 2 | 0.561 | 1.001 | 1.5 |
| 12/20/2015 | 1 | 0.034 | 0.034 | 0.5 |
| 12/21/2015 | 4 | 0.773 | 1.540 | 3.1 |
| 12/23/2015 | 4 | 0.839 | 1.451 | 14.3 |
| 12/28/2015 | 1 | 0.016 | 0.016 | 0.3 |
| **Year** | **120** | **0.568** | **6.320** | **213.6** |

**Table 9: Tritium activity in rainwater at assessment point P3 on days with precipitation**

Count: Number of hours with precipitation
Mean: Mean Tritium activity in nCi/l
Maximum: Maximum Tritium activity in nCi/l
p: Amount of precipitation in l/m²

| Date | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 1/13/2015 | 1 | 0.244 | 0.244 | 1.3 |
| 3/1/2015 | 2 | 0.428 | 0.607 | 3.3 |
| 3/24/2015 | 1 | 0.005 | 0.005 | 1.8 |
| 3/25/2015 | 2 | 0.338 | 0.664 | 2.6 |
| 4/8/2015 | 2 | 0.382 | 0.760 | 5.9 |
| 5/4/2015 | 3 | 0.938 | 1.748 | 4.8 |
| 5/5/2015 | 1 | 0.894 | 0.894 | 0.2 |
| 5/10/2015 | 1 | 0.021 | 0.021 | 1.3 |
| 5/14/2015 | 4 | 0.206 | 0.798 | 3.1 |
| 5/16/2015 | 2 | 0.027 | 0.053 | 0.6 |
| 5/26/2015 | 1 | 0.015 | 0.015 | 1.3 |
| 6/7/2015 | 3 | 0.302 | 0.661 | 10.7 |
| 6/11/2015 | 2 | 0.806 | 1.380 | 2.8 |
| 6/12/2015 | 3 | 0.058 | 0.114 | 17.9 |
| 6/13/2015 | 1 | 0.001 | 0.001 | 0.3 |
| 6/15/2015 | 1 | 0.001 | 0.001 | 4.1 |
| 6/20/2015 | 1 | 0.036 | 0.036 | 2.5 |
| 6/24/2015 | 1 | 2.330 | 2.330 | 2.5 |
| 6/25/2015 | 2 | 0.526 | 0.990 | 1.5 |
| 7/6/2015 | 1 | 0.030 | 0.030 | 15.7 |
| 7/8/2015 | 5 | 0.281 | 0.864 | 6.7 |
| 7/11/2015 | 6 | 0.469 | 1.053 | 51.2 |
| 7/12/2015 | 1 | 0.002 | 0.002 | 7.6 |
| 7/13/2015 | 1 | 0.016 | 0.016 | 5.3 |
| 7/26/2015 | 1 | 0.004 | 0.004 | 2 |
| 7/29/2015 | 1 | 0.003 | 0.003 | 1.8 |
| 8/9/2015 | 5 | 0.037 | 0.089 | 9 |
| 8/17/2015 | 2 | 0.221 | 0.404 | 7.9 |
| 8/18/2015 | 4 | 0.986 | 2.194 | 30.8 |
| 9/10/2015 | 1 | 0.005 | 0.005 | 0.8 |
| 9/11/2015 | 3 | 0.349 | 0.552 | 1.6 |
| 9/18/2015 | 1 | 0.005 | 0.005 | 0.3 |
| 10/23/2015 | 3 | 0.311 | 0.496 | 2.1 |
| 10/28/2015 | 3 | 0.691 | 2.056 | 1.6 |
| 10/31/2015 | 7 | 0.268 | 0.887 | 10.2 |
| 11/11/2015 | 2 | 0.080 | 0.160 | 2.3 |

| Date | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 11/16/2015 | 5 | 0.437 | 0.860 | 2.4 |
| 11/17/2015 | 11 | 0.165 | 0.506 | 24.6 |
| 11/18/2015 | 4 | 0.431 | 0.581 | 8.6 |
| 11/22/2015 | 1 | 0.018 | 0.018 | 0.5 |
| 11/30/2015 | 2 | 0.461 | 0.813 | 0.6 |
| 12/13/2015 | 8 | 0.325 | 0.811 | 9.2 |
| 12/16/2015 | 2 | 0.439 | 0.853 | 1 |
| 12/23/2015 | 2 | 0.252 | 0.501 | 8.2 |
| 12/26/2015 | 1 | 1.326 | 1.326 | 0.3 |
| 12/28/2015 | 1 | 0.005 | 0.005 | 0.3 |
| **Year** | **118** | **0.341** | **2.330** | **281.1** |

**Table 10: Tritium activity in rainwater at assessment point P4 on days with precipitation**

Count: Number of hours with precipitation
Mean: Mean Tritium activity in nCi/l
Maximum: Maximum Tritium activity in nCi/l
p: Amount of precipitation in l/m²

| Date | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 1/11/2015 | 1 | 0.816 | 0.816 | 0.3 |
| 1/31/2015 | 2 | 0.567 | 1.047 | 1.7 |
| 2/7/2015 | 3 | 0.343 | 1.019 | 2.3 |
| 3/1/2015 | 2 | 0.614 | 0.987 | 3.3 |
| 3/3/2015 | 2 | 0.156 | 0.210 | 2 |
| 3/29/2015 | 1 | 0.404 | 0.404 | 0.3 |
| 4/2/2015 | 2 | 0.212 | 0.327 | 1.8 |
| 4/9/2015 | 1 | 0.213 | 0.213 | 0.5 |
| 5/4/2015 | 1 | 0.155 | 0.155 | 2.5 |
| 5/8/2015 | 4 | 0.370 | 1.371 | 15.3 |
| 5/10/2015 | 2 | 0.438 | 0.829 | 1.8 |
| 5/11/2015 | 5 | 0.268 | 1.144 | 4.4 |
| 5/16/2015 | 1 | 0.048 | 0.048 | 0.3 |
| 5/25/2015 | 1 | 1.047 | 1.047 | 3 |
| 5/26/2015 | 1 | 0.033 | 0.033 | 7.6 |
| 5/29/2015 | 3 | 0.318 | 0.679 | 5.9 |
| 6/7/2015 | 8 | 0.263 | 1.452 | 27.7 |
| 6/11/2015 | 1 | 0.061 | 0.061 | 0.3 |
| 6/12/2015 | 2 | 0.584 | 1.164 | 15.3 |
| 6/13/2015 | 3 | 0.123 | 0.247 | 2.4 |
| 6/14/2015 | 1 | 0.036 | 0.036 | 1.3 |
| 6/15/2015 | 4 | 0.141 | 0.438 | 11 |
| 6/18/2015 | 1 | 1.110 | 1.110 | 2.5 |
| 6/20/2015 | 1 | 0.022 | 0.022 | 0.8 |
| 6/25/2015 | 3 | 1.062 | 1.718 | 4.8 |
| 7/6/2015 | 2 | 0.020 | 0.029 | 17.2 |
| 7/8/2015 | 1 | 0.024 | 0.024 | 0.5 |
| 7/13/2015 | 1 | 0.001 | 0.001 | 5.3 |
| 7/16/2015 | 2 | 0.163 | 0.300 | 4.1 |
| 7/20/2015 | 1 | 0.006 | 0.006 | 7.4 |
| 7/29/2015 | 2 | 8.072 | 16.100 | 2.1 |
| 8/9/2015 | 2 | 0.073 | 0.121 | 3.1 |
| 8/17/2015 | 3 | 0.052 | 0.081 | 18.8 |
| 8/18/2015 | 2 | 0.314 | 0.622 | 5.1 |
| 9/8/2015 | 4 | 0.943 | 2.928 | 16.2 |
| 9/11/2015 | 3 | 0.673 | 1.163 | 1.6 |

| Date | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 9/18/2015 | 5 | 0.456 | 1.719 | 24 |
| 9/19/2015 | 1 | 2.811 | 2.811 | 2.5 |
| 10/20/2015 | 1 | 1.110 | 1.110 | 0.8 |
| 10/21/2015 | 2 | 0.006 | 0.008 | 0.6 |
| 10/24/2015 | 1 | 0.001 | 0.001 | 1 |
| 10/28/2015 | 1 | 0.087 | 0.087 | 0.5 |
| 10/31/2015 | 1 | 0.002 | 0.002 | 1.5 |
| 11/6/2015 | 1 | 0.010 | 0.010 | 4.6 |
| 11/11/2015 | 3 | 0.008 | 0.016 | 10.7 |
| 11/22/2015 | 1 | 0.291 | 0.291 | 0.5 |
| 11/23/2015 | 1 | 1.162 | 1.162 | 0.3 |
| 11/27/2015 | 1 | 0.031 | 0.031 | 3.6 |
| 12/1/2015 | 2 | 0.827 | 1.441 | 1 |
| 12/14/2015 | 4 | 0.011 | 0.033 | 1.4 |
| 12/21/2015 | 7 | 0.034 | 0.175 | 7.7 |
| **Year** | **112** | **0.476** | **16.100** | **261.5** |

The following tables Table 11 to Table 14 show an analogous analysis, however not related to days but to months. The contents of the table are the same as the previous ones: The month, the number of hours, the average tritium activity in precipitation over these hours, the maximum tritium activity in these hours and the amount of precipitation.

