Environmental Health-Risk Assessment for Tritium Releases from the National Tritium Labeling Facility (NTLF) at Lawrence Berkeley Laboratory

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from the National Tritium Labeling Facility (NTLF) at
Lawrence Berkeley Laboratory**

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EXECUTIVE SUMMARY

This report is a health risk assessment that addresses continuous releases of tritium to the environment from the National Tritium Labeling Facility (NTLF) at the Lawrence Berkeley Laboratory (LBL). The NTLF contributes approximately 95% of all tritium releases from LBL. Established as a National Institutes of Health national resource center in 1982, the facility's role is to conduct basic research and to supply educational and tritium labeling support for biomedical researchers in North America. Facility staff and visiting researchers "label" pharmaceutical chemicals and other materials with tritium by replacing a hydrogen atom with a tritium atom in order to trace chemical and biological processes. NTLF is a unique facility that provides the technology to carry out both labeling and analysis at the same location.

Risk assessment is a process by which we evaluate information on potential health hazards of environmental contaminants and the extent of human and/or ecological exposures to these contaminants. Quantitative risk assessment is the process for identifying likely ranges of exposure, the adverse consequences of the exposures, and the associated probability of these consequences. Typically, the primary purpose of a risk assessment is to make clear the significance of a potential risk. Risk assessments are often carried out after other more direct methods, such as epidemiology studies, fail to reveal any potential impacts. In such cases the risk assessment is used to estimate the likely range of effects that could not be detected in relatively small populations with a short exposure durations.

Transport and transformation models were used to determine the movement of tritium releases from the NTLF to the air, surface water, soils, and plants and to determine the subsequent doses to humans. These models were calibrated against environmental measurements of tritium levels in the vicinity of the NTLF and in the surrounding community. Three zones were established to characterize potential impacts—Zone 1 that encompasses the immediate area around the NTLF and is roughly 30 meters (100 feet) in radius, Zone 2 that includes the entire LBL site and the UC property that encompasses the Lawrence Hall of Science, the Samuel Silver Space Sciences Laboratory, the Mathematical Sciences Research Institute, and the Botanical Gardens as well as several single family residences along Grizzly Peak Road above the NTLF, and Zone 3 that includes the UC Campus and residences that may be affected by the tritium that migrates from the NTLF site through washout from air by rain and runs off downhill via Strawberry Creek.

Risk levels were determined for human populations in each of these zones. Risk levels to both individuals and populations were calculated. A population risk level of one in a hundred per year (1×10^{-2} per year) corresponds to an expected incidence of one additional effect per hundred years of continuous exposure to that population; a population risk level of one in a million per year (1×10^{-6} per year) corresponds to an expected incidence of one additional effect per million years of continuous exposure to that population (assuming the characteristics of the population do not change over this period of time). In contrast, an individual risk corresponds to the likelihood that a representative individual within a group will experience an effect if he or she is exposed for an entire lifetime to the estimated tritium levels. Individual risk is expressed as a ratio, such as one in a hundred chance per lifetime (1×10^{-2}) or one in a million chance per lifetime (1×10^{-6}).

In this report population risks and individual risks were calculated for three types of diseases—cancer, heritable genetic effects, and developmental and reproductive effects. The estimated risk levels for these three factors in each of the zones, assuming a continuous release rate of release of 100 curies¹ per year of tritium as tritiated water, are shown below:

Zone 1 (~200 individuals at risk) 100 curies of tritium released per year	Population risk per year	Individual risk per lifetime
Cancer	2×10^{-4}	4×10^{-5}
Genetic defects	7×10^{-7}	7×10^{-8}
Reproductive and developmental effects	1×10^{-5}	1×10^{-6}

Zone 2 (~10,000 individuals at risk) 100 curies of tritium released per year	Population risk per year	Individual risk per lifetime
Cancer	8×10^{-5}	9×10^{-7}
Genetic defects	3×10^{-7}	1×10^{-9}
Reproductive and developmental effects	5×10^{-6}	2×10^{-8}

¹ Tritium releases since 1990 have been 100 curies or less due to voluntary emissions controls installed in 1989. From 1982 to 1990, typical emissions were approximately 500 curies of tritium per year. This risk assessment also address risk levels for yearly releases of 1000 curies of tritium.

Zone 3 (~100,000 individuals at risk) 100 curies of tritium released per year	Population risk per year	Individual risk per lifetime
Cancer	1×10^{-5}	7×10^{-9}
Genetic defects	4×10^{-8}	7×10^{-12}
Reproductive and developmental effects	6×10^{-7}	1×10^{-10}

In order to provide perspective on the relative magnitude of these risks, listed below are estimates of similar risks that are associated with background levels of tritium in the United States. These background levels are due to natural and human sources of tritium other than the NTLF.

Estimated risks associated with current background levels of tritium in the U.S. Assuming 100,000 individuals at risk	Population risk per year	Individual risk per lifetime
Cancer	1×10^{-2}	9×10^{-6}
Genetic defects	1×10^{-5}	1×10^{-8}
Developmental and reproductive effects	5×10^{-4}	3×10^{-7}

The process and assumptions used to calculate the above risks are described in Part 1 of this document, the health risk summary, which follows. The methods, equations, and data used for calculating the above risks are provided in Part 2 of this document and its appendices, which provide all relevant technical and mathematical details on the process by which risk is calculated.

Final Draft December, 1974

Environmental Health-Risk Assessment for Tritium Releases from the National Tritium Labeling Facility (NTLF) at Lawrence Berkeley Laboratory

PART 1: HEALTH-RISK SUMMARY

1.0 INTRODUCTION

This report provides a health risk assessment² that addresses the continuous releases to air of tritium from the National Tritium Labeling Facility (NTLF) at Lawrence Berkeley Laboratory (LBL). This risk assessment is directed both at the residents of the cities of Berkeley and Oakland who might be impacted by these releases and at members of the scientific community, who have deeper technical interests. Thus, it is divided into two sections. This first section, Part 1, provides a health risk summary in a format that is tractable by an educated but non-scientific public. Part 2 together with its appendices provide more of the technical details on how the risk assessment was actually carried out. Also provided in Part 2, are summaries of the data used in the analysis and a description of the models and methods used to quantify risk and our uncertainties about the estimated risks. Information provided in this risk assessment has been reviewed for consistency with current scientific practice and with risk assessment guidelines of the State of California, U.S. Environmental Protection Agency, the U.S Nuclear Regulatory Commission, the National Council on Radiological Protection and Measurements (NCRP), and the International Commission on Radiological Protection (ICRP).

Risk levels to both individuals and populations are calculated. Population risks and individual risks are calculated for three types of diseases—cancer, **heritable genetic effects**, and **reproductive and developmental effects**. The estimated risk levels for these three factors in each of three zones in the vicinity of the NTLF are estimated under the assumption that the NTLF releases 100 curies of tritium per year. We also look at individual and population risks that might be associated with much higher levels of continuous release.

This health risk summary is divided into six sections. The first section provides an introduction to the risk assessment process. The next section provides a general description of radiation and how it works. The third section summarizes the physical, chemical, and radiological aspects of tritium that are relevant to the risk

² Terms that are defined in the Glossary of Technical Terms are printed in bold type.

assessment. The next section describes the NTLF and explains why, how, and how much tritium is released from this facility. The fifth section summarizes the process we used to estimate how tritium is dispersed from the NTLF and what effects this could have on the surrounding population. The final section provides a glossary in which we define many of the technical terms used in the health-risk summary. Figure 1 below illustrates the framework we used for assessing risk from tritium releases at the NTLF.

1.1 WHAT IS RISK ASSESSMENT?

Reports of environmental contaminants in air, water, soil, and food result in public concern about the risks to human health posed by the byproducts of industrial societies. In order to address public concerns about the long-term residual health effects of environmental releases, regulatory agencies make use of risk assessment. Risk assessment is a quantitative evaluation of information on potential health hazards of environmental contaminants and the extent of human and/or ecological exposures to these contaminants. Risk assessment is a process for identifying adverse consequences and their associated probability. The risk assessment approach differs from other environmental protection strategies in that its principal objective is not to eliminate all risk but to quantify the risk and to assist efforts to balance the level of risk against the cost of risk reduction, against competing risks, or against risks that are generally accepted as trivial or acceptable. The end-product of a risk-based approach to environmental management is either to identify an acceptable level of exposure or prescribe a required level of technological control.

As applied to radionuclide emissions, risk assessment involves four inter-related steps—(1) determination of source concentrations and emissions characteristics, (2) exposure assessment, (3) toxicity assessment, and (4) risk characterization, which combines the information collected in steps 1 through 3 into a quantitative description of the magnitude and range of the risk. These steps can be carried out with assistance from mathematical models in order to project forward to define the risk associated with existing releases and to work backward to define how past releases might pose risk to any member of a potentially exposed population.

1.2 RADIATION AND HOW IT WORKS ³

Although radioactivity and the radiation it produces, were only discovered late in the nineteenth century, there is nothing new about radiation. Both radioactivity and the radiation it produces existed on earth long before life emerged. Radiation is a transport of energy through space. In traversing material, radiated energy is

³Material in this section is based on the book *Radiation Doses, Effects, Risks* (UNEP, 1985).

Figure 1. Outline of the strategy for tritium risk assessment

absorbed. Radioactivity is the disintegration of unstable atoms and a process that results in the production of ionizing radiation. Ionizing radiation has sufficient energy to cause the formation of electrically-charged particles (ions) in impacted matter. Elements are made up of atoms and atoms contain a cluster of particles at their center, called the nucleus, which is surrounded by a cloud of electrons. Some of the particles in the nucleus contain a positive charge and are called protons, others contain no charge at all and are called neutrons. Atoms of the same element always contain the same number of protons in their nucleus, but can have differing numbers of neutrons. Those that have differing numbers of neutrons, but the same number of protons are called isotopes of the same element. When an isotope is designated by the total number of particles (neutrons plus protons) that are clustered in its nucleus, we call it a nuclide. Some nuclides are stable and have an uneventful, unchanging existence. Most are unstable and give vent to their instability by relentlessly trying to become something else, that is another nuclide. The energy released in this transformation process is transmitted away from the atom as radiation. Thus, unstable nuclides that emit radiation as they decay to some other form are called "radionuclides."

1.2.1 Types of Radiation

As was noted above, unstable nuclides transform themselves to other nuclides in a continuing process of transformation until a stable nucleus is obtained. These transformations result in the emission of ionizing radiations. There are three types of ionizing radiation—alpha particles, which are energetic but non-penetrating particle clusters; beta particles, which are electrons released from the nucleus of an atom; and gamma rays, which are electromagnetic radiation (similar to visible light) with a very high frequency and energy and strongly penetrating. As an example, the heavy, naturally-occurring nuclide uranium-238 contains a nucleus in which the clustered particles of the nucleus are just barely able to stay together. Eventually, a chunk of two protons and two neutrons are ejected from the nucleus. As this chunk, called an alpha particle, leaves its nucleus, uranium is transformed into a different atom and nuclide, thorium-234. The resulting nuclide, thorium-234 is still unstable and releases an electron or beta particle from its nucleus in an effort to become more stable. Through this emission, it is transformed to protactinium-234. Frequently, a nuclide is in a such a highly excited state that the emission of particles is not sufficient to calm it down; it then gives off a vigorous burst of pure energy called gamma radiation. Like x-rays, gamma radiation does not involve the release of particles.

1.2.2 Measuring Radiation and Radioactive Decay

Although all radionuclides are unstable, some are more unstable than others. Some nuclides transform themselves so rapidly, that is difficult to measure their rate of transformation. Others, such as uranium-238 are so leisurely about the process, that they are almost like stable elements. Half of a lump of initially pure protactinium-234 transforms to another species in less than a minute. Whereas, it takes four and a half billion years for half of a lump of initially pure uranium-238 to be transformed to other elements. The period it takes for half of any amount of element to decay is known as its half-life. The process goes on relentlessly. After one half-life, 50 out of 100 atoms in any lump of a radioactive element will remain unchanged; during the next half life, 25 of these will decay and so on exponentially. The number of transformations that take place each second in a given amount of radioactive material is called its activity. The activity is measured in units called becquerels (Bq), after the man who discovered radioactive decay. Each becquerel equals one transformation per second. An older unit of radioactive decay still often used in the U.S. is the curie (Ci), which is equal to 3.7×10^{10} Bq.

Radiation is measured by both monitoring devices and sampling devices. A monitoring device, such as Geiger counter, is a device that measures radiation activity by counting the rate at which charged particles are produce in the vicinity of a source. A sophisticated monitoring device can be designed to measure the activity, the type of particle, and the energy deposition over some period of time. A sampling device is designed more to measure radioactivity than radiation. Sampling devices are set up to collect environmental samples, such as air, water, soil, or plants. A monitoring device is used to assess the levels of radiation coming from these samples and this is used to infer the levels of radioactivity in the samples. Since tritium is typically found at relatively low levels in the environment, its presence is often measured using environmental sampling instead of radiation monitors.

1.2.3 Radiation Doses

The different forms of ionizing radiations are emitted with different energies and have different penetrating power—and thus have different effects on living things. Alpha radiation, with is heavy chunk of neutrons and protons, is halted by a sheet of paper and can scarcely penetrate the dead outer layers of our skin. So it is not dangerous unless substances emitting it get into the body through an open wound or are eaten or breathed in—in which cases it is potentially very damaging. Beta radiation is more penetrating. Depending on the energy of the beta particle, it can go through as much as a centimeter or two (about one-half to three-quarters of an inch)

of living tissue. Gamma radiation, which travels at the speed of light, is extremely penetrating and will go through anything short of a slab of concrete or lead.

It is the energy of radiation that can cause tissue damage. The amount of energy deposited in living tissue is called the dose. Doses are expressed in different ways depending on how much of the body, and what parts of it are irradiated, whether one individual or many people are exposed and the period over which the exposure takes place. The amount of radiation energy that is absorbed per gram of tissue is called the absorbed dose and is measured in units called grays (Gy). An older unit of absorbed dose that is still often used in the U.S. is the rad, which is equal to 0.01 Gy. However, the absorbed dose does not tell the full story, because a dose of alpha radiation is much more damaging than the same dose of beta or gamma radiation. So the dose needs to be adjusted or weighted to account for its relative potential to do damage. The adjustment factor used in this damage weighting calculation is called the relative biological effectiveness (RBE). The resulting weighted dose is known as the dose equivalent and is measured in units called sieverts (Sv). An older unit of dose equivalent still often used in the U.S. is the rem, which is equal to 0.01 Sv.

1.3 TRITIUM

Tritium is a radioactive isotope of hydrogen. Tritium decays to helium through emission of an electron from its nucleus (beta decay). The radioactive half life of tritium (indicating the rate at which the nucleus disintegrates) is 12.3 years. Tritium emits low-energy beta particles which have intermediate penetrating ability. The maximum energy of its emitted electrons is insufficient for skin penetration. Thus, tritium represents little or no external dose hazard, however, once internalized, even its minimal penetration depth could be sufficient to inflict deleterious effects. Tritium has similar chemical properties to those of hydrogen, and also has similar physical properties (e.g. density) since the two differ only marginally in atomic mass.

1.3.1 Natural and Human Sources of Environmental Tritium

Tritium is continuously released to the environment from both natural and human sources. The major natural source of tritium is that produced by continuous cosmic ray reactions within the upper atmosphere. A very minor contribution of the annual natural production of tritium comes from the earth's crust. Human sources of tritium in recent decades have far exceeded natural sources. Atmospheric nuclear weapons tests in the 1950s and 1960s are estimated to have produced a large fraction of the current global inventory of tritium. However, this inventory continues to decrease by half every 12.3 years so long as there are no more additional nuclear

weapons tests. Other smaller human contributions to environmental tritium include normal and accidental releases of tritium from nuclear reactors, and consumer products. The combined natural and human emissions of tritium result in a current global inventory that is about 60 times greater than tritium levels due to natural sources alone. Based on the dilution of this quantity of tritium in the circulating water volume of the earth, we estimate that this level of tritium will result in a global average concentration of tritiated water in world's waters of 50 Bq/L.

1.3.2 How Does Exposure to Tritium Effect Humans and Other Organisms?

Tritium is found in the environment in three principal forms—molecular tritium (T_2 or HT), tritiated water, i.e., oxidized tritium (HTO), and organically bound tritium (OBT). Tritiated water is the most abundant chemical form of tritium in the environment. HTO has very similar properties to those of water. Because the relative difference in atomic mass is significantly less, the similarity of physical properties between HTO/ T_2O and H_2O is greater than that displayed between tritium and hydrogen. HTO is much more readily taken-up by organisms and environmental media than molecular tritium. Tritiated water is the principal form of tritium released from the NTLF. Following exposures to tritium gas and tritiated water, a small fraction of tritium is incorporated into organic molecules. This tritium is referred to as organically bound tritium (OBT) and will have a different metabolism than tritiated water. As a result of OBT, the retention of tritium following some exposures to HTO can be extended somewhat.

Health risks associated with exposures to tritium are attributable to the ionizing radiation emitted by tritium. In order to quantify the likely magnitude of these effects, it is necessary to address tritium metabolism and dose in terms of radiation biology and radiation protection methods. The biological effects of ionizing radiation are brought about through chemical changes in the cells caused by ionizations, excitations, dissociations, and atom displacements. The health effects of ionizing radiations—cancer, genetic defects, and reproduction effects—are proportional to the energy carried by the radiation and delivered to living cells.

Each decay of a tritium atom releases energy that is carried by the resulting beta particle.

1.4 TRITIUM RELEASES FROM THE NTLF

Only one facility at Lawrence Berkeley Laboratory (LBL) uses tritium in large quantities—the National Tritium Labeling Facility (NTLF). The NTLF is contained

within Building 75 on the LBL site. This location is identified on Map 1. The NTLF was established in 1982 as a National Institutes of Health (NIH) national resource center. The operations of this facility are currently funded in large part by the NIH. Facility users are charged a fee which is used to offset monitoring and equipment costs. This facility accounts for 95% of all tritium releases from the entire LBL complex. LBL's Environmental Health and Safety Division closely monitors all of the NTLF activities for compliance with environmental health and safety regulations. There are currently six major tritium labeling facilities in the U.S. Of these, the NTLF at LBL is the only non-commercial facility.

