SANDIA REPORT
SAND2024-12069
September 2024

Assessment of Current MACCS Capabilities for Modeling Atmospheric Physical and Chemical Transformations

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ABSTRACT

The physical and chemical transformation during atmospheric transport of radionuclides released into the environment has the possibility of impacting consequence modeling results. Accordingly, this report identifies physical and chemical transformations that may occur following release of chemically reactive radioactive species, how those transformations may affect modeling of consequences of release to the atmosphere and identifies current capabilities – in both MACCS and other state-of-practice atmospheric transport and dispersions models– to model those transformations.

It was found that the inclusion of physical and chemical transformations is currently very limited in current state-of-practice codes for atmospheric dispersion of radionuclides. State-of-practice atmospheric dispersion codes appear to be typically limited to simulating either physical-chemical transformations or radioactive transformations, but not both. A state-of-practice atmospheric dispersion code capable of performing parallel physical, chemical, and radioactive transformation was not identified. A few atmospheric dispersion codes capable of modeling physical and chemical transport of specific species such as tritium or uranium hexafluoride were identified. Consequently, there is currently no information available that clearly suggests updates to the MACCS code are needed to bring it up to state-of-practice. However, investigations concluded that the MACCS computational framework can currently accommodate multiple physical/chemical forms in one simulation. Additionally, with some major assumptions, the computational framework in MACCS can accommodate parallel physical-chemical and radioactive transformations.

ACKNOWLEDGEMENTS

The authors would like to thank the members of the larger MACCS team for their contributions to this effort, these contributors include Kyle Clavier and John Fulton. The authors would also like to acknowledge the technical contributions and input from the US Nuclear Regulatory Commission, including Keith Compton, Salman Haq, and Elena Yegorova. This research was funded by the US Nuclear Regulatory Commission under agreement number 31310020F0032.

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ACRONYMS AND TERMS

Acronym/Term	Definition
ARL	Air Resources Laboratory
CMAQ	Community Multi-Scale Air Quality
EPA	US Environmental Protection Agency
FEPs	features, events, and processes
HEDR	Hanford Environmental Dose Reconstruction
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
NOAA	National Oceanic and Atmospheric Administration
Non-LWR	Non-Light Water Reactor
NRC	US Nuclear Regulatory Commission
PNNL	Pacific Northwest National Laboratory
RADM2	Regional Acid Deposition Model, Version 2
RATCHET	Regional Atmospheric Transport Code for Hanford Emission Tracking
SCIPUFF	Second-order Closure Integrate PUFF Model
WRF-Chem	Weather Research and Forecasting-Chemistry

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1. INTRODUCTION AND PURPOSE

The US Nuclear Regulatory Commission's (NRC) Non-Light Water Reactor (non-LWR) Vision and Strategy Volume 3 report [1] identifies a need to understand the atmospheric physical and chemical transformations that can occur once a radionuclide is released into the atmosphere and how deposition behavior and dosimetry could be affected as a result. Previous work has identified that MACCS can account for variable chemical forms using alternate dose coefficients [2]. However, review of MACCS capabilities in relation to features, events, and processes (FEPs) related to atmospheric physical and chemical transformations after release and other models capable of handling these processes has not been investigated. Therefore, the objective of this report is to review FEPs of concern with regards to consequence modeling and identify current related MACCS capabilities and/or shortcomings. Additionally, other models capable of handling these identified FEPs to a varying degree are investigated.

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2. REVIEW OF POTENTIAL ATMOSPHERIC PHYSCIAL AND CHEMICAL TRANSFORMATION PROCESSES

The following discussion identifies illustrative examples of potential physical and chemical transformation processes, specifically agglomeration, hygroscopic growth, and chemical reactions, that can affect deposition and dosimetry. It is important to note, the specific transformation processes and their effect on consequence modeling discussed here is not an exhaustive list. Radionuclides can potentially undergo other transformations than what are described here, especially with varying source terms and release environment characteristics.

Agglomeration, the process of particles adhering to one another, or other substances is largely observed during containment releases due to interactions with ex-vessel core-debris [3]. Agglomeration processes are generally functions of the temperature and turbulence level of the released material and the temperature and turbulence level of the surrounding environment. Agglomeration will increase particle size, which can increase deposition velocity (at least over a certain particle size range) and affect inhalation dose coefficients.

Similarly, the tendency for a particle to absorb moisture (hygroscopic growth) is strongly dependent on the level of humidity in the atmosphere. As hygroscopic particles increase in size, the particles rate of deposition over time will change accordingly. One example of a hygroscopic radionuclide is cesium, specifically, the compounds cesium hydroxide and cesium iodine [4]. These compounds are highly hygroscopic and if released into the environment could settle very quickly affecting ground concentrations estimations in consequence modeling. Additionally, research indicates that reactions with atmospheric salts could affect hygroscopic behavior, further influencing deposition velocities [4]. However, these compound groups are typically neglected when modeling releases from traditional light water reactors and just a fraction of the mass from the compound specific to cesium is included.

Once released into the environment, radionuclides can also be susceptible to various chemical reactions depending on available reactants in the atmosphere such as pollutants, the sun (photolysis), and the release rate. For example, the reactional mechanism of iodine – a radiologically significant element released in light water reactor severe accidents [5] - in the atmosphere involves a minimum of 246 gas phase reactions including 13 photolysis reactions (see Figure 2-1 and Figure 2-2) [6].

