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Reviewing MACCS Capabilities for Assessing Tritium Releases to the Environment

Kyle A. Clavier
Daniel J. Clayton

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico
87185 and Livermore,
California 94550

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ABSTRACT

Tritium has a unique physical and chemical behavior which causes it to be highly mobile in the environment. As it behaves similarly to hydrogen in the environment, it may also be readily incorporated into the water cycle and other biological processes. These factors and other environmental transformations may also cause the oxidation of an elemental tritium release, resulting in a multiple order of magnitude increase in dose coefficient and radiotoxicity. While source term development and understanding for advanced reactors are still underway, tritium may be a radionuclide of interest. It is thus important to understand how tritium moves through the environment and how the MACCS accident consequence code handles acute tritium releases in an accident scenario. Additionally, existing tritium models may have functionalities that could inform updates to MACCS to handle tritium. In this report tritium transport is reviewed and existing tritium models are summarized in view of potential updates to MACCS.

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ACRONYMS AND DEFINITIONS

Abbreviation	Definition
EPA	United States Environmental Protection Agency
ETMOD	Environmental Tritium Model
GENII	GENeration II dose and risk assessment model
HTO	Tritiated water or oxidized tritium
LWR	Light water reactor
NRC	US Nuclear Regulatory Commission
OBT	Organically-bound tritium
RSAC	Radiological Safety Analysis Computer program
TRILOCOMO	Local compartment model for dispersion of HT and HTO

1. MOTIVATION AND SCOPE

Some radionuclides of potential concern for accident scenarios may have particularly unique physical and chemical behavior. One of the most unique radionuclides that may become relevant for advanced reactor consequence analysis is tritium, as it is highly mobile and can enter biological systems as part of water and organic molecules. As identified in the Nuclear Regulatory Commission (NRC) Non-Light Water Reactor (Non-LWR) Vision and Strategy Volume 3 report [1], additional research is necessary to identify if MACCS model or dosimetry updates are necessary to account for tritium. Andrews et al. (2021) [2] identified a potential need for tritium-specific chemistry models in MACCS. In this report, tritium was noted as a radionuclide of potential concern for:

- High-Temperature Gas Reactors (accumulated in the primary system coolant),
- Fluoride-Salt Cooled High-Temperature Reactors (the most mobile activation product, an order of magnitude larger production than LWR),
- Molten Salt Reactors (especially those with a lithium salt),
- Fast Spectrum Metallic Fuel Systems (ternary fission in fuel, diffusion through cladding)

Some of these advanced reactor types may produce larger amounts of tritium than a traditional LWR. Should the total reactor inventory and release fractions of tritium be high enough to result in significant consequences from tritium releases, it may be warranted for to update MACCS capabilities to model the fate and transport of tritium more effectively in an accident scenario. Accordingly, this document provides a review of aspects of the environmental transport of tritium and various existing tritium models. Furthermore, recommendations for how best to accommodate tritium in MACCS are provided. The primary goal of this study is not to provide detailed equations and calculations for modeling tritium, but rather focuses on MACCS core capabilities to model tritium and provides an overview of the tritium modeling approaches used by other codes to inform the potential application of those approaches to MACCS. Future research should determine if tritium is present in large enough quantities in advanced reactor source terms to warrant its inclusion in analyses of severe accident risk.

MACCS has been used in previous studies to model tritium releases. In an intercomparison study against 14 other models [3], MACCS2 was included in a recommendation list for modeling tritium source terms (model set 'F') but was rated as less preferable than other available models such as UFOTRI, COSYMA, AXAIRQ and HOTSPOT. Authors noted that models other than MACCS can differentiate between different tritium forms in transport and dose, and additionally offer environmental compartment models for transfer in food, soil to air, and plant media. Accordingly, in a model selection guide based on their findings, authors recommend that for all levels of source term energetics, model set F (which includes MACCS2) is suitable for analyzing plume passage, but authors should consider alternate models for environmental compartment analysis [3]. In a probabilistic risk assessment for a production reactor [4], a loss-of-moderator pumping accident and a loss of coolant accident were modeled using MACCS Version 1.5. Tritium related doses were noted as small relative to other radionuclides. In this scenario, a full inventory release of HTO vapor was assumed with transport properties of the noble gas fission product group. This analysis included water ingestion of tritium with the MACCS 1.5 direct deposition and run off model and neglected food ingestion because no MACCS tritium ingestion model was available. Results indicated that tritium-related doses comprised only 7% of the dose of the other fission products in the source term [4]. Velarde and Perlado (2001) used MACCS2 to model tritium releases from three types of inertial fusion reactors but provide limited details on inputs or special treatment for tritium [5]. The hazard