In addition to these tables, the average Tritium activity, the maximum Tritium activity and the number of rainy hours per month for each assessment point are shown graphically in the figures Figure 56 to Figure 59 in Appendix 3.

**Table 11: Tritium activity in rainwater at assessment point P1 per month**

Count: Number of hours with precipitation
Mean: Mean Tritium activity in nCi/l
Maximum: Maximum Tritium activity in nCi/l
p: Amount of precipitation in l/m²

| Month | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 1 | 3 | 0.423 | 1.167 | 3.8 |
| 2 | 0 | | | |
| 3 | 2 | 1.558 | 2.117 | 3.1 |
| 4 | 2 | 0.015 | 0.030 | 1.6 |
| 5 | 7 | 0.371 | 1.291 | 12.2 |
| 6 | 12 | 0.181 | 1.125 | 26.1 |
| 7 | 6 | 0.426 | 1.587 | 14.9 |
| 8 | 5 | 1.078 | 3.692 | 25.2 |
| 9 | 5 | 0.224 | 0.664 | 18.1 |
| 10 | 0 | | | |
| 11 | 2 | 0.013 | 0.022 | 2 |
| 12 | 0 | | | |
| **Year** | **44** | **0.415** | **3.692** | **107** |

**Table 12: Tritium activity in rainwater at assessment point P2 per month**

Count: Number of hours with precipitation
Mean: Mean Tritium activity in nCi/l
Maximum: Maximum Tritium activity in nCi/l
p: Amount of precipitation in l/m²

| Month | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 1 | 3 | 3.987 | 6.320 | 1.2 |
| 2 | 3 | 0.946 | 1.975 | 2.3 |
| 3 | 7 | 0.387 | 1.209 | 8.7 |
| 4 | 3 | 0.101 | 0.258 | 3.8 |
| 5 | 18 | 0.429 | 2.576 | 22.7 |
| 6 | 19 | 0.242 | 2.886 | 45 |
| 7 | 12 | 0.460 | 2.194 | 42.5 |
| 8 | 8 | 0.438 | 1.709 | 26.8 |
| 9 | 7 | 0.381 | 1.292 | 8.6 |
| 10 | 7 | 0.923 | 2.302 | 6.6 |
| 11 | 16 | 0.636 | 1.853 | 22.3 |
| 12 | 17 | 0.571 | 1.772 | 23.1 |
| **Year** | **120** | **0.568** | **6.320** | **213.6** |

**Table 13: Tritium activity in rainwater at assessment point P3 per month**

Count: Number of hours with precipitation
Mean: Mean Tritium activity in nCi/l
Maximum: Maximum Tritium activity in nCi/l
p: Amount of precipitation in l/m²

| Month | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 1 | 1 | 0.244 | 0.244 | 1.3 |
| 2 | 0 | | | |
| 3 | 5 | 0.307 | 0.664 | 7.7 |
| 4 | 2 | 0.382 | 0.760 | 5.9 |
| 5 | 12 | 0.385 | 1.748 | 11.3 |
| 6 | 14 | 0.437 | 2.330 | 42.3 |
| 7 | 16 | 0.267 | 1.053 | 90.3 |
| 8 | 11 | 0.415 | 2.194 | 47.7 |
| 9 | 5 | 0.211 | 0.552 | 2.7 |
| 10 | 13 | 0.375 | 2.056 | 13.9 |
| 11 | 25 | 0.273 | 0.860 | 39 |
| 12 | 14 | 0.379 | 1.326 | 19 |
| **Year** | **118** | **0.341** | **2.330** | **281.1** |

**Table 14: Tritium activity in rainwater at assessment point P4 per month**

Count: Number of hours with precipitation
Mean: Mean Tritium activity in nCi/l
Maximum: Maximum Tritium activity in nCi/l
p: Amount of precipitation in l/m²

| Month | Count | Mean [nCi/l] | Maximum [nCi/l] | p [l/m²] |
|---|---|---|---|---|
| 1 | 3 | 0.650 | 1.047 | 2 |
| 2 | 3 | 0.343 | 1.019 | 2.3 |
| 3 | 5 | 0.389 | 0.987 | 5.6 |
| 4 | 3 | 0.212 | 0.327 | 2.3 |
| 5 | 18 | 0.329 | 1.371 | 40.8 |
| 6 | 24 | 0.359 | 1.718 | 66.1 |
| 7 | 9 | 1.838 | 16.100 | 36.6 |
| 8 | 7 | 0.133 | 0.622 | 27 |
| 9 | 13 | 0.837 | 2.928 | 44.3 |
| 10 | 6 | 0.202 | 1.110 | 4.4 |
| 11 | 7 | 0.217 | 1.162 | 19.7 |
| 12 | 14 | 0.155 | 1.441 | 10.4 |
| **Year** | **112** | **0.476** | **16.100** | **261.5** |

## 5 Reference

[1] BMU (2007): ARTM, Entwicklung, Validierung und Bereitstellung eines atmosphärischen Ausbreitungsmodells für luftgetragene radioaktive Stoffe auf der Basis des Ausbreitungsmodells AUSTAL2000 der neuen TA Luft", GRS mbH, Köln.

[2] VDI (2017): Environmental meteorology - Atmospheric dispersion models - Determination of Klug/Manier dispersion categories. VDI Standard 3782 Blatt 6, Beuth-Verlag, Berlin.

[3] VDI (2017): Environmental meteorology - Quality assurance of meteorological data for dispersion calculation according to TA Luft and GIRL. VDI-Richtlinie 3783 Blatt 21, Beuth-Verlag, Berlin.