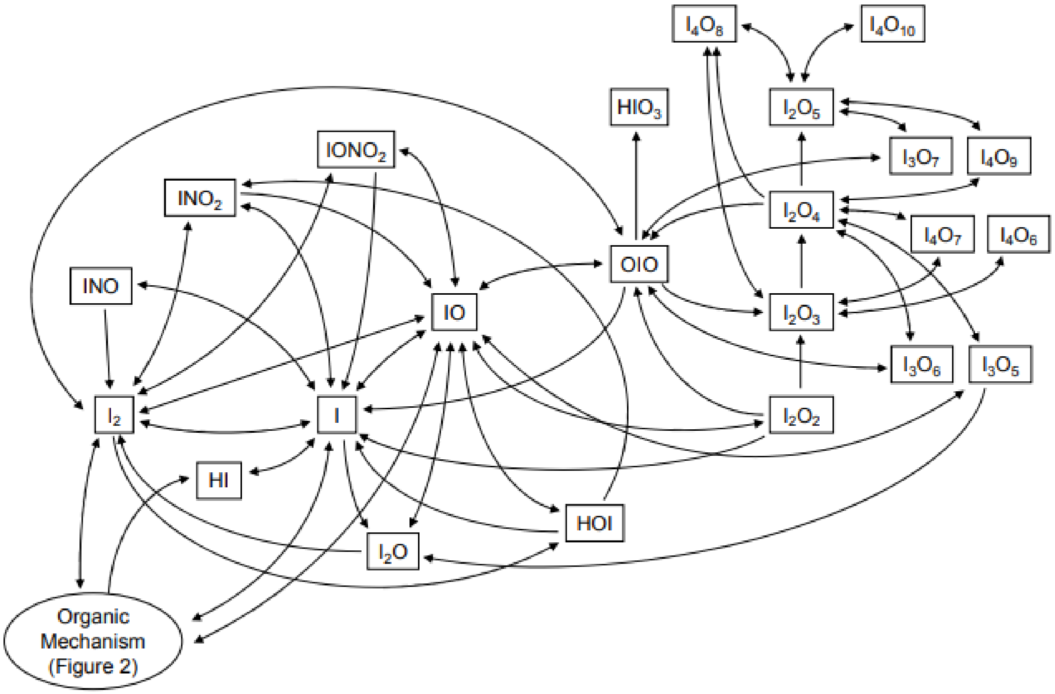

Figure 2-1. Iodine Reactional Mechanism, Inorganic[1]

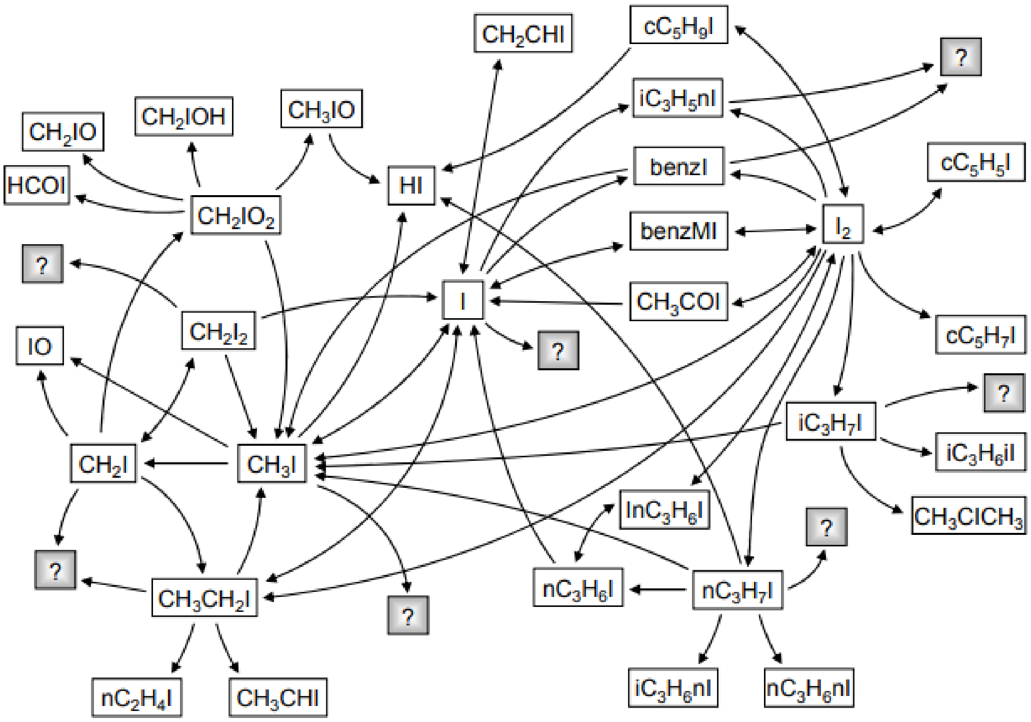

Figure 2-2. Iodine Reactional Mechanism, Organic[1]

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[1] Taken from [6]

These chemical reactions could affect the physical deposition process and dosimetry resulting in changes to the ground concentration and human health impact estimations in consequence modeling. For example, as described in [7], the variation in deposition velocity and inhalation dose coefficient across different chemical forms of iodine greatly influences ground concentration and consequence estimations. Following the Fukushima accident, release simulations of iodine have been compared to field measurements which concluded that there was a significant underestimation of radiation levels on the ground [6]. It was assumed this was due to the absence of chemical reactivity in the model. Similarly, as described in [8], photochemical oxidation is a key mechanism for tritium transformation. Using typical measured values, the dry deposition velocities of gaseous tritium and oxidized tritium vary by an order of magnitude [9].

In summary, the discussion above is just an illustrative example of some physical and chemical transformations that can affect the consequence modeling of radionuclides released into the environment. Upon this review, it is expected that these processes, among others (e.g., photolysis, hydrolysis, etc.), will affect, at a minimum, dry and wet deposition models as well as inhalation dose coefficients given particle size is a key parameter in both. Accounting for these processes in atmospheric transport and dispersion modeling would likely include the incorporation of first order transformation, fully coupled speciation models, etc., which is further investigated in the next section.

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3. REVIEW OF ATMOSPHERIC PHYSICAL AND CHEMICAL TRANSFORMATION CAPABILITIES WITHIN THE CURRENT STATE-OF-PRACTICE

The following section identifies atmospheric transport and dispersion codes with varying degrees of atmospheric physical and chemical transformation capabilities. As to be expected, most of the consideration of transformation is with respect to chemical modeling tools. While not directly applicable to nuclear power plant accident consequence modeling, these codes can be used as examples for current atmospheric physical and chemical transformation modeling practices. Additionally, the identified radionuclide atmospheric transport and dispersion models (e.g., HGSYSTEM/UF6, UFOTRI, and ETMOD) are very much radionuclide specific. A summary of capabilities in comparison to MACCS is shown in Table 3-1 below with further details discussed in the subsequent sections.

Table 3-1. Atmospheric Physical and Chemical Transformation Capabilities of Various Atmospheric Transport and Dispersion Models

Model	Models Multiple Depositing Species	Models Multiple Dosimetry Forms	Models Physical/Chemical Transformations	Models Physical/Chemical and Radioactive Transformation Together
MACCS	Yes	No	No	No
HYSPLIT	Yes	No	Some	No
CMAQ	Yes	No	Yes	No
WRF-CHEM	Yes	No	Yes	No
SCIPUFF/SCICHEM	Yes	No	Yes	No
AERMOD	Yes	No	Some	No
GENII	Yes	Yes	Tritium only	No
RATCHET	Yes	Yes	No	No
RASCAL	Yes	Yes	No	No
HGSYSTEM/UF6	Yes	No	UF_6 only	No
UFOTRI/ETMOD	Yes	Yes	Tritium only	No

As displayed in the table above, aside from chemical models with very intricate databases and radionuclide specific consequence models, the accounting of physical and chemical transformations in current atmospheric transport and dispersion modeling practices is very limited. Based on this information, it is believed that MACCS falls within the current state of practice until more information is made available on the transformation behavior of radionuclides of concern. A more detailed discussion on current MACCS capabilities is given in Section 4.

