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Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory (LLNL) Part 1. Description of Tritium Dose Model (DCART) for Routine Releases from LLNL

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ABSTRACT

DCART (Doses from Chronic Atmospheric Releases of Tritium) is a spreadsheet model developed at Lawrence Livermore National Laboratory (LLNL) that calculates doses from inhalation of tritiated hydrogen gas (HT), inhalation and skin absorption of tritiated water (HTO), and ingestion of HTO and organically bound tritium (OBT) to adult, child (age 10), and infant (age 6 months to 1 year) from routine atmospheric releases of HT and HTO. DCART is a deterministic model that, when coupled to the risk assessment software Crystal Ball®, predicts doses with a 95% confidence interval. The equations used by DCART are described and all distributions on parameter values are presented. DCART has been tested against the results of other models and several sets of observations in the Tritium Working Groups of the International Atomic Energy Agency's programs, Biosphere Modelling and Assessment and Environmental Modeling for Radiation Safety. The version of DCART described here has been modified to include parameter values and distributions specific to conditions at LLNL. In future work, DCART will be used to reconstruct dose to the hypothetical maximally exposed individual from annual routine releases of HTO and HT from all LLNL facilities and from the Sandia National Laboratory's Tritium Research Laboratory over the last fifty years.

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INTRODUCTION

Over the past fifty-three years, operations at Lawrence Livermore National Laboratory (LLNL) have released tritium, primarily as tritiated hydrogen gas (HT), to the atmosphere. Starting in 1973, doses from all releases have been calculated and published annually in the LLNL Site Annual Environmental Reports (e.g., Peterson et al. 2005), which are available to the public. Prior to 1973, LLNL was not required to calculate dose to the public from routine releases, but LLNL did report releases of tritium to the United States Atomic Energy Commission (AEC) and did publish monitoring data for nuclides in environmental reports to the AEC that date back to 1959.

Of the radionuclides released to the atmosphere by LLNL operations, tritium has been the largest contributor to dose to the public. All calculated annual doses from LLNL operations to a hypothetical maximally exposed member of the public were well below regulatory limits. The Agency for Toxic Substances and Disease Registry completed a Public Health Assessment (PHA) of the Livermore site in 2004 (ATSDR 2004) that addressed all environmental media, hazards, and risks to the public and found "No Apparent Public Health Hazard[1]." In the process of preparing the PHA, ATSDR released two documents specifically about risk to the public from LLNL tritium releases. In the first (ATSDR 2002), an expert panel reviewed LLNL's release history and monitoring program. The panel also reported on what is currently known about tritium dosimetry, with emphasis on organically bound tritium (OBT) (Osborne et al. 2001). In the second, ATSDR presented calculations of dose to the public from the two accidental HT releases in 1965 and 1970 (ATSDR 2003).

LLNL has a history of minimal dose impact on the public. The complete tritium dose reconstruction (TDR), of which this is Part 1, significantly supplements the information already publicly available.

- All doses for the TDR are predicted using one model and a known set of assumptions so that doses can be compared from year to year (Doses reported by LLNL from 1973 through the present cannot be compared on an annual basis because assumptions and models changed [Table 1 and Table 2]).
- Doses calculated for the TDR account explicitly for the additional dose contribution from ingestion of organically bound tritium[2] (OBT) (OBT is not yet included as a pathway to dose in any current regulatory model).
- Doses are predicted from releases of HT[3] that account for the conversion of HT to HTO in the environment (the regulatory approach has been either to treat HT as having negligible dose consequences or to equate it with tritiated water (HTO);

[1] This conclusion means that, although community exposures to site-related contaminants may have occurred or may be occurring, the resulting doses are unlikely to result in any adverse health effects and are consequently below levels of public health concern.

[2] The dose from ingesting 1 Bq of OBT is about 2.3 times that from ingesting 1 Bq of HTO (ICRP 1996) because OBT has a longer biological half-life than does HTO.

[3] HT converts quite rapidly to HTO in soil when the HT plume comes to ground (McFarlane et al. 1978; Brown et al. 1988). Experimental data under chronic conditions (Davis and Bickel 2000) have shown that no more than about 10% of the HT at any location gets converted to HTO. The dose consequences of this conversion are a bit higher than 10%, as will be discussed.

the first assumption greatly underestimates the dose effect of a release of HT, while the second assumption overestimates the dose effect of a release of HT.

- The TDR calculates doses for the years 1953 to 1972 (No annual doses to the public were calculated prior to 1973).
- A probabilistic set of dose predictions demonstrates that the true dose will lie within a range of results – or specifically below an upper confidence limit - based on acknowledging the uncertainty inherent in dose calculations (reported doses have been best estimates with a presumed conservative bias).
- The additional independent set of dose calculations resulting from the TDR can be compared with doses reported by LLNL (For any assessment, it is advisable that at least two independent sets of dose calculations be made. Because different assumptions and interpretations of data are often made, much greater confidence can be placed in the results if the two (or more) sets agree [Peterson et al. 1996]).
- The TDR provides a compendium of all information that could be garnered about tritium releases from LLNL.

The model used for the LLNL TDR must account for all pathways to dose from releases of HT and HTO to the atmosphere and have incorporated the latest understanding of environmental tritium transfer in the environment. At present, a few published steady-state models explicitly account for conversion of HT to HTO in the environment and for dose from OBT as well as HTO, but until very recently, most models neglected one or more important pathways (Diabaté and Strack 1990). Models that have the necessary pathways, such as the twelve that were evaluated in the **Bio**sphere **M**odeling and **Ass**essment (BIOMASS) program of the International Atomic Energy Agency (IAEA 1996), are for the most part not published. NORMTRI (Raskob 1994) is a model that has all appropriate pathways, has been published, and did participate in BIOMASS. NORMTRI has many excellent features, but its HT pathways are not based on the latest experimental information on the behavior of HT in the environment (Davis et al. 1995; Davis and Bickel 2000). Another model that includes all essential pathways is that of Murphy (1986). This model, however, uses parameter values derived from observed data at the Savannah River Site and would be difficult to apply elsewhere. A third, NEWTRIT (Peterson and Davis 2002), has been coded at the request of the Environmental Protection Agency (EPA) into a version of the GENII code (Napier et al. 1988), GENII-NESHAPs. NEWTRIT calculates doses based on empirical ratios between compartments (e.g., concentrations in plant water compared with concentrations in air moisture) that have been selected to assure the conservatism needed for regulatory compliance. NEWTRIT's predictions are too conservative for this TDR. None of these models is probabilistic.

A model has been developed at LLNL to estimate tritium doses from routine operations at LLNL. This model, DCART (**D**oses from **C**hronic **A**tmospheric **R**eleases of **T**ritium), was one of the twelve models evaluated in BIOMASS and was developed specifically to account for the conversion of HT to HTO in the environment and dose from OBT. DCART was selected as the best model to calculate doses for the LLNL TDR.

DCART is a deterministic spreadsheet code in Microsoft Excel®. When coupled to Crystal Ball 2000®[4], a risk analysis software package that provides uncertainty and sensitivity analyses for spreadsheet codes, DCART produces probabilistic results. The use of DCART to predict historical tritium doses will provide a set of consistent, defensible dose estimates based on the most recent knowledge of tritium transfer through the environment.

The equations, parameter values, and assumptions in DCART are described in this report; parameter values and assumptions are specific to California and the Livermore site when possible.

MODEL DESCRIPTION

DCART is a model that calculates doses (inhalation/skin absorption and ingestion of food and water) to adult, child (age 10 years) and infant (age 6 months to 1 year) from chronic releases of tritium gas (HT or T_2) and tritiated water (HTO) to the atmosphere. Controlling input includes the HT or HTO source terms from each facility (in Ci y^{-1}) and dilution factors (χ/Q in s m^{-3}) calculated by a dispersion model. Wet deposition (HTO) and dry deposition (HT and HTO) to soil are both calculated as if they determined the concentration of tritium in soil water. However, the concentration in soil water, due to the difficulty of accounting for emission of HTO from soil, is actually determined by empirical ratios between air and soil water concentrations. Thus, the deposition pathway, although originally included in DCART, is effectively bypassed at present because empirical ratios are more robust than any attempt to model complex processes simply. Concentrations in precipitation, however, can be calculated with DCART, as can total deposition to soil before re-emission. For an HT release, HTO concentrations in air due to emission of HTO converted from deposited HT are estimated based on empirical ratios. The model calculates concentrations of tissue free-water tritium (TFWT or plant HTO) and OBT in edible plants (leafy vegetables, root vegetables, fruit or fruit vegetables [e.g., cucumber, tomato, eggplant, beans], grain, pasture, and hay) and animal products (milk, beef, pork, poultry and eggs). Deterministic doses are calculated using dose coefficients for HTO and OBT (ICRP 1996) and HT (ICRP 1995); probabilistic doses are calculated from distributions determined by Harrison et al. (2002). Recommended parameter values and distributions of values for uncertainty analysis (see the Appendix) are specific to LLNL, to the extent possible. Dose from swimming in the LLNL pool is calculated. Concentrations of OBT in tree rings can also be calculated.

DCART has been tested within BIOMASS and EMRAS (**E**nvironmental **M**odeling for **Ra**diation **S**afety), the IAEA Coordinated Research Program that succeeded BIOMASS. Results of comparisons of predictions to observations, presented later in this paper, are among the best of all models participating. DCART has been evolving all the while, so the results presented for the BIOMASS evaluation may not be the same as would be obtained from the model described here, which has been improved based on experience

[4] Decisioneering, Inc. 1515 Arapahoe Street, Suite 1311, Denver Colorado USA 80202.

gained in BIOMASS. The version of DCART described in this report is the same as that used for the EMRAS calculations and for the LLNL tritium dose reconstruction, although, of course, some site-specific parameter values will have been changed to address the EMRAS scenario descriptions.

Very few models, and certainly no regulatory models as yet, can predict doses from releases of HT and ingestion of OBT. Most models are restricted to releases of, and doses from, HTO. Of models not restricted to HTO, DCART should do as good a job of predicting doses from LLNL releases as any and do a better job than some, particularly for HT releases (to be discussed). Being developed in-house has many advantages. One is that the model is understood completely by the developer. Also, because DCART is a spreadsheet model, its calculations are more transparent than traditional compiled computer codes. Coupled with the risk assessment software, Crystal Ball®, DCART can produce probabilistic predictions as well as deterministic predictions. The model may be modified easily.

Equations in DCART

The default parameter values used in DCART are either specific to the Livermore site or similar sites or are usually the medians of lognormal or means of normal distributions of relevant observed data from the literature. Values mentioned below when discussing the equations are deterministic; both the deterministic (best estimate) values and the distributions for each parameter may be found in Tables A1 through A8.

Deposition

Annual wet deposition of HTO is calculated:

$$\omega = \frac{\Lambda Q \exp(-\Lambda x / u)\Delta T}{xu\Delta\theta} \qquad [1]$$

where:

ω = wet deposition (Bq m^{-2} y^{-1})
Λ = washout coefficient (s^{-1}) (variable, depending on distance from source and stack height (Belot 1998)); see Table A1.
Q = release rate (Bq s^{-1})
x = downwind distance in meters from the source
u = mean wind speed (m s^{-1}) for when it rains; sector, release height and year specific data are used when available; see Table A1.
ΔT = duration of rainfall when plume is present (s y^{-1}); (calculated from fraction of time wind blows into a sector times fraction of time it rains times seconds in a year; see Table A1)
$\Delta\theta$ = sector width (radians); 0.393

The value of Λ is varied to account for the fact that the washout coefficient is not constant throughout the plume, as in the case of aerosols, but depends on the shape of the vertical profile and hence on the distance from the source. DCART can calculate wet deposition at a location from all sources.

The annual mean concentration of HTO in soil water from wet deposition is calculated.

$$C_{sw,w} = (\omega / (Precip + Irrig)) (0.001\ m^3 / L) \quad [1a]$$

where:

$C_{sw,w}$ = HTO concentration in soil water after rain or irrigating (Bq L^{-1})
Precip = mean annual precipitation ($m^3\ m^{-2}$ or m); see Table A1
Irrig = mean annual contribution of irrigation water (m) (California default 0.61 m; [Brewer 2001])

The concentration of tritium in precipitation is the same as [1a] except that the contribution from irrigation is not included.

Annual dry deposition is calculated for both HT and HTO as follows:

$$Dep_d = C_a\ v_g\ 3.15\ 10^7\ s\ y^{-1} \quad [2]$$

where:

Dep_d = dry deposition (Bq $m^{-2}\ y^{-1}$)
C_a = concentration of HT or HTO in air at the location of interest (Bq m^{-3}); see Table A2.
v_g = deposition velocity (m s^{-1}) (default for HTO: 5.3 10^{-3}; default for HT: 2.6 10^{-4}; see Table A3)

The amount of dry deposited HTO gets mixed into the amount of water entering the soil (i.e., the precipitation plus irrigation). Thus, as in [1a], the annual mean concentration of HTO in soil water (Bq L^{-1}) before re-emission from dry deposition is:

$$C_{sw,d} = (Dep_d / (Precip + Irrig)) (0.001 m^3 / L) \quad [2a]$$

The annual mean concentration in the soil water (in Bq L^{-1}) can therefore be calculated

$$C_{sw} = f_r (C_{sw,d} + C_{sw,w}) \quad [3]$$

where:

f_r = fraction retained in soil water (variable and determined by calibration)

The retained fraction of HTO from deposited HT should be larger than for deposited HTO because HT diffuses deeper in the soil than does HTO.

However, following the recommendations of the BIOMASS Tritium Working Group (IAEA 2003), DCART is calibrated independently of the above deposition equations so that the ratio of soil moisture concentration to air moisture concentration is 0.3 for a release of HTO (Table A3). Fractions have been observed up to 0.5 (Fellows et al 1990), which is the value recommended by the BIOMASS Tritium Working Group (IAEA 2003) for screening models. The effect of this calibration is, as mentioned, to bypass the deposition pathways completely.

