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Qualitatively Reviewing MACCS Capabilities for Assessing Variable Radionuclide Physical/Chemical Forms and Tritium

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Outline

- Introduction to radionuclide chemical/physical forms
- The MACCS dosimetry and deposition models
- Dose coefficient development and MACCS dosimetry lineage
- Clearance class and f_1 development
- Deposition model state of practice
- Opportunities for additional research
- Tritium overview
- Tritium transformation processes
- Existing tritium models
- Recommendations for potential MACCS upgrades

In a nuclear accident scenario, it is possible that there are multiple chemical and physical forms of a given radionuclide released

- NRC non-LWR Vision and Strategy calls for an investigation into how MACCS handles radionuclide size, shape and chemical form in atmospheric and dosimetry calculations
  - Inform potential improvements to MACCS capabilities for atmospheric transport and dosimetry
- MACCS has robust, flexible modeling capabilities that are generally well-suited to accommodate diverse forms of a given isotope
  - Understanding existing capabilities will help to inform improvements
- It is pertinent to investigate MACCS variables relevant to chemical and physical form modeling and how MACCS functionality facilitates the analysis of accident consequences from varying forms of a given isotope
  - Look into dosimetry and transport assumptions relative to state of practice
  - Technology neutral fashion

The MACCS dosimetry model

- EARLY: cloudshine, groundshine, direct/resuspension inhalation and skin deposition
    - Difference dose coefficients for each pathway read from the DC file supplied with MACCS
    - Generally, dose is a product of exposure and dose coefficient
- CHRONC: MACCS additionally considers food and water ingestion (in addition to groundshine and resuspension inhalation doses)
    - Indirect late-phase doses from ingestion may occur in different spatial elements than where the deposition processes occurred
    - Food ingestion doses will depend on farmable land area and ground concentration
    - Water ingestion will depend on direct deposition and washoff to freshwater bodies (simple secondary transport equation)
- MACCS currently does not allow for multiple chemical forms for the same isotope
    - Each isotope of a given element is attributed the same dose coefficient (for a given organ/pathway)

The MACCS deposition model

- MACCS allows user to assign radionuclides to various high-level chemical groups based on physical and chemical properties that are assumed to be identical
  - Typically defined to be consistent with accident progression codes like MELCOR
- Assuming every isotope in a group behaves the same ignores the unique physical/chemical properties of radioactive molecules
  - Particle size
  - Transformation in the environment
  - Hygroscopicity
  - Agglomeration
  - Density
- Aerosol size distribution is used to assign a dry deposition velocity
  - Dry deposition a function of ground level air concentration and dry deposition velocity
  - Particle size distribution are binned and each assigned dry deposition velocity
- Wet deposition functions independently of particle size

Other summary observations regarding the MACCS conceptual models

- The dose calculation for a given radioisotope in MACCS assumes chemical form does not change following release to the atmosphere
- No secondary environmental transport is assumed after deposition
- Risk coefficient uncertainty can vary considerably across applications
  - E.g., inhalation risk may depend strongly on availability of information on the chemical/physical form inhaled
- FGR 13: "the biokinetic, dosimetric, and radiation risk models generally have been derived from much less detailed and sometimes inconsistent databases (with) substantial uncertainties"
- Default absorption types are recommended, but information regarding this selection is often limited and, in many cases, reflects occupational rather than environmental experience
- An analyst should consider the timescale in which chemical transformations are expected to happen
  - Environmental transformation processes happen slowly
  - E.g., is a given radionuclide expected to oxidize before substantially decaying?

Conceptual accounting for alternate physical/chemical forms in dose coefficient development

- EPA regularly publishes federal guidance reports to assist with radiation protection programs
  - FGR13 provides technical accounting for risk coefficients, dependent on age, gender, metabolism, dosimetry, radiogenic risk, and competing causes of death
  - A supplement to FGR provides the basis for dose coefficients in MACCS
  - Effective dose coefficients based on ICRP60 recommendations for tissue weighting
- Absorption type/clearance class in MACCS consistent with Runkle and Ostmeyer (1985), a study discussing dosimetric data for accidental radionuclide release from nuclear reactors
  - Runkle and Ostmeyer (1985): “the most probable chemical forms of the inhaled radionuclides are used to assign clearance classes…except for Cs and I isotopes, most of the important inhaled radionuclides will be in the form of insoluble compounds (principally oxides and hydroxides)”

More on the MACCS dosimetry lineage

- FGR 12 and ICRP 72 informed external and internal dose coefficients, respectively, included in FGR 13
  - External considered radionuclides with a half-life of at least 10 minutes or occurring in the decay chain of a radionuclide that does
  - Internal excluded radionuclides with half-life less than 10 minutes and isotopes of noble gases
  - No practical differences between ICRP72 dose coefficients and those used to developed FGR 13 for isotopes of interest for consequences analysis

Clearance class and f_1 development dates back to the 1950s

- ICRP 2 (1959) – provided a foundational, single compartment model for the lung to predict deposition, retention and clearance of inhaled aerosols
- ICRP 30 (1979) – supersedes ICRP 2, improved estimations of deposition in and clearance from the respiratory tract
  - 3 anatomical compartments were used: nasopharyngeal, tracheobronchial and pulmonary
  - Introduced the “D, W, Y” shorthand
- ICRP 66 (1994) – further revises ICRP 30 model and includes morphometry, respiratory physiology, radiation biology, deposition, clearance, and dosimetry
  - Changed the “D,W,Y” clearance class convention to “F,M,S”
  - Broad nature of clearance class timelines means the risk associated with some radionuclides may be over/underestimated (actual clearance times are on lower/upper end of clearance time bin)