3.1. HYSPLIT

The National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory's (ARL) Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) is used to compute complex transport, dispersion, chemical transformation, and deposition [10]. The plume depletion processes are relatively simple in which dry deposition can be designated as a dry deposition velocity or determined using a resistance method. As for wet deposition, two different processes are used for particulates and gases. For particulates, removal from within a cloud layer and removal in rain below a cloud layer is determined using a scavenging ratio and scavenging coefficient respectively and for gases, removal is determined using Henry's Law [11]. Additionally, radioactive decay is determined using the Bateman equations and air and ground concentrations of radionuclides are adjusted accordingly. As described previously in [2], HYSPLIT does not account for variable chemical forms in plume depletion modeling. However, during transport and dispersion, some simple accounting of physical and chemical transformations can be incorporated. For two different pollutants, a conversion rate from pollutant #1 to pollutant #2 (fraction per hour) is used to track the transformation and changes in concentration at every time step [12]. Additionally, HYSPLIT does have the ability to account for the oxidation and dry and wet removal of sulfur dioxide using more complex chemistry equations. HYSPLIT also models the atmospheric fate and transport of semi volatile pollutants and includes chemical transformations due to hydroxyl radicals and photolysis [10]. However, these additional modules are not incorporated into the standard compilation [12].

It is also worth noting that the limited transformation capabilities described above cannot be modeled simultaneously with the radioactive transformation (decay). For example, Pacific Northwest National Laboratory (PNNL) conducted an analysis using HYSPLIT instead of the RATCHET code to model doses from I-131 to the public, deviating from the Hanford modeling sequence [13]. However, when considering the speciation of iodine, PNNL did note that HYSPLIT can model the transport of all these forms, but not the interchange between the forms. Consequently, PNNL used a unit source of all three forms and determined the air concentration and ground deposition of each form separately and then combined them using a weighted sum of speciation fractions [14].

3.2. CMAQ

The Community Multi-Scale Air Quality (CMAQ) model was developed by the US Environmental Protection Agency (EPA) and calculates the emission, deposition, phase transitions, physical and chemical reactions, and photolysis of several species [15]. The CMAQ modeling system contains a multipollutant, multiphase, one-atmosphere modeling approach to account for three different chemical process types; gas-phase chemistry, aqueous chemistry (formation of cloud droplets), and aerosol formation. The gas-phase chemical processes are of most importance given that pollutants are primarily emitted and react as gases, but the model includes the interactions of the gases with these other phases. There are a total of three gas-phase chemical mechanisms, one being the widely known Regional Acid Deposition Model, version 2 (RADM2) [16], that address ozone formation and acidic deposition. Each of these mechanisms contains a total number of species and the associated first order, second order reactions, etc. with the corresponding reaction rates that are then used in a mass conservation equation to determine the time-rate of change for each species concentration [15]. Dry and wet deposition of the various species is then determined using the method described in [16] and Henry's law respectively [15].

The CMAQ code can model the atmospheric transport and dispersion of radionuclides as demonstrated in [17]. However, it was noted that chemical and aerosol processes were not included because this is not well understood for radioactive materials. If more information on the physical and chemical transformation processes of radionuclides were available and something like RADM2 were to be created, it could potentially be implemented into MACCS as an additional module during the transport and dispersion simulation.

3.3. WRF-CHEM

The Weather Research and Forecasting-Chemistry (WRF-Chem) model, like CMAQ, calculates the emission, deposition, phase transitions, chemical reactions, and photolysis of several species using a fully coupled online chemistry model [18]. In fact, WRF-Chem uses the same atmospheric chemical mechanism (RADM2) for its gas-phase chemistry module and the same dry deposition method. The primary difference between CMAQ and WRF-Chem is the former is what is known as an offline model in which chemical processes are treated independently of the meteorological model while the latter is considered online [19].

Transport and chemical schemes from WRF-Chem were adapted to simulate the atmospheric transport and deposition of I-131 and Cs-137 aerosols in one study [20]. To perform this analysis, the analysts modeled different gaseous fractions of I-131 and different particle size distributions of Cs-137. It was noted that the partitioning may change during transport and deposition processes, but it was not considered in their simulations due to lack of available information [20].

3.4. SCIPUFF/SCICHEM

The Second-order Closure Integrate PUFF Model (SCIPUFF) is a Lagrangian puff dispersion model that is a collection of three-dimensional Gaussian puffs [21]. This model is very similar to HYSPLIT, CMAQ, and WRF-Chem in which it can model the dispersion and deposition of primary pollutants in both the short and long range. Historically, linear chemical transformations are accounted for in this model using exponential decay which are input from the user [21], like the process used in HYSPLIT. However, in recent years, the SCICHEM model has been developed which incorporates complete gas phase, aqueous and aerosol phase chemistry into SCIPUFF [22]. In fact, SCICHEM aerosol and aqueous-phase chemistry modules are consistent with those used in CMAQ. It appears this model is not capable of modeling radioactive species or dosimetry forms.

3.5. AERMOD

AERMOD, also a model developed by the EPA, is an air dispersion model used to estimate the dispersion of pollutants emitted into the atmosphere from industrial sources and is the preferred regulatory dispersion model. AERMOD does contain specific chemistry methods for NO_x to account for the equilibrium between NO, NO_2, and ozone in the atmosphere called the Generic Reaction Set Method [23]. This method uses reaction rates determined by a set of equations that describe the reactions between these three compounds in the atmosphere. A fifth order Runge-Kutta scheme with adaptive time-stepping is then used to solve the set of chemical reactions to give appropriate concentrations for each compound at each receptor post-chemistry [23]. It is noted that the NO_x chemistry regime can include many reactions but in the near field there are only a few dominate reactions. The model can then perform dry and wet deposition of the various compounds using the algorithms described in [23]. This model also has the capability of estimating atmospheric concentration and surface deposition of release radioactive materials.