and accident analysis for a planned Tritium Finishing Facility at the Savannah River Site used local meteorological data and MACCS2 to model tritium releases for a set of stack heights, with the intent to mitigate dose consequences to workers in the surrounding area [6]. Authors from Lawrence Livermore, Savannah River, and Los Alamos National Laboratories used MACCS to calculate early doses from inertial fusion energy assuming a ground level release using typical values for atmospheric dispersion conditions. This report showed doses below the 1 rem evacuation limit for all assessed worst case scenarios [7]. In a report outlining the technical basis for selection of a tritium dose model, MACCS was noted as defensible and reasonable for modeling acute releases of tritium from the Savannah River facility. When capabilities for modeling tritium releases were compared with AXAIRQ and UFOTRI tritium models, MACCS2 was noted as having sufficient analytical capability for scoping and preliminary hazard assessment purposes, as well as accident analysis and functional classification purposes for offsite calculations. Modelers used both a prescriptive meteorology and site-specific meteorology release of 1,000 Ci of tritium oxide. In a fire release accident sequence, MACCS provided the most conservative dose estimations [8].

2. TRITIUM ACCOUNTING

The NRC Non-LWR Vision and Strategy Volume 3 highlights a dosimetry and health effects phenomenological area focused on tritium modeling and the need to develop MACCS model and dosimetry updates to better account for the unique behavior of tritium (Task CA4). Consider that tritium may be sufficiently unique in its chemical behavior that, from a consequence analysis perspective, it may be pertinent to treat tritium as a separate chemical class. Tritium is generally grouped into the noble gas chemical group (group 1) in MACCS, however, it may be necessary to expand the available chemical groups to accommodate different chemical forms of tritium, particularly tritiated water (HTO)

Tritium can be released to the environment in different forms, and dose conversion factors might also need to be modified to account for the unique ways that HTO interacts with the human body via inhalation, ingestion, and skin absorption [1]. Environmental tritium accounting will be discussed here, along with a review of existing tritium transport model through various environmental media.

2.1. Tritium Behavior in the Environment

Tritium is a radioactive isotope of hydrogen with a half-life of approximately 12.3 years. This isotope is produced naturally in very low concentrations via atmospheric interaction with high energy cosmic rays. Tritium may also be produced anthropogenically through release during nuclear weapons tests and the operation of nuclear energy facilities. While the natural production of tritium is of low radiological risk, anthropogenic emissions of tritium from events such as nuclear reactor accidents may require attention.

Tritium behaves similarly to hydrogen in the environment and biological processes [9]. Combined with the fact that tritium is a week beta particle emitter and thus a weak external exposure hazard, the radiological hazard with tritium is primarily through human ingestion of tritiated organic molecules. This behavior of tritium makes it important to quantify physical, chemical, and biological transport processes of tritium in the event of an accident. Of particular importance to the radiological risk posed by tritium is the chemical form in which the tritium is ingested. Generally, inhaling gaseous tritium poses relatively limited radiological risk, as absorption is low (< 0.005%) and a significant fraction is exhaled from the body. Dermal contact risk associated with gaseous tritium exposure is also limited [10]. It is estimated that approximately 1 kg of tritium gas release would be required to pose a meaningful radiological risk, a quantity likely unachievable by most current reactor designs. The fraction of a given tritium release that is composed of tritium gas is understood to not impact radiological risk calculations [11]. A database of atmospheric tritium releases [12] cataloged nine releases at the Savannah River Site from a 60-meter tritium facility stack. The largest calculated individual offsite dose was 1.6 mrem, with a 46 person-rem population dose. In separate scenarios, activities of up to 479,000 Ci of tritiated hydrogen gas and 43,800 Ci of tritiated water were released [12].

Ingestion and dermal contact with HTO poses a comparatively larger risk. According to US Department of Energy guidance on safe handling practices for tritium, biological uptake of HTO is extremely efficient (up to 99% incorporated into blood stream), a process that takes place in a matter of hours. Depending on air temperature, meaningful concentrations of HTO can also be absorbed through the skin at a rate approximately half of that of inhalation (assuming an average breathing rate) [10]. As a consequence of its physical and chemical similarity to ordinary water, the biological metabolism of tritium mirrors the throughput of water through the body. This results in an effective biological half-life of tritium in the body of approximately 9-10 days in moderate temperature (decreasing with average air temperature). Tritium can be retained in organic molecules

of varying half-lives up to 550 days, but the radiological impact of these species is limited and often not accounted for in tritium exposure models.