1.4.1 What Is the Purpose of Tritium Labeling?

In the labeling process carried out at the NTLF, tritium atoms replace hydrogen atoms in a molecule of interest. There are negligible differences in physiochemical and biological properties of labeled and non-labeled compound. Successfully labeled molecules will be sources of radioactivity and therefore readily identified. Specific activity measurements are easily converted into measures of concentration. This process is unique, cost-effective, and very precise in determining reaction pathways and/or yields from chemical reactions. The labeling process has been used extensively in applications that involve the development of pharmaceutical agents. In such applications, the labeling process allows the investigation of administration, distribution, metabolism, and excretion characteristics/behavior of candidate drugs.

NTLF provides benefits to public health by helping biomedical researchers test new products that can be used in curing diseases. Collaborative research projects initiated over the past three years have shown new ways for studying cell metabolism and biomolecular structure and function. Staff members of the facility actively publish scientific articles and reports and present their work at a wide range of meetings. More than 100 users have visited the NTLF since 1982 to label upwards of 250 compounds.

1.4.2 How Much Is and Was Released?

Tritium diffuses easily through all types of materials, thus it is difficult to make any system fully resistant to tritium leakage. The NTLF has been designed and is operated in such a manner that there are multiple layers of defense to restrict the release of tritium to the environment. The NTLF currently releases approximately 100 curies per year (3.7 trillion Bq/y) which has been typical of annual releases since 1990. This value is lower than the 500 curies per year (19 trillion Bq/y) that was more typical of releases during the period 1982 to 1990.

Map 1: Selected Environmental Monitoring Locations at LBL

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1.4.3 Where Does It Go When It Is Released?

In conducting the risk assessment, we developed models that can address tritium releases from the NTLF. The first goal of these models is to determine how tritium releases could spread out from the point of release. These models were used to assist the public and ourselves in understanding how tritium will (a) remain or spread within the envelope of air to which it is released, (b) be transported to another parts of the environment by transfers that involves natural events such as wind dispersion, evaporation, precipitation, etc., and (c) be physically transformed by radioactive decay. The models were calibrated against environmental measurements of tritium levels in the vicinity of the NTLF and in the environment of Berkeley in order to provide feedback on the reliability of model predictions. These models together with site-specific data are used to characterize the contamination of air, surface water, soils, and plants adjacent to the NTLF. Based on these contamination levels, we made estimates of corresponding doses and risk to human populations.

The model includes two compartments—air and soil. Water and vegetation as exposure media are assumed to be in equilibrium with air and/or soil. The two-compartment structure is applied in each of the exposure zones. The air compartment is represented by a simple box model, which receives the NTLF air emissions and receives emissions from soil and vegetation that have retained tritium from previous emissions. Losses from air include deposition to soil; wind-driven convective losses; and radioactive decay. The soil compartment represents the surface layer of soil that receives tritium as deposition from air. Losses from soil include diffusion to air, diffusion and infiltration to deeper soil layers, runoff to surface water, and radioactive decay. Landscape, climate and hydrology parameters used in the analysis are derived from site-specific information.

1.5 POTENTIAL HEALTH EFFECTS FROM TRITIUM RELEASES AT THE NTLF

Risk levels to both individuals and populations are calculated in this report for three types of diseases—cancer, heritable genetic effects, and reproductive and developmental effects. The estimated risk levels for these three factors in each of three zones in the vicinity of the NTLF are estimated under the assumption that the NTLF releases 100 curies of tritium per year. We also look at individual and population risks that might be associated with much higher and less plausible levels of continuous release, 1000 curies of tritium per year.

1.5.1 Who Might be Affected?

Three zones were established to characterize potential impacts—Zone 1 that encompasses the immediate area around the NTLF and is roughly 30 meters (100 feet) in radius, Zone 2 that includes the entire LBL site and the UC property that encompasses the Lawrence Hall of Science, the Samuel Silver Space Sciences Laboratory, the Mathematical Sciences Research Institute, and the Botanical Gardens as well as several single family residences along Grizzly Peak Road above the NTLF, and Zone 3 that includes the UC Campus and residences that may be affected by the tritium that migrates from the NTLF site through washout from air by rain and runs off down hill via Strawberry Creek.

In Zone 1, the population at risk is taken to include the working population in this zone as well as the infants born to mothers who are part of this working population. We assume that the number of adults in this population is on the order of 200. In Zone 2, the population at risk is taken to include the working population of LBL as well as residents of single-family residences above the NTLF, who are exposed through contact with air and ingestion of homegrown foods. Also included in this group are the infants born to mothers who are part of this population. We assume that the number of people in this population is on the order of 10,000. In Zone 3, the population at risk is taken to include the working population of the main UCB campus and the city of Berkeley and the residential populations that could be exposed to tritium that is washed from the air into surface-water runoff and carried down into Strawberry Creek. We also include the infants of mothers within this population as part of the group at potential risk. We assume that the number of people in this population is on the order of 100,000.

1.5.2 How Do People Come in Contact with Tritium as It Spreads?

Because the energy of electrons emitted during the decay of the tritium nucleus is insufficient to penetrate skin, only internal dose routes (inhalation and ingestion intake and uptake through skin) are of concern. Relevant to releases from the NTLF, exposure pathways resulting in internal dose to tritium, include:

- 1) tritiated water enters the body via the lung through respiration
- 2) tritium in water, home-grown foods, and breast-milk (in the case of infants) can be ingested and absorbed into the body through the gastro-intestinal tract;
- 3) tritium from tritiated water-vapor in air can be taken through the skin, and
- 4) tritium from tritiated water in surface water can be taken through the skin during activities that involve dermal contact with the contaminated water, i.e., washing, swimming, and wading in surface water.

1.5.3 What Are the Potential Risks?

Health risks associated with exposures to tritium are attributable to the ionizing radiation emitted by tritium. The biological effects of ionizing radiation are brought about through chemical changes in the cells caused by ionizations, excitations, dissociations, and atom displacements. There are very few available studies on tritium-induced health injury in humans. Thus, health-risk estimates specific to tritium must be based on the large number of experiments with animals and cell cultures. These experiments show that exposure to tritiated water results in mutations and cell disruptions that can lead to cancer, heritable genetic effects, and reproductive and developmental effects.

When it enters the body tritiated water quickly reaches equilibrium with hydrogen in the body and imparts a uniform dose to all soft tissues. In order to properly assess combined exposures to tritiated water and organically-bound tritium in food products, we describe the retention of tritium following exposure with a model that accounts for intake of both. Based on this model we determined that the biological half-life of tritium is on the order of 15 days for the intake or uptake of tritiated water and on the order of 48 days for the ingestion of organically-bound tritium.

For releases of 100 curies per year, the predicted concentrations of tritium in air and water are below the regulatory standards for the ambient (i.e., non-occupational) environment. For the extreme case where there is a continuous release of 1000 curies per year, the concentrations in Zone 1, exceed federal limits for non-occupational level but are well below the occupational levels. For releases of either 100 or 1000 curies per year, the predicted concentrations in the air and water of Zone 2 and Zone 3 are below the regulatory standards for the ambient (i.e., non-occupational) environment.

1.6 SUMMARY OF PREDICTED RISKS

The estimated risk for tritium releases from the NTLF are quite low relative to the background incidence of the diseases considered—cancer, heritable genetic defects, and reproductive and developmental effects. The highest levels of predicted individual risks for all health endpoints are associated with the first exposure zone, Zone 1. Based on an assumed continuous release of 100 curies per year, we note the following in our calculations for Zone 1:

For individuals who are assumed to work in this area for a 40-year working life, the estimated individual risk for genetic defects and developmental and reproductive effects to their children are likely less than one chance per million.

For these same postulated individuals, we estimate that the added risk of cancer is 4 chances in a hundred thousand.

We estimate that as a result of potential fetal exposures and potential exposures through breast milk, the added lifetime risk of cancer to children born to women who work continuously in this area is on the order of 3 chances in a million. This risk calculation is based on the assumption that the infant drinks breast milk exclusively for a period of two years.

With the exception of the risk to the working populations, these estimated risks are below the lifetime individual risks associated with current background levels of tritium.

In Zone 2 and Zone 3, the estimated individual risks for all health endpoints are quite small, less than one chance in a million and are below the estimated lifetime individual risks associated with current background levels of tritium.

An uncertainty analysis was used to assess the reliability of these results. Based on the range of parameter values used as input for the predictions of risk, we assessed the precision of the model predictions by making thousands of repeated calculations of risk with all input values varied within their likely ranges. We found that 50% of the range of output values were within a factor of 2 of the reported risk and 95% of the output values were within a factor of 10 of the reported risk.

1.7 GLOSSARY OF TECHNICAL TERMS

Absorbed dose:

Energy imparted by radiation per unit mass (kg) of tissue.

Activity:

The number of transformations that take place each second in a given amount of radioactive material.

Alpha particle:

A positively charged particle, indistinguishable from a helium atom nucleus and consisting of two protons and two neutrons.

Alpha radiation:

Same as alpha particle.

Becquerel:

The special name for the unit that expresses a measure of activity. One becquerel corresponds to one disintegration per second of any radionuclide.

Beta particle:

A high-speed electron or positron, emitted in radioactive decay.

Curie:

A unit of radioactivity, equal to the amount of a radioactive isotope that decays at the rate of 3.7×10^{10} disintegrations per second.

Exposure:

The condition of having contact with a physical or chemical agent of harm.

Exposure assessment:

The process by which contact with a toxic substance is identified and quantified.

Dermal uptake:

The transfer of a contaminant from air, water, or soil into or through the outer skin layer; considered an exposure route in an exposure assessment.

Dose:

The quantity of energy or chemical agent delivered to a specific tissue living tissue following exposure.

Dose assessment:

The process by which uptake and dose to a toxic substance is identified and quantified

Dose equivalent:

An absorbed dose that is adjusted or weighted to account for the potential of different radiations to do damage.

Gamma radiation:

Electromagnetic radiation emitted by radioactive decay and having energies in a range from ten thousand (10^4) to ten million (10^7) electron volts

Gray:

The special name for the unit that expresses a measure of absorbed dose. One gray corresponds to an energy deposition of one joule per kg of living tissue.

Half life:

1. For a quantity of radiation—the time required for half of the nuclei in a sample of a specific isotopic species to undergo radioactive decay.
2. For ingested quantities—the time required for the radioactivity of material taken in by a living organism to be reduced to half its initial value by a combination of biological elimination processes and radioactive decay.

Heritable genetic effects:

Genetic damage cause by an external agent such as radiation, which manifests itself in the gene carrier and is capable of being passed on to succeeding generations.

Ingestion:

To take into the body by the mouth for digestion or absorption; considered an exposure route in an exposure assessment.

Inhalation:

To draw air into the lungs by breathing; considered an exposure route in an exposure assessment.

Ionizing radiation:

Radiation that converts the target totally or partially into ions on impact.

Isotope:

One of two or more atoms having the same atomic number but different mass numbers.

Likelihood:

The state of being probable.

Models:

A schematic description of a system, theory, or phenomenon that accounts for its known or inferred properties and may be used for further study of its characteristics

Monitoring:

To test or sample on a regular or ongoing basis the air or an object's surface for radiation intensity.

Neutron:

An electrically neutral subatomic particle, which is stable when bound in an atomic nucleus. Neutrons and protons form nearly the entire mass of atomic nuclei.

Nuclide:

A type of atom specified by its atomic number, atomic mass, and energy state, such as carbon 14.

Probability:

A number expressing the likelihood that a specific event will occur, expressed as the ratio of the number of actual occurrences to the number of possible occurrences.

Proton:

A stable, positively charged subatomic particle. Neutrons and protons form nearly the entire mass of atomic nuclei.

Radiation:

Energy emitted in the form of rays, waves, or particles by the atoms and molecules of a radioactive substance as a result of nuclear decay.

Radioactivity

Spontaneous emission of radiation, either directly from unstable atomic nuclei or as a consequence of a nuclear reaction. This radiation includes alpha particles, nucleons, electrons, and gamma rays, emitted by a radioactive substance.

Relative biological effectiveness (RBE):

An adjustment factor used to adjust an absorbed dose to account for its relative potential to do damage in biological tissue.

Reproductive and developmental effects:

Effects caused by some physical, chemical, or biological agent, such as radiation, to the formation and development of a fetus, that results in one or more observed defects, which are not passed on to succeeding generations.

Risk:

A chance of injury, loss, or detriment. A measure of the deleterious effects that may be expected as the result of an action or inaction.

Risk assessment:

The process by which the risks associated with an action or inaction are identified and quantified.

Sampling:

The act, process, or technique of selecting an appropriate environmental sample of radioactive contamination.

Sieverts (Sv):

A unit for expressing the equivalent magnitude of an absorbed dose which has been adjusted by a factor that expresses the relative tissue damage potential of absorbed doses of radiation; equivalent to one joule of x-ray energy deposited per kg of target.

Transport:

To move or be conveyed from one place to another. In the context of environmental contamination, a contaminant is transported from one location to another by advection or diffusion processes.

Transformation

Alteration of a chemical substance from one chemical form to another through a chemical or physical reaction. Radioactive decay is a nuclear transformation whereby an atom changes from one nuclide to another.

PART 2: TECHNICAL DETAILS OF THE RISK ASSESSMENT FOR TRITIUM RELEASES FROM THE NTLF

2.0 INTRODUCTION

This report provides a health risk assessment for continuous releases to air of tritium from the National Tritium Labeling Facility (NTLF) at Lawrence Berkeley Laboratory. This risk assessment is directed primarily at the residents of the cities of Berkeley and Oakland who might be impacted by these releases and thus divided into two sections. The first section above, Part 1, provides a health risk summary in a format that is tractable by an educated but non-scientific public. This section, Part 2, together with its appendices in Part 3, provide more of the technical details on how the risk assessment was actually carried out. Also provided in this part of the report are summaries of the data used in the analysis and a description of the models and methods used to quantify risk and our uncertainties about the estimated risks.

This Part of the report is divided into eight sections. This first section provides an overview of the models and data used in the tritium risk assessment. The next section summarizes the physical, chemical, and radiological properties of tritium that are relevant to the risk assessment. The third section describes the NTLF and the nature of the tritium releases from this facility. The fourth section describes how releases from the NTLF are distributed in the environment and gives details of the model we used to assess these environmental distributions. The fifth section describes human contact with these environmental levels of tritium. The sixth section illustrates the process by which exposures to tritium are linked to potential health impacts within the exposed population. The seventh section describes how we carried out the quantification of potential health risks to the populations in and near the NTLF facility. The last section provides a summary and discussion of our results.

2.1 OVERVIEW OF THE RISK ASSESSMENT MODELS AND DATA

In conducting the risk assessment process, we developed models that can address the spread of tritium releases from the NTLF, contact with human populations, metabolism within the human body, and the potential health effects. The first goal of these models is to determine how tritium releases will be spread out from the point of release. These models are used to assist the public and ourselves in understanding how tritium will (a) remain or spread within the envelope of air to which it is released, (b) be transported to another compartment by cross-media

transfer that involves dispersion or advection (i.e., volatilization, precipitation, etc.) and (c) be physically transformed by radioactive decay. The models are calibrated against environmental measurements of tritium levels in the vicinity of the NTLF and in the environment of Berkeley in order to provide feedback on the reliability of model predictions. These models together with site-specific data are used to characterize the contamination air, surface water, soils, and plants adjacent to the NTLF. Based on these contamination levels, we made estimates of corresponding doses and risk to human populations.

Three zones were established to characterize potential health impacts—Zone 1 that encompasses the immediate area around the NTLF and is roughly 30 meters [100 feet] in radius, Zone 2 that includes the entire LBL site and the UC property that encompasses the Lawrence Hall of Science, the Samuel Silver Space Sciences Laboratory, the Mathematical Sciences Research Institute, and the Botanical Gardens as well as several single family residences along Grizzly Peak Road above the NTLF, and Zone 3 that includes the UC Campus and residences that may be affected by the tritium that migrates from the NTLF site through washout from air by rain and runs off down hill via Strawberry Creek.

Risk levels were determined for human populations in each of these zones. Risk levels to both individuals and populations are calculated. A population risk level of one in a hundred per year (1×10^{-2} per year) corresponds to an expected incidence of one additional effect per hundred years of continuous exposure to that population; a population risk level of one in a million per year (1×10^{-6} per year) corresponds to an expected incidence of one additional effect per million years of continuous exposure to that population (assuming the characteristics of the population do not change over this period of time). In contrast, an individual risk corresponds to the likelihood that an representative individual within a group will experience an effect if he or she is exposed for a defined period, such as a lifetime, to the estimated tritium levels. Individual risk is expressed as a ratio, such as one in a hundred chance per lifetime (1×10^{-2}) or one in a million chance per lifetime (1×10^{-6}).

Population risks and individual risks are calculated for three types of diseases—cancer, heritable genetic effects, and developmental and reproductive effects. The estimated risk levels for these three factors in each of the zones, assuming a yearly release of 100 curie

2.2 TRITIUM PROPERTIES AND SOURCES

Tritium is a radioactive isotope of hydrogen. The particular type of radiation associated with tritium decay is ionizing radiation, which is defined as having

sufficient energy to cause the formation of ions in impacted matter. Tritium decays to helium through emission of an electron from its nucleus (beta decay)

It has one proton as does the common isotope of hydrogen (protium—the most abundant isotope of hydrogen), and thus has a similar chemical structure and an equivalent atomic number. Tritium differs from protium in its number of neutrons—it has two neutrons whereas protium has none, and deuterium has one; thus tritium has an atomic mass of 3 instead of 1 or 2. See Figure 2 below. Tritium has similar chemical properties to those of hydrogen, and also has similar physical properties (e.g. density) since the two differ only marginally in atomic mass.

The radioactivity of tritium makes it a good tracer/label. A count of nuclear decays per unit time from a known mass or volume of aliquot allows specific activity (analog of concentration) to be calculated. The nuclear disintegrations are easily monitored, thus any molecules which have incorporated tritium are easily differentiated from non-labeled forms. Thus, tritium has great utility for experiments in which the transport or fate of particular chemicals must be identified. Because tritium is so similar to hydrogen it can be incorporated, via substitution, into any molecule or compound that contains hydrogen

Figure 2. The three isotopes of hydrogen and their atomic structure.