3.6. GENII

GENII is a computer code system developed for the EPA by PNNL to calculate the radiation dose and risk from releases into the environment. Specifically, release to water or air as well as initial contamination of soil or surfaces is considered with exposure pathways including direct exposure to water, soil, and air through inhalation and ingestion [24]. The tritium model also includes conversion of gas into vapor and biological conversion of both into organically bound tritium like UFOTRI and ETMOD (described in Section 3.10 below) [25]. Other than tritium during chronic releases, GENII does not account for additional transformations of radionuclides. The dry and wet deposition processes are very similar to those used in RATCHET and RASCAL (described below) in which deposition of gases and particles is treated separately. Additionally, the model does account for radioactive transformations and uses standard dosimetry methodology.

3.7. RATCHET

The Regional Atmospheric Transport Code for Hanford Emission Tracking (RATCHET) was created by the Hanford Environmental Dose Reconstruction (HEDR) project to aid in determining the radiation doses to individuals from operations at the Hanford Site [26]. There is a version 2 of the model, named RATCHET2, that is identical to the previous version with the decay calculations disabled [27]. RATCHET treats iodine in a unique manner allowing it to be partitioned into nonreactive gas, reactive gas, and particulate components. However, the model assumes that the partitioning of iodine is independent of travel time [26]. The specific partitioning of the three groups can be made uncertain but remains constant for the entire simulation.

Therefore, RATCHET models the dry deposition of each of the three forms individually but can also model the deposition of a mixture of the forms. Separate dry deposition velocities are computed for each type of material and if the material exists simultaneously in more than one form, a weighted-average dry deposition velocity is computed. The averaging weights are based on the fraction of the iodine in each form which, again, are constant during a model run. Like HYSPLIT, wet deposition of gases and particles are treated separately. Wet deposition of gases is modeled assuming equilibrium between gas concentrations in the air and precipitation. Wet deposition of particles is modeled using washout coefficient assuming irreversible collection of particles as the precipitation falls through the puffs. There are also two methods for wet deposition of particles depending on weather conditions (rain or snow).

While the model does not have specific physical and chemical transformation capabilities like the chemical models above, there is an accounting for the chemical speciation of iodine that MACCS does not currently handle. If updating the handling of iodine in MACCS were of interest, this method could be easily adapted. Additionally, as more information on advanced reactor source terms becomes available, and if additional radionuclides are found to behave similarly to iodine in the environment, a similar process to what is used in RATCHET could potentially be utilized.

3.8. RASCAL

The RASCAL code was developed by the NRC and addresses the short-term accidental release of radioactive material. This model is used by the Protective Measures Team in the NRC operations center and is a part of the Radiation Protection Computer Code Analysis and Maintenance Program [28]. This model does account for the speciation of iodine and considers particulates, I_2 and CH_3I and models the dispersion and deposition of all these forms, but not the interchange between the forms. The RASCAL code along with other various dose assessments in the past, rely on information from the HEDR project, i.e., RATCHET. Specific algorithms from RATCHET (described in the section above) were used to upgrade the atmospheric dispersion and deposition models and dose calculations of RASCAL. However, it is noted that RATCHET includes stochastic components for treating uncertainty in parameters and RASCAL does not [28].

3.9. HGSYSTEM/UF6

The HGSYSTEM/UF_6 model is a hybrid model that combines the dispersion and HF thermodynamics aspects of HGSYSTEM with the chemistry and thermodynamic aspects of UF_6 that was originally developed to upgrade the safety analyses for various gaseous diffusion plants under the direction of the U.S. Department of Energy [29]. Accordingly, these analyses focused on the consequences of accidental release of UF_6 to the atmosphere.

In short, this model can handle transport and dispersion of dense gases, plume thermodynamics and simulate the chemical reactions of UF_6 with water vapor. However, the interactions between the three processes are complex and the three processes are solved sequentially in each integration step. The dispersion subsystem calculates the plume geometry and air entrainment rate as the plume travels down wind, the chemistry subsystem uses the air entrainment rate to calculate the composition of the plume based on the defined chemical reactions and current plume composition, and the thermodynamics system calculates the plume equilibrium temperature and density based on the newly determined plume composition. The temperature and density then affect the dispersion [29]. As for deposition, this process is applied as a post-processor and there is no feedback between dry deposition and the plume chemistry model. This is because the model assumes that the effects of deposition on dispersion, chemical reactions, and thermodynamics can be neglected [29].

If the appropriate information were to be made available for specific radionuclides of concern, a similar process could be implemented into MACCS to account for environmental transformations. However, the HGSYSTEM/UF_6 model demonstrates how complex the process is for just one species and mimicking this process for multiple species may be tedious.

3.10. UFOTRI/ETMOD

The UFOTRI and ETMOD models assess the consequences of tritium releases to the environment and accounts for exposure through inhalation, skin absorption, and contaminated foodstuffs [30][31]. For tritium, the main transformation of importance is the conversion of tritium gas into tritiated water vapor. However, this process occurs once the tritium gas has deposited and entered the soil (post atmospheric transport). Within the soil, tritium gas is converted into tritiated water vapor and subsequently re-emitted into the atmosphere through evaporation and transpiration.

To assess the impact tritium specific transformation processes can have on dose assessments, dose calculations from UFOTRI and ETMOD have already been compared to MACCS in a previous analysis documented in [32]. Nonetheless, the capabilities of these models as well as those described above further demonstrate that current state-of-practice for the inclusion of physical and chemical transformation capabilities to any degree is very much radionuclide or species specific.

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4. CURRENT CAPABILITIES OF MACCS IN RELATION TO ATMOSPHERIC PHYSICAL AND CHEMICAL TRANSFORMATIONS

Given the review of FEPs of concern and the physical and chemical transformation capabilities of other state-of-practice atmospheric transport and dispersion models, it is important to identify and understand the current capabilities of the MACCS code for handling physical and chemical transformations. As the focus of this analysis is on plume depletion and dosimetry, only modeling features affecting atmospheric transport and dispersion are assessed. Post atmospheric transport processes (e.g., resuspension and weathering) are outside the scope of this analysis and could warrant further investigation.

4.1. Radioactive Transformation and Dosimetry

It is important to consider radionuclide decay and ingrowth processes for physical and chemical transformations as decay progeny can have different chemical properties than their parents. Currently, MACCS utilizes the Bateman equation and a predefined decay chain database to account for radioactive transformation. This database includes detailed information such as half-lives, decay products, and decay fractions for each radionuclide. For user reference, the decay chain database is included in MACCS as an ASCII text file named INDEXR.DAT. For decay and ingrowth between accident initiation and plume segment release, the appropriate aerosol size and rate of deposition for progeny with different chemical properties is applied at the time of release. Depending on the fraction of decay that occurs before versus after plume segment release, the user can choose to model decay products during this period using the same release fraction and aerosol size distribution as the parent radionuclide or as those specific to the decay product. Additional decay and ingrowth after release is applied after atmospheric transport modeling is completed by adjusting the air and ground concentrations. Therefore, any ingrowth of progeny during transport with different chemical properties is not accounted for in the plume depletion model [33].