Tritium can also bind to carbon through photosynthetic processes and create non-exchangeable organically bound tritium (OBT) molecules in plants. In this form, tritium has dose coefficients nearly 3 times that of tritiated water. Thus, modeling the conversion rates of tritium gas and tritiated water into OBT in the event of a nuclear accident can be of critical importance and will be discussed in detail in the proceeding sections. Table 2-1 displays dose coefficients for various exposure pathways and chemical forms of tritium [13] and provides further evidence to suggest that the most important exposure pathways from tritium in an accident scenario may not be from the inhalation or dermal contact with airborne tritium gas, but from ingestion or inhalation of washed out, deposited, or biologically synthesized OBT and HTO molecules. The MACCS dose coefficient input file for MACCS currently assumes tritium is entirely in the form of tritiated water. A cloudshine dose coefficient of zero is assumed.

Some consequence analysts recognize that immersion in an HTO cloud will result in some amount of HTO absorption through the skin, as well as by inhalation. In the past, this phenomenon has been addressed by multiplying the inhalation dose coefficient for tritium by a correction factor greater than 1. A 1999 study analyzing the environmental impacts associated with tritium production at five commercial light water reactors, analysts altered the dose conversion factors produced by the MACCS2 dose conversion factor preprocessor to include an increase of 50% to account for skin absorption [14]. This practice is consistent with the GENII tritium model which, like MACCS, assumes that tritium is in the form of tritiated water and that the "combined total rate of intake of tritium in air is assumed to be 150% of the inhalation intake rate alone" [15].

Table 2-1. Tritium dose coefficient overview [13]

Tritium Form	Dose Coefficient (Sv/Bq)
Organically Bound Tritium (Ingestion)	4.2 E-11
Tritiated Water (Ingestion)	1.8 E-11
Tritium Gas (Inhalation, Moderate Absorption)	1.8E-15
Organically Bound Tritium (Inhalation)	4.1E-11
Tritiated Water (Inhalation)	1.8E-11

There is evidence to suggest that even large scale HTO releases cause radiation doses well below standard limits for occupational exposure. A HTO spill at Braidwood Station nuclear power plant caused an annual dose 5,000 times lower than NRC guidance for exposure limits for pregnant women and 1,000 times lower than limits for the general public [16]. For reference, a 100 mrem annual dose is the more stringent of those two regulations, corresponding to a dose about 3 times lower than average annual radiation exposure from man-made sources [16]. Nonetheless, uncertainty surrounding large-scale accidental tritium releases from advanced reactor types motivates an investigation into how tritium modelling may be incorporated into existing accident consequence models, the details of which will be described herein. The main processes include atmospheric dispersion, dry deposition to soil, dry deposition to vegetation, re-emission from soil, re-emission from vegetation, root uptake from soil, loss from top soil to deeper soil, OBT formation and loss.

These will all be discussed along with the mechanism by which various existing tritium models handle these processes.

2.2. Atmospheric Transport

Existing literature suggests that atmospheric dispersion and plume movement for tritium would be mechanically similar to commonly modeled radionuclides [17]. While re-emission of oxidized tritium is expected from both vegetation and soil to the air, evapotranspired HTO is expected to be dispersed over the course of several hours and may thus be a negligible risk pathway. Tritiated gas has a low dry deposition velocity and is not transferred to plants or washed out by rain and has a low dose per unit intake by inhalation. HTO vapor washout follows empirically derived equations available in published literature [18], with the activity of deposition represented by the spatially integrated air concentrations washed out. Due to HTO chemical similarity to water, the washout deposition is proportional to the rainfall quantity.