**Appendix 1: Two-dimensional contamination of Tritium**

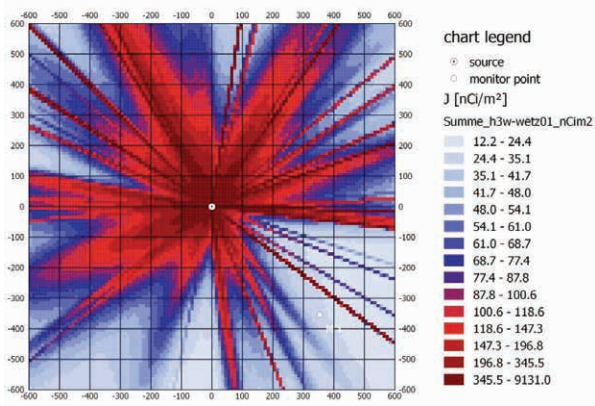


**Figure 4: Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the full year; calculation grid 1.**

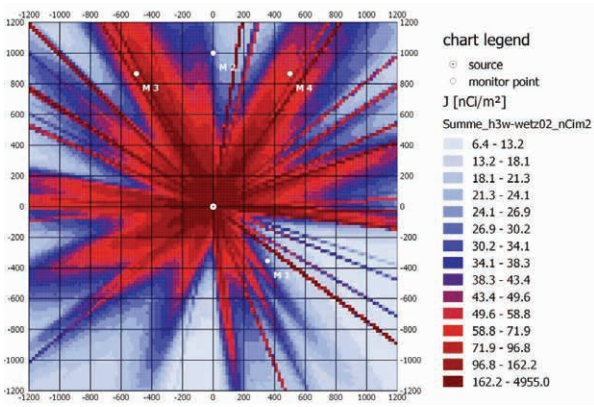


**Figure 5: Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the full year; calculation grid 2.**

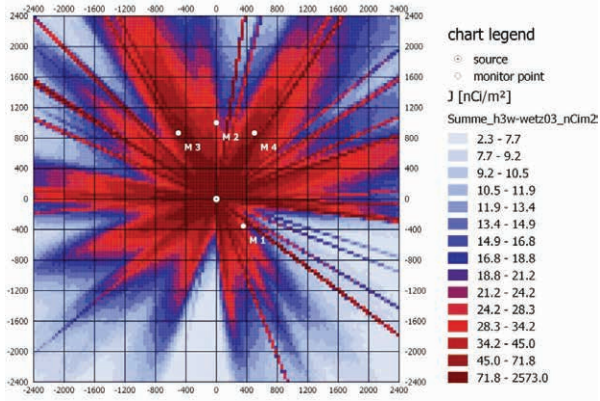


**Figure 6:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the full year; calculation grid 3.**

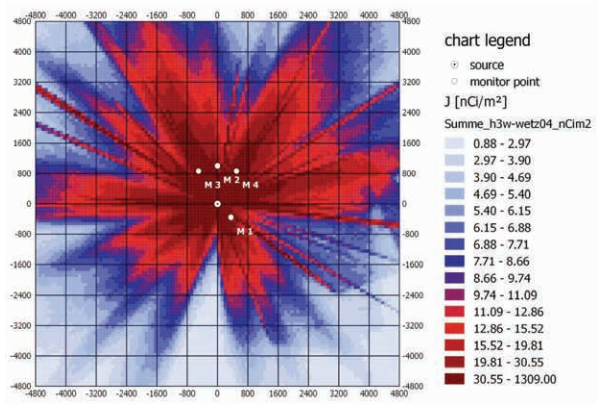


**Figure 7:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the full year; calculation grid 4.**

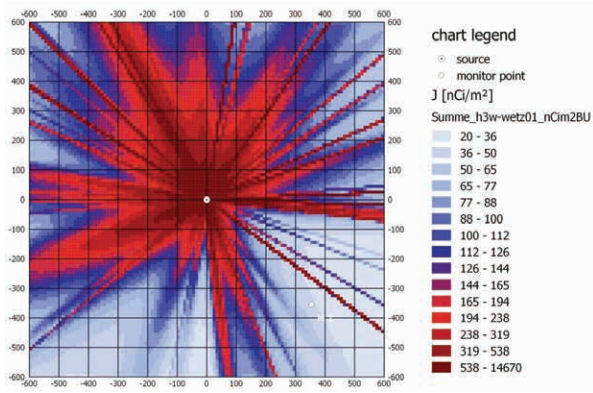


**Figure 8:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the full year; calculation grid 1.**

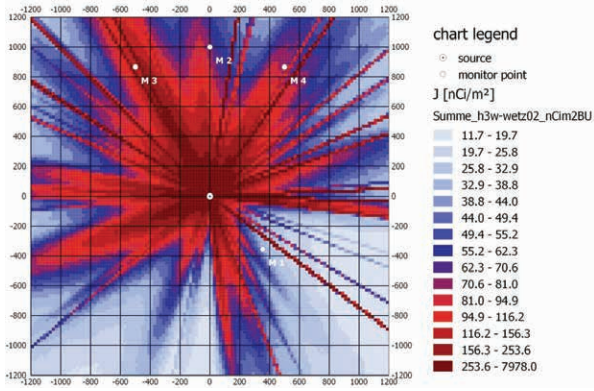


**Figure 9:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the full year; calculation grid 2.**

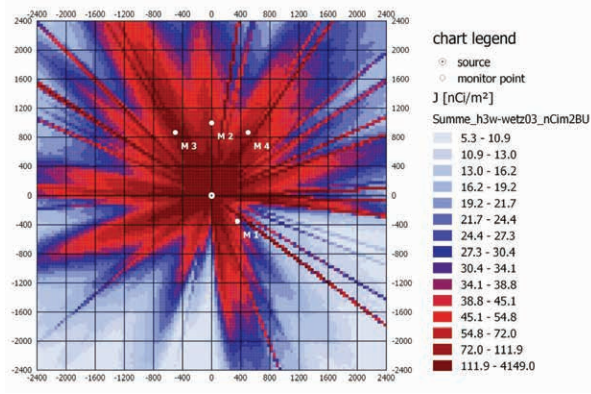


**Figure 10:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the full year; calculation grid 3.**

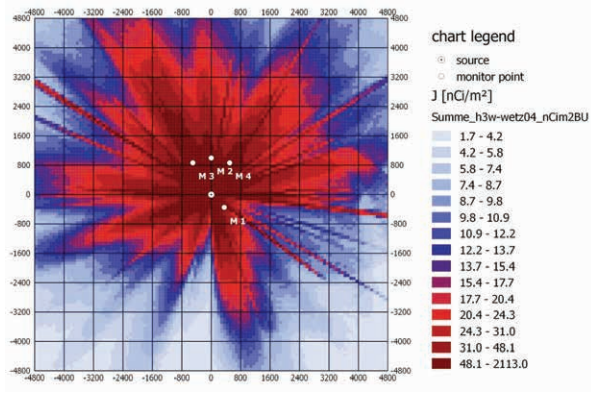


**Figure 11:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the full year; calculation grid 4.**

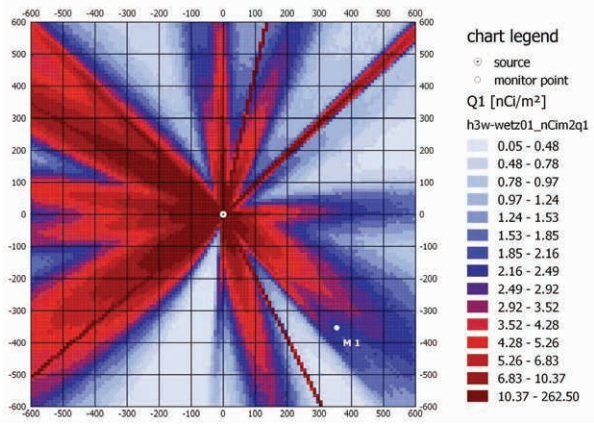


**Figure 12: Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the first quarter; calculation grid 1.**

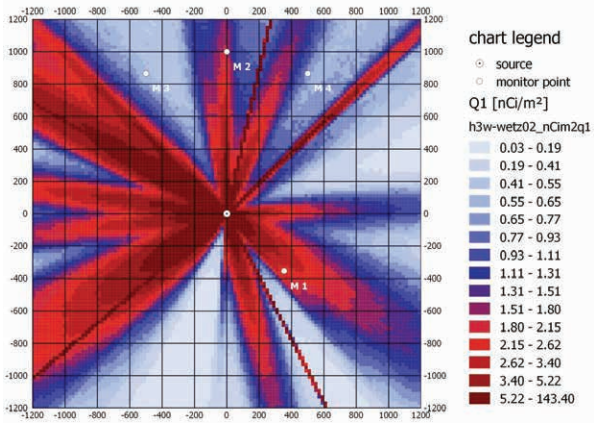


**Figure 13: Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the first quarter; calculation grid 2.**

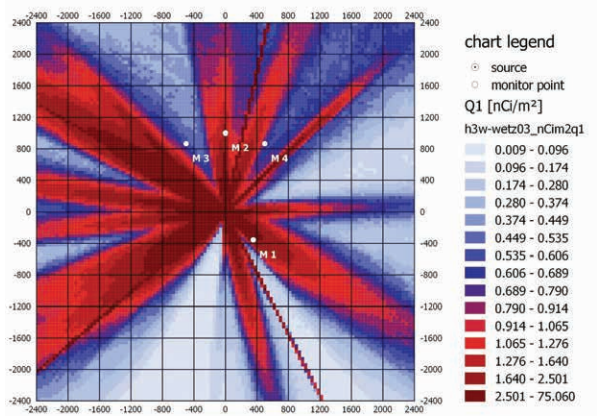