Additionally, for dosimetry purposes, the MACCS framework permits the user to define a dose coefficient factor for only one chemical form and particle size per radionuclide, without the capability to handle multiple chemical forms or particle sizes.

4.2. Dry Deposition

As described in Section 2 above, physical and chemical transformations discussed previously could positively or negatively affect particle size which will influence dry deposition velocities. However, the dry deposition model in MACCS is not capable of changing or updating the particle size distributions as a function of time to account for physical and chemical transformation processes that may occur during atmospheric transport.

Currently, the MACCS code models dry deposition using Chamberlain's source depletion method and uses a particle-size-dependent deposition velocity to determine the depletion rate from the plume. Particle size distributions for each plume segment are defined within the model (a maximum of 20 particle size bins are allowed) and the dry deposition equation is applied separately to each particle size bin. After a segment has left a radial interval, the fraction of aerosols in each particle size bin is adjusted to account for the number of aerosols removed [33]. This process is applicable to all chemical groups except for non-depositing species (e.g., noble gases). This system does not have the ability to repartition the particle size bins due to physical and chemical transformations over time.

4.3. Wet Deposition

Wet deposition handling in MACCS does not depend on particle size, but rather it is a function of precipitation duration and intensity [33]. In current modeling practices, all chemical groups except for non-depositing species (such as noble gases) are subject to wet deposition. The common modeling practice linear coefficient for washout is based on the 50th quantile for 1-micron particles [34]. This washout coefficient is applied to all chemical groups subject to wet deposition and there is no accounting for changes in particle size due to physical and chemical transformations.

5. APPLICABILITY OF INCLUDING PHYSICAL AND CHEMICAL TRANSFORMATIONS WITHIN THE CURRENT MACCS FRAMEWORK

As described in the sections above, the MACCS code is believed to be within the state-of-practice for handling physical and chemical transformations of radionuclides save for some specific cases. However, there is interest in determining if MACCS is already capable of including physical and chemical transformations if such information were to ever become available.

5.1. Modeling Multiple Physical-Chemical Forms in One MACCS Simulation

A logical first step in determining whether the current MACCS framework can incorporate transformations is to evaluate its ability to model multiple chemical forms within a single simulation, an area identified for future work in previous investigations [2]. To assess this capability, a previous analysis documented in [7] is leveraged. This analysis documents an approach to incorporating multiple forms of iodine, elemental (I_2) and organic (CH_3I) as well as particulate, using the MACCS code. However, this approach still models the three forms separately and states that for all three forms to be present in one simulation, the dose coefficient file needs to be updated. Therefore, the purpose of this investigation is to explore the feasibility of modifying relevant MACCS files to accommodate multiple physical and chemical forms for a single species. While modifying MACCS data files is generally not recommended, these investigations could yield valuable insights for future guidance aimed at expert users.

5.1.1. Modeling Approach

To perform this investigation, a similar setup as what is described in [7] was used with slight modifications. A summary of the MACCS set up used is described below. All other parameters not defined here follow the MACCS parameter guidance document [35].

- Unit release of I-131, ground-level without plume rise
- 10 plume segments each with a 1 hr release duration and 10% release fraction
- Area source consistent with 40 m x 40 m x 40 m building dimensions
- Constant weather, 5 m/s, D stability, no precipitation
- 100 ppl/km^2 population density with no emergency response actions

To represent the different forms of iodine, Table 5-1 and Table 5-2 describe the variation in deposition velocity/particle size distribution as well as inhalation dose coefficients that were used. Table 5-1 just displays information for particulate iodine since it was assumed that all elemental iodine has a deposition velocity of 2 cm/s and all organic iodine has a deposition velocity of 0.02 cm/s. As a first step, using this information, three separate MACCS simulations were run to recreate what was done in [7] to observe the impact these different forms of iodine have on the results.

Table 5-1 Particle Size Distribution and Deposition Velocities for Particulate Iodine [7]

Fraction	Particle Size (μm)	Deposition Velocity (cm/s)
6.47E-03	0.15	0.053
3.22E-02	0.29	0.049
1.21E-01	0.53	0.064
3.28E-01	0.99	0.11
3.57E-01	1.8	0.21
1.32E-01	3.4	0.43
1.85E-01	6.4	0.84
1.95E-01	11.9	1.4
3.19E-01	2.1	1.7
1.44E-03	41.2	1.7

Table 5-2 Variation in Inhalation Dose Coefficient for the Three Forms of Iodine [7]

Form	Effective dose coefficient: Inhalation, Adult (Sv/Bq)
Particulate	7.4E-09
I_2	2.0E-08
CH_3I	1.5E-08

In the next phase of this investigation, since all three simulations were modeled as unit releases, the results for each form of iodine were subsequently combined. This approach allows for a comparative analysis against a single MACCS simulation that includes all three chemical forms. For demonstration purposes, the speciation used in RATCHET and RASCAL was used. This speciation assumes particulate iodine accounts for 25%, elemental iodine accounts for 30% and organic iodine accounts for 45%. Lastly, this same speciation was used to model all forms together in one MACCS simulation. To do this, the three forms of iodine, labeled as I-131a, I-131e, and I-131o, were added to the INDEXR.DAT and DCF file with the only difference being the inhalation dose coefficient defined for each form in the table above. While updating the INDEXR.DAT and DCF file, several rules were identified (for use with MACCS 4.2) that need to be followed if one wishes to perform an analysis like the investigation described here. These identified rules are defined in Table 5-3 below. After successfully updating the files, each form was added separately to the core inventory data block and assigned to a different chemical group. The release fractions for each chemical group were consistent with the previously described speciation. Additionally, the appropriate particle size distribution and deposition velocities were assigned to each group.

Table 5-3 Identified Constraints When Altering MACCS 4.2 Files

INDEXR.DAT File	DCF File
• Any additional radionuclides/forms and their decay chains need to be added to the bottom of the list. • Each name must be unique, the same radionuclide cannot be defined twice. • Name can only be a maximum of 7 characters. • The second character in the name must be lowercase. • If there is an "m" in the 4–7-character position, it needs to be lowercase. • A maximum of 999 radionuclides are allowed.	• The title "### NUCLIDES DEFINED IN THIS FILE" at the top of the file must match the total number of radionuclides defined. • Order does not matter in this file, but the order must match between where clearance classes are defined and where the dose coefficient information is defined. • A maximum of 999 radionuclides are allowed.