Given the magnitude in difference between dose coefficients for gaseous and HTO, a key environmental transformation mechanism may be the rate at which gaseous tritium releases are oxidized in the environment. Photochemical oxidation in air during environmental transport is identified as a key mechanism by which tritium transformation happens. A 2009 study notes that the oxidation process of tritium in the atmosphere is relatively slow, with a half-life greater than 5 years. In fact, the oxidation process happens much faster at the air soil interface, yielding a tritium half-life of less than 1 hour [19]. Similar results were previously report in Burnham et al. (1988), who conducted a study on tritium release under controlled conditions and found the oxidation of tritium in the atmosphere to be minimal and that the dominant mechanism for oxidation is via soil [20]. Papagiannakopoulous and Easterly (1979) collated the conversion rate ($CiL^{-1}s^{-1}$) of gaseous tritium to oxidized tritium in various gas mixtures, either through oxidation or isotopic exchange, and dependent on the initial tritium gas concentration, the results of which are displayed in Table 2-2. Note the relatively slow rate constants.

Table 2-2. Tritium oxidation rates for various gas mixtures [21]

Mixture	Tritium Concentration Range (Ci/L)	Conversion Rate, d[HTO]/dt, ($CiL^{-1}s^{-1}$)
HT + H_2 +O_2	95-328	1.98 x 10^{-6} [HT]
HT+O_2 +Ar	0.09-90	1.7 x 10^{-8} $[HT]^{5/3}$
HT + O_2 +N_2	0.018-1	3.30 x 10^{-7} $[HT]^2$
HT + O_2 + H_2O	<1	1.20 x 10^{-6} $[HT]^2$
HT + Dry Air	0.015-0.8	1.7 x 10^{-7} $[HT]^2$
HT + H_2O + He, N_2, Ar, Kr	0.05-0.7	4.2 x 10^{-7} $[HT]^2$
HT + H_20 + Dry Air	6 x 10^{-4}-600	1.7 x 10^{-8} $[HT]^{5/3}$

2.3. Atmosphere to Soil Transfer

The air to soil transfer of tritium is largely governed by diffusion and largely depends on soil humidity. Diffusion in the vapor form is governed by concentration gradients [17], and tritium movement into soil water can generally be calculated as the mass of deposited tritium divided by the

volume of water it is being absorbed to. Short-term reemission from soil is largely negligible, with approximately half of the deposited amount reemitted to the air at the end of the release. The half-life of tritium in soil is much longer than the half-life of tritium in plants, so time integrated doses to soil exposure over time should not be ignored. Concentrations of tritium in soil water are generally lower than for those in air moisture. Once in the soil, bacterial oxidation begins to oxidize tritium, the rate at which depends on a number of factors including soil type, moisture, and temperature. Soil oxidation of molecular tritium is notably similar to the oxidation behavior of atmospheric hydrogen gas, suggesting the same reaction mechanism [22]. In one study investigating rates of oxidation for molecular tritium by surface soils, Ichimasa et al. (1988) found oxidation rate constants on the order of 0.011-0.196 min^{-1} in multiple sandy loam soil samples, with a 10-25% optimal water content for oxidation and decreasing with soil depth [23].

2.4. Atmosphere/Soil to Plant Transfer

Tritium gas has a low solubility in water, so it has minimal absorption by plants [24]. Tritium transfer to plants and crops is a highly complex process because tritium is expected to naturally take part in the water cycle and transfer of tritium is subjected to a great number of changing environmental conditions at any time, as well as the natural physiological processes of vegetation [9][17]. Tritium releases that are newly oxidized in the soil can then be utilized by plants in photosynthetic processes and become incorporated into the food chain.

The practical pathway for the air to plant transfer is an air to leaf exchange between vapor and free water in leaf stomate and cuticle, which depends on a variety of factors including leaf area index and stomatal resistance (a measure of vapor transportability into leaf pores). Consequently, transfer of HTO into plants depends on time-of-day related factors such as light, temperature, and relative air and soil humidity. Accordingly, timing of a nuclear release (night vs. day) may strongly influence the transfer of tritium through plants; the main dose contribution from plant ingestion likely occurs during the first day. The stomatal resistance is approximately 300 s/m during a sunny day (open stomata) and 3000 s/m during the night (closed stomata).

When considering the mathematical relationship between water exchange velocity and activity of leaf free water, and accounting for stomatal resistance and activity of leaf free water, the order of magnitude difference between stomatal resistance during day and night illustrates the significant influence of release timing for atmosphere to plant transfer. Generally, equilibrium between plant and soil activity is reached in approximately 2 or 3 days, at which point the plant free water concentration decreases with the same half-life as the soil concentration. Evapotranspiration reduces the soil water activity, and the daily rate of emission is proportional to the evapotranspiration rate. Tritium concentrations are approximately 10-100 times less in the transfer process from soil to plant. Accordingly, limiting the consumption of affected crops in the days immediately following will largely avoid this exposure pathway.