**Figure 14:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the first quarter; calculation grid 3.**

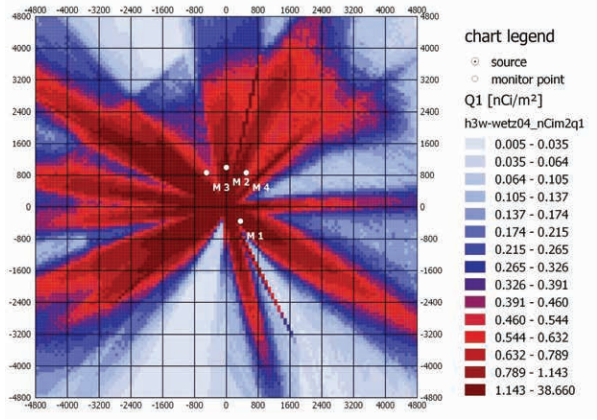


**Figure 15:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the first quarter; calculation grid 4.**

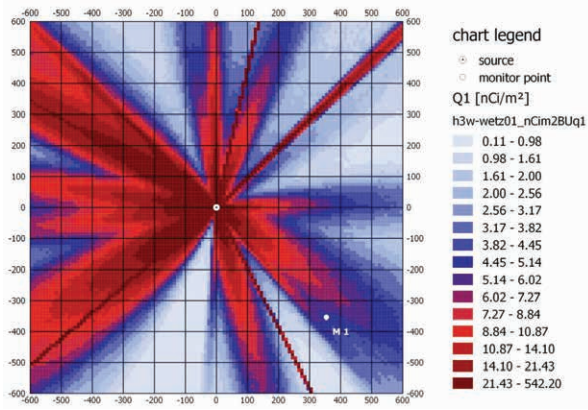


**Figure 16: Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the first quarter; calculation grid 1…4.**

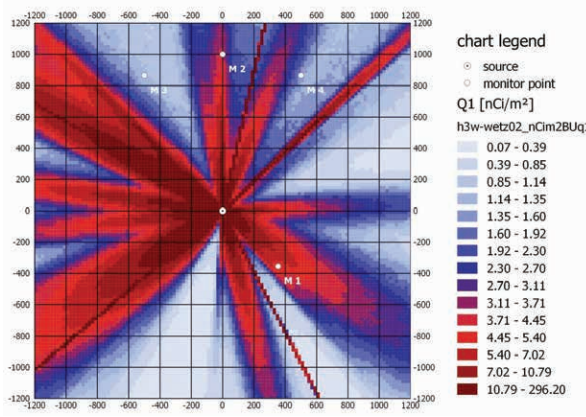


**Figure** 17**: Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the first quarter; calculation grid 2.**

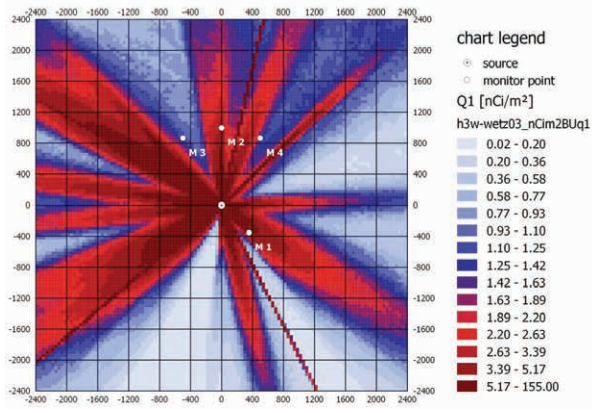


**Figure 18:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the first quarter; calculation grid 3.**

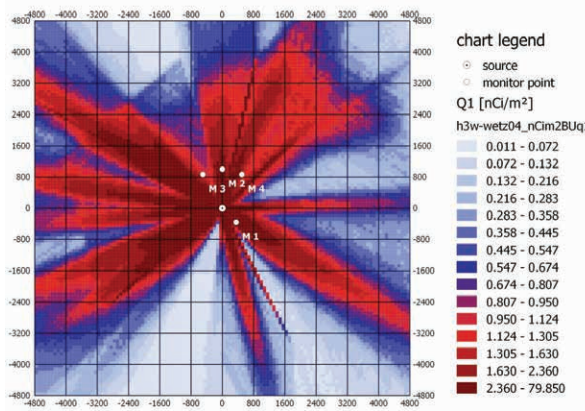


**Figure 19:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the first quarter; calculation grid 4.**

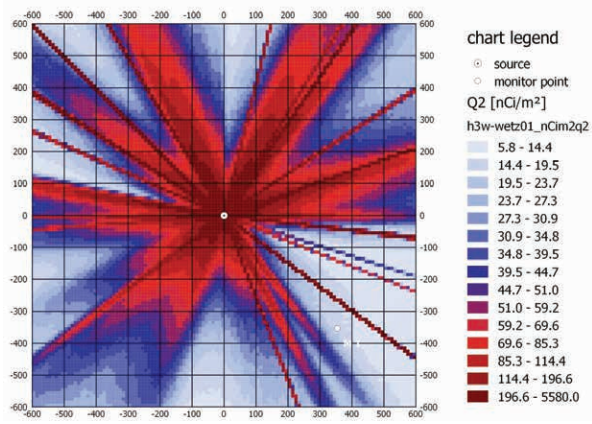


**Figure 20:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the second quarter; calculation grid 1.**

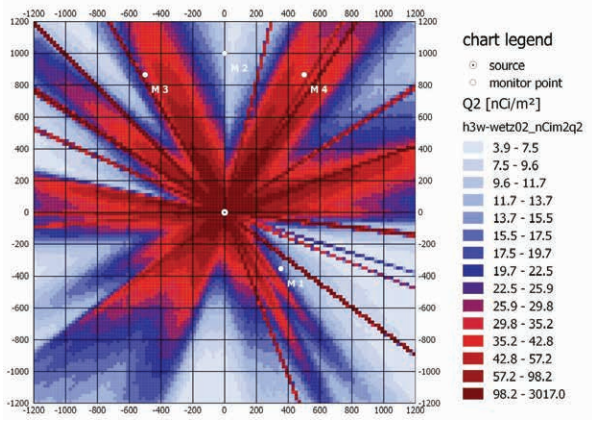


**Figure 21:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the second quarter; calculation grid 2.**

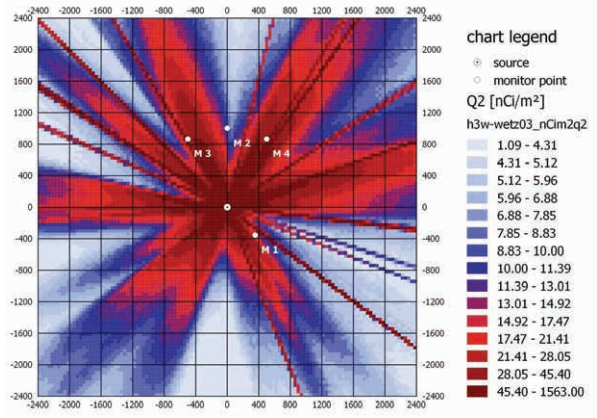


**Figure 22:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the second quarter; calculation grid 3.**

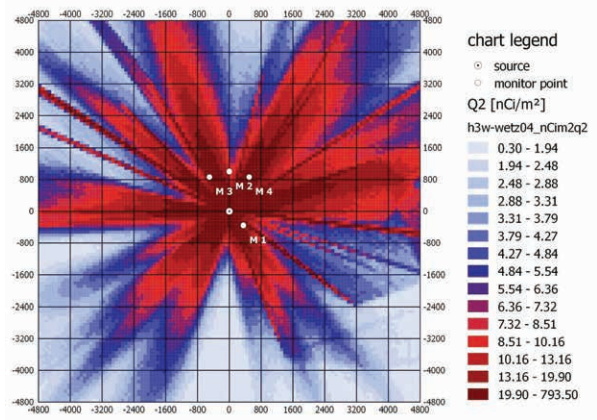


**Figure 23:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the second quarter; calculation grid 4.**

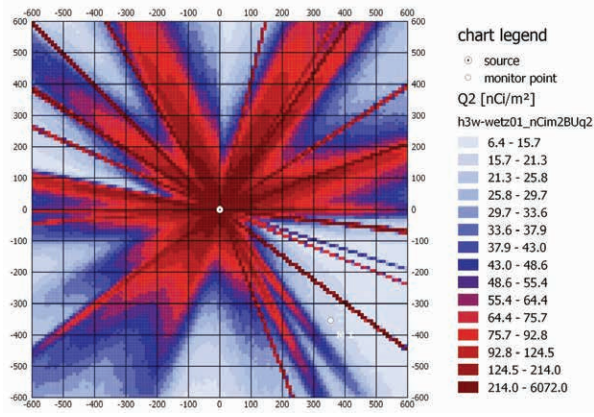