Tritium emits low-energy beta particles which have intermediate penetrating ability. The maximum energy of its emitted electrons is insufficient for skin penetration (it has an estimated penetration depth of 6 micro-meters (μm) versus the 10 to 100 μm thick stratum corneum (non-living outer skin layer); the 100-200 μm thick epidermis; and the 1000-3000 μm dermis. Thus, tritium represents little or no external dose hazard, however, once ingested, even its minimal penetration depth could be sufficient to inflict deleterious effects. The radioactive half life of tritium is 12.3 years. The radiological units commonly used to characterize exposure, dose, dose equivalent and radiation protection standards for tritium are listed in Table 1.

2.2.1 Physical-Chemical Properties of Tritium

Tritium is found in the environment in three principal forms—molecular tritium (T_2 or HT), tritiated water, i.e., oxidized tritium (HTO), and organically bound tritium (OBT). Tritiated water is the most abundant chemical form of tritium in the environment. HTO has very similar properties to those of water. Because the relative difference in atomic mass is significantly less, the similarity of physical properties between $\text{HTO}/\text{T}_2\text{O}$ and H_2O is greater than that displayed between tritium and hydrogen. HTO is much more readily taken-up by organisms and environmental media than molecular tritium. Tritiated water is the principal form of tritium released from the NTLF. Following exposures to tritium gas and tritiated water, a small fraction of tritium is incorporated into organic molecules. This tritium is referred to as organically bound tritium (OBT) and will have a different metabolism than tritiated water. As a result of OBT , the retention of tritium following some exposures to HTO can be extended somewhat. The conversion of T_2 to organically bound tritium (OBT) may be more likely than the conversion of HTO to OBT (Murphy, 1984). Table 2 provides a list of the physical and chemical characteristics of tritium that are relevant for a dose and risk assessment.

2.2.2 Sources of Tritium Release in the Environment

Tritium is continuously released to the environment from both natural and anthropogenic sources. The major natural source of tritium is that produced by continuous cosmic ray reactions within the upper atmosphere. A very minor contribution of the annual natural production of tritium comes from the earth's crust as a result of neutron capture reactions by ^6Li in rocks. The steady-state global inventory of tritium attributable to all natural sources of tritium is estimated to be in the range of 1 to 1.3×10^{18} Bq (Okada and Momoshima, 1993). Based on a 12.3 y half-life, this corresponds to an annual production of 0.062 Bq/y. If this inventory

Table 1. Common Radiological Units

Unit or quantity	Symbol	Brief description	Comment
Becquerel	Bq	1 nuclear disintegration per second	SI unit of radioactivity
Curie	Ci	3.7×10^{10} nuclear disintegrations per second	Older unit of radioactivity often still used in the U.S.
Roentgen	R	2.58×10^{10} coulombs per kg (photons in air)	Special unit of exposure
Gray	Gy	1 joule/kg	SI unit of radiation dose
Rad	rad	100 erg/g (= 0.01 Gy)	Older unit of radiation dose often still used in the U.S.
Dose equivalent	H	Dose $\times$ Q $\times$ any other modifying factors	Used in radiation protection
Quality factor	Q	Biological Effectiveness of radiation	Used in radiation protection
Sievert	Sv	Gy $\times$ Q $\times$ any other modifying factors	SI unit of dose equivalent
Rem	rem	rad $\times$ Q $\times$ any other modifying factors	Older unit of dose equivalent often still used in the U.S.

were uniformly diluted in all of the world's land and ocean waters— 1.4×10^{18} L (van der Leeden, 1990)—then the average tritium concentration in the world's waters would be 0.8 Bq/L. Measurements taken before nuclear testing began (before 1950) of HTO in water for a few water-bodies in the U.S. ranged from 0.14-7.9 Bq/L for Chicago rain water, 0.16-0.21 Bq/L for Lake Michigan water, and 0.30-0.77 Bq/L for the Mississippi (Okada and Momoshima, 1993).

Anthropogenic sources of tritium in recent decades have far exceeded natural sources. Atmospheric nuclear weapons tests in the 1950s and 1960s are estimated to have produced from 185 to 240×10^{18} Bq of tritium (Okada and Momoshima, 1993). It is estimated that the legacy of the nuclear weapons test of the 1950's and 1960's is a current global inventory of 52×10^{18} additional Bq (Okada and Momoshima, 1993).

Table 2. Physical and chemical characteristics of tritium

Property	Value	Reference
Radioactive half life ($T_{1/2}$)	12.35 y	NCRP (1985)
Decay constant ($= 0.693/T_{1/2}$)	$5.6 \times 10^{-2} \text{ y}^{-1}$ $1.780 \times 10^{-9} \text{ s}^{-1}$	NCRP (1985)
Average beta decay energy	5.685 keV	NCRP (1985)
Maximum beta decay energy	18.6 keV	NCRP (1985)
Average track length (water)	0.56 μm	Okada and Momoshima (1993)
Maximum track length (water)	6.0 μm	Okada and Momoshima (1993)
Maximum track length (air)	5 mm	Okada and Momoshima (1993)
1 g of T_2 (gas)	$3.59 \times 10^{14} \text{ Bq}$ $9.7 \times 10^3 \text{ Ci}$	Okada and Momoshima (1993)
1 atom T per 10^{18} atoms H = 0.118 Bq HTO per L (water) = 3.2 pCi HTO per L (water)	1 tritium unit (TU) or 1 tritium ratio (TR)	Okada and Momoshima (1993)

However, this inventory continues to decrease by half every 12.3 years so long as there are no more additional nuclear weapons tests. Normal releases of tritium from nuclear facilities are estimated to produce $0.02 \times 10^{18} \text{ Bq/y}$ and off-normal releases an additional $0.001 \times 10^{18} \text{ Bq/y}$ with a resultant steady-state build up of $0.4 \times 10^{18} \text{ Bq}$ globally (Okada and Momoshima, 1993). Based on total previous levels of production, the legacy of luminous products (such as watch dials) is estimated to currently release $0.4 \times 10^{18} \text{ Bq/y}$ with a resultant steady-state build up of $7.4 \times 10^{18} \text{ Bq}$ globally (Okada and Momoshima, 1993). However, the luminous-dial source is expected to decrease in time.

The combined natural and anthropogenic emissions of tritium result in a current global inventory of approximately $61 \times 10^{18} \text{ Bq}$, which is about 60 times greater than tritium levels due to natural sources alone. Based on the dilution of this quantity of tritium in the circulating water volume of the earth, we estimate that this level of

tritium will result in a global average concentration of tritiated water in world's waters of 50 Bq/L. Based on the energy deposition in water from this level of tritium activity, we estimate that this tritium concentration produces an average radiological dose in humans that is less than 0.2% of the average background radiation dose of 0.0024 Sv (UNSCEAR, 1993) that is attributable to all natural sources of radioactivity—cosmic rays, terrestrial gamma rays, natural radionuclides in the body, and radon and its decay products.

The NTLF currently releases on the order of 3.7×10^{12} Bq/y (100 Ci/y) of tritiated water. This number is typical of annual releases since 1990 and is significantly lower than the 500 Ci/y (19×10^{12} Bq/y) that was more typical of releases during the period 1982 to 1990.

2.3 TRITIUM RELEASES FROM THE NTLF

The NTLF was established in 1982 to centralize tritium labeling activities which had been conducted at LBL since the early '70s. The operations of this facility are funded in part by the National Institutes of Health (NIH). Facility users are charged a fee which is used to offset monitoring and equipment costs. This facility accounts for 95% of all tritium releases from the entire LBL complex. There are currently six major tritium labeling facilities in the U.S. Of these, the NTLF at LBL is the only non-commercial facility.

Tritium diffuses easily through all types of materials, thus it is difficult to make any system fully resistant to tritium leakage. The NTLF has been designed and is operated in such a manner that there are multiple layers of defense to restrict the release of tritium to the environment.

Tritium comes to the NTLF facility in uranium beds, which are contained in sealed metal cylinders that are heated to release the chemically-bound tritium from the uranium in these beds. The beds are attached to a closed system inside a glove box. All formation reactions take place in the closed system within the glove box. There are three main components of this system that are designed to trap and re-circulate tritium so as to keep it from being released to the environment. These are (1) the glove-box ventilation system, the tritium recovery system, and the silica-gel traps; (2) the reaction manifold and the secondary U-bed; and (3) the chemistry lab hoods.

There are three major pathways by which tritium can enter the ambient air compartment from the NTLF. These pathways can be identified from Figure 3 below (the NTLF Exhaust Schematic). These release pathways include:

Figure 3 - NTLF Exhaust Schematic

- (1) Tritium in air drawn through the glove-box ventilation system, is directed through silica-gel traps, after which booster blowers direct it into the main stack trunk-line. The box-air would be contaminated mainly by fugitive losses through gaskets as well as some diffusional losses through the steel tubing.
- (2) Tritium in the reaction manifold which fails to sorb out in the secondary U-bed, is directed through a HTO recovery system. The gaseous residual from this process is then sent through silica-gel traps, onwards to a tributary pipe which ultimately joins the main stack-trunk-line. This tritium is most likely to be in oxidized form (i.e., tritiated water).
- (3) Off-gases passing through the chemistry lab hoods (i.e., laboratory air) as well as any off-gases from the pump-shed (including pumps, copper oxide furnaces and storage tank) are directed into the main stack trunk-line.

All postulated releases from the NTLF are to the air compartment in the form of tritiated water. When tritium is released to the environment—whether air, water, or soil—it spreads rapidly.

2.4 ENVIRONMENTAL DISTRIBUTION OF TRITIUM RELEASES FROM NTLF

The predominant influences on the relative distribution of HTO in the environment are the hydrogen cycling processes interacting between the various media (compartments) of the release environment, the water content of these media, and finally the radioactive decay rate of tritium (Murphy, 1993). The first two influences suggest the importance of understanding the water budget of the release site. The environmental cycling of tritium follows quite closely that of natural hydrogen as it occurs in gas, water, and organic molecules. Perhaps because of hydrogen abundance, no sequestering processes (bioaccumulation) in biota have evolved for hydrogen/tritium, and thus there is no mechanistic reason to expect biomagnification, which is a metabolically mediated build up in biological tissues. However, because hydrogen can bind to organic molecules, we might expect some accumulation as a result of organically bound tritium.

To gain some general insight regarding the principal factors affecting the distribution of HTO subsequent to a continuous atmospheric release, we conducted a literature review of field studies relating to the fate and transport of HTO in the environment. This insight was then employed in the development of a customized HTO fate and transport model. In the sections below, we summarize some of the major components of this model. More specific details on the model structure and the rationale for the model components are provided in Appendix A of this report.

2.4.1 Model Structure

We use a two-compartment model to estimate the steady-state distribution of tritiated water between air and soil media resulting from NTLF's continuous HTO emissions to the atmosphere. This structure is an idealization, constructed to allow the imposition of a mass balance on the released HTO in accordance with some of its well-studied affinities, as well as site-specific characteristics including physical properties (e.g., soil characteristics and relative humidity) and observed environmental processes (e.g., precipitation, runoff, and infiltration). Using a systematic accounting of the gains and losses for each compartment in isolation and for the whole system in concert, the model provides an algorithm for predicting steady-state HTO specific activities (concentrations) in the soil and air compartments. The overall model structure is illustrated in Figure 4.

The premises of our modeling approach are that HTO will distribute itself primarily in the aqueous phase of a compartment, that non-aqueous tritium (i.e., OBT) in any compartment can be accounted for by increasing the effective residence time and that under equilibrium conditions water-phase tritium activities in each compartment will be equal.

Figure 4. Compartment model for assessing the environmental fate of tritiated water emissions from the NTLF.

2.4.1.1 Steady State Mass Balance Equations

The steady-state equations describing gains and losses in the two compartments are used to solve for the steady-state inventory in each compartment. In Table 3, we list the gains and losses considered for each compartment. The following equations express gains and losses for the air and soil compartments respectively.

$$S + T_{sa} N_s = L_a N_a \quad (\text{air}) \quad (1)$$

and,

$$T_{as} N_a = L_s N_s \quad (\text{surface soil}) \quad (2)$$

In the equations above, an N represents a compartment's HTO inventory (note a compartment's bulk inventory and its water phase inventory are equivalent) and the T_{ij} ($i, j = a$ or s) are transfer rate constants, with units of day^{-1} , that express fraction per unit time of the inventory of compartment i that is transferred to compartment j . Here the total compartment activity in Bq is used to represent inventory (activities in Bq are proportional to molar inventories). The compartment abbreviations are a for air, and s for surface soil. The product of an N term and a T term is the rate of change of inventory in Bq/d. $L_i N_i$ represents all losses from compartment i , Bq/d. The term S in Equation 1 represents the rate of

Table 3. Summary of the processes by which HTO is exchanged between and lost from air and soil.

Compartment	Gains	Losses
(1) Air (both the gas and vapor phase — particulate phase ignored)	evapotranspiration from Biota and soil HTO load carried in with outside air (negligible for occupational volume) <i>NTLF HTO release</i>	precipitation scavenging radioactive decay loss to "outside" air via advective flushing
(2) Soil	diffusion from air (ignored) washout (scavenged portion) from precipitation loading in runoff entering from outside the 'unit world' (neglected since we calculate added risk)	diffusion to air (included in evapotranspiration) diffusion to deeper soil (lumped in with advection) advection to deeper soil loss carried away in runoff radioactive decay

HTO input (i.e., the NTLF's HTO emission rate) into the air compartment, Bq/d. Transfer-rate constants are functions of landscape characteristics and environmental mass transfer rates. Details on the derivation of transfer rate constants are provided in Appendix A.

2.4.1.2 Air Compartment (a)

The air compartment is represented by a simple box model in which losses include deposition to soil; convective losses; and radioactive decay. In the box model used for air, the inventory, N_a in Bq, is described by solving Equations 1 and 2. L_a is the sum of all loss-rate constants from the air compartment,

$$L_a = T_{as} + T_{ao} + \lambda , \quad (3)$$

$L_a N_a$ is the sum of all losses from the air compartment, Bq/d; T_{as} is the rate constant for advection and diffusion losses to soil, day^{-1} , and accounts for rain-water washout from air to ground-surface soil; T_{ao} is the rate constant for convective dispersion losses in air, day^{-1} and λ is the rate constant for losses due to radioactive decay, day^{-1} .

2.4.1.3 Soil Compartment (s)

The soil compartment represents the surface layer of soil in which losses include diffusion to air, diffusion and infiltration to deeper soil layers, runoff to surface water, and radioactive decay. The mass balance that defines the inventory, N_s , in Bq of HTO, is described by Equation 2. L_s is the sum of all loss-rate constants from the soil compartment,

$$L_s = T_{sa} + T_{\text{recharge}} + T_{\text{runoff}} + \lambda , \quad (4)$$

where L_s represents the total loss rate constant from the soil compartment, day^{-1} ; T_{sa} is the soil to air transfer rate primarily representing evapotranspiration processes, day^{-1} ; T_{recharge} and T_{runoff} are the rate constants for recharge losses to ground water and runoff losses to outside the unit world, respectively, day^{-1} ; and λ is the radioactive decay rate, day^{-1} . These loss-rate constants are given

2.4.2 Solutions for the Tritium Inventories

Equations 1 and 2 represent a system of two equations, with two unknowns and can be solved using substitution to determine the steady-state inventories, N_i , of contaminant in the soil and air compartments. This solution yields the following relationships,

$$N_s = \frac{S}{\left[\frac{L_s}{T_{as}} \times L_a - T_{sa} \right]} \quad \text{and} \quad (5)$$

$$N_a = \frac{S}{\left[L_a - T_{sa} \times \frac{T_{as}}{L_s} \right]} \quad (6)$$

These solutions are used in later sections to determine the inventory of tritium in the air, soil, and runoff from the site.

2.5 HUMAN CONTACT WITH TRITIUM RELEASES

Because the energy of electrons emitted during the decay of the tritium nucleus is insufficient to penetrate skin, only internal dose routes (inhalation and ingestion intake and uptake through skin) are of concern. Relevant to releases from the NTLF, pathways resulting in internal dose to tritium, include:

- 1) tritiated water enters the body via the lung through respiration
- 2) tritium in water, breast milk, or foods can be ingested and absorbed through the gastro-intestinal tract;
- 3) tritium from tritiated water-vapor in air can be taken through the skin, and
- 4) tritium from tritiated water in water can be taken through the skin during activities that involve dermal contact with the contaminated water, i.e. bathing, swimming, and wading.

2.5.1 Intake and Uptake

When environmental concentrations of tritium are constant over an exposure duration, ED, the population-averaged intake (for ingestion or inhalation routes) or uptake (for dermal contact) is the average daily exposure rate (AED_{ijk}), in Bq/kg-d and is given by

$$AED_{ijk} = \left[\frac{C_i}{C_k} \right] \times \left[\frac{IU_{ij}}{BW} \right] \times \frac{EF \times ED}{AT} \times C_k \quad (7)$$

In this expression $[C_i/C_k]$ is the intermedia-transfer ratio, which expresses the ratio of tritium concentration in the *exposure* medium i (i.e., personal air, tap water, milk, soil, etc.) to the tritium concentration in an environmental medium k (ambient-air, soil, surface water, and ground water, etc.) $[IU_{ij}/BW]$ is the intake or uptake factor per unit body weight associated with the exposure medium i and route j . For exposure through the inhalation or ingestion route, $[IU_{ij}/BW]$ is the daily intake rate per unit body weight of the exposure medium such as $m^3(\text{air})/\text{kg-d}$, $L(\text{milk})/\text{kg-d}$, or $\text{kg}(\text{soil})/\text{kg-d}$. For uptake through the dermal route, $[IU_{ij}/BW]$ is the uptake factor per unit body weight and per unit initial concentration in the applied medium, $(L(\text{water})/\text{kg-d}$ or $m^3(\text{air})/\text{kg-d}$). EF is the exposure frequency for the exposed individual, in days per year. ED is the exposure duration for the exposed population, in years. AT is the averaging time for the exposed population, in days; and C_k is the tritium concentration in environmental medium k .

2.5.2 Exposure factors

In constructing dose models one needs to define the characteristics of individuals in various age/sex categories and the characteristics of the microenvironments in which they live or from which they obtain water and food. This section defines both the types of anatomical and activity data needed to carry out the dose assessment and provides representative values for these parameters. Appendix B describes how these data are obtained and evaluated. For all exposure factors used in this assessment, we define both an arithmetic mean value and a coefficients of variation (CV), which is the arithmetic standard deviation divided by the arithmetic mean.