2.5. Production of Tritiated Organic Matter

HTO exposure following an acute release may come from the consumption of food with high concentrations of organically bound tritium. A function of its chemical similarity to water, HTO has a tendency to incorporate into plant organic matter during the photosynthesis process. Tritium bound to carbon undergoes limited decrease in concentration during the photosynthetic process and is often referred to as non-exchangeable OBT. Tritium bound to oxygen, nitrogen, or sulfur freely exchanges with hydrogen in free water in a matter of hours and is commonly referred to as exchangeable OBT. Soil half-lives are long, and thus the production of OBT may continue until the harvest and ingestion of plants and the incorporation rate is largely a function of the water needs

of the plant at harvest. Different plant organs incorporate tritium at different rates, and thus the careful selection of which parts of the plant are ingested after an accident may limit radiological risk; the calculation of actual tritium incorporation is challenging for plants other than leafy vegetables. Deriving this figure for grains or fruits may require empirically derived equations due to variable plant physiology. Additionally, as animals ingest tritium contaminated plants, OBT may be formed in animal meat and milk. Large differences may exist in how models calculate OBT formation at night; some may assume no tritium uptake at night, while others may make a conservative approximation of no differences between daytime and nighttime.

2.6. Tritium Transfer to Animals

In the aftermath of a tritium release, animals are expected to ingest grass or other plants containing exchangeable OBT. This incorporation of tritiated organic matter via grass ingestion is expected to continue for several months, with additional tritium ingestion from the soil and water. One protective measure is therefore to consider relocating animals to uncontaminated grass. The concentration is a function of transfer coefficients derived for each animal product and the corresponding dose to man following ingestion. The concentration in the animal product is a function of the integrated concentration of HTO and OBT in that product, the dose coefficient, and the amount of that product consumed. Previous studies have indicated that milk and meat are not major exposure pathways in accident cases, due to the speedy biological turnover of HTO and OBT.

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3. EXISTING TRITIUM MODELS

The following discussion identifies a select group of codes that have been used to model the consequences of tritium releases. These codes were identified by a review of the literature and other large benchmarking studies for tritium modeling and should not be considered an exhaustive list of available tritium models. Of the codes listed in this section, UFOTRI is considered to best reflect state-of-practice for tritium modeling and thus be suitable as a candidate for MACCS code upgrades. The other codes identified in the review are summarized for completeness but are generally used for research purposes, are still under development, are limited in scope, or do not have sufficient documentation to further consider them as candidates for MACCS code upgrades.

3.1. UFOTRI

UFOTRI is a tritium transport and dispersion model most frequently mentioned in the literature and is the most comprehensive regarding tritium releases, dispersion, deposition, and subsequent movement and transformation through the environment. UFOTRI is a code suite developed to assess the off-site consequences from accidental tritium releases [25]. To evaluate the atmospheric transport of such a release, this model employs a simple 2-d, Gaussian dispersion, segmented plume model that accounts for meteorological conditions at the time of an accident scenario (e.g., wind speed, direction, precipitation) to calculate radioactive plume trajectories; in this regard, the functionality of UFOTRI mirrors MACCS functionality for common LWR source terms.

The UFOTRI first-order compartment module describes a release of tritium as it moves first through the atmosphere (dispersion, deposition, re-emission), the air and ground surface concentrations resulting from this transport, and the direct exposure pathways as a result (e.g., inhalation, skin absorption) [26]. The module then describes the movement of tritium in the food chain and the associated transfer processes in soil, plants, and animals.

UFOTRI is based on a modified version of the MUSEMET atmospheric dispersion model [26]. This model divides source terms into hour-long intervals and uses an area source model to calculate tritium re-emission from soil and plants. In addition to accounting for milk and beef production, this model also accounts for tritium gas to tritiated water conversion, tritium transport into deep soil, and tritium uptake by plant and subsequent conversion to OBT. At the conclusion of the transport and transfer processes (steady-state), the calculated concentrations are then input into an ingestion module. Similar to MACCS, outputs of UFOTRI include inhalation, skin absorption, and ingestion doses.