**Figure 24:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the second quarter; calculation grid 1.**

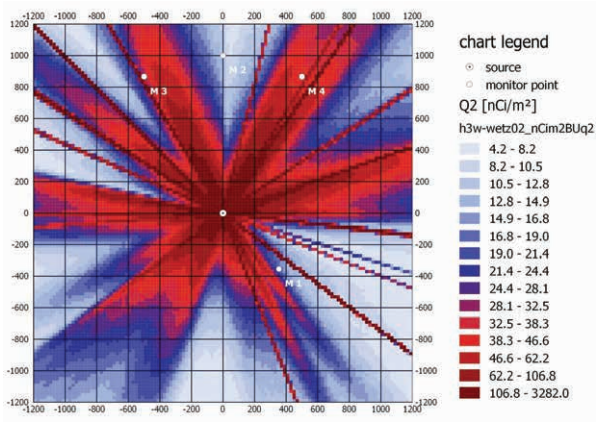


**Figure 25:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the second quarter; calculation grid 2.**

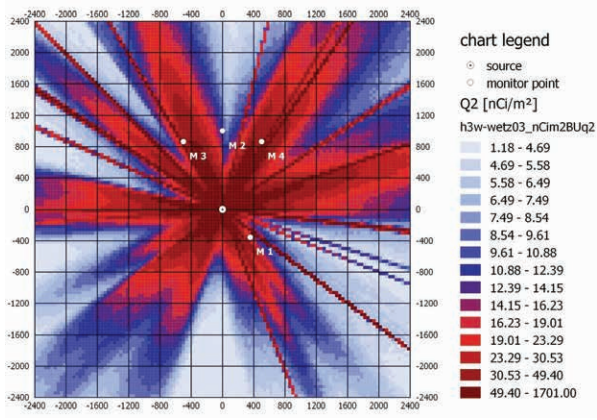


**Figure 26:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the second quarter; calculation grid 3.**

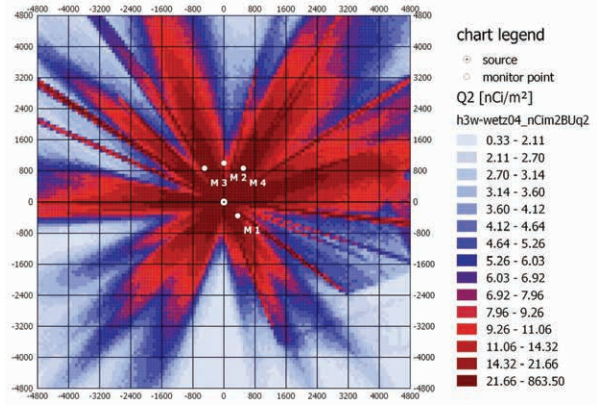


**Figure 27:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the second quarter; calculation grid 4.**

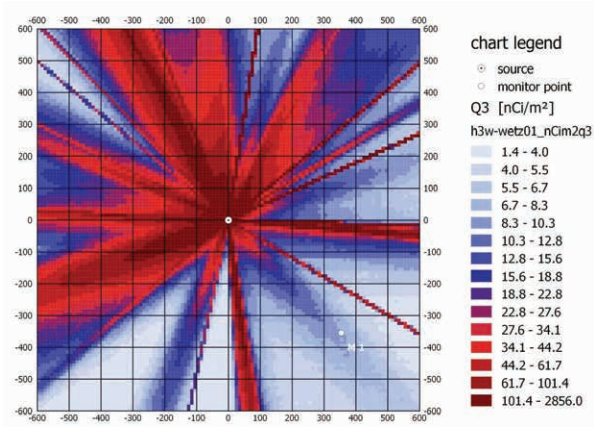


**Figure 28: Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the third quarter; calculation grid 1.**

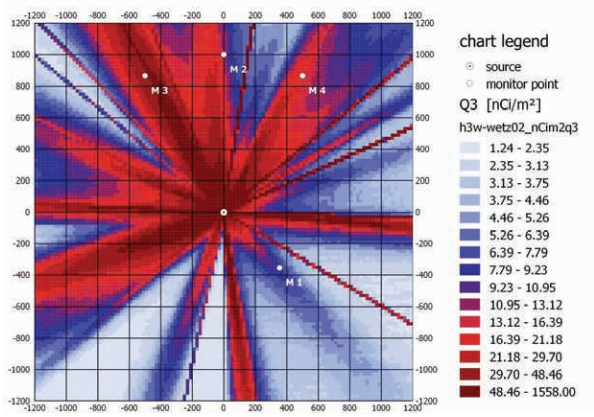


**Figure 29: Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the third quarter; calculation grid 2.**

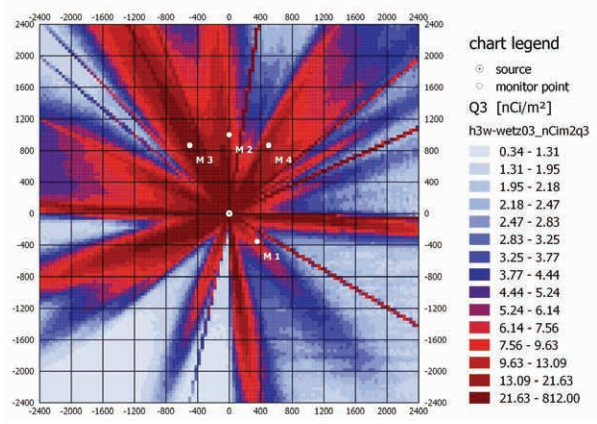


**Figure 30:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the third quarter; calculation grid 3.**

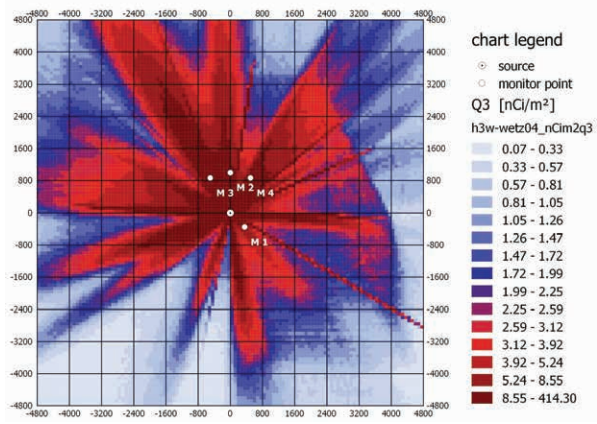


**Figure 31:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the third quarter; calculation grid 4.**

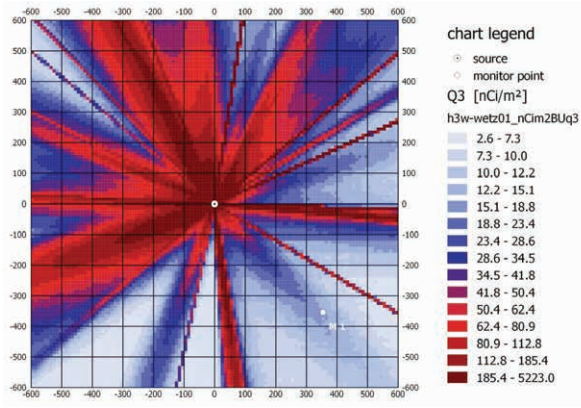


**Figure 32:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the third quarter; calculation grid 1.**

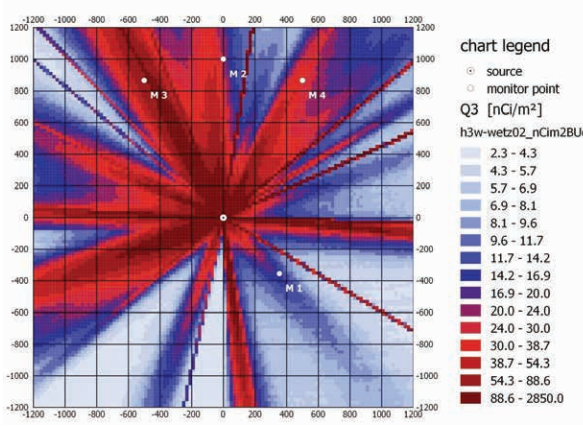


**Figure 33:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the third quarter; calculation grid 2.**

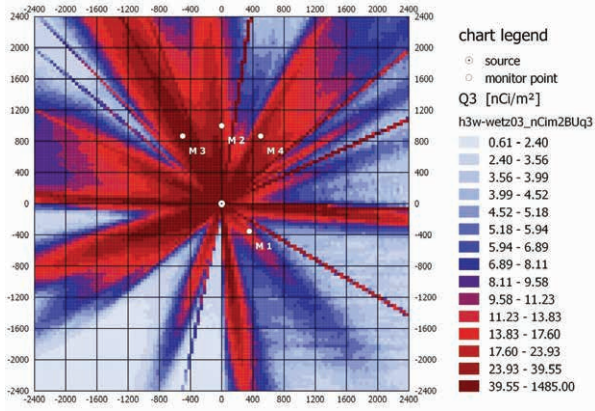