2.5.2.1 Exposure Duration and Averaging Time

In the basic exposure model, the exposure duration (ED) is the amount of time, in years, that the exposed population is assumed to be in contact with a specified environmental contaminant. The averaging time (AT) is the period, in days, over which exposure is averaged. The estimates of risk provided below are based on population exposures and the committed dose in a defined population on an annual basis. Thus, for this risk assessment ED and AT represent the same period of time and the ratio ED/AT in Equation 7 is $[1 \text{ y}]/[365 \text{ d}]$.

2.5.2.2 Anatomical Properties, Inhalation, and Ingestion

In Appendix B, we calculate the arithmetic-mean and CV of body weight and body surface area for three age groups—infant, child, and adult. The body weight for infants is needed for estimating exposures to contaminants in breast milk. We also evaluate available data to assess values for population breathing rates, ingestion of water during recreational activities, ingestion of breast-milk by infants, and ingestion of home-grown fruits vegetables and grains. The descriptions, parameter symbols, and values we obtained for these inputs are summarized in Table 4.

2.5.2.3 Activity Patterns and Exposure Times

Activity patterns define the frequency and amount of times that individuals allocate to activities that could result in exposures to tritium contaminated media—air, water, soil. For occupational exposures the exposure time per day is typically in the range of 8 to 10 hours and the exposure frequency (EF) is in the range of 200 to 300 days per year, assuming 50 working weeks per year and from 4 to 6 working days per week. For non-occupational exposures, the U.S. EPA (1989a) reports the average time spent at home indoors as 8 to 16 h/d and the time spent at home outdoors is 0.3 h/d. We estimate the CV in these latter numbers to be 0.14.

In order to assess doses that might occur during water-recreation activities associated with creeks running down from the Berkeley Hills, we need to estimate the number of days per year and hours per day spent in contact with surface water during swimming or other water recreation. The U.S. EPA (1992a) reports that for recreational swimming, the time spent swimming ranges from a typical value of 0.5 h/d to an upper bound of 1 h/d and the frequency ranges from a typical value of 5 d/y to an upper bound of 150 d/y. From this we obtain a mean exposure time of 0.5 h/d with a CV of 0.5, and because of climate and proximity to water, we use 15 d/y (CV of 4) as representative of exposure frequency in California. Values and units of activity patterns and exposure times are listed in Table 5.

2.5.3 Dermal Uptake

As was noted above, for dermal exposures, the term $[IU_{ij}/BW]$ in Equation 7 represents the uptake factor per unit body weight and per unit initial concentration in the applied medium, that is (L(water)/kg-d from water or m³(air)/kg-d) from air. In the subsections below, we derive these factors for tritium.

Table 4. Values of human anatomical and intake properties used in the exposure calculations.^a

Parameter, symbol	Child ^b		Adult ^b		Combined ^b		Units
Body weight of infants age 0 to 1 y, BW	7.2	(0.3)	--		--	--	kg
Body weight, BW	29	(0.24)	71	(0.2)	62	(0.2)	kg
Surface area, SA _b	0.032	(0.09)	0.024	(0.06)	0.026	(0.07)	m ² /kg
Working breathing rate, BR _w	--	--	0.030	(0.3)	--	--	m ³ /kg-h
Active breathing rate, BR _a	0.023	(0.3)	0.018	(0.3)	0.019	(0.3)	m ³ /kg-h
Resting breathing rate, BR _r	0.008	(0.3)	0.006	(0.2)	0.0064	(0.2)	m ³ /kg-h
Breast milk intake, I _{bm} ^c	0.11	(1)	--		--		kg/kg-d
Water intake during during recreation, I _{flr}	0.0007	(1)	0.0007	(1)	0.0007	(1)	L/kg-h
Ingestion of homegrown exposed produce, I _{ep}	0.0016	(0.7)	0.00078	(0.7)	0.00096	(0.7)	kg/kg-d
Ingestion of homegrown unexposed produce, I _{up}	0.00095	(0.7)	0.00053	(0.7)	0.00062	(0.7)	kg/kg-d

^a Listed are the arithmetic-mean value and (in parentheses) the estimated coefficient of variation (CV), equal to the standard deviation divided by the mean. Body weights are from Najjar and Roland (1987), breathing rates are from ICRP (1975), tap water intakes are from Yang and Nelson (1986) and Ershow and Cantor (1989) and food intakes are from Yang and Nelson (1986).

^b The child category covers ages 0 to 15, the adult category covers ages 16 to 70, the combined category is used to represent lifetime equivalent exposure and is obtained by multiplying the child category by 15/70, the adult category by 55/70, and then summing these products.

^c Breast-milk intakes are from Butte et al. (1984) and Whitehead and Paul (1981).

2.5.3.1 Dermal Uptake from Air

For individuals who are at rest during the time of exposure, the amount of tritium entering the body by dermal uptake is approximately equal to that entering the body by inhalation (Hill and Johnson, 1993, Pinsom and Langham, 1980). For an individual at rest, the ratio of inhalation intake per unit body weight (Bq/kg-h) to air concentration (Bq/m³) is 0.0064 m³/kg-h as defined in Table 4. For dermal

Table 5. Values and units of exposure times and exposure frequencies used to characterize activity patterns.

Activity pattern	Mean value	Range	Units	Reference
Exposure time at work, ET_w	9	8 to 10	h/day	this report
Exposure frequency at work, EF_w	250	200 to 300	day/y	this report
Exposure time, at home indoors, ET_h	12	8 to 16	h/day	U.S. EPA (1989a)
Exposure time, at home outdoors, ET_o	0.3	0.02 to 0.4	h/day	U.S. EPA (1989a)
Exposure frequency at home, EF_h	365	n/a	day/y	assumed
Exposure time in surface water, ET_{sw}	0.5	0 to 1	h/d	U.S. EPA (1992a)
Exposure frequency to surface water, EF_{sw}	15	5 to 60	day/y	U.S. EPA (1992a)

uptake, the ratio of dermal uptake to air concentration is the product of the effective skin permeability (m/h) and the mass-specific surface area of the body,

$$\text{Dermal uptake rate (Bq/kg-h)} = C_{\text{air}} (\text{Bq/m}^3) \times K_p^{\text{air}} (\text{m/h}) \times SA_b (\text{m}^2/\text{kg}) \quad (8)$$

$$\frac{\text{Dermal uptake rate}}{C_{\text{air}}} = 0.0064 \text{ m}^3/\text{kg-h} = K_p^{\text{air}} \times SA_b \quad (9)$$

where K_p^{air} is the effective permeability of tritium in air through human skin and SA_b is the ratio of body surface area to body mass as described above and listed in Table 4. The implication of this relationship is that K_p^{air} is on the order of 0.25 m/h which is somewhat high but not inconsistent with permeabilities that have been measured for organic vapors (EPA, 1992).

2.5.3.2 Dermal Uptake from Water

Dermal uptake of tritium from water is used to address recreational scenarios, such as wading and swimming in surface waters. As was the case for air, the dermal

uptake of tritium from air is based on the measured ratio of uptake to concentration. Based on such experiments, the permeability of water on skin has been measured as 1.5×10^{-5} m/h (EPA, 1992). Since the diffusion coefficients of tritiated water (T_2O) in air and water are essentially the same as those of protiated water (H_2O) (NCRP, (1985), this value is assumed to be appropriate for tritiated water.

2.6 THE RELATION BETWEEN TRITIUM EXPOSURE, RADIOLOGICAL DOSE AND HEALTH EFFECTS

The purpose of this section is to illustrate the process by which exposures to tritium are linked to potential health impacts within the exposed population. We begin with a review of the potential health effects associated with tritium and establish the relation between tissue concentration and radiological dose. We next look at metabolism and dosimetry in order to establish the appropriate relation between exposure and dose. We consider regulatory standards that have been established to limit exposure and dose. We conclude with a derivation of the risk factors that can be used to estimate the incidence of health impacts within the exposed population.

2.6.1 Health Risks Associated with Tritium

Health risks associated with exposures to tritium are attributable to the ionizing radiation emitted by tritium. In order to quantify the likely magnitude of these effects, it is necessary to address tritium metabolism and dose in terms of radiation biology and radiation protection methods. The biological effects of ionizing radiation are brought about through chemical changes in the cells caused by ionizations, excitations, dissociations, and atom displacements. The health effects of ionizing radiations—cancer, genetic defects, and reproduction effects—are proportional to the energy carried by the radiation and delivered to living cells.

Each decay of a tritium atom releases energy that is carried by the resulting beta particle. The rate of decay is expressed as the product of the number of atoms N and the decay constant which is equal to 0.693 divided by the half-life, $T_{1/2}$,

$$\text{decays per unit time} = N \times 0.693/T_{1/2} \quad (10)$$

Decay rates are expressed using becquerels (Bq). One becquerel is equal to one decay per second. Decay rates may also be expressed using a curie (Ci) which is equal to 3.7×10^{10} decays per second. The energy released by these decays and absorbed in living tissue is equal to the rate of decay times the average beta decay energy E_β (in

joule/decay). The energy absorbed in a tissue is equal to the number of beta particles deposited in the tissue per unit time divided by the tissue mass, M_t

$$\text{energy absorption rate} = \frac{E_\beta \times N \times 0.693}{T_{1/2} \times M_t} \quad (11)$$

The gray (Gy) is a unit of measure used to quantify energy absorption in living tissues. One Gy is taken to represent one joule of energy deposited per kilogram (kg) of tissue. In the past, energy absorption was reported using a *rad* which is equal to 0.01 Gy.

There are very few available studies on tritium-induced health injury in humans. Thus, health-risk estimates specific to tritium must be based on the large number of experiments with animals and cell cultures (Straume, 1993). These experiments show that exposure to tritiated water results in mutations and cell disruptions that can lead to cancer, heritable genetic defects, and reproductive and developmental effects (Straume, 1993). These effects are qualitatively similar to the injuries caused by low linear-energy-transfer (LET) radiations produced by cesium-137 and cobalt-60 such as x-rays and gamma-rays for which a significant amount of human data is available. The quantitative difference between tritium beta emissions and a standard low-LET radiation is expressed by the relative biological effectiveness (RBE). The RBE for tritium ranges from 1 to 3 (Straume, 1993). This means that although the health effects of the beta doses from tritium are believed to be qualitatively similar to those of gamma rays, quantitatively these beta rays are believed to be somewhat more potent per unit energy deposition than gamma rays.

2.6.2 Tritium Metabolism and Dosimetry

There is general consensus that the current understanding of the metabolism of HTO is sufficient for radiation protection purposes (Hill and Johnson, 1993). When it enters the body, tritiated water quickly reaches equilibrium with hydrogen in the body and imparts a uniform dose to all soft tissues. The ICRP (1978) recommends the assumption that internalized HTO is completely absorbed and mixed rapidly with total body water so that the concentration in all body waters—blood, urine, sweat, sputum, insensible perspiration, and exhaled water vapor—is the same. However, following HTO exposures, a small fraction of tritium will eventually be incorporated into organic molecules. This tritium is referred to as organically bound tritium (OBT) and will have different metabolic rates than those for total body water. There is no preferential distribution of OBT to any specific body tissue.

Therefore, both HTO and OBT can be assumed to be uniformly distributed throughout the body, once taken in by some exposure route.

Information gained from human studies indicates that the retention of tritium can generally be characterized by a two or more exponential functions. These three components have biologically half-lives in the respective ranges of 6 to 12 days, 10 to 34 days, and 130 to 550 days with respective median values of 9, 22, and 340 days (Hill and Johnson, 1993). When HTO is ingested, the attached tritium atom is rapidly distributed to both body water and to other hydrogen containing organic molecules. The first of the biological half-lives described above corresponds to the body water metabolism of hydrogen and reflects the turnover of water within the pool of body water. The second component appears to be related to tritium involved in carbon-tritium bonds and the last component represents tritium incorporated into organic molecules having very slow turnover rates (Hill and Johnson, 1993).

Tritium can also enter the body as organically bound tritium (OBT) through food products. The fate of OBT after ingestion is dependent primarily on the hydrogen metabolism of the human body. Until recently, direct uptake and metabolism of OBT was generally ignored in models of tritium dosimetry. However, recent reports using more advanced environmental tritium models indicate that ingestion doses from OBT in food could be much higher than doses from equivalent quantities of tritium ingested or inhaled as HTO (Etnier et al., 1984). In order to properly assess combined exposures to HTO and OBT, we describe the retention of tritium following exposure with a model that accounts for intake of both. Following the recommendation of Hill and Johnson (1993), we use the four-compartment model proposed by Etnier et al., (1984) to describe tritium retention. One compartment is used to represent the retention of tritium in body water and three compartments are used to represent the retention of tritium in different types of organic molecules. This tritium metabolism model is illustrated in Figure 5.

In this model, the λ parameters represent rate constants for the transfer of tritium among compartments. These parameters are measured by studying the retention and loss of tritium in human and animal subjects. Some of these parameters can be determined directly from curves of tritium retention in the body. For example, λ_{wo} is equal to $0.693/T_{1/2}^w$, λ_{lw} is equal to $0.693/T_{1/2}^l$, and λ_{no} is equal to $0.693/T_{1/2}^n$; where $T_{1/2}^w$, $T_{1/2}^l$, and $T_{1/2}^n$ are the biological half-lives which reflect, respectively, the turnover of hydrogen within the pool of body water, labile tritium involved in non-water bonds, and tritium incorporated into organic molecules having very slow turnover rates. Other λ parameters are derived from steady-state distributions of

Figure 5. Four-compartment model for tritium metabolism as applied to the intake or uptake of tritiated water (HTO) and organically-bound tritium (OBT). The four compartments are body water (w); the rapid turnover organically bound tritium compartment, which contains labile tritium (l); the fast-fat compartment in which there is rapid turnover of non-labile, organically-bound tritium (f); and the slow-turnover compartment in which there is slow turnover of non-labile, organically-bound tritium (n). The λ parameters represent rate constants for the transfer of tritium among compartments and relate to the half-lives in these compartments.

tritium activity. Etner et al., (1984) report the following values (with units of inverse day), λ_{wl} is equal to 0.0005463, λ_{wf} is equal to 0.00416, λ_{fw} is equal to 1.5472, and λ_{fn} is equal to 0.10328. Under steady-state conditions, these parameters can be used to assess the body burden associated with a continuous input of tritium as HTO or OBT by estimating the effective tritium half life, $T_{1/2}^{tl}$, for a given intake. When HTO is ingested, based on the ranges of compartment half-lives discussed above, the model in Figure 5 gives an estimated mean value of $T_{1/2}^{tl}$ equal to 15 days with a coefficient of variation of 0.2. When OBT is ingested into either the labile or fast fat compartment, the model in Figure 5 gives an estimated mean value of $T_{1/2}^{tl}$ equal to 48 days with a coefficient of variation of 0.3. Based on the appropriate effective half-

life $T_{1/2}^{\text{trit}}$, the total amount of tritium retained in the body, Q_{total} , Bq is calculated from

$$Q_{\text{total}} \text{ (Bq)} = \frac{\text{IU} \times T_{1/2}^{\text{trit}}}{0.693} \quad (12)$$

From this steady state inventory, and the body weight, BW, in kg, we calculate the total body burden (in Bq/kg) and the corresponding dose rate in Gy per year as

$$\text{burden (Bq/kg)} = \frac{\text{IU} \times T_{1/2}^{\text{trit}}}{0.693 \times \text{BW}} \quad (13)$$

$$\text{Dose rate (Gy/y)} = \text{burden} \times 5.685 \frac{\text{keV}}{\text{dist}} \times 3.15 \times 10^7 \frac{\text{sec}}{\text{year}} \times 1.6 \times 10^{-16} \frac{\text{Joule}}{\text{keV}} \quad (14)$$

2.6.3 Regulatory Guidance Relevant to Tritium

International, federal, and state agencies have all set standards for controlling human exposures to tritium in both occupational and nonoccupational settings.

For the purposes of radiation protection in an occupational setting, the ICRP (1978) has recommended the use of both an annual limit on intake (ALI) and a derived air concentration (DAC) for tritiated water (HTO). The DAC is the air concentration that would result in the ALI through inhalation and dermal routes as result of exposure times and exposure frequencies typical in the work environment (2000 hours of exposure per year and 2400 m³ of air as a annual breathing volume). The 1978 recommendation of the ICRP is that the ALI for tritiated water be 3 billion Bq (0.08 Ci) and that the corresponding DAC be 800,000 Bq/m³ (22 µCi/m³). This number corresponds to a committed annual effective dose to workers of 0.05 Sv (5 rem). Based on the risk factor of 0.04 fatal cancers per Sv recommended for low-dose exposures to working populations by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 1993), 0.05 Sv corresponds to a fatal cancer risk of 0.002 for the working population exposed to these levels. Recently, the ICRP (1991) has recommended the reduction of the committed effective dose limit for workers from 0.05 Sv to 0.02 Sv. With this recommendation, the ALI and DAC for tritiated water become respectively 1 billion Bq and 300,000 Bq/m³. These new

values correspond to a fatal cancer risk of 0.0008 for the working population exposed to these levels.

Currently, the Code of Regulations of the United States (10 CFR-835, 1994) follows the 1978 ICRP guidelines and specifies that exposure at U.S. Department of Energy (DOE) facilities not exceed a committed annual effective dose to workers of 0.05 Sv. Based on this standard, the code recommends the same ALI and DAC for tritium as are recommended by the ICRP and listed above. In addition, the federal code specifies that the dose limit for the embryo/fetus from the period of conception to birth as a result of occupational exposure of a pregnant worker is 0.005 Sv (0.5 rem); that any minor exposed to radiation or radioactive material during direct on-site access at a DOE site or facility shall not exceed 0.001 Sv (0.1 rem) total effective dose equivalent; and that any member of the public exposed to radiation and/or radioactive material during direct on-site access at a DOE site or facility shall not exceed 0.001 Sv (0.1 rem). To guarantee compliance with these latter guidelines requires the use of a limit on tritium air concentrations of roughly 8,000 Bq/m³ of HTO.