For a release of HT, the soil water concentration is calibrated in DCART so that the ratio of HTO in soil (Bq L^{-1}) to that of HT in air (Bq m^{-3}) is 6.0 (Table A3), which is the median ratio observed over natural soil during an experimental HT release of 1994 (Davis et al 1995; Davis and Bickel 2000).

Although both wet and dry deposition pathways in DCART are bypassed when the calibrated ratios are used, when necessary, the precipitation sub-model can be used by itself to estimate concentrations of tritium in rainfall.

Concentrations of TFWT and OBT in plants

For both HTO and HT releases, the HTO annual mean concentrations in plant water of leafy vegetables and pasture (Raney and Vaadia 1965; Murphy 1984) are given by:

$$C_{pw} = 1/\gamma \ [R_H \ C_{a\_HTO} / H_a + (1 - R_H) \ C_{sw}] \qquad [4]$$

where:

C_{pw} = concentration of tritium in the plant water (Bq L^{-1} or Bq kg^{-1})
γ = ratio of vapor pressure between HTO and H_2O (0.909)
R_H = relative humidity (LLNL annual mean default 0.69; Table A4)
C_{a_HTO} = concentration of HTO in air (Bq m^{-3})
H_a = absolute humidity (kg m^{-3}) (LLNL annual mean default 0.0078); Table A4.
C_{sw} = concentration of tritium in soil moisture (Bq L^{-1})

Concentration of TFWT in Bq kg^{-1} fresh weight (fw) is obtained by multiplying C_{pw} by the fraction of the fresh weight plant that is water (1 – dry mass fraction; Table A4). Equation [4] is used to predict TFWT concentrations in most tritium research models (IAEA 2003), although it routinely appears to overestimate TFWT concentrations by about 20% when compared with observations (IAEA 2003).

For both HTO and HT releases, concentration of OBT in all plants in Bq L^{-1} water equivalent (i.e., the amount of water produced when the dry matter is combusted; L kg^{-1}) equals the concentration in plant water (as calculated for leafy vegetables and pasture, eqn [4]) reduced by a discrimination factor that arises from isotopic effects in OBT formation. This isotopic discrimination results in a lower value for the specific activity (T/H - tritium atom/hydrogen atom) in the water of combustion compared with the

specific activity in the plant water (McFarlane, 1976; Garland and Ameen, 1979). An isotopic discrimination factor of 0.7 has been chosen (Kim and Baumgärtner 1994). The annual mean concentration of OBT in kilograms fresh weight plant is obtained from:

$$C_{veg,OBT} = ID_p\, C_{pw}\, M_D\, W_{eq} \qquad [5]$$

where:

$C_{veg,OBT}$ = concentration of OBT in 1 kg of fresh weight plant
ID_p = isotopic discrimination factor for plants (0.7); Table A4
M_D = dry mass per kg fresh weight of plant material; Table A4
W_{eq} = water equivalent of dry plant matter (L kg^{-1}); Table A4

The annual mean concentrations of HTO in fruits, fruit vegetables[5] and grain are calculated assuming 60% of the water in the fruit, fruit vegetable, or grain comes from air moisture and the other 40% comes from soil water (Davis 2003; Davis et al. 2002; Dinner et al. 1980; Fellows et al 1990; Table A4).

$$C_{fv,HTO}\ (Bq\ L^{-1}) = (0.6\ C_{a\_HTO}/H_a) + (0.4\ C_{sw}) \qquad [6]$$

where:

C_{fv_HTO} = concentration of tritium (Bq L^{-1}) in water of fruits, fruit vegetables and grain

OBT in fruit, fruit vegetables and grain is calculated just as OBT in other vegetables (see eqn [5]), assuming that OBT is the product of photosynthesis and is translocated from leaves to fruit.

The annual mean concentrations of HTO in below-ground plant products (e.g., potatoes and root crops) are calculated as they are in NORMTRI (Raskob, 1994). This assumes that the water in the root crop is nearly in equilibrium with the soil water (Davis 2003; Davis et al. 2002; Table A4).

$$C_{rc,HTO} = 0.95\ C_{sw} \qquad [7]$$

where:

$C_{rc,\,HTO}$ = the HTO concentration in root crops in Bg L^{-1}

OBT in root crops is calculated just as OBT in other vegetables (see eqn [5]), assuming that OBT is the product of photosynthesis and is translocated from leaves to roots.

Default parameters for relative humidity are the averaged annual values for the Livermore site for 1999 and 2001 - 2003[6] or year-specific. Default parameters for

[5] Fruit vegetables are above-ground non-leafy vegetables such as tomato, cucumber, squash, beans, etc.
[6] Fifteen-minute data for all meteorological parameters are available for 1997 to the present at http://www.metdat.llnl.gov. An exceptionally high number of 15-minute readings of relative humidity were greater than 94% for the years 1997, 1998, and 2000, so the relative humidity for these years is not considered reliable.

absolute humidity are derived from the mean tritium monitoring data (pCi m^{-3} / pCi L^{-1} = L m^{-3}) for 1974 through 2003 or are year-specific[7]. Default parameters for fresh and dry weight fractions and water equivalent are mean values from Geigy (1981) (Table A4).

Resuspension/emission of HTO

The contribution of reemitted HTO to the HTO concentration in air for an HTO release is not modeled in DCART because reemitted HTO's contribution to air concentrations is insignificant except within 500 m of an elevated source (IAEA 2003). For a ground level release, reemission is not an issue if the model does not deplete the plume in the first place, as, for example, CAP88-PC does not for HTO. However, because it is necessary to estimate HTO concentrations in air after a release of HT, emission is modeled for deposited HT that has been converted to HTO. At present, the HTO concentration in air moisture that is being inhaled or absorbed through the skin after a release of HT is calibrated to an observed ratio of 4.0 Bq L^{-1} HTO / Bq m^{-3} HT from an experimental HT release (Davis and Bickel 2000). The concentration of HTO in air moisture from a release of HT at 20 cm (plant height) is calibrated to a ratio of 6.0 Bq L^{-1} HTO / Bq m^{-3} HT observed during the same release (Davis and Bickel 2000; Table A4).

Concentrations of HTO and OBT in animal products

Data that describe the transfer of tritium from the environment to animals are minimal, but, for equilibrium conditions, the best evidence (Evans 1969; Moghissi et al. 1987) indicates that the tritium to hydrogen ratio in animal water and organic matter is the same as the environment to which the animal is exposed. Although tritium is too mobile to truly equilibrate with the environment, it is not unreasonable to develop a model for concentrations of tritium in animal products that assures that specific activity is maintained. This has been done for NEWTRIT (Peterson and Davis 2002) and for NORMTRI (Raskob 1994). The approach used for DCART is the same as that in NEWTRIT, except that DCART calculates the fractions of water contributed from plant water, plant organic matter, drinking water, and inhalation and skin-absorption based on user-provided diets rather than default diets, as in NEWTRIT.

The specific activity model followed in DCART is contained in the following calculation that gives the concentration of either HTO in water or OBT in water equivalent (Bq L^{-1} or Bq kg^{-1}).

[7] The decision to use air tritium monitoring data to estimate absolute humidity for DCART was based primarily on its having the longest time series and less uncertainty than using the absolute humidity calculated from temperature and relative humidity from the LLNL Meteorological Station. Although absolute humidity may be underestimated based on the results of silica gel sampling (Paulus et al. 2003), the use of lower humidity in DCART will increase the dose. The error, if any, is thus conservative.

$$C_{aw} = \Sigma_{g,h,p}(C_{pw}\,(F_{pw} / T_w)) + \Sigma_{g,h,p}(C_{pweq}\,(F_{pweq} / T_w)) + C_a\,(F_{inh,s} / T_w) + C_{adw}\,(F_{dw} / T_w) \quad [8]$$

where:

- C_{aw} = tritium concentration in animal water (HTO) or water equivalent (OBT) (Bq L^{-1})
- $\Sigma_{g,h,p}$ = summation of the contributions of grain, hay or pasture to diet
- C_{pw} = concentration of HTO in the water fraction of the plant ingested (Bq L^{-1}) (see eqn [4])
- F_{pw} = kg of total water in the daily diet obtained from grain, hay or pasture (kg kg^{-1}); kg d^{-1} food type *x* times fresh weight fraction for food type *x*
- T_w = total water ingested daily by the animal from all sources. This includes the water directly available from all foods, the water available after digested foods, the water obtained through skin absorption and breathing, and the water obtained from drinking water.
- C_{pweq} = concentration (Bq L^{-1}) in the water produced from foods after organic matter has been digested
- F_{pweq} = kg of water obtained daily after digestion of organic matter (kg kg^{-1}) in grain, hay or pasture; kg d^{-1} food type *x* times dry matter content of food type x times water equivalent factor of food type *x*
- $F_{inh,s}$ = amount of water in L obtained from inhalation and skin absorption each day
- C_{adw} = concentration of tritium in the animal's drinking water (Bq L^{-1})
- F_{dw} = kg of water obtained daily from drinking water

As for the various plants, HTO and OBT concentrations for animal products (Bq kg^{-1} in milk, beef, pork, poultry, and eggs, as eaten) are calculated separately. To obtain the concentration of HTO in Bq kg^{-1} of animal product, each result of eqn [8] (C_{aw}) is multiplied by the fresh water fraction (1 minus the dry matter fraction) of the particular animal product (Table A5). Similarly, to obtain the concentration of OBT in Bq kg^{-1} of animal product, each C_{aw} must be multiplied by the dry matter fraction times the water equivalent factor (Table A5) for the particular animal product.

Default parameters for fraction dry weight and water equivalent are mean values from Geigy (1981). Other parameter values for these equations (Table A5) have been obtained primarily from National Academy of Sciences data on animal nutrition (NAS 1994, 1996, 1998, 2001) supplemented with information from agricultural and veterinary colleges obtained from the World Wide Web (e.g., Ely and Guthrie 2001). In the case of cows, the diet is based on what is known about the milk production (24.1 L d^{-1}) of the average California dairy cow (UCD 1998). The annual diet of dairy and beef cows (Table A5) is averaged over the diet obtained from fresh pasture (four months of the year) and the diet of hay and grain for the rest of the year. Concentrations in animal products are quite insensitive to the type of feed ingested, but they are sensitive to the quantity of water ingested by each type of animal. Modern dairy cows need to drink large amounts of water to support modern (increased) milk productivity, and this will reduce the

concentrations in milk by a few percent in DCART, because drinking water concentrations are assumed less than concentrations in plants (see next section).

Recently, a model for tritium concentrations in animal products based on hydrogen metabolism has been developed (Galeriu et al. 2001). This model accounts separately for each possible transfer from plants to animals. As mentioned, there is a paucity of data about the transfer of tritium to animals. The data that are available, however, indicate that some transfers occur preferentially when the system is not in equilibrium. These observations are supported by knowledge of hydrogen metabolism. For example, the bound hydrogen in the organic matter of plants that is digested to carbohydrates, proteins, and lipids by the animal is more likely to be synthesized into the organic matter of the animal than is the tritium atom that enters the body as water. The likelihood of transfers from diets to animals in decreasing order of occurrence is

- hydrogen in water to hydrogen in water
- hydrogen bound in organic matter to hydrogen bound in organic matter
- hydrogen bound in organic matter to unbound hydrogen in water
- unbound hydrogen in water to bound hydrogen in organic matter

A model like this is quite elegant and helps improve our understanding of the processes involved. Furthermore, it is finely tuned to all aspects of the animal, from the diet to the temperature of the ambient air to the activity level of the animal. However, the specific activity model may be preferred for simplicity and robustness. A comparison of the results of the DCART animal model with those of the Galeriu model is presented below.

Concentration of HTO in drinking water

If small surface water bodies are the source of the drinking water, as they might be for animals, the concentration of tritium in drinking water should be about that of soil moisture. Concentrations of tritium in small bodies of water are expected to have come from the atmosphere, just as soil tritium does, from an atmospheric release. Deposition rates of HTO to water and soils are reasonably similar. Thus concentrations in soil water and ponds may be similar. Exchange between soil water and pond water, although unlikely, would only serve to bring the concentrations closer together. For a screening level model (Peterson and Davis 2002), an assumption that the drinking water for animals is 50% that of air moisture is considered conservative. This assumption is similar to the screening level model assumption that soil water concentrations are 50% those of air moisture (IAEA 2003) but is additionally based on observed tritium concentrations in small bodies of water relative to that in air moisture. More realistically, soil water has been found to have about 30% the concentration of air moisture (IAEA 2003); equally, the concentration of surface drinking water concentrations for animals could be 30% of air moisture for a release of HTO. However, because atmospheric HTO depositing on the water surface is diluted by mixing with the mass of water, a water concentration of 30% or higher than that of air moisture is likely to be found only in very shallow bodies of water. The LLNL swimming pool could serve as a surrogate for a local small body of water. Between 1986 and mid-2005 when the pool closed, the concentration of tritium in

the pool water was sampled; air tritium concentrations have been sampled adjacent to the pool since mid-1990. As seen in Table 3, annually (e.g., median of all pool samples divided by the median of all air tritium samples), the pool water/air moisture ratio does approach 30%. However, the overall mean and median of the ratios are about 16%, which probably represent the dynamics of tritium behavior in the Livermore Valley, given that releases are variable, have been dropping over time, and that loss of tritium can occur from bodies of water when wind direction changes, etc. Thus, the concentration of drinking water for animals after a release of HTO is assumed in DCART be 16.5% the concentration of HTO in air moisture (Table A5).

For a release of HT, the soil concentrations (HTO) predicted by DCART are the same as those in air moisture close to the ground due to the conversion of HT to HTO in the soil. It is very unrealistic, however, to assume that, for an HT release, the concentration in a small pond will equal that of soil water, because HT deposited on water is not converted to HTO as it is when it lands on soil. The pond concentration/HTO in air moisture ratio would be about the same after a release of HT as after a release of HTO, because most pond tritium comes from air HTO (in this case, emitted from the soil before being deposited on the water) even for an HT release. For this reason, in DCART, the drinking water for animals after an HT release is assumed to have a concentration 16.5% that of predicted air moisture.