**Figure 34:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the third quarter; calculation grid 3.**

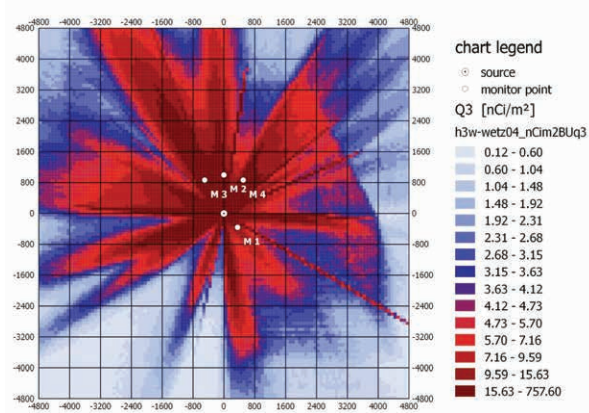


**Figure 35:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the third quarter; calculation grid 4.**

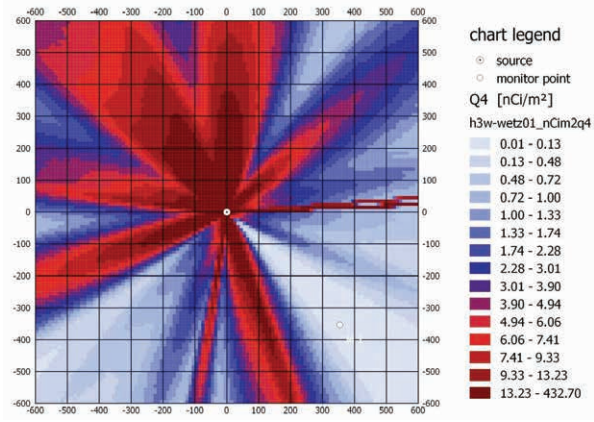


**Figure 36: Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the fourth quarter; calculation grid 1.**

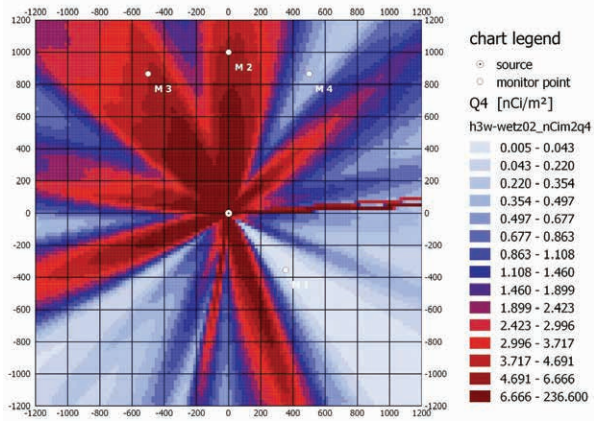


**Figure 37: Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the fourth quarter; calculation grid 2.**

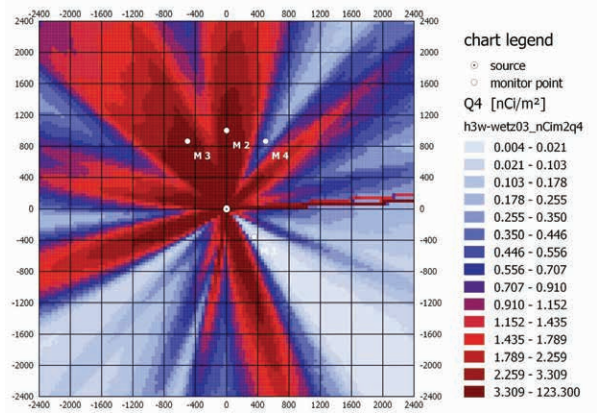


**Figure 38:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the fourth quarter; calculation grid 3.**

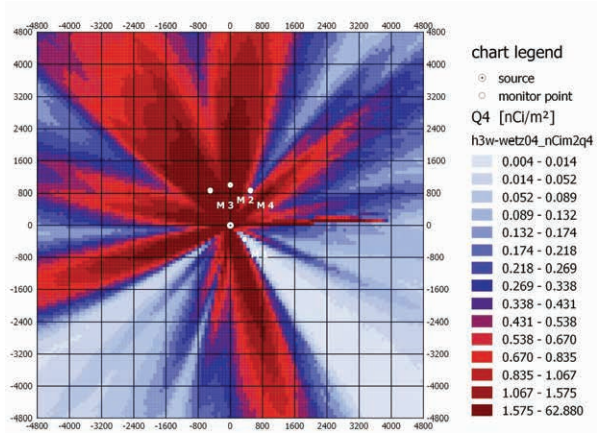


**Figure 39:** **Continuous release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the fourth quarter; calculation grid 4.**

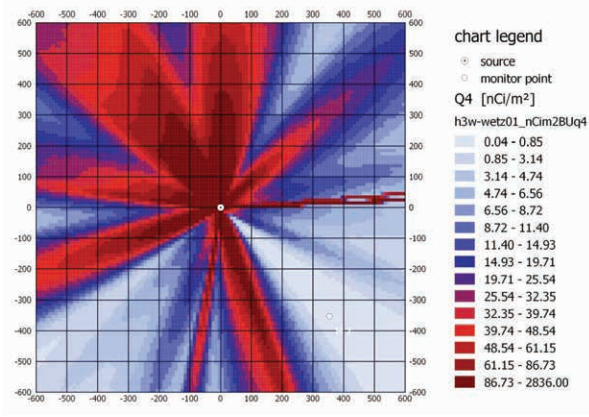


**Figure 40:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the fourth quarter; calculation grid 1.**

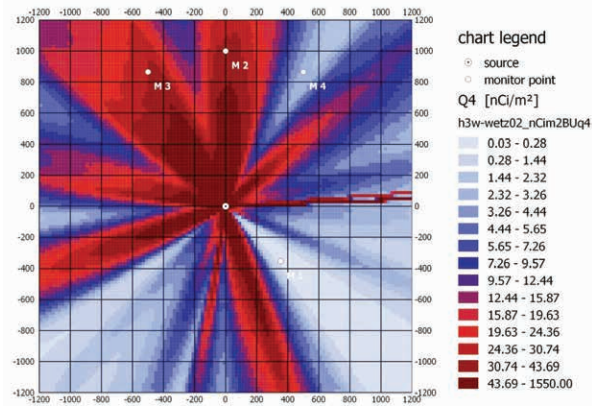


**Figure 41:** **Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the fourth quarter; calculation grid 2.**

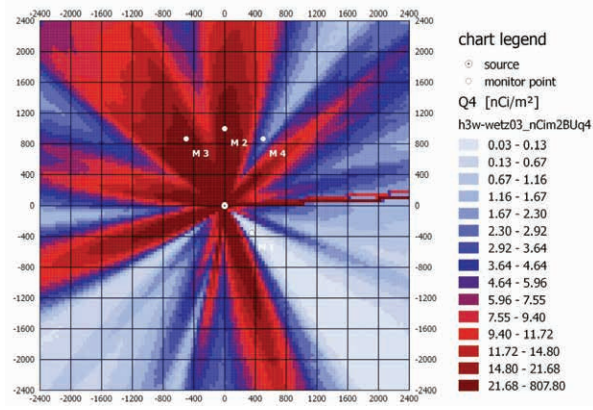


**Figure 42: Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the fourth quarter; calculation grid 3.**

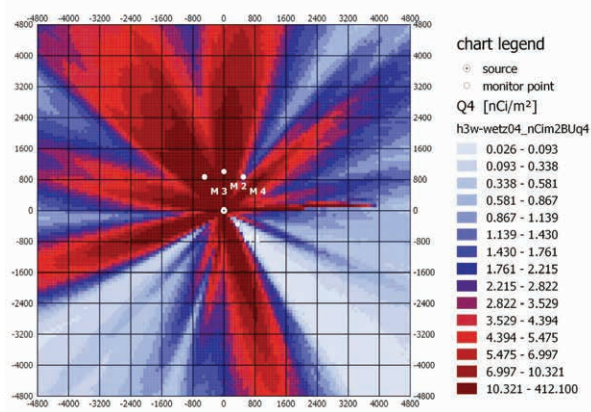


**Figure 43: Sum of continuous release and batch release: two-dimensional contamination of Tritium [Ci/m²] due to precipitation for the fourth quarter; calculation grid 4.**