The Code of Regulations of the United States also specifies maximum contaminant levels in air (40 CFR-61, 1993) and water (40 CFR-141, 1994) for beta-particle activity from man-made radionuclides released to unrestricted areas. For emissions to air, the code of regulations specifies that emissions of radionuclides to the ambient air from Department of Energy facilities should not exceed those amount that would cause any member of the public to receive in any year an effective dose equivalent of 0.0001 Sv (0.01 rem). Based on scaling the DAC for working environments from a risk of 0.05 to 0.0001 Sv and adjusting for an annual exposure time of 8,760 h (i.e., 24 h/d for 365 d/y) versus 2,000, we estimate that the ambient concentration limit corresponding to continuous exposure for a member of the public is on the order of 365 Bq/m³ (10,000 pCi/m³). The maximum contaminant levels for tritium in water is specified in the federal code of regulations as 740 Bq/L (20,000 pCi/L). The State of California uses federal standards for tritium concentration limits in air and water (California Environmental Regulations Section 64443. Man-Made Radioactivity, 1993).

2.6.4 Risk Factors: The Relation Between Radiological Dose and Health Effects

Detrimental effects of toxic chemicals on humans, animals, and plants can be classified as "stochastic" or "nonstochastic" effects (ICRP, 1977). Stochastic effects are those for which the probability of an effect occurring, rather than the severity of effect is proportional to dose; without threshold. Nonstochastic effects are those for which the severity of effect is a function of dose and for which a threshold may exist.

For example the human health effects of carcinogens and many types of genetic effects are assumed to be stochastic. In contrast, the effects of neurotoxins, such as lead and mercury, are assumed to be nonstochastic. The detrimental effects of body burdens of tritiated water are stochastic and include cancer, genetically transmitted defects, and developmental and reproductive effects. Risk factors that can be used to estimate the likely occurrence of these effects in exposed populations are presented in this section

Over the last decade, there have been a large number of radiobiological studies, which together with previous studies, indicate that tritium in body water produces the same portfolio of radiogenic effects observed following whole-body exposure to penetrating ionizing radiations such as x-rays and gamma rays (Straume and Carsten (1993). This portfolio of effects includes cancer, heritable genetic defects, and reproductive and developmental effects. Nevertheless, it has been observed and reported that the beta rays produced by tritium have greater biological effectiveness (i.e., damage potential) than x-rays or gamma rays of the same energy (Straume and Carsten, 1993).

As is illustrated above, an annual dose equivalent from tritium exposure can be calculated from tritium concentration in body water by converting this body-water concentration to an estimate of equivalent whole-body energy deposition in Gy. Based on the average dose in Gy for a population the risk factors described in the sections below can be used to predict the potential incidence of detrimental effects.

2.6.4.1 The Risk Factor for Cancer, R_{HTO}^{can}

Exposures to tritiated water appear to induce DNA changes, indicating that HTO is an initiator of cancer. In addition, there is evidence that HTO can cause transformations in mutant stem cells and thus enhance later stages of the progression from normal to cancer cells, thus indicating that HTO might also be considered a promoter of cancer. Types of cancers associated with HTO dose include leukemia and nonleukemia (primarily soft-tissue carcinomas) (Straume, 1993).

Straume (1993) has developed a stochastic dose-response model of fatal cancer risk for HTO doses based on the human dose-response data for human cancer data and a modification for the biological effectiveness of HTO relative to gamma rays. This model expresses the probability of cancer per Gy of dose and is illustrated in Figure 6. In this model the risk factor for cancer incidence within a population has respective median and mean values of 0.080 and 0.089 fatal cancers per Gy of dose. The range of this distribution is from 0.018 to 0.580, and the coefficient of variation is approximately 1.3.

Figure 6. The stochastic cancer risk factor for population doses of tritiated water as derived from the model of Straume (1993).

2.6.4.2 The Risk Factor for Heritable Genetic Effects, R_{HTO}^{gen}

To estimate genetic risk to humans from low-level HTO exposures, we use a model developed by Straume (1993), who has used ranges and distributions of gamma-ray-induced risks to construct a stochastic-dose response function for the number of heritable genetic defects expected per Gy of population dose. The distribution of defects per unit dose obtained from this model is shown in Figure 7. This distribution has respective median and mean values of 0.006 and 0.0066 defects per live-born Gy of population dose in the first generation. The range of this distribution is from 0.0014 to 0.033, and the CV is approximately 1.3.

2.6.4.3 The Risk Factor for Developmental and Reproductive Effects, R_{HTO}^{rep}

To estimate reproductive and developmental risk in humans from low-level HTO exposure, we use the model of Straume (1993) who has used ranges and distributions of gamma-ray-induced risks to construct a range of likely values for the dose-response function for THO doses. From these data we developed a stochastic-dose response function for the number of reproductive and developmental effects expected per Gy of dose to the live-born infant. The probability density function of this model is shown in Figure 8. This distribution has respective median and mean values of 0.15 and 0.17 defects live-born Gy of population dose. The range of this distribution is from 0 to 0.4, and the CV is approximately 0.45.

Figure 7. The stochastic distribution of the genetic risk factor expressed as the number of heritable defects live-born Gy of population dose in the fetus.

Figure 8. The distribution of reproductive and developmental effects associated with population doses of tritiated water expressed per average dose in Gy within that population. This model is based on recommendations of Straume (1993).

2.7 ESTIMATED HEALTH RISKS FOR TRITIUM RELEASES FROM THE NTLF

In this section, we carry out the quantification of potential health risks to the populations in and near the NTLF facility. The purpose of this assessment is to determine how doses from tritium and potential health effects might be distributed among the occupational and nonoccupational populations of LBL, the University of California (UC), and the cities of Berkeley and Oakland. Three geographic "zones" and associated populations are defined in order to carry out this analysis (see Figure 9). The first zone includes the natural bowl that surrounds the NTLF facility and its tritium release stack. The population potentially at risk within this zone is the working population and their off-spring. The next zone includes an area that encompasses the entire LBL site and the UC property that encompasses the Lawrence Hall of Science, the Samuel Silver Space Sciences Laboratory, the Mathematical Sciences Research Institute, and the Botanical Gardens as well as the single-family residences along the ridge above the site, which is part of Oakland. The third zone includes the main UC campus, the city of Berkeley and the associated populations that could be exposed to tritium that is washed from the air into surface-water runoff and carried down into Strawberry Creek.

2.7.1 Tritium Concentrations in the Air, Water, and Soil of the NTLF Environment

This first step of our analysis is to characterize the concentration of tritium in the air, water, and soil of the proximate NTLF environment. These tritium concentrations provide the basis for estimated tritium concentrations in each of the three exposure zones. These concentrations are estimated using the environmental transport model described above and are then compared to site-specific measured values.

Landscape, climate and hydrology parameters used in the analysis are derived from site-specific information. These parameter values are summarized in Table 6. The references and methods used to obtain these values are describe in some detail in Appendix C of this report.

2.7.1.1 Source Term

The NTLF currently releases approximately 100 Ci/y (3.7 trillion Bq/y) which has been typical of annual releases since 1990. This value is lower than the 500 Ci/y (19 trillion Bq/y) that was more typical of releases during the period 1982 to 1990. According to the 1990 LBL Site Environmental report, essentially all of LBL's tritium emissions originate from the NTLF (LBL Site Report, 1990). The total inventory of

Table 6. Landscape properties for the NTLF Environment.

Parameter description	Symbol	Mean value	CV	Reference(s)
Yearly average wind speed, m/s	v_w	3	0.3	Merry (1991) (NOAA, 1974)
Yearly average temperature, °C	T	12.5	0.2	Merry (1991) (NOAA, 1974)
Yearly average relative humidity, %	RH	79	0.04	Merry (1991) (NOAA, 1974)
Soil compartment depth, m	d_s	2.5	0.5	(Thibodeaux, 1994) (Murphy, 1993)
Moisture content of soil, L(water)/L(soil)	ϕ_s	0.3	0.1	van der Leeden et al. (1991)
Annual average precipitation, m/y	$rain$	0.64	1.0	Merry (1991) (NOAA, 1974)
Annual average evapotranspiration, m/y	$evapotrans$	0.45	0.14	van der Leeden et al. (1991)
Annual average runoff, m/y	$runoff$	0.13	0.26	van der Leeden et al. (1991)
Infiltration to ground water	$recharge$	0.06	0.26	this report
Scavenging efficiency	AQ_{rain}	0.4	0.2	(Murphy, 1993)

tritium which is attached to the U-beds is roughly 10,000 Ci—the U-beds are designed with a stoichiometric capacity for 25,000 Ci of tritium, and thus are always under-utilized. The release of the entire inventory, even in the case of an extreme event, is unlikely. In an effort to be highly conservative, this study also evaluates the risk that would be associated with a continuous release ten times the current release rate. It is highly unlikely that such a release rate, 1000 Ci/yr, could ever occur. It is even less likely that it would persist for an extended period (i.e., more than a few days). Note that a catastrophic accident would be a one time release and certainly would not persist.

2.7.1.2 Source Area and Mixing Height of the Atmosphere Near the NTLF

The base of the NTLF stack is at an elevation of 1040 ft (315 m) and sits two thirds of the way up in a natural bowl or amphitheater. We used the volume of this bowl to characterize the initial mixing volume of a stack release (see Figures 9 and 10). We define the top lip of the bowl to be the Lawrence Hall of Science, which has an

Figure 10

elevation of roughly 1150 ft (350 m). The lower extent of the bowl is formed by the western extent of the parking lot for buildings 75, 75A, 75B, and 69, which has an elevation of roughly 980 ft (300 m). Note from Figure 10 that the radius defining our occupational volume extends west beyond the parking lot including a partial area having a lower elevation than that of the parking lot. A mixing height appropriate for the Zone-1 volume is taken to be the difference in elevation between the top lip of the bowl (LHS) and the lower extent of the parking lot, resulting in a height of 50 m.

2.7.1.3 Predicted Concentrations in the Vicinity of the NTLF, Zone 1

The total volume of air for characterizing exposure in Zone 1 is estimated to be 125,000 m³ and the volume of soil in this zone is estimated to be 6,250 m³. Based on these volumes, the landscape properties defined in Table 6, and the model equations in Appendix A, we estimated the following tritium turnover rates for the air and soil compartments in Zone 1.

Air

Turnover rate from air to soil, T_{as}	0.29 per day
Turnover rate out of the air volume by wind, T_{ao}	1190 per day
Effective environmental half-life of tritium in air, $1/L_a$	0.00058 day

Soil

Turnover rate from soil to air, T_{sa}	0.0016 per day
Turnover rate out of the soil by leaching, T_{so}	0.00070 per day
Effective environmental half-life of tritium in soil, $1/L_s$	280 days

Table 7 lists the air, soil, and runoff water concentrations of tritium that are predicted from our model for Zone 1 based on these turnover rates used in Equations 5 and 6 under the assumption that there are continuous stack releases of 100 Ci/y and 1000 Ci/y. We note that for releases of 100 Ci/y, the predicted concentrations are below the regulatory standards for the ambient (i.e., nonoccupational) environment, 365 q/m³ in air and 740 Bq/L in water as discussed in Section 2.6.3 above. For the extreme case where there is a continuous release of 1000 Ci/y, the concentrations exceed federal limits for nonoccupational level but are well below the occupational levels.

Table 7. Tritium concentrations and inventories in Zone 1.

Compartment	Tritium concentration, Bq/m ³	Tritium inventory, Bq
Assuming a release of 100 Ci/y		
Air	70	8×10^6
Soil	1×10^5	1×10^9
Runoff water	5×10^5 (500 Bq/L)	n/a
Assuming a release of 1000 Ci/y		
Air	700	8×10^7
Soil	1×10^6	1×10^{10}
Runoff water	5×10^6 (5,000 Bq/L)	n/a

2.7.1.4 Predicted Concentrations in the Second Exposure Zone, Zone 2

Zone 2 includes an area that encompasses the entire LBL site and the UC property that encompasses the Lawrence Hall of Science, the Samuel Silver Space Sciences Laboratory, the Mathematical Sciences Research Institute, and the Botanical Gardens as well as the single-family residences along the ridge above the site. The Berkeley Hills reach an elevation of 2000 ft (610 m) which is used in defining a mixing height for the residential or regional volume. Temperature inversions have a frequency of 66%. According to Merry (1991) the LBL site is either within an inversion layer or below one located at the top of the Berkeley Hills 40 % of the time. Based on this information, we use 300 m as the mixing height for Zone 2. The main LBL site occupies 134 acres ($550 \times 10^3 \text{ m}^2$) and this is roughly half the area of the second zone. Thus, we use $1 \times 10^6 \text{ m}^2$ to represent the area of Zone 2.

The total volume of air for characterizing exposure in Zone 2 is estimated to be $3 \times 10^8 \text{ m}^3$ and the volume of soil in this zone is estimated to be $2.5 \times 10^6 \text{ m}^3$. Based on these volumes, the landscape properties defined in Table 6, and the model equations in Appendix A, we estimated the following tritium turnover rates for the air and soil compartments in Zone 2.

Air

Turnover rate from air to soil, T_{as}	0.15 per day
Turnover rate out of the air volume by wind, T_{ao}	60 per day
Effective environmental half-life of tritium in air, $1/L_a$	0.012 day

Soil

Turnover rate from soil to air, T_{sa}	0.0016 per day
Turnover rate out of the soil by leaching, T_{so}	0.00070 per day
Effective environmental half-life of tritium in soil, $1/L_s$	280 days

Table 8 lists the air, soil, soil runoff water, vegetation HTO, and vegetation OBT concentrations of tritium that are predicted from our model for Zone 2 based on these turnover rates used in Equations 5 and 6 under the assumption that there are continuous stack releases of 100 Ci/y and 1000 Ci/y. The HTO levels in vegetation are estimated under the assumption that vegetation water is a mix of 75% atmospheric water and 25% soil water (Murphy, 1993; Anspaugh et al., 1973). The OBT levels in vegetation are based on the estimate of Diabate and Strack (1993) that the OBT compartment contains about 3% of the hydrogen in fresh-plant material such as vegetables. We assume that the OBT level in fresh vegetables is 0.03 times HTO levels in plant water.

Table 8. Tritium concentrations and inventories in Zone 2.

Compartment	Tritium concentration, Bq/m ³	Tritium inventory, Bq
Assuming a release of 100 Ci/y		
Air	0.6	2×10^8
Soil	4×10^3	1×10^{10}
Runoff water	1×10^4 (10 Bq/L)	n/a
Vegetation (HTO levels)	30 Bq/kg (fresh mass)	n/a
Vegetation (OBT levels)	0.9 Bq/kg (fresh mass)	n/a
Assuming a release of 1000 Ci/y		
Air	6	2×10^9
Soil	4×10^4	1×10^{11}
Runoff water	1×10^5 (100 Bq/L)	n/a
Vegetation (HTO levels)	300 Bq/kg (fresh mass)	n/a
Vegetation (OBT levels)	9 Bq/kg (fresh mass)	n/a

We note that for both releases of 100 Ci/y and 1000 Ci/y the predicted concentrations are below the regulatory standards for the ambient (i.e., nonoccupational) environment, 365 q/m³ in air and 740 Bq/L in water as discussed in Section 2.6.3 above.

2.7.1.5 Predicted Concentrations in the Third Exposure Zone, Zone 3

The third zone for which an analysis is carried out is the area of the main UCB campus and the city of Berkeley and the associated populations that could be exposed to tritium that is washed from the air into surface-water runoff and carried down into Strawberry Creek. Only a small fraction of the water flowing down Strawberry Creek originates from the Grizzly Area, which coincides well with our first "occupational volume". Assuming most of the stream flow stems from runoff, this fraction could be estimated as:

$$F_{S,bwl} = A_{bwl} / A_{S,Ctchmt} \quad (15)$$

where $F_{S,bwl}$ is the fraction of Strawberry Creek flow originating from the "bowl area", A_{bwl} square area of the Zone-1 volume and $A_{S,Ctchmt}$ is the total area of the

Strawberry-Creek catchment. We have not made a precise determination of $A_{S,Ctchmt}$, thus we estimate it as being one half of the LBL site area. The main LBL site occupies 134 acres ($550 \times 10^3 \text{ m}^2$), thus $F_{S,bwl}$ is found to be:

$$F_{S,bwl} = 25,000 / (275 \times 10^3) = 0.10 \quad (16)$$

The above is a really rough estimate and could be markedly improved with some effort. Maps clearly showing the Strawberry Creek catchment area could be used to better estimate, $A_{S,Ctchmt}$. In addition, the assumption that all creek flow arises from runoff should be confirmed.

We estimate that the tritium concentration in Strawberry Creek would be some 10% of the tritium water concentration found in Zone 1. Based on a mass-balance model of water evaporation in the area of the UC Campus, we estimate that evaporation from Strawberry Creek could contribute no more than about 0.01% to the total water inventory in the air mass associated with the campus. This analysis is carried out as follows. We assume that the flow path of Strawberry Creek through the UC Berkeley campus and city is on the order of 2000 m (1.3 mi) and that it has an

average width of 1 meter and depth of 0.5 meter. For evaporation of water from fast-moving water bodies, Southworth (1979) has shown that the rate of water evaporation is limited by the rate of mass transfer of saturated vapor in the air above the water. Furthermore, he has shown that this evaporation rate depends on the wind speed over the water, the water flow velocity, and difference between the saturated and average relative vapor concentrations in air and is given by the following expression,

$$\text{evaporation rate (m/d)} = 0.00316 \times 86400 \text{ s/d} \times (v_w + \text{current}_w) \times [\phi_a(100\%) - \phi_a], \quad (17)$$

where v_w is the yearly average wind speed (3 m/s from Table 6), current_w is the flow rate of the stream (1 m/s assumed), $\phi_a(100\%)$ is the water content of the air at saturation and ϕ_a is the water content of the air at the average relative humidity. Using Equations A2 and A3 in Appendix A with an assumed daytime temperature of 293 K and a yearly average daytime relative humidity of 70%, we obtain $\phi_a(100\%) = 1.7 \times 10^{-5}$, $\phi_a = 1.2 \times 10^{-5} \text{ m}^3(\text{water})/\text{m}^3(\text{air})$ and an evaporation rate of 0.005 m/d. Based on the stream dimensions given above, this amounts to an total mass evaporation of 5 m³ per day into the surrounding volume. If we assume the volume of air associated with the stream is 1 km square by 100 m high and use the residence time of this air mass based on the box model described above (0.02 day) then the water coming from Strawberry Creek is on the order of 7.4×10^{-5} of the water in this air volume. The uncertainty in this estimate is within a factor of 5 higher or lower but most likely lower. Table 9 lists our estimates of tritium inventories in air and tritium concentrations in the air and water of Zone 3.