Accordingly, a MACCS calculation that appropriately accounts for tritium may utilize the existing MACCS dispersion model to calculate airborne, vegetation, and ground surface tritium concentrations as a function of distance. However, this model would also need to include an additional model to account for tritium dispersion and transfer relationships with plants (uptake and evaporation), exchanges at the soil-atmosphere interface, deep soil transport, conversion of tritium transport into OBT, and intake by grazing animals. In UFOTRI, the secondary model does *not* include a calculation for the conversion of tritium gas to tritiated water in the atmosphere, a process with a half-life on the order of years [27].

In a previous assessment, MACCS was compared to UFOTRI for basic modeling functions relevant to tritium. A table summarizing that assessment, adapted directly from [8], is recreated in Table 3-1. The table elucidates the main differences between the two models being:

- the specific differentiation between forms of tritium in the environment,
- a detailed reemission physics model,

- accounting for the conversion of tritium to HTO,
- uptake of tritium by plants, and
- conversion into OBT.

In summary, MACCS maintains all of the basic atmospheric transport functionalities of UFOTRI but lacks the detailed tritium accounting of a UFOTRI's tritium-specific model.

A related code, NORMTRI, models atmospheric releases and behavior of tritium in the environment for routine releases of tritium. NORMTRI considers tritium gas and water vapor and the conversion of tritium gas into tritiated water and subsequent re-emission and conversion into organically bound tritium. Inputs for NORMTRI are closely modeled after UFOTRI [28].

Table 3-1. Comparison of basic model features between UFOTRI and MACCS

Model Feature	UFOTRI	MACCS/Gaussian Model
Gaussian release model	Yes	Yes
Sensible heat/release duration	Yes	Yes
Wet and dry deposition	Yes	Yes
Stratified random sampling of site meteorology	Yes (smaller sample size than MACCS)	Yes
Prescribe meteorology	Yes	Yes
HTO-HT differentiation	Yes	No
Reemission from soil/vegetation	Detailed reemission physics model	Resuspension model
Tritium conversion from HT to HTO	Yes	No
Update of HTO by vegetation	Yes	No
Conversion of HTO into OBT	Yes	No
1st order compartment model for long-term behavior in food chain	Yes, for HT and HTO	Yes

Source: Adapted from Table 1 of [8]

3.2. GENII

The "GENeration II" GENII model is a well-documented dose and risk assessment model developed by the US Environmental Protection Agency (EPA) Office of Radiation and Indoor Air [29]. The model is also capable of calculating accumulation in the environment and radiation doses from surface water, groundwater, and soil. GENII contains an atmospheric transport and deposition module that accounts for the transport, diffusion, deposition, depletion and decay of radionuclides in the atmosphere [29]. This module releases on the straight-line Gaussian plume and Lagrangian puff models. The surface water transport module in GENII calculates transport through surface waters and can be used to estimate the water concentration from radionuclide releases. GENII also contains the capability to calculate near-field exposures for receptors near to the initial

contamination. GENII utilizes special tritium and carbon-14 models within the near-field exposure module that assumes root uptake pathways directly from the soil are zero, based on an assumption that tritium and carbon-14 are in specific activity equilibrium with air or water [29].

In the acute exposure module, GENII provides estimates of short-term contamination exposure via air inhalation, external exposure to contaminated air, water ingestion, showering, swimming and boating [29]. GENII implements special considerations for tritium that assume acute releases of tritium will have comparatively lower levels of contamination to vegetation and animals than chronic releases. GENII assumes that a tritium contaminated plume is only present for a few hours, resulting in a fractional specific-activity model with a exposure time equal to the time of plume passage and the application of an equilibrium factor to account for the ratio of exposure time and growing period. GENII assumes an 8-hour desorption half-time [29].

The chronic exposure module in GENII calculates exposure media concentrations from groundwater, surface water, and airborne contamination pathways [29]. Exposure pathways include air, crops, animal products, soil, aquatic foods, and water. Tritium concentrations in the chronic exposure pathways depends on the concentration in contaminating media, and the fractional content of hydrogen in the plant or animal being contaminated. GENII uses the hydrogen content of the wet and dry portions of the food product to calculated tritium concentrations. OBT generation is also calculated for plants and animals [29]. In the tritiated water special model, air moisture concentrations are calculated based upon humidity and air concentration. A reduction factor is applied for plant products to account for dilution from soil water. In animals, HTO is assumed equal to the average tritium concentration in ingested water (from food and drinking water). GENII assumes that the uptake of elemental tritium does not readily occur by plants but elemental tritium is oxidized by soil microbes [29].