**Appendix 2: Daily Impact of Tritium at four assessment points**

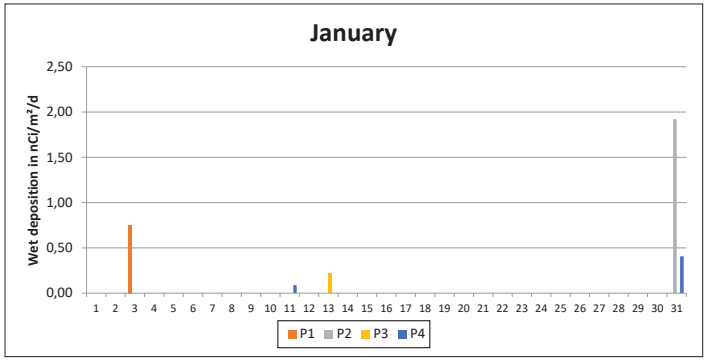


**Figure 44: Maximum daily mean values to be expected for January at the four examination points.**

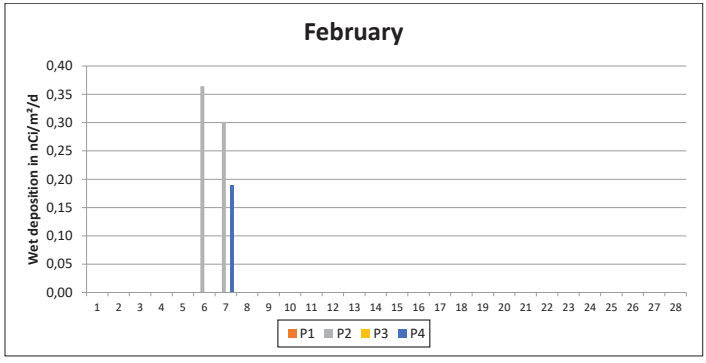


**Figure 45: Maximum daily mean values to be expected for February at the four examination points.**

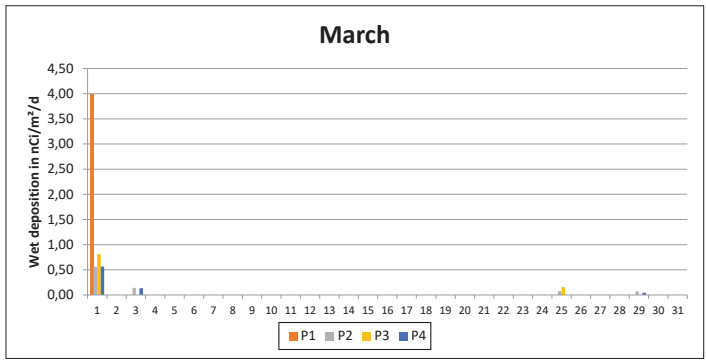


**Figure 46:** **Maximum daily mean values to be expected for March at the four examination points.**

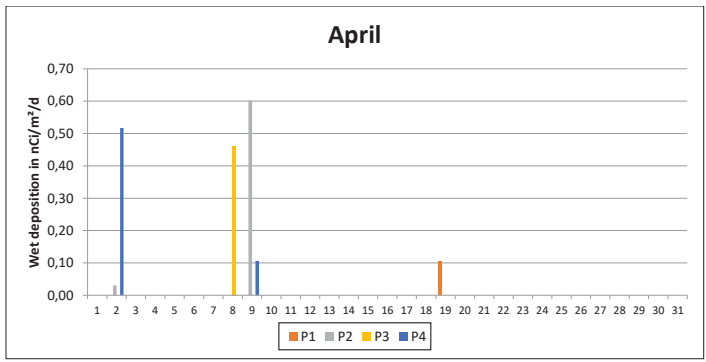


**Figure 47:** **Maximum daily mean values to be expected for April at the four examination points.**

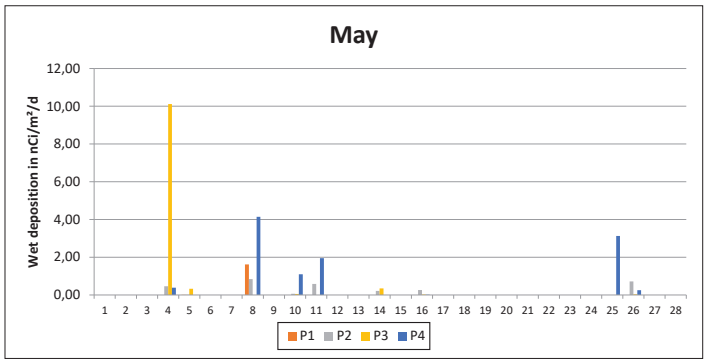


**Figure 48:** **Maximum daily mean values to be expected for May at the four examination points.**

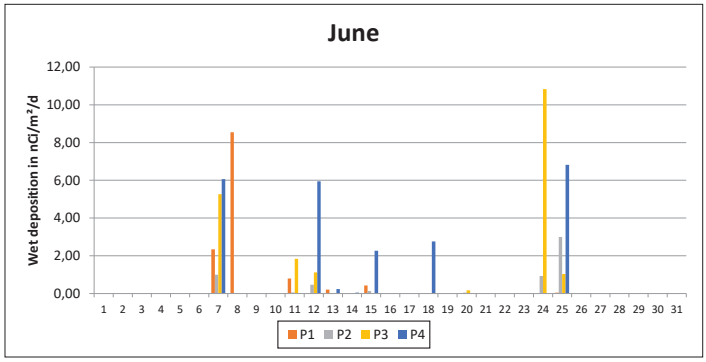


**Figure 49:** **Maximum daily mean values to be expected for June at the four examination points.**

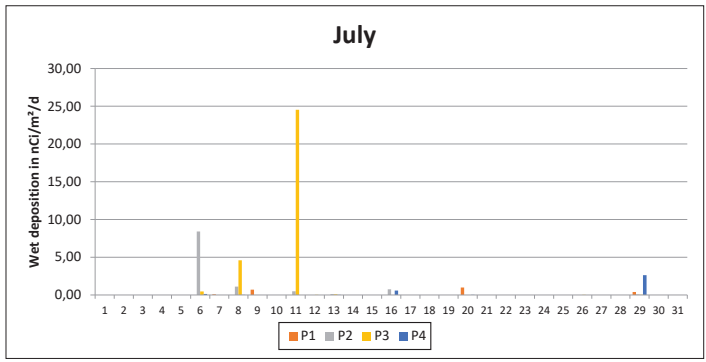


**Figure 50:** **Maximum daily mean values to be expected for July at the four examination points.**

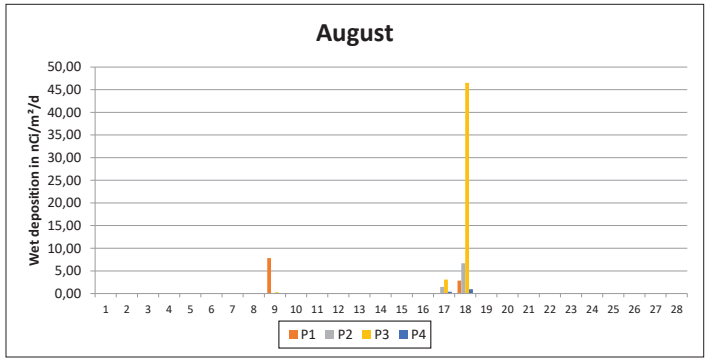


**Figure 51:** **Maximum daily mean values to be expected for August at the four examination points**

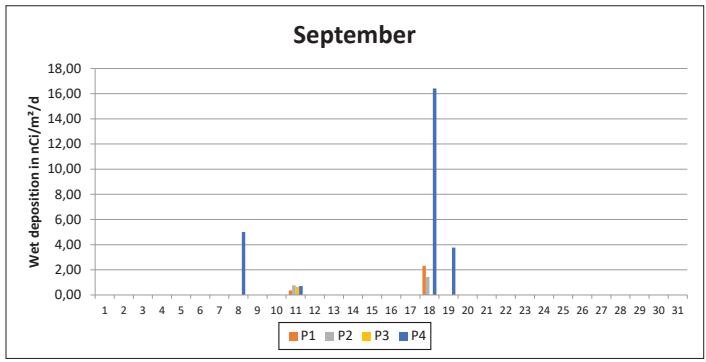


**Figure 52:** **Maximum daily mean values to be expected for September at the four examination points.**

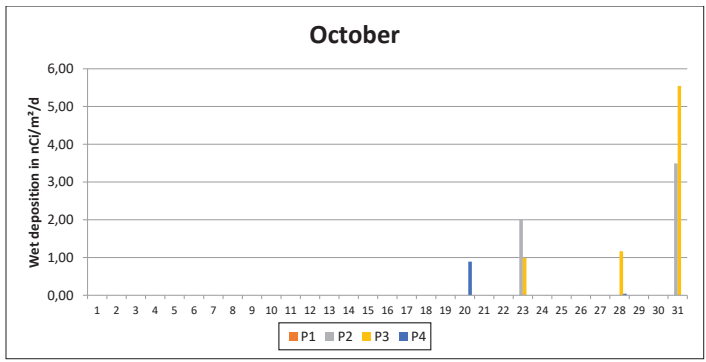


**Figure 53:** **Maximum daily mean values to be expected for October at the four examination points.**

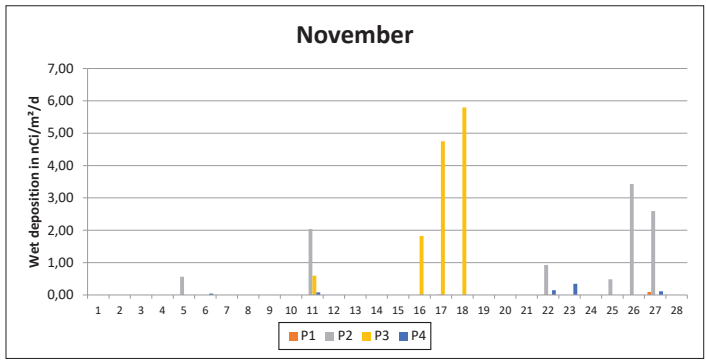


**Figure 54:** **Maximum daily mean values to be expected for November at the four examination points.**

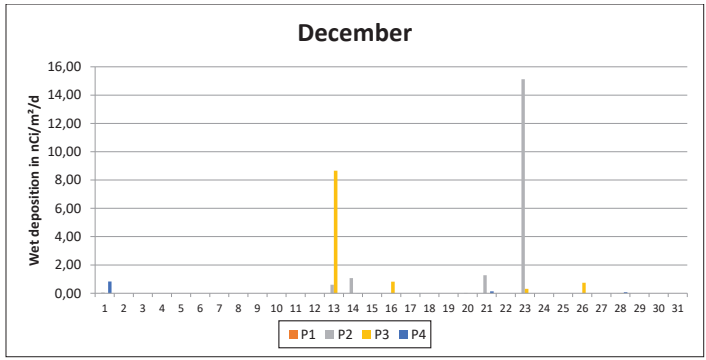


**Figure 55:** **Maximum daily mean values to be expected for December at the four examination points.**

**Appendix 3: Tritium activity in rainwater at four assessment points**

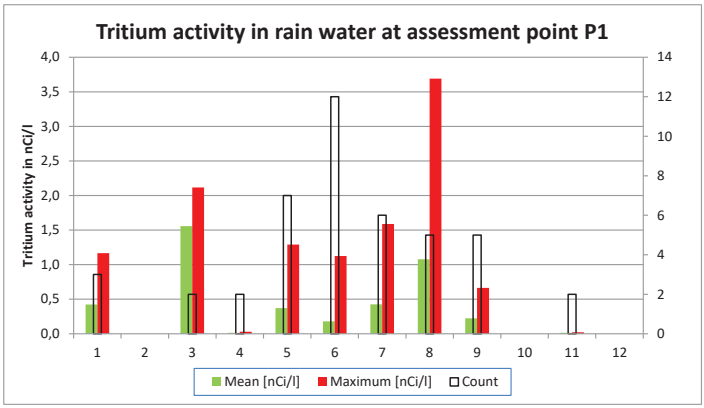


**Figure 56: Tritium activity in rainwater at assessment point P1 per month**

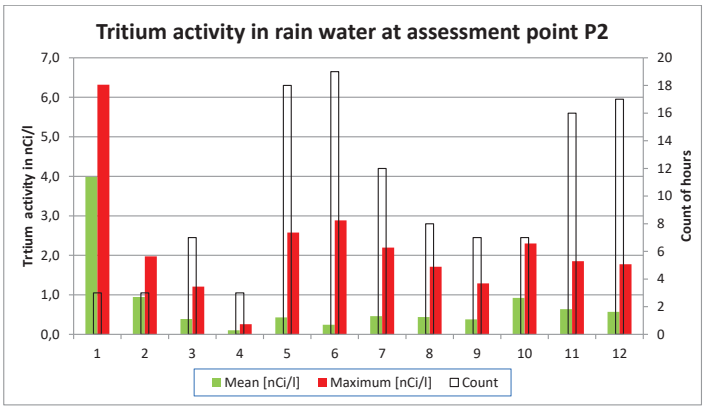