Other relevant, newer documentation exists for dosimetry but are often associated with occupational intakes

- ICRP 68 supersedes ICRP 61 and provides dose coefficients for occupational intake of radionuclides
  - Used the lung model from ICRP 66
  - Tabulates effective dose coefficients for inhalation for varying isotopes, clearance classes, and particle sizes
  - Provides recommendations for clearance types and f_1 values for various chemical compounds
- Beginning in 2015, Occupational Intakes of Radionuclides (OIR) reports published by ICRP to replace the ICRP 30 series, ICRP 54, 68, and 78
  - Contains more up-to-date and detailed information than ICRP 68
  - Detailed information on chemical forms commonly encountered in an occupational setting and associated clearance class and f_1 values
  - An update to FGR 13 may logically include a reference to the OIR series

Other deposition models exist, but MACCS is consistent with state-of-practice specifically for modeling variable chemical/physical forms

- EPA AERMOD model includes algorithms for both dry and wet deposition for particulates and gaseous emissions and relies on the resistance model
  - Particulate deposition calculation method based on particle size
  - Wet deposition depends on washout coefficient and precipitation rates
- NOAA HYSPLIT uses either a user specified velocity or calculated using a known particle diameter, air density and particle density
  - Users can optionally select the resistance model
  - Wet deposition model assumes a scavenging ratio to account for rainout and washout (removal constant)
- Generally, the impact of chemical form on wet and dry deposition is not currently account for in any state-of-practice model for particulate deposition, including MACCS

Additional work may be warranted to produce data necessary to inform model improvements

- Identify whether non-LWR accident releases contain chemical forms other than insoluble oxide or hydroxide forms, and what those forms are
- Conduct a sensitivity analysis for dose coefficients for variable chemical forms
- Expand the dose coefficient file and allow the user to define which chemical form should be used
- Enhance MACCS to allow a user to specify release fractions for different chemical forms of the same isotope
- Review non-LWR accident progression analyses to determine whether significant gaseous releases are likely
- Benchmark the MACCS dry and wet deposition models against alternate state of practice models

Tritium has highly unique chemical behavior in the environment and may become important for advanced reactor consequence analyses

- If released in large enough quantities, it may be warranted to update MACCS capabilities to model tritium fate and transport more effectively in an accident scenario
- Updates may be informed by existing modeling capabilities from other tritium models
- MACCS has been used in previous studies to model tritium releases

Tritium behaves similarly to hydrogen in the environment and biological processes

- Weak beta emitter – primary radiological hazard is through ingestion of tritiated organic molecules
- Chemical form can heavily influence radiological risk posed by tritium
  - Inhaling gaseous tritium poses relatively limited radiological risk (low absorption, significant exhale). Dermal contact also limited
  - Ingestion and dermal contact with HTO poses a comparatively larger risk (high biological uptake)
  - Meaningful concentrations of HTO can also be absorbed through the skin at a rate approximately half that of inhalation
  - Can also bind to carbon through photosynthetic processes and create OBT

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

Complex environmental processes govern tritium transformation processes

- Gaseous tritium oxidation in the atmosphere (slow process)
  - Photochemical oxidation in the air
  - Isotopic exchange
  - Depends on gas mixture and tritium concentration
- Oxidation via soil microbes
  - Air to soil transfer governed by diffusion, depends on soil humidity and/or concentration gradients
  - Oxidation depends on soil type, moisture and temperature
- Transformation in plants
  - Tritium expected to naturally take part in the water cycle and thus subject to a great number of changing environmental conditions at any time and physiological processes of vegetation
  - Air to plant transfer depends on leaf exchange between vapor and free water in leaf organs
  - Timing of release (day vs. night) may strongly influence the transfer of tritium through plants
  - Eventually incorporated into plant organic matter (OBT)
- Transfer to animals
  - Animals ingest grass or other plants containing exchangeable OBT
  - Consider relocating animals to uncontaminated grass
  - Milk and meat likely not a major exposure pathway (fast biological turnover of HTO and OBT)

Existing tritium models

- UFOTRI – the most comprehensive model regarding tritium releases, dispersion, deposition and the subsequent movement and transformation through the environment
  - Similar Gaussian dispersion model to MACCS
  - Differentiates between different forms of tritium in the environment, has a detailed reemission physics model, accounts for the conversion of tritium to HTO, uptake by plants, and conversion into OBT
- GENII – well documented dose and risk assessment model developed by EPA
  - Utilizes special tritium models for acute and chronic exposures
  - Chronic module depends on hydrogen content of plant/animal being contaminated
  - Also accounts for OBT generation
- RSAC – Radiological Safety Analysis Computer program
  - Uses different equations for calculating ingestion doses from tritium
  - Assumed ratios of plant water and tritium concentration in plant water vs atmospheric water
- Other less documented/research models

Multiple pathways may exist for updates to MACCS to accommodate acute tritium releases

- The atmospheric transport processes are largely similar, but MACCS may benefit from updates to longer-term environmental process models
- Multiple pathways for updates:
  - 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. Reduces the need to have an exact accounting of the chemical form of tritium releases
  - Develop the capabilities for MACCS to identify multiple different chemical forms of tritium and associated transformation. Variable tiers of complexity for which this might be accomplished (e.g., incorporating a simple transfer rate model)
  - Introduce tritium accounting capabilities similar to those in UFOTRI and the more complex capabilities of other models. This would require substantial effort but would provide more detailed estimations of tritium in an accident scenario
- Consider examining advanced reactor inventories first to determine if model updates are worthwhile (i.e., is tritium even an issue?)

Thank you!

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