Table 9. Tritium concentrations and inventories in Zone 3.

Compartment	Tritium concentration, Bq/m ³	Tritium inventory, Bq
Assuming a release of 100 Ci/y		
Air	0.00006	5,000
Surface water (Strawberry Creek)	$\sim 1 \times 10^5$	n/a
Assuming a release of 1000 Ci/y		
Air	0.0006	50,000
Surface water (Strawberry Creek)	$\sim 1 \times 10^6$	n/a

2.7.2 Generic Calculations of Intake, Uptake, Body Burdens, and Dose

This section summarizes the mathematical expressions used to estimate inhalation, dermal uptake, and ingestion exposures to tritium in each of the three exposure zones.

2.7.2.1 Average Intake by Inhalation

The average intake of tritium in Bq/d per kg body weight by inhalation in a working population is calculated from the yearly average tritium concentration of the air of each zone, $C_{air}(Z_i)$ Bq/m³; the working inhalation rate per unit body weight, BR_w m³/kg-h; the exposure time at work, ET_w h/d; the exposure frequency at work, EF_w days/y.

$$\text{Intake(inh)} = \frac{BR_w \times ET_w \times EF_w}{365 \text{ d/y}} \times C_{air}(Z_i) \quad (\text{Bq/kg-d}) \quad (18)$$

The average intake of tritium in Bq/d per kg body weight by inhalation in nonoccupational population is calculated from the yearly average tritium concentration of the air of each zone, $C_{air}(Z_i)$ Bq/m³; the active and resting inhalation rates per unit body weight, BR_a and BR_r in m³/kg-h; the exposure time at home, ET_h h/d; the exposure frequency at home, EF_h days/y; and the assumption that of the time at home, 8 h are spent resting.

$$\text{Intake(inh)} = \frac{[BR_r \times 8 + BR_a \times (1 - ET_h) \times EF_h]}{365 \text{ d/y}} \times C_{air}(Z_i) \quad (\text{Bq/kg-d}) \quad (19)$$

Values used for the parameters in these expression are discussed in Section 2.5 and Appendix B.

2.7.2.2 Average Dermal Uptake

The average uptake of tritium in Bq/d per kg body weight by dermal uptake in the working population is calculated from the yearly average tritium concentration of the air of each zone, $C_{air}(Z_i)$ Bq/m³; the surface area per unit body weight, SA_b m²/kg; the exposure time at work, ET_w h/d; the exposure frequency at work, EF_w days/y; and the permeability for tritium from air through skin, K_p^{air} m/h.

$$\text{Uptake(drm)} = \frac{SA_b \times ET_w \times EF_w}{365 \text{ d/y}} \times K_p^{air} \times C_{air}(Z_i) \quad (\text{Bq/kg-d}) \quad (20)$$

Equation 20 is used for non-occupational exposures to air with ET_w and EF_w replaced by the exposure time and exposure frequency at home, ET_h and EF_h . Equation 20 is used for recreational exposures to water with ET_w and EF_w replaced by the exposure time and exposure frequency for recreational uses of surface water, ET_{sw} and EF_{sw} ; with the air permeability K_p^{air} replaced by that for water K_p^{water} ; and with $C_{air}(Z_i)$ replaced by $C_{water}(Z_i)$. Values used for the parameters in this expression are discussed in Section 2.5 and Appendix B.

2.7.2.3 Average Intake by Ingestion of Homegrown Produce

The average intake of tritium in Bq/d per kg body weight by ingestion of homegrown produce is calculated from the yearly average tritium concentration of the vegetation of each zone, $C_{veg}(Z_i)$ Bq/kg and the ingestion rates of exposed and unexposed produce, I_{ep} and I_{up} kg/kg-d.

$$\text{Intake(ing)} = (I_{ep} + I_{up}) \times C_{veg}(Z_i) \quad (\text{Bq/kg-d}) \quad (21)$$

Values used for the parameters in this expression are discussed in Section 2.5 and Appendix B.

2.7.2.4 Average Ingestion Intake of Surface Water During Recreation

The average intake of tritium in Bq/d per kg body weight by ingestion during water recreation is calculated from the yearly average tritium concentration in the water of a zone, $C_{water}(Z_i)$ Bq/m³; the ingestion rate during the activity, I_{flr} L/kg-h; the exposure time and exposure frequency for recreational uses of surface water, ET_{sw} h/d and EF_{sw} d/y; and a conversion factor from m³ to L

$$\text{Intake(ing)} = \frac{I_{flr} \times ET_w \times EF_w}{365 \text{ d/y}} \times 10^{-3} \text{ m}^3/\text{L} \times C_{water}(Z_i) \quad (\text{Bq/kg-d}) \quad (22)$$

Values used for the parameters in this expression are discussed in Section 2.5 and Appendix B.

2.7.2.5 Total Body Burden of Tritiated Water

The total body burden in Bq/kg from all exposure routes for an average individual within the population is the sum or intakes dermal uptake in Bq/day multiplied by

the removal rate. The removal rate is equal to the effective biological half-life, $T_{1/2}^{\text{tlt}}$, divided by 0.693.

$$\text{Burden}(\text{adult}) = \frac{[\text{Intake}(\text{inh}) + \text{Intake}(\text{ing}) + \text{Uptake}(\text{drm})] \times T_{1/2}^{\text{tlt}}}{0.693} \text{ (Bq/kg)} \quad (21)$$

As discussed above, $T_{1/2}^{\text{tlt}}$ is 15 days for THO and 48 days for OBT.

2.7.2.6 Estimated Live-Born Infant Body Burden

The body burden of tritium in a fetus is assumed to be the same as the mother.

$$\text{Burden}(\text{fetus}) = \text{Burden}(\text{adult}) \quad (22)$$

2.7.2.7 Potential Infant Intake and Body Burden through Breast Feeding

For estimating infant exposure through breast milk, the tritium concentration in breast milk is assumed to equal the tritium burden in the mother. The infant burden is then equal to the breast-milk intake, I_{bm} kg(milk) per kg body weight per day, times the removal rate in the infant.

$$\text{Burden}(\text{infant}) = \frac{\text{Burden}(\text{adult}) \times I_{\text{bm}} \times T_{1/2}^{\text{tlt}}}{0.693} \text{ (Bq/kg)} \quad (23)$$

2.7.2.8 Total Dose, Population Risk, and Individual Risk

The total dose, in Gy, associated with ingestion, inhalation, and dermal uptake is calculated from the product of body burden times a conversion factor that translates from Bq/kg to Gy/y.

$$\text{Dose [Gy/y]} = \text{Burden [Bq/kg]} \times 5.685 \frac{\text{keV}}{\text{dist}} \times 3.15 \times 10^7 \frac{\text{sec}}{\text{year}} \times 1.6 \times 10^{-16} \frac{\text{Joule}}{\text{keV}} \quad (24)$$

$$\text{Dose [Gy/y]} = \text{Burden [Bq/kg]} \times 2.86 \times 10^{-8} \text{ [Gy/y per Bq/kg]} \quad (25)$$

The population risk for cancer represents the committed cancers per year of exposure in the population and includes exposures to adults and infants. The magnitude of the population for cancer depends on the total number of adults and number of infants born to exposed adults.

$$\begin{aligned}
 & \text{Population Risk(cancer) [cancers per year]} \\
 &= [\text{Burden(adult)} \times N_{\text{adults}}(Z_i) + \text{Burden(infant)} \times N'_{\text{infants}}(Z_i) \times 2 \text{ y}] \\
 &\quad \times 2.86 \times 10^{-8} [\text{Gy/y per Bq/kg}] \times R_{\text{HTO}}^{\text{can}} \quad (26)
 \end{aligned}$$

where $N_{\text{adults}}(Z_i)$ is the total number of adults in the population, $N'_{\text{infants}}(Z_i)$ the number of infants born per year to adults in this population, and 2 y is the time that this infant is assumed to be breast fed. The individual lifetime cancer for an average individual is estimated as the product of the population risk and the exposure duration divided by the number of adults in the population.

$$\text{Individual Lifetime Risk(cancer)} = \frac{\text{Population Risk(cancer)} \times \text{ED}}{N_{\text{adults}}(Z_i)} \quad (27)$$

The population risk for genetic effects represents the committed hereditary effects per year per live-born child within the exposed population. The magnitude of the population risk for heritable defects depends on the total number of live births and the exposure to the parents of a child.

$$\begin{aligned}
 & \text{Population Risk(genetic) [genetic effects per year]} \\
 &= [\text{Burden(adult)} \times N'_{\text{infants}}(Z_i) \times 1 \text{ y}] \\
 &\quad \times 2.86 \times 10^{-8} [\text{Gy/y per Bq/kg}] \times R_{\text{HTO}}^{\text{gen}} \quad (28)
 \end{aligned}$$

Assuming one birth per adult per lifetime, we obtain

$$\begin{aligned}
 & \text{Individual Lifetime Risk(genetic) [genetic effects per lifetime]} \\
 &= \text{Burden(adult)} \times 2.86 \times 10^{-8} [\text{Gy/y per Bq/kg}] \times R_{\text{HTO}}^{\text{gen}} \quad (29)
 \end{aligned}$$

The population risk for reproductive and developmental effects represents the expected number of effects per year per live-born child within the exposed population. The magnitude of the population risk for reproductive and developmental effects depends on the total number of live births and the exposure to the fetus during development.

$$\begin{aligned}
 & \text{Population Risk(rep-dev) [effects per year]} \\
 &= [\text{Burden(fetus)} \times N'_{\text{infants}}(Z_i) \times 1 \text{ y}] \\
 & \quad \times 2.86 \times 10^{-8} [\text{Gy/y per Bq/kg}] \times R_{\text{HTO}}^{\text{rep}}
 \end{aligned} \tag{30}$$

$$\begin{aligned}
 & \text{Individual Lifetime Risk(rep-dev) [effects per live birth]} \\
 &= \text{Burden(fetus)} \times 2.86 \times 10^{-8} [\text{Gy/y per Bq/kg}] \times R_{\text{HTO}}^{\text{rep}}
 \end{aligned} \tag{31}$$

2.7.3 Estimated Risks in Zone 1

The population at risk is taken to include the working population in this zone as well as the infants born to mothers who are part of this working population. We assume that the number of adults in this population $N_{\text{adults}}(Z_1)$ is on the order of 200. We estimate the number of live-born infants per year associated with this population, $N'_{\text{infants}}(Z_1)$ by assuming that half of the working population are women and that each woman bears 2 children over a twenty year period.

$$N_{\text{adults}}(Z_1) = 200 \text{ working adults} \tag{32}$$

$$N'_{\text{infants}}(Z_1) = \frac{N_{\text{adults}}(Z_1)}{2} \times \frac{2 \text{ infants}}{20 \text{ years}} = N_{\text{adults}}(Z_1) \times 0.05 \text{ infants/y} \tag{33}$$

The for the purposes of calculating lifetime individual risk, the exposure duration is assumed to be a working lifetime of 40 years. Based on the equations presented in Section 2.7.2 above for estimating intake, uptake, body burdens, dose, and risk combined with tritium concentrations listed in Section 2.7.1, we estimated population risk and individual risks. The results of these calculations for assumed releases of 100 Ci/y and 1000 Ci/y are listed in Table 10.

Table 10. Predicted risks for Zone 1.

Health end point	Population risk per year (expected effects per year)	Individual risk per lifetime (probability in a lifetime)
Assuming a release of 100 Ci/y		
Cancer	2×10^{-4}	4×10^{-5}
Genetic defects	7×10^{-7}	7×10^{-8}
Developmental and reproductive effects	1×10^{-5}	1×10^{-6}
Assuming a release of 1000 Ci/y		
Cancer	2×10^{-3}	4×10^{-4}
Genetic defects	7×10^{-6}	7×10^{-7}
Developmental and reproductive effects	1×10^{-4}	1×10^{-5}

2.7.4 Estimated Risks in Zone 2

The population at risk is taken to include the working population in this zone as well as the infants born to mothers who are part of this working population. The working population is assumed to number about 9,000. In addition the population is assumed to contain some 1,000 residents who occupy the single-family homes near the LBL site. All of these individuals are assumed to consume food from home gardens. For purposes of assessing tritium metabolism and dose, children over the age of two are represented as adults. We assume that the total number of individuals in this population $N_{\text{adults}}(Z_2)$ is on the order of 10,000. We estimate the number of live-born infants per year associated with this population, $N'_{\text{infants}}(Z_2)$ by assuming that half of the working population are women and that each women bears 2 children over a twenty-year period. Thus, we have

$$N_{\text{adults}}(Z_2) = 9,000 \text{ working adults} + 1,000 \text{ residents} = 10,000 \text{ individuals} \quad (33)$$

$$N'_{\text{infants}}(Z_2) = \frac{N_{\text{adults}}(Z_2)}{2} \times \frac{2 \text{ infants}}{20 \text{ years}} = N_{\text{adults}}(Z_2) \times 0.05 \text{ infants/y} \quad (34)$$

For the purposes of calculating lifetime individual risk, we only report the risk to the residential population, whose individual risk is about twice that of the occupational population. The exposure duration for the residential population is assumed to be 70 years. Based on the equations presented in Section 2.7.2 above for estimating of intake, uptake, body burdens, dose, and risk combined with tritium

concentrations listed in Section 2.7.1, we estimated population risk and individual risks. The results of these calculations for assumed releases of 100 Ci/y and 1000 Ci/y are listed in Table 11.

2.7.5 Estimated Risks in Zone 3

The third zone for which an analysis is carried out is the area of the main UCB campus and the city of Berkeley and the associated populations that could be exposed to tritium that is washed from the air into surface-water runoff and carried down into Strawberry Creek. As is illustrated in Figure 11 and discussed in Section 2.7.1.5 above, we consider inhalation, dermal uptake from air and dermal uptake from water contact as potential exposures in Zone 3.

The population at risk is taken to include the working population of the main UCB campus and the city of Berkeley and the residential populations that could be exposed to tritium that is washed from the air into surface-water runoff and carried down into Strawberry Creek. For purposes of assessing tritium metabolism and dose, children over the age of two are represented as adults. We assume that the total number of individuals in this population, $N_{\text{adults}}(Z_3)$, is on the order of 100,000. Half of this population is assumed to have occupational activity patterns and the

Table 11. Predicted risks for Zone 2.

Health end point	Population risk per year (expected effects per year)	Individual risk per lifetime (probability in a lifetime)
Assuming a release of 100 Ci/y		
Cancer	8×10^{-5}	9×10^{-7}
Genetic defects	3×10^{-7}	1×10^{-9}
Developmental and reproductive effects	5×10^{-6}	2×10^{-8}
Assuming a release of 1000 Ci/y		
Cancer	8×10^{-4}	9×10^{-6}
Genetic defects	3×10^{-6}	1×10^{-8}
Developmental and reproductive effects	5×10^{-5}	2×10^{-7}

Figure 11. Exposure scenarios for the vicinity near Strawberry Creek. This diagram illustrates dermal uptake from water as a result of wading in the creek and inhalation and dermal uptake of tritium from the envelope air surrounding the creek contains elevated levels of tritium as a result of tritium in the creek.

other half is assumed to have residential activity patterns. We estimate the number of live-born infants per year associated with this population, $N'_{\text{infants}}(Z_3)$ by assuming that half of the population is women and that each woman bears 2 children over a twenty-year period.

$$N_{\text{adults}}(Z_3) = 100,000 \text{ individuals} \quad (35)$$

$$N'_{\text{infants}}(Z_3) = \frac{N_{\text{adults}}(Z_3)}{2} \times \frac{2 \text{ infants}}{20 \text{ years}} = N_{\text{adults}}(Z_3) \times 0.05 \text{ infants/y} \quad (36)$$

The for the purposes of calculating lifetime individual risk, the exposure duration is assumed to be 70 years. Based on the equations presented in Section 2.7.2 above for estimating intake, uptake, body burdens, dose, and risk combined with tritium concentrations listed in Section 2.7.1, we estimated population risk and individual risks. The results of these calculations for assumed releases of 100 Ci/y and 1000 Ci/y are listed in Table 12.

Table 12. Predicted risks for Zone 3.

Health end point	Population risk per year (expected effects per year)	Individual risk per lifetime (probability in a lifetime)
Assuming a release of 100 Ci/y		
Cancer	1×10^{-5}	7×10^{-9}
Genetic defects	4×10^{-8}	7×10^{-12}
Developmental and reproductive effects	6×10^{-7}	1×10^{-10}
Assuming a release of 1000 Ci/y		
Cancer	1×10^{-4}	7×10^{-8}
Genetic defects	4×10^{-78}	7×10^{-11}
Developmental and reproductive effects	6×10^{-6}	1×10^{-9}

2.8 SUMMARY AND DISCUSSION

The estimated risk for tritium releases from the NTLF are quite low relative to the background incidence of the diseases considered—cancer, heritable genetic defects, and reproductive and developmental effects. Figure 12 provides a comparison of the estimated health impacts of the NTLF in each of the three population zones. In Figure 12, the estimated individual health risks of the NTLF under the *mean* (100 Ci/y) and *high* (1,000 Ci/y) release scenarios are compared here to the current incidence of three diseases categories in the U.S. and to the health risks attributable to the current natural background levels of tritium in the U.S. In this diagram, the scale is in factors of ten—each tick mark indicates a factor of ten change in incidence. From Figure 12, we can see that the estimated cancer risk of the NTLF facility is between four and seven orders of magnitude lower than the cancer incidence risk within the exposed population from all causes, and comparable to and as much as three orders of magnitude lower than the cancer incidence attributable to current background levels of tritium in the U.S. and in the Bay Area. Within Zone 3, the projected levels of tritium in the environment and the estimated population risk are so low relative to the background levels of tritium, that the neither the levels of tritium present or the potential health impacts could be detected by even the most sensitive measuring devices or health surveys.