Drinking water for people in the Livermore Valley is obtained either from groundwater or surface water from distant sources, both of which are not contaminated with locally produced tritium (Moran et al. 2002; Beller et al. 2005; Moran 2005). Thus, in DCART it has been assumed that all drinking water for people is uncontaminated by LLNL tritium.

Doses

The annual dose from inhalation and skin absorption of HTO is calculated:

$$Dose_{inh\_HTO} = g_a \, C_{a\_HTO} \, I_{inh} \, 1.5 \, DC_{inh\_HTO} \quad [9a]$$

where:

- g_a = fraction of the consumed air arising from the contaminated source (assumed, at least initially, to be 1)
- I_{inh} = inhalation rate of adult, child or infant in $m^3 \, y^{-1}$ (Table A6)
- 1.5 = the factor that includes the dose from water intake through skin absorption relative to inhalation rate
- DC_{inh_HTO} = ICRP (1996) dose coefficient for inhalation of HTO for adult, child, or infant (Sv Bq^{-1}; Table A7)

The annual dose from inhalation of HT is calculated:

$$Dose_{inh\_HT} = g_a \, C_{a\_HT} \, I_{inh} \, DC_{inh\_HT} \quad [9b]$$

where:

DC_{inh_HT} = ICRP (1995) dose coefficient for inhalation of HT for adult, child, or infant (Sv Bq^{-1}; Table A7)

In addition to direct inhalation of HT or HTO, there is also inhalation of HTO from HT that has been converted to HTO in the soil and emitted back to the atmosphere. The concentration of HTO from a release of HT at head-height is 4 (Bq L^{-1} HTO / Bq m^{-3} HT) (Davis and Bickel 2000; Table A6). Dose from HTO converted from HT is, of course, calculated as in eqn 9a.

The annual dose from drinking water is calculated:

$$Dose_{dw} = g_{dw}\, C_{dw}\, I_{dw}\, DC_{HTO} \qquad [10]$$

where:

g_{dw} =the fraction contaminated (0 for the Livermore Valley)

I_{dw} = drinking water consumed by adult, child or infant (L y^{-1}; Table A6)

DC_{HTO} =the ICRP (1996) dose coefficient for ingestion of HTO for adult, child, or infant (Sv Bq^{-1}; Table A7).

The annual ingestion doses are calculated:

$$Dose_{ing} = (\Sigma(g_f\, I_f\, C_{f,HTO})\, DC_{HTO}) + (\Sigma\, (g_f\, I_f\, C_{f,OBT})\, DC_{OBT}) \qquad [11]$$

where:

g_f = fraction of the consumed food arising from the contaminated source (assumed, at least initially, to be 1)

I_f = consumption rate of the appropriate foodstuff (kg fw y^{-1}) by adult, child or infant (Table A6)

C_f = concentration of HTO or OBT in the appropriate foodstuff (leafy vegetables, fruit or fruit vegetables, root crops, grain, milk, beef, pork, poultry, and eggs) (Bq kg^{-1} fw)

DC_{OBT}= the ICRP (1996) dose coefficient for ingestion of OBT for adult, child and infant (Sv Bq^{-1}; Table A7)

Default parameter values are based on diets based on intakes in g per kg-day for different age groups as presented in the Exposure Factors Handbooks (EFH) of the US EPA (1999 and 2002). Each diet contains about 80% of the mean caloric intake of an average adult, child age 10, and infant age 6 months to 1 year. The diet does not account for fish, nuts, and essential oils, which will not be affected to any degree by tritium releases in the Livermore Valley. The diet is thus relatively complete and, if it were assumed entirely contaminated with tritium, it should over-compensate for variations in the actual hypothetical diet of the Site-Wide Maximally Exposed Individual (SW-MEI[8]). Default

[8] A hypothetical member of the public who receives the greatest LLNL-induced effective dose equivalent (summed over all pathways) from all sources of radionuclides released at a site.

water consumption rates are intake rates for tap water from the EFH; inhalation rates are also from the EFH; Table A6).

Dose coefficients (ICRP 1995, 1996) are summarized in Table A7.

In addition to the major contributors to dose (i.e., inhalation and ingestion), there was another pathway at LLNL to dose – swimming in the LLNL pool. The contribution from this is negligible compared with doses from inhalation and ingestion. The calculation is included for the sake of completeness.

The annual dose from immersion in water (Osborne 1968) can be expressed as:

$$Dose_{imm} = C_p\, A_{bs}\, I_s\, H_s\, T\, DC_{HTO} \qquad [12]$$

where:

- C_p = concentration of HTO in pool water (Bq/g) (observed or computed from known air concentrations)
- A_{bs} = surface area of the body (m^2); see Table A8.
- I_s = intake rate of HTO through skin (5.1 L/min m^2)
- H_s = humidity at skin surface (0.02724 g per L at mean temperature (28° C) of LLNL pool)
- T = duration of time swimming in minutes
- DC_{HTO} = the ICRP (1996) dose coefficient for skin absorption for adult, child and infant (Sv Bq^{-1}; Table A7)

Although concentrations in pool water were not measured for some of the years the pool was in use, pool water concentrations can be estimated. Air moisture concentrations measured at the LLNL Discovery Center (previously the Visitors Center) are quite well correlated with air moisture concentrations measured adjacent to the pool. Pool water concentrations are in turn correlated with air moisture concentrations adjacent to the pool (Table 3). Thus, although with increased uncertainty, the annual mean air moisture concentrations measured at the Discovery Center can be used to estimate pool water concentrations. Pool water concentrations estimated from the values for the two ratios in Table A8, however, tend to overestimate tritium concentrations in the swimming pool when tested against the years with observed data.

Seasonal vs annual mean concentrations in vegetables

Regulatory models calculate doses based on annual average release rates and annual average concentrations in foodstuffs and in air, because the calculations are relatively easy to do and err on the side of conservatism. Food, either for animals or people, that is not eaten fresh, is assumed stored and eaten later. Radiological decay accounts for loss of activity during this time. DCART, like regulatory models, assumes that the vegetables and animal products contain the annual average concentrations when they are ingested but does not account for radiological decay because of the 12.32-year half-life of tritium

and the assumptions that no foodstuff is stored for more than eight months and that most foods are eaten within days of harvest.

In the Livermore Valley, vegetables can grow nearly year round, although certain types of vegetables may only grow in particular seasons. In summer, if things are to grow, they need to be watered. To account for drought conditions, an irrigation pathway was added to DCART. However, as DCART evolved, the ratios of tritium in soil moisture to either HTO in air moisture for an HTO release or HT in air for an HT release were set to experimentally observed values. Because of this calibration, soil tritium concentrations have to be the same regardless of water content of the soil or the source of the soil water. The irrigation pathway is therefore unnecessary unless the irrigation water were to contain tritium from LLNL operations, and it has been disabled for the TDR.

If the DCART deposition pathways (eqn [1] – [3]) were to be activated and the ratio (0.3 soil concentration/air moisture concentration) maintained by loss of tritium from the soil (eqn [3]), the model requires that at least 8 cm of rain or irrigation water be input so that the fraction of HTO retained by the soil will not become negative (a physical impossibility). This problem is academic, however, because edible vegetation needs more than 15 cm of water per year to grow, and because more than 15 cm of rain has fallen every year since the laboratory opened.

If DCART were used to predict seasonally rather than annually, any differences between predicted winter and summer concentrations in vegetation would be due to different assumptions about absolute and relative humidity and different wind speeds and direction. Summer absolute humidity is higher and summer relative humidity is lower than during the winter, but obviously the annual average accounts for both, and the use of mean concentrations in vegetables will account for differences in concentrations depending on the season. The uncertainty is such that it would be pointless to attempt to model winter vegetables differently from summer vegetables, even though some vegetables only grow well at certain times of year. In addition, if predictions were to be made for periods of less than one year, input files based on seasonal meteorological data would have to be generated to replace the files from annual data that are normally used in dispersion models when calculating dose from routine releases.

The above discussion of seasonal changes in absolute and relative humidity only affects concentrations in plants after a release of HTO. For an HT release, the concentrations in vegetation are independent of absolute and relative humidity because the HTO in air moisture is set equal to the observed ratio of 6 Bq L^{-1} in air moisture to Bq m^{-3} HT in air at plant height (Davis and Bickel 2000; Table A4). Thus, the only seasonal effect on tritium concentrations at a particular location would be due to differences in wind patterns.

UNCERTAINTY AND SENSITIVITY ANALYSIS OF DCART

All uncertain parameters that are not source, receptor or time specific[9] have been given distributions (see the Appendix) to be sampled using the Latin Hypercube Sampling (LHS) option in Crystal Ball® to assess the 95% confidence interval on doses to adult, child, and infant and to determine the parameters to which the model is sensitive. Obviously, the uncertainty about the input that drives the model (source term, dilution factor estimated from the dispersion model, or the average annual observed air concentrations at a particular location) will have a major effect on the uncertainty on the doses. As well, these same parameters, when varied, may be those to which the model is most sensitive. Thus, the results of an actual uncertainty and sensitivity analysis must be unique to each annual LLNL input, given that the magnitude of the sources at each location may vary considerably each year. Assessments of the uncertainty for each year of the LLNL dose reconstruction will be calculated and reported when the dose reconstruction is carried out; assessments of the sensitivity will be calculated and reported for a few of the years of the dose reconstruction. For now, the sensitivity and uncertainty analyses presented here apply only to the parameters in the Appendix and to the specific distributions assigned to them. The results in Tables 4 and 5 were calculated using Crystal Ball® assuming an input of unit air concentration (1 Bq/m^3 of HT or HTO) and running DCART 10,000 times with LHS. Because the precipitation pathway is bypassed in DCART and these calculations are driven by concentrations of tritium in air rather than by source terms, only parameters listed in Tables A3 – A7 are included in the analysis. The analysis carried out in this way will be referred to as the generic analysis.

Uncertainty analysis

Assuming unit concentration of HT or HTO in air, the magnitude of the 95% confidence interval when all generic parameters are varied is shown in Table 4. In addition, the ratio between the mean of the output distribution and the deterministic dose is shown for all generic parameters.

The uncertainty is greater on doses from HT releases than on doses from releases of HTO because of the large uncertainty on the conversion of HT to HTO in the environment (see Tables A3 and A4). Uncertainty is highest for infant doses, in large part due to uncertainty in infant diet. The magnitude of the 95% confidence interval is reduced significantly when either the uncertainty on the dose coefficients or the uncertainty on diet is removed. If uncertainty on both the dose coefficients and diet is removed, the magnitude of the 95% confidence limit for a dose from a release of HT is reduced to a factor of 3.5; for a release of HTO, the overall uncertainty is less than a factor of 2.

Removing the uncertainty on the dose coefficients not only reduces the overall uncertainty on all doses considerably, it also brings the mean of the probabilistic doses close to that of the deterministic doses. The difference of more than a factor of two between the probabilistic and deterministic doses is due to the fact that distributions of

[9] These distributions have been estimated for Part 2 of the TDR.

dose coefficients are skewed very high compared with recommended ICRP values. Yet, until the ICRP or other regulatory body[10] agrees to higher deterministic values for a dose coefficient, the values currently recommended must be used.

As mentioned, more uncertainty will be introduced when the distributions on source terms, and dilution factors are included.

In some circumstances, knowing the confidence that can be placed in the predictions of concentrations in rainfall is important. To test uncertainty associated with predicted concentrations of tritium in rain, relatively high, hypothetical HTO release rates from both stacks and area sources were chosen (2000 Ci from the Tritium Facility, 4.4 Ci from the Building 612 yard and 7.3 Ci from the Building 331 waste accumulation area [WAA]). Meteorological data for LLNL for 2002 were used as input. For the precipitation sub-model, rainfall concentrations must be calculated at a particular location, so tritium concentrations in rain from each source were calculated at the Discovery Center, which is adjacent to the location of LLNL's SW-MEI for compliance with 40 Code of Federal Regulations (CFR) Part 61, Subpart H (National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities – NESHAPs). The parameters that were sampled are those from Table A1. Uncertainty on rainfall concentrations is high, ranging from a ratio (upper confidence limit divided by lower confidence limit) of 7.7 for predicted rain concentrations when the source of the tritium was the Building 331 WAA to 10.7 when the source was the Building 331 stacks. LLNL has monitored concentrations in rain for many years at the Discovery Center and elsewhere, and it should be possible to test DCART's precipitation sub-model using the LLNL data.

Sensitivity analysis

The sensitivity of predictions to specific parameters strongly depends on the set of input parameters and the endpoint. The input to DCART for this sensitivity analysis, as for the uncertainty analysis above, was unit concentration of HT or HTO in air (Bq m^{-3}). The endpoint of this analysis was the total dose to adult, child and infant.

The results of this generic sensitivity analysis are shown in Table 5. The four most important parameters to each dose endpoint, as determined by the rank correlation coefficient, are shown.

All of the parameters in Table 5 have relatively large uncertainties that are either due to lack of knowledge, natural variability, or a combination of both. The negative correlation coefficient for leafy vegetable intake (I_l) is due to the fact that I_l is correlated with fruit

[10] The EPA (Eckerman et al. 1999) recommends slightly different dose coefficients. For inhalation of HT or HTO and ingestion of OBT, the ICRP and EPA dose coefficients are within 2% of each other; for ingestion of HTO, the EPA's values are higher than those of the ICRP by 15% for a 1-year old and by 7% for a 10-year old and an adult. Both EPA and ICRP dose coefficients are about a factor of two lower than the geometric means of the distributions (Harrison et al. 2002) (Table A7).

vegetable intake (I_f; Table A4). The dose is more sensitive to I_f than to I_l, so if I_l increases, then I_f decreases, as does the dose.

When a prediction is sensitive to a parameter, the parameter should be examined to see if its uncertainty can be reduced. In the case of the parameters in Table 5, nothing at this point in time can reduce the magnitude of the uncertainty (Tables A4, A6, and A7). More experiments could refine the uncertainty on the ratio between HTO in plant water and HT in air, but the predictions of dose from an HT release would remain sensitive to the ratio. When the ICRP or other regulatory body addresses the uncertainty in the dose coefficients, sensitivity to the dose coefficients should be reduced. The uncertainties in ingestion rates are due to natural variability and presumably cannot be reduced. Sensitivity of the model to some parameters will always exist, regardless of how well the values are known and described.