**Figure 57: Tritium activity in rainwater at assessment point P2 per month**

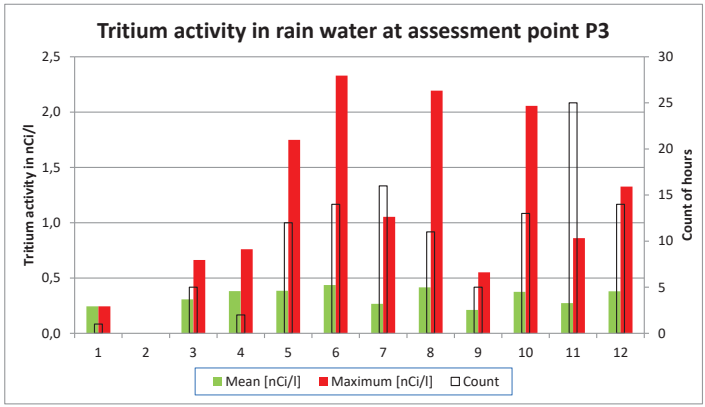


**Figure 58:** **Tritium activity in rainwater at assessment point P3 per month**

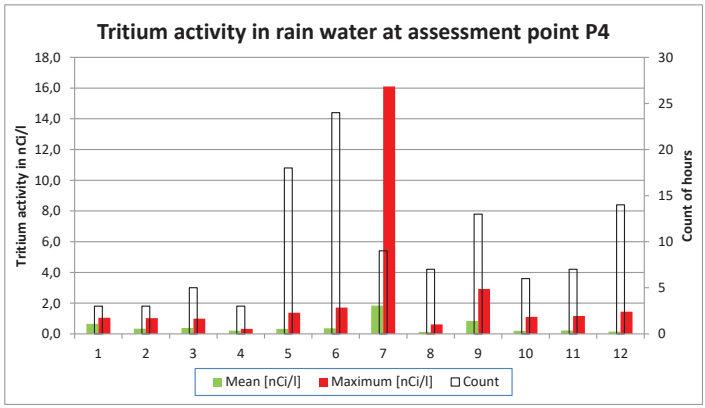


**Figure 59:** **Tritium activity in rainwater at assessment point P4 per month**

. . .extremely important and timely, especially considering the present plans for the large-scale release of radioactive, tritium-contaminated water from the damaged Fukushima-Daiichi Nuclear Power Plant. . . .

**Dr. Robert Richmond**
*Research Professor and Director,*
*Kewalo Marine Laboratory, University of Hawaii at Manoa.*

Rigorously independent and critical analyses in the field of ionising radiation and health, that draw upon and update evidence-based conclusions, like this one, are vital. . . .

**Tilman Ruff, MB, BS (Hons.)**
*Co-President, International Physicians for the Prevention of Nuclear War*

This book is a must-read for anyone concerned about the impact of tritium on the environment. Written in an easy-to-read style, it challenges conventional wisdom and provides a comprehensive overview of the large variety of effects of tritium beyond the commonly understood link to cancer. . . .

**Dr. Ferenc Dalnoki-Veress**
*Scientist-in-Residence and Adjunct Professor,*
*James Martin Center for Nonproliferation Studies,*
*Middlebury Institute of International Studies at Monterey*

**Arjun Makhijani** has a Ph.D. in Engineering (specialization: nuclear fusion) from the University of California at Berkeley. He has written widely about the impacts of nuclear weapons production and testing, nuclear power, and nuclear waste for over four decades. He is president of the Institute for Energy and Environmental Research (www.ieer.org), which provides the public with accurate scientific information to promote the democratization of science and a safer, healthier environment.

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