5.1.2. Investigation Results

Similar plots as what is shown in [7] (Figure 5-1 and Figure 5-2) were created to compare the results of this investigation. As can be seen in Figure 5-1 and Figure 5-2 below, the surface deposition and effective dose during the 7-day emergency phase period were used for comparison. Given that only unit-releases were modeled, and these results are for demonstration purposes only, units on the y-axis were not included. In each figure, the blue, green, and yellow lines represent the three separate MACCS simulations for each form of iodine and the red line represents the results after all three simulations were combined assuming the partitioning of each form of iodine defined above. Finally, the black x's represent the results when all three forms of iodine are included in one MACCS simulation after updating the INDEXR.DAT and DCF file. As can be seen in both figures below, the post-process manual combination and combination simulated in MACCS produce almost identical results (differences were less than 0.41%) demonstrating the modeling of multiple physical/chemical forms in MACCS was successful. However, modifying MACCS files to include multiple chemical forms should be approached with extreme caution as modifying these files incorrectly could introduce additional complications. Such modifications should only be undertaken by expert users and be subject to thorough scrutiny to ensure that no code errors are inadvertently introduced.

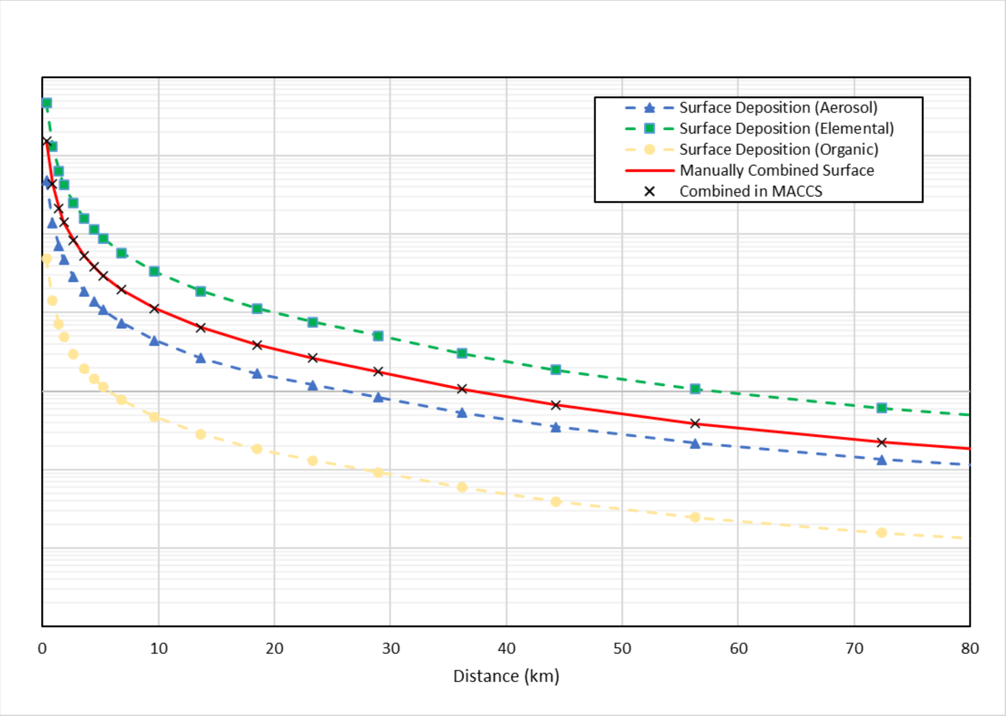

Figure 5-1 MACCS Surface Deposition Results for Multiple Forms of Iodine[2]

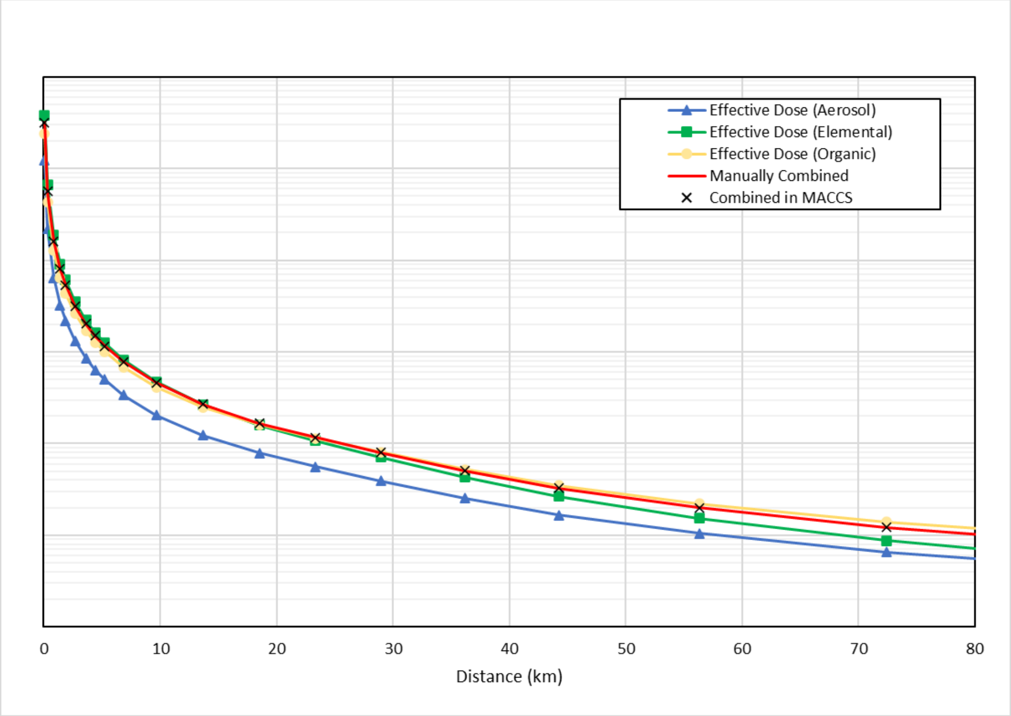

Figure 5-2 MACCS Emergency Phase Effective Dose (7 Days) for Multiple Forms of Iodine[2]

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[2] Vertical axes are unlabeled because only the relative results are of interest.