3.3. RSAC

The Radiological Safety Analysis Computer (RSAC) Program calculates consequences associated with a radionuclide release to the atmosphere [30]. Similar to other radionuclide transport and consequence models, RSAC calculates transport, decay and ingrowth through the environment and the corresponding doses from inhalation, air immersion, ground surface, ingestion and cloud gamma. RSAC treats all radionuclides similarly for the atmospheric transport and deposition compartments of the model [30]. However, RSAC uses different equations for calculating ingestion doses from tritium, compared to other radionuclides [30]. Tritium doses in vegetation from chronic releases are calculated accounting for the release rate of tritium, an assumed fraction of total plant mass that is water (0.75), an assumed ratio of tritium concentration in plant water to tritium concentration in atmospheric water (0.5) and the absolute humidity [30]. Notably, a similar calculation is used for C-14 that instead accounts for the fraction of total plant mass that is carbon and the natural carbon concentration in the atmosphere. For acute releases, RSAC assumes tritium is released from vegetation following an acute release with a nominal 1 day half time and additionally assumes that harvest occurs at a constant rate beginning when the release is initiated [30]. No other special equations or considerations for tritium are used in RSAC.

3.4. Other Models

Other models included are those that are used for research purposes, are still under development, are limited in scope, or do not have a standalone software package available for general use. Often these models are without user guides or theory manuals and include only brief mentions in the literature or model validation and comparison studies. Those models are briefly summarized here.

3.4.1. ETMOD

The Environmental Tritium Model (ETMOD) model developed by Ontario Hydro is used to calculate tritium behavior and fate for short timescale atmospheric release of tritium. ETMOD simulates atmospheric dispersion, deposition, oxidation of tritium in soil, re-emission of oxidized tritium to the atmosphere, and movement of oxidized tritium in soil, plants, animals, and milk [31]. ETMOD uses a resistance model for deposition that considers atmospheric turbulence resistance, canopy resistance, and soil surface; re-emission additionally accounts for stomatal resistance [32]. Soil transport is calculated accounting for diffusion, groundwater flow advection and transpiration; users may select up to 25 soil layers. Re-emission considers concentration gradients between the soil surface and the air, with a similar model used for plant re-emission. ETMOD is documented and may be useful for informing updates to MACCS calculations for soil deposition, HT conversion and remission. Previous documentation has also noted good agreement between model outputs to field measurements [33].

3.4.2. TRITRAJ

TRITRAJ models tritium transfer though the environment and is based on UFOTRI [32]. Using a fixed transfer rate, TRITRAJ accounts for tritium transport into edible plants, milk and beef and includes a compartment model for variable soil depths (0-5 cm, 5-15 cm, and 15-30 cm). For each of these pathways, transfer of tritium from and between variable soil depths is modeled [32]. For the plant pathway, transfer of HTO from each soil layer into edible plants and eventually OBT is considered. For the milk pathway, transfer from the top layers of soil into pasture grass, into dairy cattle, and eventually into milk (in the form of oxidized tritium or organically bound tritium) is considered. A similar pathway is considered for beef cattle, with the transfer of HTO into pasture grass and eventually into beef cattle as either OBT or HTO. These compartment models rely on transfer coefficients, calculated based on the hydrogen equilibrium for each of the compartments. The transfer rates for most major processes are fixed. This model accounts for the difference between daytime and night-time stomatal resistance for plants on 12-hour cycles [32]. There has been some research interest in the TRITRAJ model, but no detailed documentation was located.

3.4.3. TOCATTA

TOCATTA is a simple research model that uses a dynamic compartment model for evaluating tritium behavior in agricultural soil and plants exposed to tritium contaminated water and gaseous HTO releases from nuclear facilities [9]. TOCATTA uses a single soil layer and a resistance model paired with a concentration gradient to calculate deposition of HTO to soil. Re-emission flux also depends on concentration gradient. TOCATTA does not account for non-oxidized tritium. TOCATTA requires meteorology and a known concentration of HTO in the air and rainwater [34].

3.4.4. TRILOCOMO

The local compartment model for dispersion of HT and HTO (TRILOCOMO) is a research model that was developed alongside two 1980's tritium field studies in France for accident dose assessment. TRILOCOMO uses a resistance deposition model for soil and plants and uses a concentration gradient to calculate re-emission from plants and soil. TRILOCOMO calls a sub-model called COWTRI to model OBT formation in animals and milk [32].