A sensitivity analysis was also run on the precipitation sub-model. The model is very sensitive (correlation coefficients between 0.57 and 0.75 depending upon the source of tritium) to the washout coefficient and to the fraction of time the wind blows into a sector when raining (Table A9).

TESTING DCART

BIOMASS scenarios - concentrations of tritium in plants, soil, and rain

Over the past twenty years, several international efforts have been directed at testing models that calculate doses from releases of various radionuclides to the environment. The last completed model testing program was the International Atomic Energy Agency's BIOMASS (BIOspheric Modeling and ASSessment) program (IAEA 2003). One of the groups, the Tritium Working Group, analyzed the results of the following five scenarios.

- Scenario 1: Modeling of the steady-state behavior of HT and HTO in the environment when atmospheric releases are assumed to be on average nearly constant and a steady-state equilibrium has been reached.
- Scenario 2: Model-model intercomparison exercise for predicting the rise of tritium from contaminated ground waters.
- Scenario 3: A test of chronic atmospheric release models using Canadian data.
- Scenario 4: A test of chronic atmospheric release models using Russian data.
- Scenario 5: A test of chronic atmospheric release models using French data.

DCART has been tested in all scenarios except Scenario 2. DCART was under development during the period of the BIOMASS program, and the current version will give somewhat different results. Differences between the version tested and the current version are mentioned below. For Scenarios 1, 4 and 5, air concentrations used as input to DCART had to be predicted using dispersion models. Two different models were used

for BIOMASS depending upon the form of the meteorological input data. One was the dispersion model from CAP88-PC (Parks 1997) and the other was the Canadian Standard N288.1 (CSA 1987). Results of these dispersion models will not be discussed here, except as how they affect concentrations predicted by DCART.

Predictions for BIOMASS scenarios had to be submitted before any observations were released. This way, modelers could not tune their results. However, once the observations were revealed, modelers could revise their predictions based on having found mistakes in the codes or on having misunderstood the scenario description. The final results presented in the reports (IAEA 2003) are revisions, as are the results presented below. (If a participant chose not to revise obviously wrong predictions, they were omitted from the analysis.)

Those aspects of DCART's performance in BIOMASS that are relevant to using DCART to reconstruct doses from routine releases at LLNL are presented below.

Scenario 1 – Model Intercomparison

Scenario 1 assumed a continual release of a gram of either HT or HTO from a 60 m stack over the course of a year. Frequency of occurrence of stability classes and mean wind speeds for each stability class were provided, as were deposition velocities for HT and HTO. Endpoints asked for were tritium concentrations in air, soil water, plant water and combustion water (OBT) (all in Bq L^{-1}) at every 100 m, from 100 to 1000, and every 1,000 meters thereafter to 10,000 meters. The Canadian dispersion model (N288.1), with a vertical dispersion parameter recommended in the scenario description, was used to calculate air concentrations.

Because this exercise was a model intercomparison, no answer could be right or wrong. Model intercomparisons simply reveal the degree of consensus (or lack thereof) between modelers. Eleven models participated in the HTO scenario, while nine participated in the HT scenario. LLNL joined BIOMASS after this scenario closed, but the scenario was used as the first test of DCART early in its development. Among all models tested, the variability in model results was higher close to the source due to the use of different dispersion assumptions. For example, some modelers included the contribution from resuspended HTO in their estimates of air concentrations. Agreement was better at distance. Because LLNL's major interest in a dose reconstruction will be dose to an individual living offsite, results between the participating models and DCART are compared in Table 6 for 1,000 and 5,000 m from the source. For the release of HTO, results have been normalized to the median air concentration of each data set so that, in effect, all models are starting with the same air concentration, thus nullifying the effects of dispersion modeling. For the HT release, HT concentrations in air cannot be normalized because this endpoint was not requested, so the effects of dispersion modeling carry through the other predictions. The estimation of HTO in air after a release of HT is different for each model.

The modelers agree quite well on how to model HTO, because the spread in predictions is less than a factor of 2.5, except for soil, which has a spread greater than a factor of 5.9. DCART's results are very close to the median concentration predicted by the other modelers, except for soil. DCART's soil water/air moisture ratio was 0.44 (now calibrated at 0.30), which is low compared with other models in this scenario, which were using higher (i.e., screening) levels (IAEA 2003). High soil water/air moisture ratios are due to high deposition velocities coupled with explicit or implicit low emission rates. In general, soil pathways are not considered very important to a tritium dose model.

The uncertainty in the predictions of concentrations of HTO in the environment from a release of HT is much greater, ranging up to a factor of 36 between the lowest and highest predictions for soil concentrations at 1000 m. Between the time when the participants submitted results and when DCART's predictions were calculated, much had been learned about the concentrations of HTO in the environment after a release of HT because of ongoing analysis of the data from the 1994 HT release at Chalk River Laboratories, Canada (Davis and Bickel 2000). These studies revealed that more HTO is emitted than had previously been thought. Thus, as can be seen, DCART's prediction for HTO in air moisture is the highest (although this may be partially due to the results of dispersion modeling), and DCART's predictions for the other media are amongst the highest. The ratio of HTO in air moisture to HT in air for this comparison was 5.7 based on Chalk River observations. As mentioned, this ratio in DCART is now based on 6.0 (with uncertainty) due to improved understanding of the uncertainties on the Chalk River data.

Scenario 3 – Chalk River Laboratories, Chalk River, Canada

Scenario 3 (IAEA 2003), based on observations at Chalk River Laboratories (CRL), Ontario, Canada, was the first opportunity to test DCART against field observations. Modelers were provided with comprehensive meteorological data measured at CRL for June 5 – August 10, 1995. These included hourly and daily averages of relative humidity, air, leaf and soil temperatures, rainfall and rain intensity, wind speed, wind direction and stability class. Source terms (Bq s^{-1}) were provided for the reactor stack, the reactor building and a contaminated lake. In addition, measured daily concentrations of tritium in air (Bq m^{-3}) were provided for about half the days of the study for three locations at CRL. Average observed air concentrations were used to drive DCART, as was the intention of the scenario developer, to limit the uncertainty arising from the use of various dispersion models.

The observational data at three sampling locations against which the models were tested included:

- TFWT sampled at 9:00 (representing nighttime processes) and 15:00 (representing daytime processes) daily,
- Soil water concentrations sampled at 9:00 and 15:00 daily,
- OBT in plants sampled on June 28, July 12 and August 9, and

- Concentrations of tritium in rainfall at one site.

As well as calculating the above endpoints, plus a 24-hour mean concentration in plant water, modelers were asked to provide the 95% confidence intervals on all predictions.

DCART's predicted to observed (P/O) ratios for the chronic endpoints (Table 7) were similar to those of the other participants. With the exceptions of rain and OBT on August 9, DCART overestimates the observations by a small amount. If predictions were made again with the present DCART, P/O ratios would be closer to 1.0. The low P/O ratio for rain was due to the observed ratio (rain/air moisture in Bq L^{-1}) being about 0.8, which is higher than any model result. In fact, only DCART and one other model had uncertainty bounds that overlapped the uncertainty on the observed concentration in rain. As seen in Table 7, the P/O ratio for OBT in grass drops over time. This is because the observed OBT in plants increased over time relative to the air concentrations. This too is an unexpected result and was not predicted by any model.

The various ratios predicted by DCART varied depending upon the location and the time of day. The soil/air moisture ratio ranged between 0.34 and 0.44; the TFWT/air moisture ratio ranged from 0.85 to 1.0; and the rain/air moisture ratio ranged from 0.19 to 0.28; the OBT/TFWT ratio was 0.8. In the version of DCART developed for the LLNL TDR, the soil/air ratio for HTO is 0.3 and the OBT/TFWT ratio is 0.7.

At the time Scenario 3 was modeled, DCART could not be run with Monte Carlo uncertainty analysis, so parameter perturbation was used to assess uncertainty on the predictions. The median values of various parameters, plus or minus one standard deviation, were calculated from a database of variables used in DCART. These were then combined as a set of values that would produce a low average predicted concentration and a set of values that would produce a high average predicted concentration. The three sites are very different, so the uncertainty on the predictions varied with the site. Nevertheless, in general, the uncertainty on the plant water concentrations and OBT was about a factor of 3.3 (high/low). Uncertainty in soil concentrations was roughly a factor of 50. Because these uncertainty estimates were extremes, the demonstrated uncertainty was higher than the 95% confidence interval that would have been predicted using Monte Carlo analysis.

In the stochastic version of DCART to be used for the TDR, the ratios of the upper confidence limit divided by the lower confidence limit of the output distributions are about two, three and six for HTO in pasture, OBT in pasture, and soil water concentrations, respectively. Uncertainty in soil water concentrations in the TDR version of DCART is driven by the assumption that the soil water to air moisture ratio is always 0.30.

Scenario 4 - Russian Federal Nuclear Center, Sarov, Russia

The Sarov Scenario (IAEA 2003) asked for concentrations in air, plant water, soil water, snow, and rain at 500, 1,700, 2,800, 3,000, 5,000 and 7,600 m from a source that annually emitted equal quantities of HT and HTO. The modelers were provided with annual emissions in arbitrary tritium units for each year after the start-up of operations. Emissions peaked in the 5th year of operations at 120 arbitrary tritium units; in the 10th year, 12 units were emitted; in the 15th year, 4 units were emitted; and for years 16-20 after start-up (1985-1989), 1 unit per year was emitted. The actual release rate from Sarov was kept secret this way, so all results that the modelers discussed were the normalized ratios of measured or predicted concentrations to the measured or assumed release rates.

Unfortunately, the only long-term average concentrations of HTO measured were those in air moisture[11]. As a result, the only model predictions that could be compared meaningfully with observations were air concentrations based on assumptions about absolute humidity. Thus the Sarov scenario really only tested the dispersion model used (CSA-N288.1). Nevertheless, this scenario has been described here in detail because of one of the conclusions of the BIOMASS Tritium Working Group (IAEA 2003). At Sarov, there was some evidence for tritium retention in biosphere media after a significant decrease in emission rates (in this case, over 2 orders of magnitude during 20 years of releases). The observed tritium air concentrations showed a slight increase in time when normalized by the release rates. However, the magnitude of the increase was comparable to experimental uncertainties, which demonstrates a need for further study to determine if retention really does occur and, if necessary, to determine those processes responsible for it so that it can be taken into account in long-term assessment models. Two of the participating models attempted to account for memory effects, but results were not distinguishable from those of DCART or the other models.

The fraction of HTO in air contributed by the HT releases could not be distinguished from the HTO in air contributed by HTO releases. Therefore, there was no discussion about the fraction calculated in the different models.

Scenario 5 – Centre Energie Atomique, Valduc, France

The Valduc Scenario (IAEA 2003) described three sources of tritium with annual releases of HTO and HT from 1983 to 1998. At specified locations, modelers were asked to predict annual average tritium concentrations in air, plant water, and rainwater. Six models participated. Concentrations of OBT in tree rings of birch for 1983 to 1988 and OBT concentrations in oak leaves were also endpoints. The dispersion modeling for this scenario was complex because of the three sources, uneven terrain and numerous receptor sites. CAP88-PC was used as the dispersion model, and it performed as expected

[11] Plant and soil water samples were collected on only one day each year, so model predictions could not be compared meaningfully with the observations.

(Peterson 2003, 2004[12]). Stations 1-4 ranged from 3,500 to 6,900 m from the sources. The distances from the sources to the birch tree ranged from 9,000 to 9,500 m.

Vegetation was sampled monthly at four locations for eleven years. For this comparison of modeling results, concentrations in vegetation were normalized based on observed air concentrations to remove the bias of different dispersion models. Normalized concentrations of vegetation are compared with observations in Table 8. DCART's predicted TFWT concentrations lie for the most part midway in the range of the other modeling results. All participants overestimated TFWT concentrations.

Rainfall was collected under oil monthly to provide time-integrated concentrations of tritium in rain at the same locations as vegetation (Table 9). Because DCART calculates concentrations in rain independently of any dispersion model (eqn [1]), there is no reason to normalize rain concentrations to observed concentrations of tritium in air. On average, DCART shows a small tendency to underestimate concentrations of tritium in rain (Table 9).

DCART's predictions of OBT in birch tree rings ranged from 0.82 to 3.2 of the observed values; DCART's average P/O ratio for the six years was 1.8. DCART generally overestimated the concentrations in tree rings, but it was on average closer to the observations than all other models (with overall average P/O ratios of 2.4 –3.3). It was impossible to normalize the predictions of concentrations of OBT in the beech tree rings to air because no air concentrations had been measured at the locations of the trees. DCART's overestimation of OBT in tree rings was probably not due to an overestimation of HTO in air by CAP88-PC (see footnote 12).

Modelers also had to predict concentrations of OBT in oak leaves at 36 locations at the Valduc site. These locations ranged from 1.7 to 15 km from the vicinity of the tritium sources. P/O ratios of DCART ranged from 0.26 to 1.8, with a mean of 0.92 and a median of 0.83. The average results of the other five models ranged from 1.4 – 2.2. No measurements of tritium in air were taken at the location of the oak trees, so data could not be normalized to known air concentrations. DCART's small underestimation of observations is probably due to underestimated air concentrations at distance by CAP88-PC (Peterson 2003).

Average ratios and range of ratios of various endpoints to air moisture are shown in Table 10 for DCART. DCART's OBT/TFWT ratio for grass and oak leaves was 0.8 (now 0.7 for the LLNL TDR); for beech tree rings it was 0.4 (Kalin et al. 1995).

[12] All predictions for the Valduc locations fell within a factor of three of the observations. Only about 37% of the predictions were higher than the observations, which is similar to CAP88-PC's performance at locations similarly distant from LLNL.