5.2. Incorporation of Transformation Processes Within the Current MACCS System

Given the results discussed in the section above, the MACCS information files (specifically the INDEXR.DAT and DCF files) can be successfully modified to include multiple physical/chemical forms for one radionuclide species. However, what still needs to be demonstrated is whether transformations between these different physical/chemical forms can be included as well to better prepare the code when such information becomes available. Therefore, the investigation discussed here uses the following reactions with ozone and hydroperoxyl radicals in the atmosphere as an initial example. Given that this is for demonstration purposes only, the radionuclide of concern is denoted as X.

$$O_3 + X \rightarrow XO + O_2$$
$$HO_2 + XO \rightarrow XOH + O_2$$
$$XOH + hv \rightarrow OH + X$$

In this scenario, there are three chemical species to consider: X, XO, XOH. One thing to note is that MACCS can only have one equation per radioisotope in the INDEXR.DAT file. This necessitates the use of a placeholder radionuclide when XOH transforms back into X. In this example, it was decided to denote Xtemp-123 as a placeholder for of X-123 in the third transformation. The Xtemp-123 radioisotope is then treated identical to X-123.

For this demonstration, the transformation rates are treated like what is currently done in HYSPLIT for pollutant conversions, where species #1 is transformed into species #2 at a certain fraction per hour. These rates assume a first-order reaction rate, which lends itself to a rate equation in the same format as radioactive decay. Chemical transformation reaction rates that depend on higher order reaction rates or other factors (e.g., humidity) would not be able to be represented in this manner. It is assumed for this demonstration that X transforms into XO at a rate of 30% per hour (giving a half-life of 116 minutes), XO transforms into XOH at a rate of 60% per hour (giving a half-life of 45 minutes) and XOH transforms back into X at a rate of 75% per hour (giving a half-life of 30 minutes). Radioactive decay must also be considered in parallel, and it is assumed the half-life of radioisotope X-123 is 4 hours and all the chemical forms of X decay into radioisotope Y-123.

A set of equations incorporating the above defined chemical transformations and radioactive decay in parallel is shown in Table 5-4. Hand calculations using the set of equations and rates defined in Table 5-4 were performed to demonstrate that this set of parallel chemical transformations and radioactive decay was reasonable. Figure 5-3 shows the calculated change in mass over time for each species using the Bateman equations. As a confirmatory comparison, the radioactive transformation of X-123 into Y-123 over time without the inclusion of chemical transformation was calculated to ensure conservation of material and is denoted in the figure as the curves labeled "X simp" and "Y simp". As can be seen in Figure 5-3, the sum of all X species over time and the generation of Y over time behaves as expected as these values are equal to X simp and Y simp respectively at all time steps. As can be seen with the set of equations shown in Table 5-4 and results in Figure 5-3, if such transformation information is available, there is a potential for the INDEXR.DAT file to include this information with some appropriate adjustments.

Table 5-4. Incorporating Chemical Transformation Into MACCS Example

Transformation	Rates and Fractions
X-123 → XO-123 (X_{R1}) + Y-123 ($1-X_{R1}$)	CT_1 → Half-life – 116 min (λ_{CT1} = 9.96E-05) $X_{R1} = \lambda_{CT1} / (\lambda_D+\lambda_{CT1})$ = 6.74E-01 Rate for CT_1 → Half-life – $\ln(2)/(\lambda_D+\lambda_{CT1})$ = 78.2 min
XO-123 → XOH-123(X_{R2}) + Y-123 ($1-X_{R2}$)	CT_2 → Half-life – 45 min (λ_{CT2} = 2.57E-04) $X_{R2} = \lambda_{CT2} / (\lambda_D+\lambda_{CT2})$ = 8.42E-01 Rate for CT_2 → Half-life – $\ln(2)/(\lambda_D+\lambda_{CT2})$ = 37.9 min
XOH-123 → Xtemp-123(X_{R3}) + Y-123($1-X_{R3}$)	CT_3 → Half-life – 30 min (λ_{CT3} = 4.33E-04) $X_{R3} = \lambda_{CT3} / (\lambda_D+\lambda_{CT3})$ = 8.89E-01 Rate for CT_3 → Half-life – $\ln(2)/(\lambda_D+\lambda_{CT3})$ = 26.7 min
Xtemp-123 → Y-123	Half-life – 4 hours (λ_D = 4.81E-05)

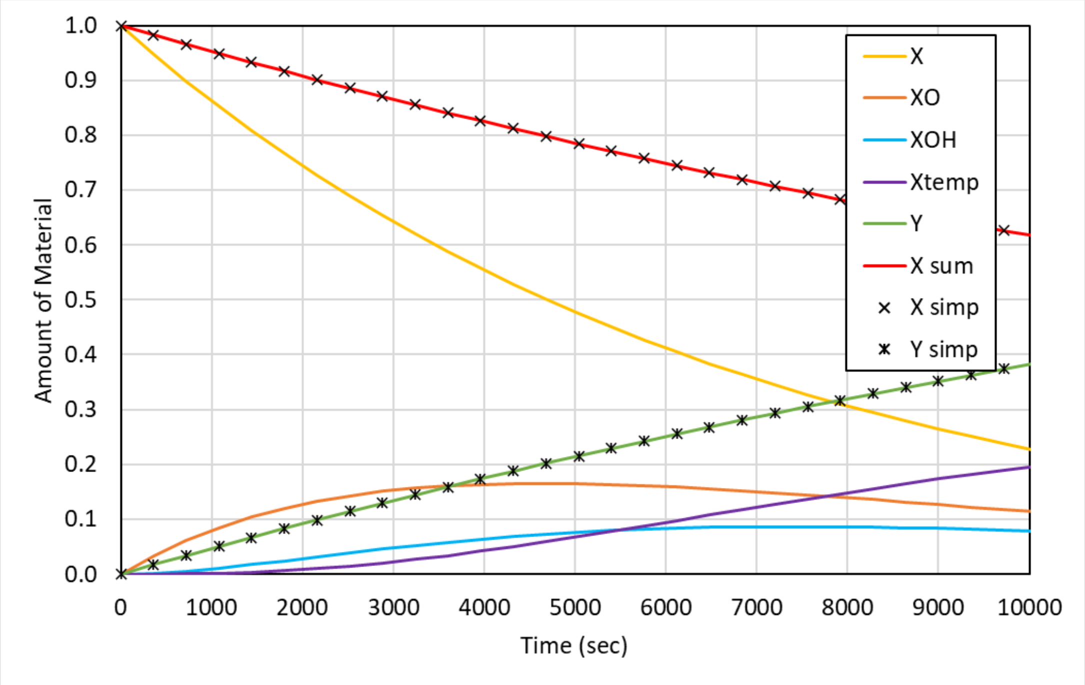

Figure 5-3. Accounting for Chemical and Radioactive Transformations Simultaneously Using the Bateman Equation Example

The equations and rates shown in Table 5-4 were then implemented into the INDEXR.DAT file to conduct a MACCS calculation. MACCS was then used to demonstrate the impact including chemical transformations can have on dose assessments, especially if deposition velocity and dose coefficient information varies significantly for the different physical/chemical forms of concern. Using the general MACCS set up described below and the representative deposition velocities and inhalation dose coefficients defined in Table 5-5, the peak dose is compared when chemical transformation is and is not included.