Figure 12. Comparison of the estimated cancer risks of the NTLF in each of the three population zones to risks from current THO background levels and to the risks associated with the current total cancer incidence within the U.S. population.

The scale of the risk numbers estimated here are difficult to put in perspective. For example, the highest estimated annual population cancer risk is estimated here for the population working in then vicinity of the NTLF to be on the order of 3 chances per million per year. These means that, if the facility continued to operate for a very long period of time, we would have to wait on average about 150,00 years before we would see an additional cancer attributable to this facility. It should also be noted that in the other environmental zones, even though the populations at risk are larger, the population risks are significantly lower. This means that in these populations the "waiting times" on average for the first expected cancer will be greater than 150,000 years and could approach more than several million years.

An uncertainty analysis was used to assess the reliability of these results. The uncertainty analysis was based on a tiered approach which included the following steps: Tiered Approach to Addressing Uncertainty:

1. Variance in the input values is clearly stated. At a minimum, this is done by listing the estimation error or the experimental variance associated with the parameters when these values or their estimation equations are listed in tables. Summary and justification of the assumptions used for each aspect of the model are provided. In addition, it is stated whether these assumptions are likely to result in representative values or conservative (upper bound) estimates of risk.
2. A sensitivity analysis was used to assess how model predictions are impacted by model reliability and data precision.
3. Variance propagation methods (Monte-Carlo methods) were used to map how the overall precision of risk estimates is tied to uncertainty associated with the model inputs.

Based on the range of parameter values used as input for the predictions of risk, we assessed the precision of the model predictions by making thousands of repeated calculations of risk with all input values varied within their likely ranges. For the estimations of cancer risk in Zone 1, we found that 50% of the range of output values were within a factor of 2 of the reported risk and 95% of the output values were within a factor of 10 of the reported risk.

PART 3: APPENDICES

APPENDIX A: TWO-COMPARTMENT ENVIRONMENTAL TRANSPORT MODEL FOR TRITIUM

The physical-chemical properties of HTO imply that the fate and transport tendencies of HTO are best predicted by modeling it as having infinite affinity for the water phase of any compartment and relatively negligible affinity for other phases, e.g., organic phase of biota or mineral phase of soil. These properties include the complete miscibility of HTO in water; the absence of any known sequestering tendencies for HTO, i.e., no preferential uptake of HTO by any organisms or other components of the environment; the similarity of HTO properties (such as diffusion coefficient and vapor pressure) to those of water; and the observed uniform distribution of HTO among the aqueous phases of all interacting compartments in those field studies approximating steady-state conditions (Murphy, 1984, 1993).

A.1 General Issues

We use a two-compartment model to estimate the steady-state distribution of tritiated water between air and soil media resulting from NTLF's continuous HTO emissions to the atmosphere (air compartment). Using a systematic accounting of the gains and losses for each compartment in isolation and for the whole system in concert, the model provides an algorithm for predicting steady-state HTO specific activities (concentrations) in the soil and air compartments.

A.1.1 Tritium in Air

On a local scale, the transfer of HTO from air to the surface of the earth occurs mainly by precipitation with a minor role played by diffusional vapor exchange (NCRP, 1979). On a global scale, where the tritium concentration in air is relatively uniform and, on average higher than that in soil, the reverse appears to be the case. Deposition of HTO in precipitation can result from both incorporation into rain drops as they form and washout, which is the scavenging of HTO by rain drops as they fall through a plume of contaminated HTO vapor (Murphy 1993). HTO deposited from air to ground does not necessarily represent a definitive loss from the air compartment since deposited HTO can re-emerge back into the air compartment via evapotranspiration or simple evaporation. Only HTO which finds its way deep into the ground (well below the root zone) represents a true air compartment loss.

A.1.2 Tritium in Soil

When it enters soil, HTO follows the same transport processes as H₂O (Murphy, 1993). Thus, important transport mechanisms in the terrestrial environment include; bulk flow (i.e., due to pressure gradient such as hydraulic pressure head); vapor gradient; temperature gradient; and finally HTO concentration gradient. The physical half-life of tritium in soil is generally longer than in other components of the ecological system; and this relatively longer half-time extends the half-time of tritium in vegetation rooted in that soil. High-water-use plants may affect the movement of a tritium pulse in soil. The observed physical half-life of tritium in soil appears to be made up two components—a shorter one reflecting the bulk movement of water; and a longer one reflecting tritium retained in more stationary water, such as chemically bound water (Koranda and Martin, 1973).

A.1.3 Tritium in Surface Waters

Under steady-state conditions, the concentration of tritium in surface waters would be the same as the tritium concentration in the water-phase of the atmosphere. When the source of tritium is air and the residence-time of the surface water body is long relative to the half-life of tritium—which is the case for the oceans, the Great Lakes, San Francisco Bay, etc.—the HTO concentration in the surface water will be lower than that in atmospheric water on average. However, near the top of a surface water column the steady-state concentration is likely to exist. HTO concentrations in aquatic organisms closely follow those in the water (Murphy, 1993).

A.1.4 Tritium Uptake in Biota

Tritium in the form of HTO is taken up by plants both from water vapor in air through plant respiration and from water in soil through transpiration (Koranda and Martin, 1973). For plants grown in tritium-free soil, the tritium concentration in plant leaves is on the order of 0.3 to 0.7 times the tritium concentration in atmospheric water (Murphy, 1993). Some of the tritiated water taken into plant tissues can be converted to organically-bound tritium (OBT), which has a longer residence time in plants than molecular water. Photosynthesis is the primary process by which HTO is converted to OBT in plants. However, a very low fraction of T moving through the plant is transformed to OBT. Thus, Murphy (1993) has noted that OBT in plants can be neglected as a sink for T, however the effective residence time of tritium in plants may be affected by even small amounts of OBT.

A.1.5 Why Only Two Compartments?

As discussed in the previous section tritium activity in the terrestrial biota compartment is readily inferred once the soil and air activities are known, thus the activity in the biota compartment need not be predicted explicitly. Indeed adding a biota compartment would require the quantification of inputs such as the uptake rate from soil into plants: there is insufficient information for this quantification. Because the current application of the model does not include the estimation of exposure due to the food ingestion pathway, the biota concentrations are not required for our purposes. Biota need only be considered in as much as it effects the ultimate steady state HTO inventories in air and soil water (soil water effects runoff concentration which will be subject to dermal contact). In addition the fraction of land covered by surface-water is minimal allowing the surface water compartment to be neglected for the purposes of fate and transport modeling (streams are considered in the dose assessment). Thus a two compartment model is sufficient. The premises of our modeling approach are that HTO will distribute itself primarily in the aqueous phase of a compartment, that non-aqueous tritium (i.e., OBT) in any compartment can be accounted for by increasing the effective residence time and that under equilibrium conditions water-phase tritium activities in each compartment will be equal.

For convenience, we call the water-phase HTO specific activity of compartment i , its "equivalence", which will be denoted AQ_i . In a closed system that is in equilibrium, the equivalences of all compartment are the same. Similar terminology has been used in modeling the fate and transport inorganic and organic chemical species (Mackay and Diamond, 1989; Diamond et. al., 1992; McKone, 1993).

We propose here a model for a system that has achieved steady state in terms of mass exchange by balancing gains and losses. However, in terms of chemical thermodynamics this system is not assumed to be at equilibrium. The simultaneous balancing of gains and losses for both soil and air compartments allows non-equilibrium equivalences of the two compartments to be calculated. The bulk compartment's specific activity is easily calculated once its equivalence is quantified. Because only the aqueous phase of a compartment is allowed to hold HTO, both the total compartments inventory and water-phase inventory of tritium are one and the same. The bulk specific activity (i.e., inventory normalized by the aqueous as well as non-aqueous phase volumes) of compartment i , is calculated as a function of its volumetric water content, denoted $\varnothing_i$, and its equivalence (AQ_i) as:

$$C_i = \varnothing_i \times AQ_i \quad (A1)$$

where, C_i , denotes compartment i 's bulk specific activity, and all other terms are defined above.

Thus, the bulk specific activities of HTO in adjacent compartments may differ even under equilibrium conditions since the two compartments may differ in their volumetric water contents. Under equilibrium conditions compartment i 's capacity for holding HTO is effectively dictated by its volumetric water content, ϕ_i .

The volumetric fraction of water in air is calculated based on the observed relative humidity (RH—expressed as the fraction of saturation) at LBL, and the saturation vapor pressure. The saturated vapor pressure (VP_{sat}) in Pascals (Pa) is calculated as a function of temperature using the following Antoine equation (Weast et al., 1986),

$$\log VP_{sat} = 11.28 - 2319.25/T \text{ Pa} \quad (A2)$$

where T is the ambient absolute air temperature in kelvins. From VP_{sat} , we calculate the volume fraction of air that is water, ϕ_a , as

$$\phi_a = RH \times \frac{VP_{sat}}{R \times T} \times \frac{MW_{H_2O}}{\rho_{H_2O}} \quad (A3)$$

where R is the universal gas constant, $8.314 \text{ Pa}\cdot\text{m}^3/\text{mol}\cdot^\circ\text{K}$; MW_{H_2O} is the molecular weight of water, 18 g/mol ; and ρ_{H_2O} is the density of water, 10^6 g/m^3 .

The volumetric water fraction of the soil compartment is determined directly from field data (See Appendix A).

A.2 Steady State Mass Balance Equations

The steady-state equations describing gains and losses in the two compartments are used to solve for the steady-state inventory in each compartment. The following equations express gains and losses for the air and soil compartments respectively.

$$S + T_{sa} N_s = L_a N_a \quad (\text{air}) \quad (A4)$$

and,

$$T_{as} N_a = L_s N_s \quad (\text{surface soil}) \quad (A5)$$

In the equations above, an N represents a compartment HTO inventory (note a compartments bulk inventory and its water phase inventory are equivalent) and the T_{ij} ($i, j = a$ or s) are transfer rate constants, with units of day^{-1} , that express fraction per unit time of the inventory of compartment i that is transferred to compartment j . The compartment abbreviations are a for air, and s for surface soil. The product of an N term and a T term is the rate of change of inventory in Bq/d. $L_i N_i$ represents all losses from compartment i , Bq/d. The term S in Equation A4 represents the rate of HTO input (i.e., the NTLF's HTO emission rate) into the air compartment, Bq/d. Transfer-rate constants are functions of landscape characteristics and environmental mass transfer rates.

In terms of equivalence the balance in Bq/d is expressed as a loss from a compartment i and transfer to a compartment j in the form

$$\text{loss} = \text{Area} \times v_{ij} \times \emptyset_{ik} \times A Q_i = T_{ij} N_i \quad (\text{A6})$$

where Area in m^2 is that across which mass exchange occurs, v_{ij} is the advection velocity from i to j at the exchange boundary, and $\emptyset_{ik}$ is the moisture content of the moving phase k from i to j , (in this analysis the moving phase is always water, for example recharge or runoff, and therefore $\emptyset_{ik}$, generally equals, $\emptyset_{\text{water}}$, i.e., unity) and $A Q_i$ represents the equivalence of compartment i . The relationship in Equation A6 implies that

$$N_i = \emptyset_i A Q_i V_i, \text{ and} \quad (\text{A7})$$

Compartment inventory N_i is calculated using Equation A1 and the compartment volume. Combining this result with Equations A6 and A7 and rearranging terms yields the following for T_{ij} ,

$$T_{ij} = \frac{\text{Area} \times v_{ij} \emptyset_{ik}}{V_i \emptyset_i} = \frac{v_{ij} \emptyset_{ik}}{d_i \emptyset_i}, \quad (\text{A8})$$

where V_i is the compartment volume, d_i is the compartment depth or height, and $\emptyset_i$ is the volumetric fraction of water in compartment, i . This is the general approach used in the paragraphs below to obtain the transfer-rate constants.

A.2.1 Transfer Rate Constants

In the box model used for air, the inventory, N_a in Bq, is described by solving Equations A4 and A5. L_a is the sum of all loss-rate constants from the air compartment,

$$L_a = T_{as} + T_{ao} + \lambda , \quad (A9)$$

$L_a N_a$ is the sum of all losses from the air compartment, Bq/d. The rate constant, T_{as} , accounts for rain-water washout from air to ground-surface soil.

$$T_{as} = \frac{\text{rain} \times \phi_{\text{water}}}{\phi_a \times d_a} . \quad (A10)$$

The air-compartment mixing depth is analogous to the mixing height parameter used in Gaussian plume modeling. For the NTLF analysis this parameter depends on the box-model volume used in the analysis. For the Zone 1 risk characterization, the mixing depth is roughly 50 m (see Site Characteristics subsection below). ϕ_{water} in this expression describes the scavenging factor for rain drops passing through air.

The factor T_{ao} on a box model for atmospheric dispersion that is applied to the natural bowl in which the NTLF is located. According to Benarie (1980), the long-term average pollutant concentration in a region bordered by a box model with volume V_a and pollution source, S in Bq/d, is given by

$$C_{\text{air}} = N_a / V_a = \frac{c S}{\text{Area} \times v_w} , \quad (A11)$$

where c is a unitless proportionality constant; Area is the area of the region being modeled, and v_w is the long-term average wind speed in m/d. This implies that the inverse of the rate constant, T_{ao} , is the convective residence time and is given by the expression, $c d_a / v_w$, where d_a is the atmospheric mixing height. Based on a model for area sources developed by Turner (1970), the constant c can be estimated as $4.3 \sqrt{\text{Area}} / d_a$, where $\sqrt{\text{Area}}$ is the cross-sectional length of an assumed square area containing the source S . Making the appropriate substitutions gives the following expression for the convective loss-rate constant in the air compartment:

$$T_{ao} = \frac{0.23 v_w}{\sqrt{\text{Area}}} . \quad (A12)$$

The mass balance that defines the soil compartment inventory, N_s , in Bq of HTO, is described by Equation A5. L_s is the sum of all transfer-rate constants from the soil compartment,

$$L_s = T_{sa} + T_{recharge} + T_{runoff} + \lambda , \quad (A13)$$

where T_{sa} is the soil to air transfer rate primarily representing evapotranspiration processes, day^{-1} ; $T_{recharge}$ and T_{runoff} are the rate constants for recharge losses to ground water and runoff losses to outside the unit world, respectively, day^{-1} ; and λ is the radioactive decay rate, day^{-1} . These loss-rate constants are given by

$$T_{sa} = \frac{\text{evapotrans} \times \phi_{\text{water}}}{\phi_s \times d_s} , \quad (A14)$$

$$T_{recharge} = \frac{\text{recharge} \times \phi_{\text{water}}}{\phi_s \times d_s} , \quad (A15)$$

$$T_{runoff} = \frac{\text{runoff} \times \phi_{\text{water}}}{\phi_s \times d_s} , \text{ and} \quad (A16)$$

$$\text{recharge} = \text{rain} - \text{evapotrans} - \text{runoff} . \quad (A17)$$

The parameter *recharge* is the yearly average ground-water recharge at the site, m/d; *rain* is the yearly average rainfall, m/d; *evapotrans* is the yearly average evapotranspiration, m/d; and *runoff* is the yearly average runoff; m/d.

A.2.2 Solutions for the Compartment Inventories

Equations A4 and A5 represent a system of two equations, with two unknowns and can be solved using substitution to determine the steady-state inventories, N_i , of contaminant in the soil and air compartments. This solution yields the following relationships,

$$N_s = \frac{S}{\left[\frac{L_s}{T_{as}} \times L_a - T_{sa} \right]} \quad \text{and} \quad (A18)$$

$$N_a = \frac{S}{\left[L_a - T_{sa} \times \frac{T_{as}}{L_s} \right]} \quad (A19)$$

These solutions are used to determine the inventory of tritium in the air, soil, and runoff from the site.

APPENDIX B: HUMAN ANATOMICAL AND INTAKE FACTORS USED IN THE EXPOSURE ASSESSMENT CALCULATIONS

In constructing dose models one needs to define the characteristics of individuals in various age/sex categories and the characteristics of the microenvironments in which they live or from which they obtain water and food. This appendix defines both the types of anatomical and activity data needed to carry out the exposure/dose assessment and explains how these data are obtained. For all factors used in this assessment, we define both an arithmetic mean value and a coefficients of variation (CV), which is the arithmetic standard deviation divided by the arithmetic mean.

B.1 Body Weight

We calculate the arithmetic-mean body weight and CV of body weight for three age groups—infant, child, and adult. The body weight for infants is needed for estimating exposures to contaminants in breast milk. Body weight for adults and children are needed to calculate lifetime-average contact rates per unit body weight and contact rates for an exposure duration of less than a full lifetime. The child age category applies to ages 0 to 15 years, but excludes breast-fed babies. General data on body-weight distributions by age and sex are available from the ICRP (1975), the U.S. EPA (1985, 1989a) and Najjar and Roland (1987). Because it provides more details on age-specific variations, we used the Najjar and Roland (1987) data set to develop the mean value and CV of body weight for ages 0 to 1, for ages 0 to 15, 15 to 70, and for lifetime. These values are listed in Table B1.

B.2 Body Surface Area

Information on the relation between human body weight and surface area has been published by the ICRP (1975) and the U.S. EPA (1989b). The EPA(1989b) reports that surface area (SA) in m² can be estimated as 0.1 times body weight (BW) in kg raised to the 2/3 power. Using this formula, along with methods described in Bevington (1969), we estimate the mean value and standard deviation of surface area per unit body weight, SA_b in m²/kg, for children and adults using the formula

$$SA_b = \left[\frac{SA}{BW} \right] = 0.1 \times BW^{-1/3} \pm \left(0.1 \frac{\sigma_{BW}}{3} \right) BW^{-4/3} , \quad (B1)$$

where the second term is the standard deviation of the surface area-body weight ratio. The resulting surface-area values and CVs are given in Table B1.

B.3 Breathing Rate

General data on breathing rates by age and sex are available from the EPA (1985, 1989a) and the ICRP (1975). Values in Table B1 are taken primarily from the ICRP (1975) with variances estimated by McKone and Daniels (1991). The working breathing rate is for 8 hours of work and, when combined with 8 hours of breathing at the active rate and 8 hours at the resting rate, gives a daily equivalent intake of 30 m³ for an adult (EPA, 1989b). Layton (1992) has derived breathing rates that are consistent with the quantities of oxygen needed to metabolize reported dietary intakes of fats, carbohydrates, and proteins. He has shown that the values in Table B1 could be high by a factor of from 20 to 30%. But this is within the CV reported here.