BIOMASS scenarios – summary

Out of four scenarios, only a small fraction of the data was appropriate to test chronic release models. Although there were many measurements of concentrations of HTO in air, these data are primarily only useful to test dispersion models. At Chalk River, TFWT samples were taken frequently enough to be averaged meaningfully; the monthly TFWT samples taken at Valduc may not be quite as representative of an annual average. Because it is expensive and time-consuming to sample OBT, oftentimes OBT measurements are too few to test models adequately. At Chalk River, it would have been better to have had more OBT measurements, but at least those Chalk River OBT measurements corresponded to tritium measurements in air. In contrast, at Valduc, although there were many OBT measurements, there were no corresponding air concentrations with which to compare them. Samples for model testing need to be collocated in time and space. At Valduc, samples of rainfall and air concentrations met this criterion, thus providing reliable ratios for model testing. In contrast, the rainfall data for Chalk River were not collected at the same location as any of the air samplers, although distances between them were not great.

EMRAS scenarios - concentrations of tritium in plants, animals, and soil

The current Coordinated Research Program of the International Atomic Energy Agency, similar in form and content to BIOMASS, is EMRAS (Environmental Monitoring for Radiation Safety). EMRAS began in 2003 and will terminate in 2007. The Tritium/Carbon-14 Working Group has been very active and expects to complete nine scenarios. Eight of these scenarios test models against observations, while one is a model intercomparison; seven address tritium and two concern ^{14}C; five are scenarios for acute releases, and four are for equilibrium conditions; two of the scenarios are for aquatic environments, while seven are for terrestrial environments. DCART has been tested in the two EMRAS scenarios for terrestrial pathways at equilibrium.

Pickering Scenario, Canada

For the Pickering Scenario, crops, animal products, and soil moisture were sampled at three farms and an experimental garden at various distances from the Pickering Nuclear Generating Station in the province of Ontario. Modelers were asked to predict OBT concentrations in vegetation (fodder, fruit, root crops, raspberry leaves), in animal products (milk, calf and chicken meat, eggs), and in soil moisture using observed concentrations in air moisture at each location provided as input.

DCART's results were either similar to or better than those of the six other participants. DCART's predictions of concentrations in soil and vegetation were similar to those of the other models. DCART's predictions of soil moisture concentrations were slightly higher than observed, but the observations were included in the 95% confidence interval. DCART's predictions of concentrations in most vegetation were so high that only the

2.5% confidence limit overlapped the confidence interval on the observations; that, however, was a better result than was achieved by several of the models. DCART was noticeably better at predicting concentrations in animal products than the other models. For example, DCART was the only model to correctly predict concentrations of OBT in eggs and was one of two to correctly predict HTO concentrations in eggs and OBT concentrations in calf flesh. All observations for animal products fell within DCART's predicted 95% confidence interval. In this scenario, concentrations of tritium in drinking water were supplied. Because drinking water is a major contributor to tritium body burden, the effect of the over-predictions of fodder concentrations was damped. DCART's drinking water contributed about 60% and 70% of the cow and the chicken's body burden, respectively.

This scenario presented a valuable opportunity to test DCART against animal data for the first time. Results are summarized in Table 11, and details may be found in Peterson (2005).

Pine Tree Scenario, Japan

For the Pine Tree Scenario, modelers were supplied with monthly emission data for 1981 through 1987 from four stack sources of tritium and asked to predict the mean monthly concentrations of tritium in air moisture, rain, and pine needles, at two locations and all of these plus tree rings at a third location. Hourly meteorological data were supplied. The dispersion model used with DCART for these calculations was CAP88-PC. Predictions for DCART were annual rather than monthly means, because an annual mean is a better test of both CAP88-PC's dispersion modeling and DCART's dose predictions.

The Japanese data set unfortunately only included air moisture concentrations for one location. It was assumed that air moisture concentrations at the other locations would be very similar, but this assumption may have introduced considerable uncertainty into the test. All air moisture concentrations were underestimated by DCART, and the upper confidence limits on the predictions did not include the observations; the mean of the P/O ratios, without one high value, was 0.23. These results were surprising given that CAP88-PC when tested with LLNL data (Peterson 2003, 2004) or the Valduc data (see above) were within a factor of 3 and overestimated air concentrations more than half the time. Only two other modelers participated in this scenario (and both submitted monthly predictions), but only one of them (a Japanese) predicted air moisture concentrations close to the observed values. The Japanese meteorological data were provided with ten stability classes. Models such as CAP88-PC need data for just six stability classes. The failure of non-Japanese models to predict the observed air moisture concentrations may have been due to errors that occurred while converting from ten to six stability classes.

Using the low predicted air moisture concentrations as input, DCART underestimated the concentrations in pine needles and tree rings. However, when the observed air moisture concentrations were used as input, the predictions of tritium concentrations in needles and tree rings were very good (Table 12); all observations were included within

DCART's 95% confidence interval. The model in DCART for predicting rain concentrations (eqn. [1]) is independent of CAP88-PC's dispersion model and performed adequately (Table 12); most of the observations were included within DCART's 95% confidence interval.

A complete discussion of the scenario results may be found in Peterson (2006).

Models for animal products compared

Experiments (Moghissi et al. 1987) and monitoring of equilibrated environments (Evans 1969) have shown that the T/H ratio in animals equals that of their environment when all compartments of that environment have the same T/H ratio. The calculation of tritium concentrations in meat and eggs in DCART is based on these observations, as is the calculation in NORMTRI (Raskob 1994). The models in DCART and NORMTRI are very similar. With the same input, concentrations of OBT in beef and poultry are the same in both models; all other concentrations calculated by DCART are about 10% higher than those of NORMTRI, except for HTO concentrations in eggs, which are about 20% higher. Thus DCART does not underestimate concentrations in animal products compared with NORMTRI, nor does it overestimate significantly.

Recently Galeriu et al. (2001) developed a model based on the hydrogen metabolism of animals. This model uses the type of transfer factor (d kg^{-1}) found in some regulatory models to calculate concentrations in animals from intakes of feed and water. However, because each possible transfer from diet HTO or OBT to animal HTO or OBT does not have an equal chance of occurring, four different transfer factors were developed[13]. These transfer parameters vary depending upon various assumptions about the metabolic status of the animal. When diets are not uniformly contaminated, this approach potentially gives a better estimate of fractions of HTO or OBT in the animal and thus a better estimate of dose than does the specific activity model used in DCART.

Concentrations in animal products have been compared between the Galeriu model and DCART assuming the same ingestion rates and concentrations of tritium in the diet. In Figure 1, the deterministic predictions of the Galeriu model are compared with the 95% confidence intervals of DCART for all animal products. Most results from the Galeriu model fall midway between the uncertainty bounds of DCART, indicating that the model predictions are comparable. The Galeriu predictions of OBT in milk and pork fall within the DCART's confidence interval but are not centered. This indicates less agreement between the models. Nevertheless, although the Galeriu model is potentially more accurate, in this comparison the results may be considered indistinguishable.

[13] HTO in vegetation to HTO in animal; HTO in vegetation to OBT in animal; OBT in vegetation to HTO in animal; OBT in vegetation to OBT in animal

Doses compared: DCART vs NEWTRIT

DCART has also been compared with NEWTRIT, a relatively simple model geared to regulatory compliance (Peterson and Davis 2002). Doses will differ because of the different diets used in each model, so doses will not be compared here. Other differences stem from the greater scope of DCART (more detailed modeling) and the more realistic (vs. conservative) choice of parameter values, coupled with uncertainty analysis, in DCART.

The mean and the 2.5 and 97.5 percent confidence limits of DCART's stochastic predictions are compared graphically with NEWTRIT's deterministic prediction in Figure 2 (for HT) and Figure 3 (for HTO). Initial air concentrations were the same (1 Bq m^{-3}), as was the absolute humidity (8 g m^{-3}).

In Figure 2 (for HT), the inherent conservatism of NEWTRIT stands out. NEWTRIT's prediction is either above the upper confidence limit of the DCART prediction or is not far below it. Parameters for transfer of HT in the environment were chosen deliberately high so that NEWTRIT would be more acceptable to regulators who had been requiring that a release of HT be modeled as if it were a release of HTO. Concentrations in vegetables are much lower in DCART than NEWTRIT because DCART is calibrated to an HTO in air moisture to HT in air volume ratio of 6.0, while NEWTRIT is calibrated to 8.0. Furthermore, the plant water concentrations in NEWTRIT are set at 1.5 times air moisture, while in DCART, plant water concentrations are only about 1.1 times air moisture. The large difference between DCART's mean concentration for root crops and NEWTRIT's is due to the fact that DCART assumes that the root crop is nearly in equilibrium with the soil water, which is 30% the concentration of air moisture. In contrast, NEWTRIT, having no soil compartment, assumes conservatively that the concentration in root crops is the same as that of fruit vegetable.

In Figure 3 (for HTO), the mean of DCART's stochastic predictions is not very different than NEWTRIT's predictions for vegetables, with the exception of root crops (explained above). These differences are accounted for by the assumptions about the relationship between concentrations in air moisture and plant water. In NEWTRIT, the concentration in plant water is assumed 90% that in air moisture; in DCART, the concentration in plant water can be decreased relative to air moisture due to the contribution of soil water, but it is also increased by the pressure difference between HTO and H_2O (γ in eqn [4]). Furthermore, the discrimination factors (OBT/TFWT) are different. In NEWTRIT the discrimination factor for OBT is 0.9 or 0.8 depending upon the vegetable, while in DCART it is 0.7.

NEWTRIT's higher predictions for animal products, obvious in both Figure 2 and Figure 3, are due to the conservative assumption in NEWTRIT that the concentration of tritium in the drinking water of animals is 50% that of air moisture (compared with 16.5% in DCART). NEWTRIT's concentrations of milk and beef, that lie above DCART's upper confidence limit, are due to a diet that is primarily pasture grass, rather than the mixture of pasture grass, hay and grain that is found in DCART.

CONCLUSION

DCART is a steady-state spreadsheet model for transfer of tritium from the atmosphere to dose. Developed at LLNL, DCART is adaptable, flexible, and transparent. It can be modified easily to account for new information. All doses are predicted as the mean and 95% confidence interval. DCART has been tested against observations and predictions of other models within international model validation groups. Some of the pathways are still untested because of the dearth of observational data; when transfers are not known well (as for animals), DCART errs on the conservative side. Because of this inherent conservatism, doses are highly unlikely to exceed the upper confidence limit of DCART's predictions.

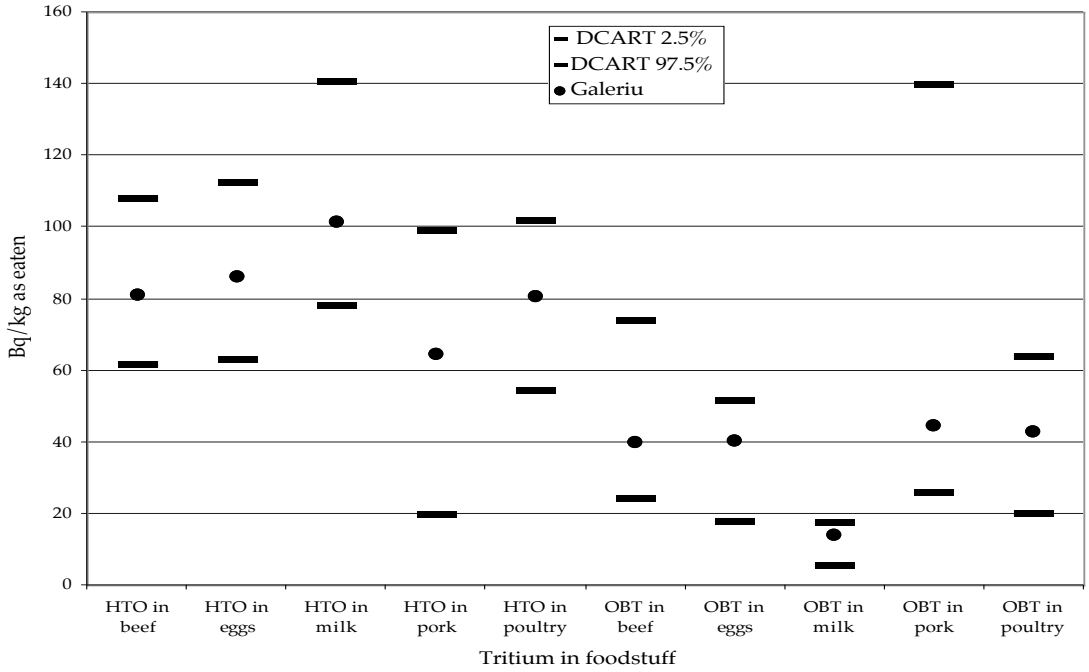

Figure 1. Deterministic predictions of the Galeriu metabolic model compared with the confidence interval on DCART's predictions for HTO and OBT in animal products. Only parameters for animal intake were varied in DCART, and all concentrations in pasture, hay, grain, and water were the same for both model calculations.