3.4.5. TRIMOVS

TRIMOVS is a research model that describes the deposition, conversion of tritium to oxidized tritium, and the conversion of HTO to OBT. Deposition to soil uses the resistance model. TRIMOVS also calculates exchange velocities of oxidized tritium. These velocities are calculated at the soil surface/atmosphere interface by considering turbulent mixing and surface resistance. TRIMOVS incorporates submodels for growth of plant storage organs and OBT assimilation and translocation [32]. Up to 150 soil layers may be considered. This model has a simple OBT model that only accounts for the average growth of plants.

3.4.6. TRICAROM

TRICAROM is a research model used to determine tritium transfer rates. This model uses plant physiology, soil physics and crop modeling to inform model outputs [32]. This model has limited relevance to the accident consequence modeling space, as its utility is largely limited to the calculation of transfer rates, rather than the movement of tritium through the environment and the associated consequences. OBT formation in plants depends on environmental conditions and animals and milk follow a retention function.

3.4.7. TRINIRBU

TRINIRBU models the movement of tritium from air to soil into roots and leaves, and the transfer into milk and beef as OBT or HTO. Fixed transfer rates are assumed for all processes including deposition to soil and plants (zero at night), re-emission, and OBT formation in plants, animals, and milk [32].

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4. CONCLUSIONS AND RECOMMENDATIONS

A review of tritium transport and tritium-specific models suggests that MACCS is fundamentally flexible enough to accommodate the atmospheric transport of tritium, as the atmospheric transport processes are very similar. The major differences are in the identification of the variable chemical forms of tritium and the transformation of those forms in the environment, deposition, reemission, and conversion to OBT. These are areas in which MACCS may benefit from updates to longer-term environmental process models, especially for the transformation processes of tritium in the atmosphere, soil, plants, and animals. Based on existing models, multiple pathways may exist for these updates including:

- Simply continue to use the most conservative dose coefficient for HTO in the existing MACCS code with no changes to the atmospheric transport or longer-term processes. This approach reduces the need to have an exact accounting of the chemical form of tritium releases, which may additionally require updates to precursor codes such as MELCOR. This approach, which is the current practice in MACCS, is likely conservative because not all tritium releases will be in the oxidized form, the oxidation of tritium in the environment takes time, the water ingestion model in MACCS assumes ingestion of all directly deposited and washoff radionuclides despite tritium gases' low solubility in water and additionally assumes that the land does not act as a sink for deposited radionuclides, and food ingestion depends on ground concentration and ignores any transformation (e.g., organically bound tritium). A simple update to supplement this pathway might include the addition of the 50% inhalation dose correction factor to account for HTO skin absorption, as used by previous studies and other tritium models.

- Similar to the models discussed above, develop the capabilities for MACCS to identify multiple different chemical forms of tritium and the associated transformation. There are variable tiers of complexity for which this might be accomplished, whereby the simplest option from a MACCS standpoint requires knowledge of the chemical forms of tritium expected in a release, with an option for the analyst to input these releases directly into MACCS. Additional complexity might be introduced by incorporating a simple transfer rate model for plants, animals, and milk, like those in the TRITRAJ or TRINIRBU. These updates might increase the fidelity of tritium accounting in MACCS with minimal effort.

- The most complex option would be to introduce tritium accounting capabilities similar to those in UFOTRI and the more complex capabilities of other models. This would include detailed calculations for atmospheric transport of tritium as they currently exist, updating the deposition model to the resistance model, providing a more detailed reemission model, accounting for tritium uptake and transformation in plants, animals and milk, and general conversion of tritium in the atmosphere and soil. While the effort involved in these updates is likely substantially higher, this would provide the most detailed estimation of tritium in an accident scenario.

Overall findings suggest that existing capabilities in MACCS are likely sufficient for calculating individual doses from tritium, but more complex, longer-term phenomena (e.g., food chain pathways) might benefit the most from model updates. Before any updates to MACCS are considered, advanced reactor inventories should be examined in order to ensure that time investment for tritium updates is worthwhile. For instance, tritium inventories and release fractions for advanced reactors may be low enough that they would not pose an environmental or human health risk even under the most conservative release scenarios. In this scenario, the current MACCS

model is likely sufficient to accommodate accident scenarios involving tritium. However, if there is potential for significant tritium releases from a risk perspective, then further modeling updates may be advisable.

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