B.4 Water Ingestion

Tap-water intake includes all household tap water that is consumed as a beverage or used to prepare foods and beverages. Yang and Nelson (1986) have published tap-water and total-fluid intakes in mL/d for the U.S. population by age, sex, and region of the country. Ershow and Cantor (1989) have published population-based estimates of sex-, region-, and season-averaged tap-water intakes per unit body weight by the U.S. population in mL/kg-d. From these two papers, we have derived intakes of tap water in L/kg-d for children, adults, and lifetime equivalent. We also need to determine the amount of incidental ingestion that occurs during water recreation. Based on EPA (1989b) estimates of the ingestion rate of surface water by swimmers, we use 0.0007 L/kg-h (CV equal to 1) as the ingestion rate of any surface water during recreational use. The mean values and CVs of tap-water intake and surface water intake are listed in Table B1.

B.5 Breast-Milk Ingestion

Data on ingestion of breast milk by infants are available in Butte et al. (1984) and Whitehead and Paul (1981). We calculate the breast-milk ingestion per unit body weight for infants ages 0 to 12 months as 0.11 kg/kg-d with a CV of 0.2. This CV is based on other ingestion factors.

B.4 Ingestion of Homegrown Fruits, Vegetables, and Grains

Ingestion of homegrown foods that are obtained from gardens in the vicinity of the NTLF facility are included in the risk assessment. For the food intakes included in the exposure assessment, we calculate here the arithmetic mean and standard deviation of homegrown food intakes per unit body weight for children, adults, and

Table B1. Values of human anatomical and intake properties for used in the exposure calculations.^a

Parameter, symbol	Child ^b		Adult ^b		Combined ^b		Units
Body weight of infants age 0 to 1 y, BW	7.2	(0.3)	--		--	--	kg
Body weight, BW	29	(0.24)	71	(0.2)	62	(0.2)	kg
Surface area, SA _b	0.032	(0.09)	0.024	(0.06)	0.026	(0.07)	m ² /kg
Working breathing rate, BR _w	--	--	0.030	(0.3)	--	--	m ³ /kg-h
Active breathing rate, BR _a	0.023	(0.3)	0.018	(0.3)	0.019	(0.3)	m ³ /kg-h
Resting breathing rate, BR _r	0.008	(0.3)	0.006	(0.2)	0.0064	(0.2)	m ³ /kg-h
Fluid intake, I _f	0.029	(0.2)	0.020	(0.2)	0.022	(0.2)	L/kg-d
Breast milk intake, I _{bm} ^c	0.11	(1)	--		--		kg/kg-d
Water intake during during recreation, I _{flr}	0.0007	(1)	0.0007	(1)	0.0007	(1)	L/kg-h
Total ingestion of fruits and vegetables, I _f	0.0075	(0.2)	0.0042	(0.2)	0.0049	(0.2)	kg/kg-d
Total ingestion of grains, I _g	0.0070	(0.2)	0.0028	(0.2)	0.0037	(0.2)	kg/kg-d
Ingestion of homegrown exposed produce, I _{ep}	0.0016	(0.7)	0.00078	(0.7)	0.00096	(0.7)	kg/kg-d
Ingestion of homegrown unexposed produce, I _{up}	0.00095	(0.7)	0.00053	(0.7)	0.00062	(0.7)	kg/kg-d

^a Listed are the arithmetic-mean value and (in parentheses) the estimated coefficient of variation (CV), equal to the standard deviation divided by the mean. Body weights are from Najjar and Roland (1987), breathing rates are from ICRP (1975), tap water intakes are from Yang and Nelson (1986) and Ershow and Cantor (1989) and food intakes are from Yang and Nelson (1986).

^b The child category covers ages 0 to 15, the adult category covers ages 16 to 70, the combined category is used to represent lifetime equivalent exposure and is obtained by multiplying the child category by 15/70, the adult category by 55/70, and then summing these products.

^c Breast-milk intakes are from Butte et al. (1984) and Whitehead and Paul (1981).

lifetime equivalent exposure, all on body-weight basis. We consider *homegrown* foods those foods produced on the land associated with a household and consumed within that household. The following food groups are considered in the exposure assessment:

leafy vegetables, which include exposed produce such as cabbage, cauliflower, broccoli, celery, lettuce, and spinach;

other exposed produce, such as apples, pears, berries, cucumber, squash, grapes, peaches, tomatoes, string beans, etc.;

protected produce or root crops, such as carrots, beets, turnips, potatoes, legumes, melons, citrus fruits, etc.; and

grains such as wheat, corn, rice, barley, millet, etc.

Total intake of foods in each of these groups is obtained from data compiled by the Nationwide Food Consumption Survey (NFCS) (USDA, 1983). In this survey, the U.S. Department of Agriculture (USDA) used a stratified probability sample of households in the 48 conterminous states and the District of Columbia in each of four seasons from April 1977 through March 1978. The samples were designed to be representative of the United States and were classified according to geographic regions of the country; geographic divisions within each of the regions; and central city, suburban, and non-metropolitan populations. We calculate total annual average food intakes using results compiled by Yang and Nelson (1986), who analyzed the data from the USDA survey. The variance of intake-per-body-weight ratio used to determine the CV is calculated under the assumption that food intake correlates with body weight to the two-thirds power. The mean and variance of body weight used in this estimate are taken from Table B1. Listed first in Table B1 is our estimated total annual average population intake of fruits and vegetables and of grains expressed on a body-weight and age-specific basis.

Limited data are available on the amount of home-grown food that is produced and consumed in California or in the U.S. The U.S. EPA (1989a) has compiled for U.S. households data on the fractions of consumed produce that come from home gardens. For all categories of fruits and vegetables reported in this study, the fraction that is homegrown is in the range 0.04 to 0.75, and for the one grain listed (corn), the average fraction that is homegrown is 0.45 for the 25% of the individuals surveyed who consumed homegrown corn. From these data we estimated that the fraction of consumed fruits and vegetables that are homegrown is 0.24 with CV of 0.7, and that the fraction of consumed grains (mostly corn) that are homegrown is 0.11 also with a CV of 0.7. These values represent households with home gardens and not

necessarily the average of total homegrown consumption in either the U.S. or California population.

According to Yang and Nelson (1986), 47% of all consumed produce (fruits and vegetables) consists of leafy vegetables and other exposed produce, which intercept contaminants from the atmosphere. The remaining 53% of fruits and vegetables consists of protected produce or root crops, in which contaminant transfer to the edible portion is primarily by root uptake. All grain crops are assumed to be exposed primarily to air contaminants.

Based on the information provided in the previous paragraphs, we estimated the mean and CV of the ingestion of exposed (above-ground) and unexposed homegrown produce in households near the NTLF. Table B1 lists our estimates of the mean average annual ingestion of both exposed and unexposed homegrown foods on a body-weight basis for children, adults, and a lifetime of exposure. As is the case for other contact rates, both the mean value and the CV are provided.

APPENDIX C: LANDSCAPE, CLIMATE AND HYDROLOGY PARAMETERS

The purpose of this appendix is to describe how values and ranges of inputs for the landscape, climate, and hydrology parameters used in the risk assessment model are obtained. Table C-1 lists the landscape properties for the NTLF environment derived in this appendix.

C.1 Wind Speed

A summary report of the LBL site meteorology indicates that the average annual wind speed in the vicinity of Building 75 is 2.4 m/s (Merry, 1991). This average reflects measurements taken from a tower located on Building 4 at LBL during the period beginning March 1963 and ending February 1968. However Merry (1991) notes that wind speeds could reach up to 13 m/s during summer periods. In

Table C-1 Landscape properties for the NTLF Environment

Parameter description	Symbol	Mean value	CV	Reference(s)
Yearly average wind speed, m/s	v_w	3	0.3	Merry (1991) (NOAA, 1974)
Yearly average temperature, °C	T	12.5	0.2	Merry (1991) (NOAA, 1974)
Yearly average relative humidity, %	RH	79	0.04	Merry (1991) (NOAA, 1974)
Soil compartment depth, m	d_s	2.5	0.5	(Thibodeaux, 1994) (Murphy, 1993)
Moisture content of soil, L(water)/L(soil)	ϕ_s	0.3	0.1	(Jury et al., 1983)
Annual average precipitation, m/y	$rain$	0.64	1.0	Merry (1991) (NOAA, 1974)
Annual average evapotranspiration, m/y	$evapotrans$	0.45	0.14	van der Leeden et al. (1991)
Annual average runoff, m/y	$runoff$	0.13	0.26	van der Leeden et al. (1991)
Infiltration to ground water	$recharge$	0.06	0.26	this report
Scavenging efficiency	AQ_{rain}	0.4	0.2	(Murphy, 1993)

addition Merry expressed concern that the Building 4 location may have been shielded from the wind. If so, this average may be an under-estimate for Building 75 and the NTLF stack. The mean monthly wind speed for San Francisco for a 44-year period was 4.7 m/s and had a coefficient of variation (CV) of 0.26 (NOAA, 1974). The average annual wind speed reported by Oakland Airport at a elevation of 10 m was 4.2 m/s (NESHAPS Datafile). The yearly average wind speed measured at Lawrence Livermore National Laboratory (LLNL) in 1989 is 3.0 m/s (Sims et al., 1989). A wind speed of 3.0 m/s with a CV of 0.3, is adopted as a nominal wind speed for LBL's site. Some recent site-specific wind speed data have become available. However because these data have yet to be examined for quality control, they were not considered. When these site-specific wind speeds have been determined they should supersede the preliminary values adopted for this analysis.

C.2 Temperature

The mean monthly temperature for San Francisco is 14 °C and has a coefficient of variation (CV) of 0.21 (NOAA, 1974). The average annual temperature employed in the 1994 NESHAPS study for LBL (derived from Oakland airport data) was 10 °C (283 K) while Merry (1991) reports average summer and winter temperatures for the LBL site of 17 °C and 9 °C respectively (implying an approximate average of 12.5 °C, or 285 K). The latter average annual temperature compares well with that observed at LLNL (Sims et al., 1989). We use 12.5 °C with a CV of 0.21 to represent the LBL site.

C.3 Humidity

Merry (1991) reports average relative humidity ranges of 85-90% and 65-75% in the early morning and afternoon, respectively, for the LBL site. The mean humidity obtained from these values is 79%. Mean monthly relative humidity for San Francisco over a 12-year period has a mean annual value (and CV in parentheses) of 84% (0.03) at 7 am, 70% (0.08) at 1 PM and 77% (0.04) overall (NOAA, 1974). We use a mean annual humidity of 77% and a CV of 0.04 to represent the LBL site.

C.4 Depth of Soil Compartment

For the purposes of modeling, the soil compartment represents only the vertical extent of soil interacting with the air compartment. Theoretically, when HTO reaches a critical depth in soil it has such a small likelihood of re-emerging to air that it can effectively be treated as sequestered in soil compartment. In our model, this critical depth is used to delimit the vertical extent of the soil compartment. The Damkohler Number (Thibodeaux, L.J., personal communication) is used to define

a "maximum depth of penetration". When the Damkohler Number is one, radioactive decay and diffusion/advection are balanced. The depth which corresponds to a unit Damkohler number is the maximum depth of penetration for that chemical. We estimate the depth corresponding to the unit Damkohler number for tritium as roughly 2.5 meters. This depth is adopted as the vertical extent of the soil compartment. Murphy has noted that the vertical extent of soil interacting with the air compartment depends to a great extent on the depth of roots of the native vegetation since transpiration plays an important role in the interaction of the soil and air compartments (Murphy, 1993). A typical depth of root penetration for the U.S. is 1 m, which is compatible with the 2.5 meter depth adopted for this study. However, site-specific field measurements of soil penetration could help identify a better choice of depth for defining the soil compartment.

Vadose zone monitoring well observations of HTO levels at various depths in the soil are compatible with a 2.5 meter penetration depth. The measurements show a leveling-off of HTO levels beyond the 7 ft (2.2 m) depth ("Tritium Results for Soil Samples", BSVM-TS-1). However, the observations originating from just one monitoring well are not necessarily representative of the entire soil body within the study area.

C.5 Moisture Content of Soil

The average mass percent of water measured in soil samples taken from boreholes made in the vicinity of the Grizzly Area is 17 percent (including only those samples taken between 0 and 8 ft deep, i.e., to get a water fraction representative of the soil compartment). The equivalent mass fraction can be converted to a volumetric fraction assuming the ratio of aggregate soil (i.e., including soil water and air fractions) to that of water to be 1.6. Using this factor a nominal volumetric fraction water for the soil compartment is calculated as, $0.30 \text{ L(water)/L(composite-soil)}$. We estimate the CV is this value to be 0.10.

C.6 Precipitation

The annual average precipitation rate reported for the LBL site is 0.64 m/y (Merry, 1991). Monthly distributions of average precipitation for San Francisco give a mean annual rainfall of 0.47 m and a coefficient of variation (CV) of 1.0 (NOAA, 1974). Based on these values, we use an average annual rainfall of 0.64 m/y and a CV of 1.0 to model the LBL site.

C.7 Evapotranspiration

No site-specific data for evapotranspiration are available. According to the Water Encyclopedia (van der Leeden et al., 1991), evapotranspiration is roughly 60 % of rainfall in North America and is 65% worldwide with a CV of 0.14. For dryer climates (such as Australia and Africa) however, evapotranspiration is as much as 90 percent of the precipitation. Assuming the Berkeley climate to be somewhat dryer than the average North American climate, we set the nominal evapotranspiration to be 70 percent of the local precipitation rate or 0.45 m/y with a CV of 0.14.

C.8 Runoff

No estimates of runoff are available for the LBL site. Merry (1991) notes that rainfall intensities at LBL are rarely greater than 1.3 cm/hr. With this relatively low intensity then one would expect a lower than average runoff, since runoff basically results when the rainfall rate exceeds the infiltration rate. However, the presence of parking lots around the facility should increase the rate of runoff above average. The Water Encyclopedia (van der Leeden et al., 1991) reports that runoff generally represents between 10 and 30 % of the precipitation rate with a global average of 25% and global CV of 0.26 and North American average of 30%. We adopt 20% as the representative value for LBL, which corresponds to a yearly average runoff of 0.13 m/y with a CV of 0.26.

C.9 Infiltration to Ground Water

Because no infiltration data are available for the LBL site, the infiltration rate is taken as the difference between the precipitation rate and the sum of the runoff and evapotranspiration rates.

C.10 Rain Scavenging Efficiency

The scavenging efficiency is the fraction of HTO in air that is transferred to rain. Transfer occurs by one of two processes—1) incorporation during the formation of raindrops in the atmosphere and 2) rain-out of HTO as rain drops fall through the HTO vapor and adsorbing some fraction of HTO as they fall. A scavenging efficiency can be inferred from observed ratios of AQ_{rain} to AQ_{air} . Murphy (1993) notes that this ratio is generally less than unity implying lower HTO equivalence in rain than in airborne vapor and reports the minimum, average, and maximum ratios as 0.20, 0.40, 0.55, respectively. In the analysis here, this average is assumed to be equivalent to a scavenging efficiency with nominal scavenging of 0.40 and a CV of 0.2.

APPENDIX D: EVALUATING THE POTENTIAL LEGACY OF PAST ELEVATED HTO EMISSIONS

The NTLF emitted HTO at greater rate in the past. Prior to 1989 the emissions were five times the current release rates. Depending on the effective residence time of HTO in the soil compartment, the pre-1989 releases may have left a legacy of non-negligible soil levels over and above those predicted under the premise of the current release rates. Here we investigate the potential significance of such a legacy. This analysis requires a dynamic treatment of the compartmental inventories because, the HTO source term is dynamic. From the literature review and based on experience with multi-media fate and transport modeling of other contaminants the effective residence time of the soil can be assumed to be far greater than that in air. Thus only the soil compartment must be treated as dynamic. Based on Equations 1 and 2, the following revised system of equations can be defined:

$$S + T_{sa} N_s = L_a N_a \text{ (air)} \quad (D1)$$

and,

$$\frac{dN_s}{dt} = T_{as} - L_s \times N_s \text{ (surface soil)} \quad (D2)$$

The above system of equations can be solved to determine the time dependent inventory of the soil compartment, $N_s(t)$, as:

$$N_s(t) = \left[N_s(0) - \frac{T_{as} \times S}{L_a \times Y} \right] \times \exp(-Y t) + \frac{T_{as} \times S}{L_a \times Y} \quad (D3)$$

where,

$$Y = \left[L_s - T_{sa} \times \frac{T_{sa} \times T_{as}}{L_a} \right] \quad (D4)$$

and, $N_s(0)$ is the HTO inventory (Bq) in the soil compartment at, $t = 0$, and all other terms are as previously defined. Assuming that emissions prior to 1989 were

continuous at five times the current level, a steady state inventory in soil due to the pre-1990 emissions can be expressed as:

$$N_{s(1989)} = \frac{5 \times S \times T_{as}}{L_a \times Y} \quad (D5)$$

Equation D5, can be found by either allowing $t \rightarrow \text{infinity}$, in Equation D3, or by substituting Equation D4, into Equation 5. Equation D5 can be substituted into Equation D3, as the initial concentration, i.e., for $N_s(0)$, to calculate a time dependent inventory in soil reflecting the cumulative result of both the initial conditions established by the legacy of the early elevated emission rate and the subsequent post-1990 continuous release of, S . The resultant inventory at 1994 due to both legacy and continuous release will be denoted $N_{s(t=1994)}$. The ratio of the latter to N_s , as calculated using Equation 5 (i.e., under steady state conditions and neglecting the legacy of former operations) can be expressed as:

$$\frac{N_{s(t=1994)}}{N_s} = \left[\frac{5 \times S - S}{S} \right] \times \exp(-Y t) + 1 \quad (D6)$$

$$\text{or, } \frac{N_{s(t=1994)}}{N_s} = 4 \times \exp(-Y t) + 1 \quad (D7)$$

The above ratio is used to evaluate the relative significance of the legacy from past elevated release in terms of affecting the current HTO inventory in soil. This analysis does not take into account the enhanced residence time in soil which would be affected by OBT. However Murphy (1993) asserts that the amount of HTO which would be converted to and reside in the organically bound form is in terms of quantity as negligible.

PART 4: REFERENCES

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