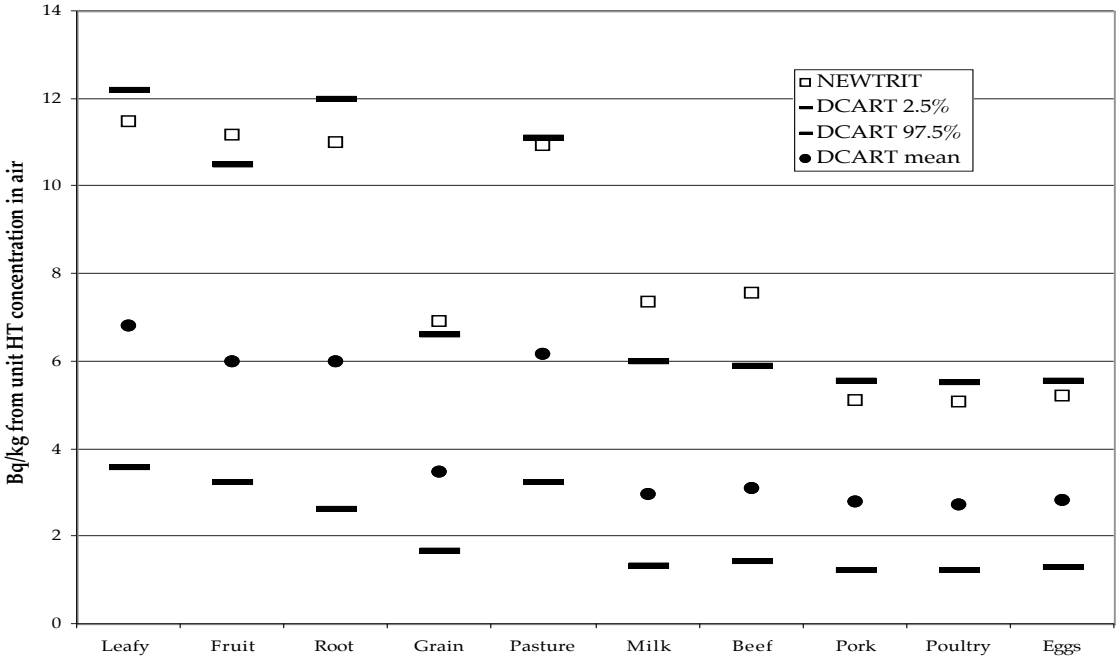

Figure 2. Concentrations of tritium in foodstuffs from a unit air concentration of HT predicted deterministically by NEWTRIT and probabilistically by DCART.

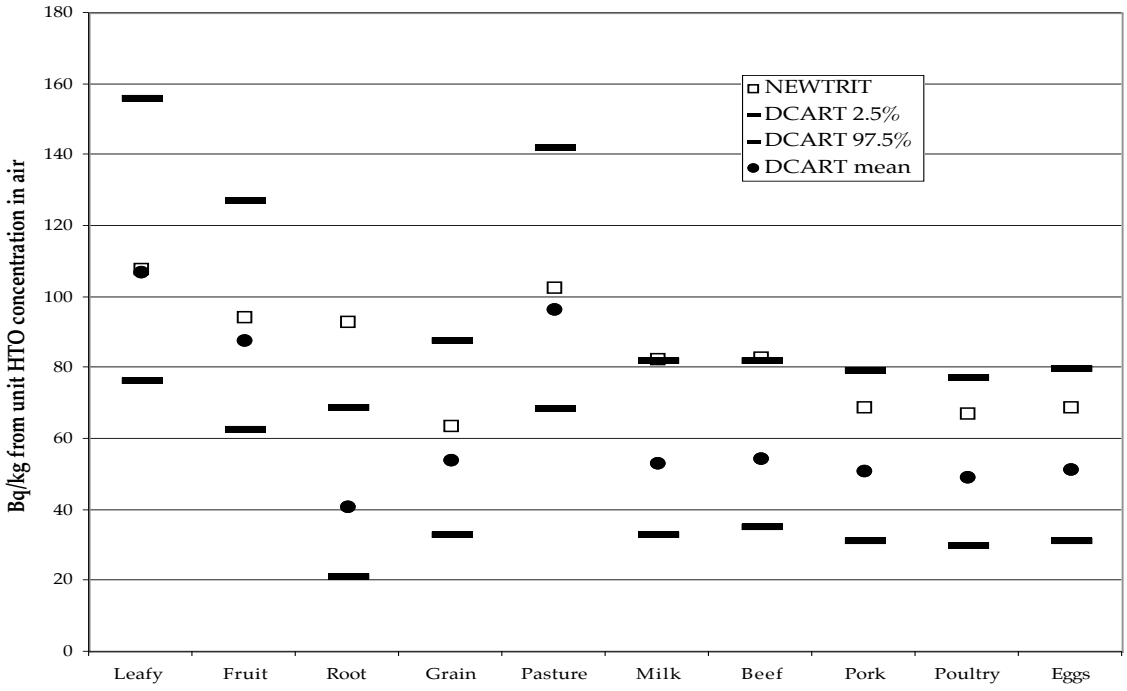

Figure 3. Concentrations of tritium in foodstuffs from a unit air concentration of HTO predicted deterministically by NEWTRIT and probabilistically by DCART.

Table 1. Comparison by year of types of doses (inhalation, ingestion) predicted by LLNL from species of tritium released (HT, HTO and HT + HTO modeled as HTO)

	Model	Inhalation	Ingestion	Release HT	Release HT as HTO	Release HTO	Ingestion from HTO	Ingestion from OBT
'53 -'72								
1973	A	X			X	X		
1974	B	X			X	X		
'75 -'83	C	X			X	X		
1984	C	X				X		
1985	C	X				X		
'86 & '87	C & D	X				X		
1988	C & D	X	X (D)			X	X	
'89 -'91	C & D	X	X (D)		X	X	X	
'92 -'97	D	X	X			X	X	
'98 -'05	D	X	X		X	X	X	

Blank spaces indicate that no tritium dose to a member of the public was reported by LLNL. Only DCART can account for dose from releases of HT and ingestion of OBT. The TDR will provide doses for missing years and pathways.
Model A calculated dose from the highest observed air tritium measurement from the LLNL perimeter.
Model B calculated dose from a meteorological diffusion model based on Gifford and Hilsmeier (1961).
Model C calculated dose using the Continuous Point Source model (Peterson 1976).
Model D calculated dose using AIRDOS-EPA (US EPA 1989)/CAP88-PC (Parks 1992, 1997); ingestion rate assumptions were changed in 2004.

Table 2. Maximum predicted annual dose in mrem at the site boundary from 1986 through 1991 from HTO released from the Tritium Facility at LLNL. Numbers were obtained from LLNL annual environmental reports (UCRL-50027-yr).

	Clean Air Act Code (AIRDOS-EPA [US EPA 1989])	Continuous Point Source (CPS) Code (Peterson 1976)
1986	0.03	0.04
1987	0.07	0.12
1988	0.55	0.10
1989	0.52	0.26
1990	0.27	0.099
1991	0.20	0.062

Note: LLNL only calculated dose using two different dispersion models between 1986 and 1991 as the transition was made to using the Clean Air Act Code exclusively.

Table 3. Ratios of tritium in pool water (PW) to tritium in air moisture (AT) for corresponding time periods of sampling between 1990 and 2000. Only data greater than the lower limit of detection were used.

Year	Mean PW/Mean AT	Median PW/ Median AT	Number of samples
1991	0.156	0.281	11
1992	0.248	0.272	11
1993	0.102	0.132	7
1994	0.138	0.128	12
1995	0.151	0.231	8
1996	0.0776	0.0969	9
1997	0.143	0.148	6
1998	0.137	0.193	4
1999	0.102	0.163	3
2000	0.150	0.200	2
All data	Overall mean	Overall median	All ratios
'90 – '00	0.166	0.162	79

Table 4. Magnitude of 95 % confidence intervals and comparison between means of DCART's probabilistic and its deterministic dose predictions for unit air concentrations of HT and HTO. Results are shown for HT and HTO when all generic parameters are varied, when all are varied except the dose coefficients (DC), and when all are varied except distributions on diet.

	Upper / lower confidence limits			Mean of probabilistic dose / deterministic dose		
	All	Less DC	Less diet	All	Less DC	Less diet
HT - Adult	6.8	6.1	4.0	2.6	1.2	2.4
HT - Child	6.3	4.9	4.5	2.3	1.2	2.2
HT - Infant	12.	9.8	4.8	2.3	1.2	2.3
HTO - Adult	4.4	3.7	2.4	2.4	1.1	2.3
HTO - Child	4.1	2.9	3.0	2.2	1.1	2.1
HTO - Infant	7.7	6.4	3.2	2.2	1.1	2.2

Table 5. The four most significant parameters and their rank correlation coefficients (r^2) for doses to adult, child, and infant derived from unit air concentrations of HT and HTO in DCART.

Parameter	r^2	Parameter	r^2
Adult dose from HT		Adult dose from HTO	
[a]HTO in plant / HT in air	0.49	Fruit or fruit vegetable intake	0.52
Fruit or fruit vegetable intake	0.49	HTO dose coefficient	0.38
Leafy vegetable intake	-0.32	Absolute humidity	-0.35
[a]HTO in soil / HT in air	0.31	Leafy vegetable intake	-0.34
Child dose from HT		Child dose from HTO	
[a]HTO in plant / HT in air	0.54	HTO dose coefficient	0.50
HTO dose coefficient	0.35	Absolute humidity	-0.36
Fruit or fruit vegetable intake	0.34	Fruit or fruit vegetable intake	0.35
[a]HTO in soil / HT in air	0.31	OBT dose coefficient	0.34
Infant dose from HT		Infant dose from HTO	
Milk intake	0.50	Milk intake	0.57
[a]HTO in plant / HT in air	0.44	Fruit or fruit vegetable intake	0.39
Fruit or fruit vegetable intake	0.40	HTO dose coefficient	0.37
HTO dose coefficient	0.28	Leafy vegetable intake	-0.29

[a] Units are Bq L^{-1} / Bq m^{-3}

Table 6. DCART predictions of HTO in air, plant water, and soil, and OBT in plants at 1,000 and 5,000 m from the 60 m source of HTO or HT divided by the median, maximum and minimum predicted concentrations submitted by participants in Scenario 1 of BIOMASS. Results from the HTO release have been normalized to the same starting air concentrations.

Ratios DCART/other models - continuous release of one g HTO over a year						
	Median		Maximum		Minimum	
	1000 m	5000 m	1000 m	5000 m	1000 m	5000 m
HTO in air	[1.0]	[1.0]	[1.0]	[1.0]	[1.0]	[1.0]
Plant water	1.0	0.99	0.81	0.78	1.2	1.2
OBT in plants	0.96	0.95	0.66	0.66	1.7	1.3
HTO in soil	0.73	0.68	0.39	0.34	2.3	2.3
Ratios DCART/other models - continuous release of one g HT over a year						
	Median		Maximum		Minimum	
	1000 m	5000 m	1000 m	5000 m	1000 m	5000 m
HTO in air	2.9	2.2	1.0	1.0	15.	9.8
Plant water	1.2	1.0	0.52	0.53	12.	5.4
OBT in plants	1.4	1.1	0.42	0.16	12.	5.4
HTO in soil	0.93	0.90	0.33	0.28	12.	5.1

Table 7. Predicted-to-observed ratios for DCART for the endpoints of the CRL Scenario

Endpoint	Location 1	Location 2	Location 3
Rain, 24 hour	0.30	------	------
Soil, morning	1.4	1.4	1.7
Grass, HTO, morning	1.1	1.1	1.1
Grass, HTO, afternoon	2.6	1.2	1.4
OBT, June 28	2.5	1.9	2.9
OBT, July 12	1.8	------	2.6
OBT, August 9	1.0	0.69	1.2

Table 8. Predicted-to-observed (P/O) ratios for concentrations of TFWT in grass at four locations at Valduc. Predictions were normalized to air moisture concentrations at each location. DCART's normalized means for 1988 through 1998 are compared with the range of normalized predictions submitted by the other participants.

Year	DCART's Normalized P/O Values			
	Station 1	Station 2	Station 3	Station 4
1988	3.4	2.6		2.3
1989	2.0	2.6		2.9
1990	1.0	2.2		1.7
1991	2.3	1.9		3.3
1992	1.2	1.8		1.5
1993	4.6	1.5		3.0
1994	1.4	1.6	2.1	1.8
1995	0.80	1.3	2.1	1.5
1996	2.7	1.9	2.0	1.8
1997	3.8	3.6	1.8	3.1
1998	1.3	1.7	1.6	2.2
DCART normalized mean	2.2	2.1	1.9	2.3
Modeler's Range – normalized means	1.7 – 2.6	1.8 – 3.3	1.7 – 2.6	1.8 – 2.9

Table 9. Predicted-to-observed (P/O) ratios for DCART and the range of the mean P/O ratios for all models for tritium in rain at Valduc.

Year	DCART's Normalized P/O Values			
	Station 1	Station 2	Station 3	Station 4
1988	0.50	0.86		0.24
1989	0.72	1.2		0.57
1990	0.63	0.65		0.48
1991	1.9	1.3		1.3
1992	0.72	0.71		0.45
1993	1.0	0.84		0.52
1994	1.5	0.87	0.82	1.1
1995	0.69	0.84	0.56	0.62
1996	1.4	1.2	0.56	0.91
1997	0.78	0.99	0.37	0.49
1998	0.79	1.2	0.66	0.56
DCART mean	**0.97**	**0.97**	**0.59**	**0.66**
Modeler's Range	**0.12 – 2.2**	**0.32 – 3.9**	**0.39 – 1.4**	**0.18 – 1.5**

Table 10. TFWT and rain concentrations predicted by DCART normalized to air moisture concentrations predicted by DCART for four locations at Valduc. The mean and range of DCART results for 1988-1998 are presented.

	Station 1	Station 2	Station 3	Station 4
Mean TFWT	1.0	1.3	1.0	1.1
Range TFWT	0.46 – 2.4	0.66 – 1.8	0.64 – 1.5	0.60 – 1.7
Mean Rain	0.24	0.69	0.14	0.13
Range Rain	0.12 – 0.57	0.36 – 0.92	0.10 – 0.20	0.08 – 0.18

Table 11. DCART's predicted-to-observed (P/O) ratios for mean HTO concentrations in soil moisture and mean concentrations of OBT in crops and animal products, Pickering, Canada.

	P/O Ratio
Soil moisture	1.2
Fodder at Farm A	1.6
Fodder at Farm B	1.9
Fodder at Farm C	3.7
Vegetables from Garden, Farm C	2.5
Vegetables from Garden D	1.7
Milk (Farms A and B)	1.1
Calf meat (Farms A and B)	1.0
Chicken and eggs (Farm C)	1.8

Table 12. Mean predicted-to-observed ratios for endpoints of the Pine Tree Scenario at three locations. Concentrations in pine needles and wood were calculated using observed air moisture concentrations at Location 1.