- Point source, ground-level, non-buoyant unit release of X-123
- Constant weather, 4 m/s wind speed, D stability, no rain
- 100 ppl/km^2 population density with no emergency response actions

Table 5-5. Dose Coefficient Information for Chemical Transformation Example

Chemical Form	Deposition Velocity	Effective dose coefficient: Inhalation, Adult (Sv/Bq)
X-123 (and Xtemp-123)	0.002 m/s	1.0E-09
XO-123	0.0002 m/s	2.0E-08
XOH-123	0.02 m/s	1.0E-08

Figure 5-4 shows the peak dose as a function of distance for the MACCS runs conducted with and without the chemical transformations. As can be seen in Figure 5-4 below, with the consideration of chemical transformation along with radioactive transformation as defined in Table 5-4, the peak dose output from MACCS is noticeably influenced given the deposition velocities and inhalation dose coefficients used above for each chemical form. This demonstrates that MACCS is potentially capable of including physical/chemical transformations within the current framework.

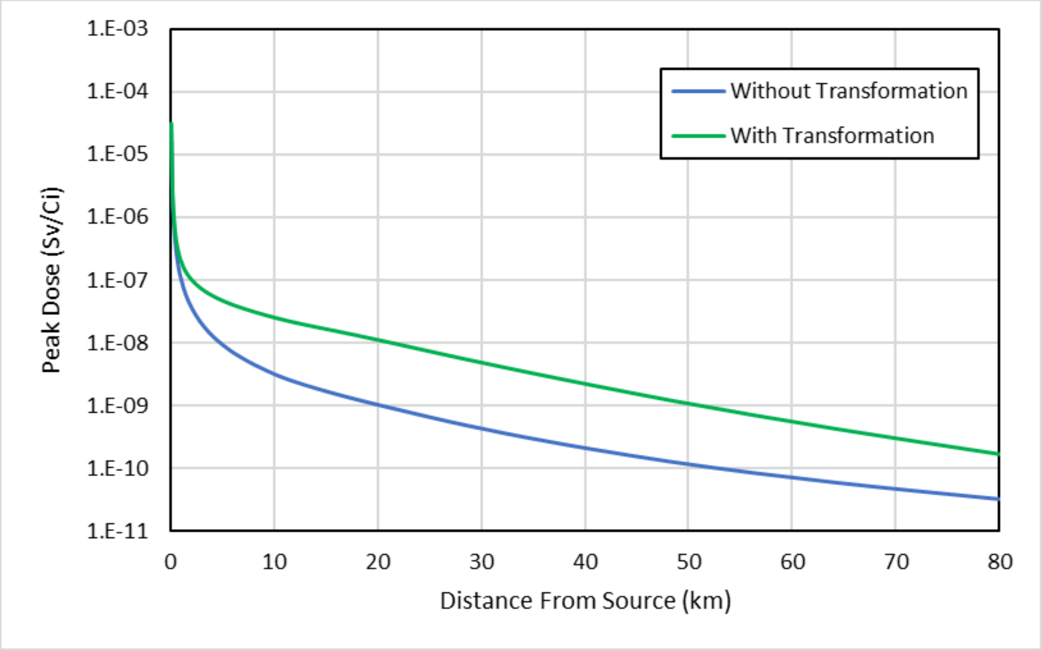

Figure 5-4. MACCS Peak Dose Output When Chemical Transformation is Included

However, while this demonstration was successful, as the situation gets more complex (more transformation reactions and/or more radionuclides to consider), it becomes harder to manage. Additionally, this is dependent on transformation rates in the form of a fraction or percentage per unit of time being available for such transformations. However, if such information were to become available in the future, a logical next step would be to investigate updating the current MACCS framework to allow for parallel physical/chemical and radioactive transformations to be a more user-friendly process.

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6. CONCLUSIONS AND RECOMMENDATIONS

To reiterate, the objective of this analysis was to identify physical and chemical transformation FEPs that may occur following a release of chemically reactive radioactive species and evaluate current MACCS capabilities for handling these transformations. Upon review of possible transformations, it is expected that physical and chemical transformation processes will mainly affect plume depletion (specifically particle size) and dosimetry. However, within the current state-of-practice, the inclusion of physical and chemical transformations in atmospheric transport and dispersion modeling is limited to chemical transport and only for specific radionuclides. Furthermore, a model capable of performing parallel physical/chemical and radioactive transformation was not found. As a result, there is no clear evidence suggesting that updates to the MACCS code are necessary. In a broader sense, incorporating generic (i.e., not species-specific) physical and chemical transformations for radionuclides appears to be beyond the current state-of-practice.

However, investigations discussed in Section 5 demonstrate the MACCS code architecture can accommodate multiple physical and chemical forms in a single simulation, as well as model the transformation of one form to another with some adjustments. However, it is crucial to reiterate that user modification to the INDEXR.DAT and DCF files should be approached with caution. This analysis does not generally recommend user modification of these files; the intent was solely to demonstrate that the MACCS code architecture is fundamentally flexible enough to handle both multiple physical and chemical forms in one simulation, as well as modeling the transformation of one form to another. Given this, a logical next step could include continuing the previous work described in [2] and updating the MACCS framework to give users the flexibility to model multiple physical and chemical forms in one simulation, such as iodine. Additionally, with some major assumptions, it was demonstrated that MACCS could simulate parallel physical/chemical and radioactive transformations using the Bateman equation and current set up of the INDEXR.DAT file. If reaction rate data for chemically reactive radioactive species becomes available, it would be worthwhile to investigate whether parallel physical/chemical and radioactive transformations can be implemented more feasibly. Lastly, alternative dry and wet deposition modeling techniques were identified during the investigation. Since MACCS currently models only particle deposition, while other models include both particles and gases, it may be worth exploring updates to MACCS.

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