	Location 1	Location 2	Location 3
Rain	0.60	0.47	0.52
Needle HTO	1.6	0.73	0.67
Needle OBT	0.69	0.97	NA
Tree rings	NA	1.7	NA

APPENDIX - All Parameters and Distributions in DCART

Table A1. Input parameters for the precipitation pathway; specific input is required for specific sources and receptors. Generic values are provided as rough guidance. (GM = geometric mean; GSD = geometric standard deviation)

Parameter	Units	Reference	Recommended value	Type of distribution	Generic values
Release rate	Bq s^{-1}	Facility records	Year-dependent	normal	NA
Distance source to receptor	m	GPS measurements	Receptor-specific	NA	NA
Fraction of year that it rains		15 minute LLNL met data (1997 – 2003[1])	Annual, if known	normal	0.018 ± 0.0060
Washout coefficient	s^{-1}	Belot (1998)	For specific source to receptor	lognormal (GM ± GSD)	$1.6\ 10^{-4}$ ± 2.0
Average rainfall rate when wind towards receptor from specific source[2]	mm h^{-1}	Mean of 1997 – 2003[1] data or year specific	For specific source to receptor	lognormal	2.1 ± 0.14
Wind speed while raining at approximate height of release	m s^{-1}	Mean of 1997 – 2003[1] data or year specific	For specific source to receptor	normal	2.8 ± 0.37 (10 m) 3.9 ± 0.41 (40 m)
Fraction of time wind into sector when raining		Mean of 1997 – 2003[1] data or year specific	For specific source to receptor	normal	See Table A9
Precipitation	m y^{-1}	LLNL annual records	Known annual	lognormal	0.34 ± 0.12 for 1952 - 2003[1]
Δθ	radians	LLNL wind rose	0.393		

[1] Because the precipitation model is not being used for the LLNL TDR, data preparation was discontinued after 2003.
[2] This parameter is not used directly in DCART; however, it is used to estimate the washout coefficient.

Table A2. Controlling parameters: air concentrations

Parameter	Units	Reference	Recommended value	Type of distribution	Range of values
HT or HTO concentration in air	Bq m^{-3}	Dispersion model (e.g., CAP88-PC)	Year-dependent	normal	Year specific
HTO concentration in air	Bq L^{-1} or Bq m^{-3}	Annual average observed air tritium sampling locations	Year-dependent	normal	Year specific

Table A3. Input parameters for soil pathways (GM = geometric mean; GSD = geometric standard deviation)

Parameter	Units	Reference	Recommended value	Type of distribution	Range of values
Dry deposition velocity, HTO	m s^{-1}	Tritium Data Base[a]	5.3 10^{-3}	lognormal (GM ± GSD)	5.3 10^{-3} ± 3.8
Dry deposition velocity, HT	m s^{-1}	Tritium Data Base[a]	2. 6 10^{-4}	lognormal (GM ± GSD)	2.6 10^{-4} ± 3.5
Fraction HTO retained by soil	Bq L^{-1} soil HTO / Bq L^{-1} air moisture	IAEA 2003	0.3 x air moisture	triangular	0.1 – 0.3 – 0.5
Fraction HTO in soil from HT release	Bq L^{-1} HTO / Bq m^{-3} HT in air	Davis and Bickel 2000	6.0 x HT in air	lognormal (GM ± GSD)	6.0 ± 1.5

[a] The Tritium Data Base contains experimental values of deposition velocities that have been published in the open literature

Table A4. Input parameters for plant pathways (GM = geometric mean; GSD = geometric standard deviation)

Parameter	Units	Reference	Recommended value	Type of distribution	Range of values
Absolute humidity	kg m^{-3}	Silica gel data; 1974 – 2005	Year-specific or 0.0078	normal	0.0078 ± 0.00040
Relative humidity		1999, 2001 - 2005 met data	Year-specific or 0.69	normal	0.69 ± 0.07
HTO concentration in plant water from HT	Bq HTO L^{-1} / Bq HT m^{-3}	Davis and Bickel 2000	6.0	lognormal (GM ± GSD)	6.0 ± 1.5
HTO/H_2O ratio of vapor pressure	γ	Raney and Vaadia 1965	0.909		
Dry mass:	kg dry weight / kg fresh weight	Geigy 1981			
Leafy vegetables			0.094	Uniform	0.03 – 0.15
Fruit, Fruit vegetables			0.136	extreme value	m = 0.11; scl = 0.05
Root crops			0.15	lognormal (GM ± GSD)	0.14 ± 1.56
Grain			0.882	normal	0.88 ± 0.02
Pasture –fresh			0.253	uniform	0.18 – 0.32
Hay			0.902	extreme value	md = 0.89; scl = 0.01
Tree Rings			1.0		
Water equivalent:	L kg^{-1}	Geigy 1981			
Leafy vegetables			0.6	lognormal (GM ± GSD)	0.600 ± 1.03
Fruit, Fruit vegs.		Calculated from fresh	0.581	lognormal (GM ± GSD)	0.581 ± 1.02
Root crops		weights, dry weights, and	0.575	logistic	m = 0.575; s = 0.003
Grain		hydrogen content;	0.577	uniform	0.566 – 0.581
Pasture			0.590	uniform	0.586 – 0.594
Hay			0.583	uniform	0.580 – 0.587
Tree rings		Measured, CRL	0.57		
Isotopic discrimination		Kim & Baumgärnter 1994; Kalin et al 1995	0.7 (plants) 0.4 (tree rings)	extreme value triangular	md= 0.67; scl = 0.14 0.2 - 0.4 - 0.7
Fraction of tritium from air moisture			0.6 (fruit and grain) 0.05 (root crops)	triangular triangular	0.5 – 0.6 –0.7 0.0 –0.05 –0.20

Table A5. Input parameters for animal pathways. (Correlations: -0.9 pasture with hay; -0.9 pasture with grain; 0.90 and 0.95 grain intake of pigs and poultry, respectively, with water intake) (GM = geometric mean; GSD = geometric standard deviation)

Parameter	Units	Reference	Recommended value	Type of distribution	Range of values
Diet, pasture:	kg fresh weight d^{-1}	NAS 1996; 2001			
Milk cow			25.0	normal	25 ± 5.0
Beef cow			13.0	normal	13 ± 2.6
Diet, hay:	kg fresh weight d^{-1}	NAS 1996; 2001			
Milk cow			11.6	normal	11.6 ± 2.32
Beef cow			6.2	normal	6.2 ± 0.88
Diet, grain:	kg fresh weight d^{-1}	NAS 1994; 1996; 1998; 2001			
Milk cow			3.0	normal	3.0 ± 1.0
Beef cow			1.5	normal	1.5 ± 0.5
Pigs			3.4	normal	3. 34 ± 0.34
Chicken			0.18	normal	0.18 ± 0.021
Laying hen			0.11	normal	0.11 ± 0.017
Concentration of animal drinking water: fraction of air		Empirical data from LLNL pool, 1990 - 2000	0.165	lognormal (GM ± GSD)	0.16 ± 1.5
Water intake:	L d^{-1}	NAS 1994; 1996; 1998; 2001			
Milk cow			103.	normal	103 ± 1.82
Beef cow			44.	normal	44.3 ± 1.9
Pig			8.5	normal	7.5 ± 1.1
Chicken			0.36	normal	0.35 ± 0.042
Laying hen			0.22	normal	0.22 ± 0.034

Table A5 continued

Parameter	Units	Reference	Recommended value	Type of distribution	Range of values
Inhalation rate:	$m^3\ d^{-1}$	Assorted values, mostly from models			
Milk cow			158	truncated normal	144 ± 67.0 (75 – 300)
Beef cow			127	truncated normal	127 ± 72.0 (60 – 300)
Pigs			43.0	truncated normal	43 ± 18 ((20 – 70)
Chicken			1.0	truncated normal	1.0 ± 0.60 (0.3 – 2.)
Laying hen			1.0	truncated normal	1.0 ± 0.60 (0.3 – 2.)
Water from inhalation	$kg\ d^{-1}$	Raskob 1994	1.5 x inhalation rate ($m^3\ d^{-1}$) x absolute humidity (kg/m^{-3})		
Dry mass of:	kg dry weight / kg fresh weight	Geigy 1981			
Milk			0.103	uniform	0.09 – 0.12
Beef			0.332	triangular	0.28 - 0.32 - 0.44
Pork			0.5	custom	70% 0.28-0.48; 30% 0.8 – 1
Chicken			0.33	uniform	0.27 – 0.39
Egg			0.26	uniform	0.25 – 0.27
Water equivalent:	$L\ kg^{-1}$	Geigy1981 Calculated from fresh weights, dry weights, and hydrogen content;			
Milk			0.711	triangular	0.592 – 0.711 – 0.746
Beef			0.795	triangular	0.72 – 0.8 – 0.90
Pork			0.904	uniform	0.786 – 1.0
Chicken			0.796	uniform	0.73 – 0.85
Eggs			0.835	uniform	0.834 – 0.836
Uncertainty on transfer to animal OBT			1.0	truncated normal	1.0 ± 0.4; (0.8 - ∞)

Table A6. Inhalation (Myers et al. 2000) and ingestion intake rates (EPA 1999, 2002) for adult, child (age 10) and infant (6 m - 1 y); correlations: -0.75 leafy vegetables and fruit/fruit vegetables; -0.8 root crops and grain; -0.8 beef and pork. All distributions are lognormal and consist of a geometric mean and a geometric standard deviation.

Parameter	Units		Recommended value	Range of values
Inhalation rate	$m^3\ y^{-1}$	Adult	4860.	4604 ± 1.37
		Child	4930.	4689 ± 1.37
		Infant	1640.	1346 ± 1.87
HTO concentration in air from HT	Bq HTO L^{-1} / Bq HT m^{-3}	Davis and Bickel 2000	4.0 for inhalation	4.0 ± 1.5
Drinking water	$L\ y^{-1}$	Adult	552.	510 ± 1.55
		Child	356.	318 ± 1.66
		Infant	120.	107± 2.03
Leafy vegetables	$kg\ y^{-1}$	Adult	15.9	11.0 ± 2.37
		Child	9.53	6.51 ± 2.40
		Infant	1.17	0.43 ± 4.12
Fruit, fruit vegetables	$kg\ y^{-1}$	Adult	117.	99.8 ± 2.2
		Child	107.	92.4 ± 1.79
		Infant	64.8	46.4 ± 2.23
Root crops	$kg\ y^{-1}$	Adult	27.6	20.5 ± 2.32
		Child	24.5	16.3 ± 2.62
		Infant	6.17	0.61 ± 10.1
Grain	$kg\ y^{-1}$	Adult	80.0	69.3 ± 1.78
		Child	85.1	75.5 ± 1.64
		Infant	23.4	20.1 ± 2.04
Milk (and products)	$kg\ y^{-1}$	Adult	91.2	69.1 ± 2.22
		Child	177.	157 ± 1.71
		Infant	208.	152 ± 2.17
Beef	$kg\ y^{-1}$	Adult	17.7	14.1 ± 2.12
		Child	14.5	12.2 ± 2.02
		Infant	3.13	1.62 ± 3.06

Table A6 continued

Parameter	Units		Recommended value	Range of values
Pork	kg y^{-1}	Adult	5.68	1.83 ± 4.79
		Child	4.66	1.54 ± 4.58
		Infant	0.967	0.259 ± 6.29
Chicken	kg y^{-1}	Adult	12.9	7.98 ± 2.74
		Child	11.5	7.37 ± 2.70
		Infant	2.71	0.591 ± 5.92
Eggs	kg y^{-1}	Adult	6.24	4.90 ± 2.88
		Child	5.49	4.52 ± 3.12
		Infant	2.63	2.89 ± 2.33

Table A7. Dose coefficients (ICRP (1995, 1996); uncertainty from Harrison et al. 2002). All distributions are lognormal and consist of a geometric mean and a geometric standard deviation.

Parameter	Units		Recommended value	Range of values
HT inhalation	Sv Bq^{-1}	Adult	$1.8\ 10^{-15}$	$3.82\ 10^{-15}$ ± 1.23
		Child	$2.3\ 10^{-15}$	$4.43\ 10^{-15}$ ± 1.32
		Infant	$4.8\ 10^{-15}$	$9.22\ 10^{-15}$ ± 1.33
HTO inhalation/ingestion	Sv Bq^{-1}	Adult	$1.8\ 10^{-11}$	$3.82\ 10^{-11}$ ± 1.23
		Child	$2.3\ 10^{-11}$	$4.43\ 10^{-11}$ ± 1.32
		Infant	$4.8\ 10^{-11}$	$9.22\ 10^{-11}$ ± 1.33
OBT ingestion	Sv Bq^{-1}	Adult	$4.2\ 10^{-11}$	$8.34\ 10^{-11}$ ± 1.34
		Child	$5.7\ 10^{-11}$	$1.02\ 10^{-10}$ ± 1.47
		Infant	$1.2\ 10^{-10}$	$2.22\ 10^{-10}$ ± 1.49

Table A8. Parameters used in the sub-model for dose from swimming. (GM= geometric mean, GSD = geometric standard deviation)

Parameter	Units	Reference		Recommended value	Type of distribution	Range of values
Ratio air moisture POOL / VIS air tritium samplers		Empirical from LLNL data		2.1535	normal	2.1535 ± 0.215
Ratio pool water / air moisture at POOL		Empirical from LLNL data		0.165	lognormal (GM ± GSD)	0.16 ± 1.5
Skin area	m^2	EPA 1999	Adult	1.82	normal	1.82 ± 0.25
			Child	1.12		1.12 ± 0.12
			Infant	0.425		0.425 ± 0.045
Humidity at skin/water temperature	mg L^{-1}	Weight of saturated water vapor		0.02724	triangular	0.0224 – 0.02724 – 0.02878
Intake rate for skin	L min^{-1} m^{-2}	Osborne 1968		5.1	normal	5.1 ± 1.5
Minutes spent swimming per year		EPA 1999, 2002	Adult	150	triangular	0 – 150 – 9000
			Child	720		0 – 720 – 2500
			Infant	510		0 – 510 - 2500

Table A9. Fraction of time the wind blows towards all sectors when raining (average of data 1997 – 2003). Uncertainty is shown for directions from sources towards the Visitors Center.

Direction towards	10 m winds	40 m winds
N	0.079	0.096
NNE	0.14 ± 0.027	0.15 ± 0.024
NE	0.17	0.16
ENE	0.11 ± 0.031	0.12 ± 0.039
E	0.074	0.068
ESE	0.036 ± 0.012	0.030 ± 0.012
SE	0.019	0.015
SSE	0.019	0.020
S	0.036	0.033
SSW	0.036	0.040
SW	0.061	0.048
WSW	0.066	0.047
W	0.035	0.057
WNW	0.030	0.037
NW	0.026	0.028
NNW	0.059	0.055

REFERENCES

ATSDR. Health Consultation, Tritium Releases and Potential Offsite Exposures. Agency for Toxic Substances and Disease Registry. U.S. Department of Health and Human Services, Atlanta, GA: 99 pp; March 2002.

ATSDR. Public Health Assessment, Community Exposures to the 1965 and 1970 Accidental Tritium Releases. Agency for Toxic Substances and Disease Registry. U.S. Department of Health and Human Services, Atlanta, GA: 142 pp; July 11, 2003.

ATSDR. Public Health Assessment, Lawrence Livermore National Laboratory, Main Site (U.S. DOE). Agency for Toxic Substances and Disease Registry. U.S. Department of Health and Human Services, Atlanta, GA: 108 pp; June 29, 2004.

Beller, H.; Eaton, G.; Ekwurzel, B; Esser, B.K.; Hu, Q.; Hudson, G.B.; Leif, R.; McNab, W.W.; Moody-Bartel, C.; Moore, K.; Moran; J.E. California GAMA Program; Sources and transport of nitrate in groundwater in the Livermore Valley Basin, California. Lawrence Livermore National Laboratory, Livermore, CA. UCRL-TR-217189, November 2005; 30 pp.

Belot, Y. Predicting the washout of tritiated water from atmospheric plumes. Presented at the Workshop of the IAEA Task Group on Tritium Safety and Environmental Effects, held at AECL, Chalk River, Canada, on May 11-12, 1998.

Brewer, B. Irrigation expenses threaten vineyards. Contra Costa Times, January 21, 2001.

Brown, R.M.; Ogram, G.L.; Spencer, F.S. Field studies of HT behaviour in the environment: 1: Dispersion and oxidation in the atmosphere. Fusion Technol. 14: 1165-1169; 1988.

Canadian Standards Association (CSA). Guidelines for Calculating Derived Release Limits for Radioactive Material in Airborne and Liquid Effluents for Normal Operations of Nuclear Facilities. CAN/CSA-N288.1-M87. 69 pp; 1987.

Davis, P.A. Unpublished data. 2003.

Davis, P.A.; Bickel, G.A. Environmental HTO / HT ratios arising from a chronic atmospheric HT release. *In*: Proceedings of the International Workshop on the Environmental Behavior of Tritium, Osaka, Japan; 2000.

Davis, P.A., Galeriu, D.C.; Spencer, F.S.; Amiro, B.D. Evolution of HTO concentrations in soil, vegetation and air during an experimental chronic HT release. Fusion Technology 28: 833-839l; 1995.

Davis, P.A.; Kotzer, T.G.; Workman, W.J.G. Environmental Tritium Concentrations due to Continuous Atmospheric Sources. Fusion Science and Technology 41:453-457; 2002.

Diabate, S. and S. Strack. Doses due to tritium releases by NET-data base and relevant parameters on biological tritium behaviour. Prepared for NET under Contract No. NET 89-195, Kernforschungszentrum Karlsruhe GmbH, Karlsruhe; 1990.

Dinner, P.J.; Gorman, D. J.; Spencer, F.S. Tritium dynamics in vegetables: experimental results. In: Tritium technology in fission, fusion, and isotopic applications, Dayton, Ohio. American Nuclear Society. 1980.

Eckerman, K.F.; Leggett, R.W.; Nelson, C.B.; Puskin, J.S.; Richardson, A.C.B. Cancer Risk Coefficients for Environmental Exposure to Radionuclides. Federal Guidance Report No. 13. Office of Radiation and Indoor Air, United States Environmental Protection Agency, Washington, D.C. EPA-402-R-99-001. 335 pp. 1999.

Ely, L.O. and Guthrie, L.D. University of Georgia College of Agricultural & Environmental Sciences. Cooperative Extension Service. Feeding the Dairy Herd. (http://www.ces.uga.edu/pubcd/b816-w.htm); taken from web December 2001.

Evans, A.G. New dose estimates from chronic tritium exposures. Health Phys. 16: 57-63; 1969.

Fellows, R. J., D.A. Cataldo, M.W. Ligotke, and B.A. Napier. Transfer of atmospheric tritiated water to foliage and fruit of crops. A research report for Westinghouse Hanford Company. Pacific Northwest Laboratory, Richland, WA. PNL-7521, UC-602; 1990.

Galeriu, D.; N.M.J. Crout; A. Melintescu; N.A. Beresford; S-R. Peterson; M. Van Hees. A Metabolic derivation of tritium transfer in animal products. Radiation and Environmental Biophysics Vol 40: 325-334; 2001.

Garland, J.A. and M. Ameen. Incorporation of tritium in grain plants. Health Physics 36: 35-38; 1979.

Geigy. Geigy Scientific Tables, Vol 1, 8th Edition. Units of measurement, body fluids, composition of the body, nutrition. Basel, Switzerland: Ciba-Geigy Ltd; 1981.

Gifford, F.A., Jr.; Hilsmeier, W.F. Graphs for estimating atmospheric dispersion. United States Weather Bureau Report, ORO-545; 1961.

Harrison, J.D.; Khursheed, A.; Lambert, B.E. Uncertainties in dose predictions for intakes of tritiated water and organically bound forms of tritium by members of the public. Radiation Protection Dosimetry 98(3):299-311; 2002.

Hilsmeier, W.F. and F.A. Gifford, Graphs for Estimating Atmospheric Dispersion. U.S. Weather Bureau Report ORO-545, Oak Ridge, TN. 1962.

International Atomic Energy Agency. International programme on biosphere modelling and assessment methods (BIOMASS). Vienna: IAEA; 1996.

International Atomic Energy Agency. Modelling the environmental transport of tritium in the vicinity of long-term atmospheric and sub-surface sources sources. Report of the Tritium Working Group of the Biosphere Modelling and Assessment (BIOMASS) Programme, Theme 3. International Atomic Energy Agency, Vienna: IAEA-BIOMASS-3; March 2003.

International Commission on Radiological Protection. Age dependent doses to members of the public from intake of radionuclides, Part 4, Inhalation Dose Coefficients. Oxford: Pergamon Press; ICRP Publication 71; Ann. ICRP 25(3&4); 1995.

International Committee on Radiation Protection. Age-dependent doses to members of the public from intake of radionuclides: Part 5. Compilation of Ingestion and Inhalation Dose Coefficients. .Annals of the ICRP Vol. 26 No. 1. ICRP # 72 ISSN 0146-6453, Pergamon Press, Oxford; 1996.

Kalin, R.M., C.E. Murphy Jr., and G. Hall. 1995. Reconstruction of tritium release history from contaminated groundwater using tree rings. Fusion Technology Vol. 28: 883-887.

Kim, M.A.; Baumgärtner, F. Equilibrium and non-equilibrium partition of tritium between organics and tissue water of different biological systems. Appl. Radiat. Isot. Vol. 45(3): 353-360; 1994.

McFarlane, J.C. Tritium fractionation in plants. Environ. Exp. Botany 16: 9-14; 1976.

McFarlane, J.C.; Rogers, R.D.; Bradley, D.V., Jr. Environmental tritium oxidation in surface soil. Environmental Science and Technology 12(5): 590-593; 1978.

Moghissi, A.A.; Bretthauer, E.W.; Patzer, R.G. Biological concentration of ^{3}H. Health Phys. 53(4); 385-388; 1987.

Moran, Jean E. Personal communication, December 2005.

Moran, J.E.; Hudson, G. B.; Eaton, G.F.; Leif, R. A Contamination Vulnerability Assessment for the Livermore-Amador and Niles Cone Groundwater Basins. Report to the California State Water Resources Control Board. Lawrence Livermore National Laboratory, Livermore, CA. UCRL-AR-148831, July 2002; 36 pp.

Murphy, C.E. Jr. The relationship between tritiated water activities in air, vegetation and soil under steady-state conditions. Health Physics 47: 635-639; 1984.

Murphy, C.E. Jr. Modelling tritium transport in the environment. Rad. Prot. Dosimetry 16: 51-58; 1986.

Myers, L.; Riggs, M.; Lashley, J.; Whitmore,R.; Moya, J. Options for development of parametric probability distributions for exposure factors. U.S. Environmental Protection Agency, Washington, D.C.;EPA/600/R-00/058; July 2000.

Napier, B.A.; Peloquin, R.A.; Strenge, D.L.; Ramsdell, J.V. GENII – the Hanford environmental radiation dosimetry software system. Richland, WA: Pacific Northwest Laboratory, PNL-6584 Vol. UC-60; 1988.

National Academy of Sciences. Nutrient Requirements of Poultry. Ninth Revised Edition. National Academy Press, Washington D.C. 1994.

National Academy of Sciences. Nutrient Requirements of Beef Cattle. Seventh Revised Edition. National Academy Press, Washington D.C. 1996.

National Academy of Sciences. Nutrient Requirements of Swine. Tenth Revised Edition. National Academy Press, Washington D.C. 1998.

National Academy of Sciences. Nutrient Requirements of Dairy Cattle. Seventh Revised Edition. National Academy Press, Washington D.C. 2001.

Osborne, R.V. Intake of tritium when skin is splashed with tritiated water. Health Physics Vol. 15: 155-156; 1968.

Osborne, R.V.; Hamby, D.; Johnson, J.; Kahn, B.; Straume, T. Environmental tritium evaluations at SRS and LLNL with emphasis on monitoring and dosimetery of organically-bound-tritium. Final report of an expert panel convened by the Agency for Toxic Substances and Disease Registry. U.S. Departmen of Health and Human Services,Atlanta, GA. July 2001.

Parks, B.S. User's Guide for CAP88-PC, Version 1.0. U.S. Environmental Protection Agency, Office of Radiation Programs, Las Vegas, NV (EPA 402-B-92-001); 1992.

Parks, B.S. CAP88-PC Version 2.0 User's Guide. Germantown, MD. U.S. Department of Energy ER-8/GTN; 1997.

Paulus, L.R.; Walker, D.W.; Thompson, K.C. A comparison of dessicant materials used for monitoring atmospheric tritium concentrations in a semi-arid climate. Health Physics 85(3):348-356; 2003.

Peterson, K.R.; Crawford, T.W.; Lawson, L.A. CPS: A continuous-point-source computer code for plume dispersion and deposition calculations. Lawrence Livermore National Laboratory, Livermore, CA; UCRL-52049; 1976.

Peterson, S-R. Testing CAP88-PC's Predicted Air Concentrations Against LLNL Historical Air Tritium Monitoring Data, 1986 – 2001. UCRL-ID-155505; September 2003.

Peterson, S-R. Test of CAP88-PC's Predicted Concentrations of Tritium in Air at Lawrence Livermore National Laboratory. UCRL-JRNL-201833. *Health Physics* 87(6): 583-595; 2004.

Peterson, S-R. Modeling the Pickering scenario. Submission to the Tritium Working Group of the International Atomic Energy Agency's Coordinated Research Program, "Environmental Modeling for Radiation Safety" (EMRAS). UCRL-MI-214957. August 4, 2005.

Peterson, S-R. Modeling the Pickering scenario. Submission to the Tritium Working Group of the International Atomic Energy Agency's Coordinated Research Program, "Environmental Modeling for Radiation Safety" (EMRAS). UCRL-MI-218056. January 2006.

Peterson S-R.; P.E. Althouse; N.A. Bertoldo; R.A. Brown; C.G. Campbell; Clark, L.M.; Gallegos, G.M.; Grayson, A.R. Harrach, R.J.; Jones, H.E.; MacQueen, D.H.; Mathews, S.; Nisbet, B.A.; Noyes, C.D.; Paterson, L.; Revelli, M.A.; Ruepell, D..; Sanchez, L,; Taffet, M.J.; Tate, P.J.; Wilson, K. *Environmental Report for 2004*, Lawrence Livermore National Laboratory, Livermore, CA (UCRL-50027-04), Livermore CA; 2005.

Peterson, S-R. and P.A. Davis. Tritium Doses from Chronic Atmospheric Releases: A New Approach Proposed for Regulatory Compliance. UCRL-JC-141535. Health Physics 82(2):213-225; 2002.

Peterson, S-R.; Hoffman. F.O.; Köhler, H. Summary of the BIOMOVS A4 Scenario: Testing Models of the Air-Pasture-Cow Milk Pathway Using Chernobyl Fallout Data. Health physics 71(2):149-159; 1996.

Raney, R., and Vaadia, Y. Movement and Distribution of THO in Tissue Water and Vapor Transpired by Shoots of Helianthus and Nicotiana. Plant Physiology 40, 383-388 (1965).

Raskob, W. Description of NORMTRI: A computer program for assessing the off-site consequences from air-borne releases of tritium during normal operation of nuclear facilities. Report KfK-5364, Kernforschungszentrum Karlsruhe, Germany. 1994.

University of California, Davis. Dairy Care Practices. Part of the Animal Care Series, Dairy Workgroup, UC Cooperative Extension; second edition, 1998. (http://www.vetmed.ucdavis.edu/vetex/INF-DA_CarePrax.html#Dairy)

U.S. Environmental Protection Agency. User's Guide for AIRDOS-PC, Version 3.0. Washington, D.C. EPA 520/6-89-035; 1989.

U.S. Environmental Protection Agency. Exposure Factors Handbook (EFH). National Center for Environmental Assessment. Office of Research and Development, Washington, D.C. EPA/600/C-99/001; February 1999.

U.S. Environmental Protection Agency. Child-Specific Exposure Factors Handbook. National Center for Environmental Assessment. Office of Research and Development, Washington, D.C. EPA-600-P-00/002B; Interim report, September 2002.