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Methodology for Selection of Proper Material Form for Specification of Inhalation Dose Conversion Factors at Savannah River Site

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Abstract

Consequence analysis of postulated accident releases requires that assumptions be made on the material form being released. This paper documents the general methodology for the selection of the proper material form (i.e., chemical compound) and its associated solubility category that is being used at Savannah River Site (SRS). The solubility characteristics of a radionuclide inhaled into the body directly influences the uptake and retention of that radionuclide, and consequently, the committed effective dose equivalent (CEDE). The CEDE per unit uptake is reflected in the inhalation dose conversion factor (DCF) of a given radionuclide. The methodology that is presented here provides a strategy that is used in SRS applications for specifying reasonably conservative inhalation DCFs based on consideration of chemical compound forms that are expected from SRS-based source terms arising from postulated accident conditions.

Introduction

The inhalation DCFs are key input data of the dosimetry calculations of MACCS2[1] and similar radiological consequence codes used for safety analysis applications. Sometimes, bounding default DCFs are used without questioning the validity of these DCFs to the inventory challenged by the accident condition being analyzed. The approach used at SRS recognizes that CEDEs are chemical compound-dependent and many radionuclides can have multiple inhalation DCF values depending on the chemical and physical nature of the radionuclide under consideration. Specifically, the methodology for DCF selection makes use of the characterization of most SRS source terms as being similar in chemistry to the inventory challenged by the accident condition. In general, this can be interpreted as material in oxide powder, oxide metal or nitrate solution forms.

The methodology at the SRS for specifying inhalation DCFs has been applied over recent years to the DCF database set from Federal Guidance Report (FGR) 11[2] and more recently to the set that is based on Publications 68 (Worker)[3] and 72 (General Public)[4] of the International Commission on Radiological Protection (ICRP). This paper will present the methodology in terms of the ICRP 68/72 DCFs, but the methodology presented here is easily adaptable to the FGR-11 based DCFs. The ICRP 68/72 inhalation DCFs are based on ICRP 1990

recommendations on radiation protection standards in Publication 60[5] as well as the revised kinetic and dosimetric model of the respiratory tract in Publication 66[6]. Since the issuance of ICRP Publications 68 and 72, the ICRP has issued a compact disc with a DCF database,[7] using the same models. The database gives both individual organ and effective dose coefficients. Additionally, the database gives the user greater flexibility by including inhalation DCFs for ten particle sizes.

Since plutonium in the source term is often the main contributor to dose, specifying the appropriate inhalation DCFs for plutonium radionuclides is especially important and thus given particular emphasis. Recent guidance from ICRP publications on plutonium lung absorption has focused on the various oxide forms of plutonium and the importance of the material's history, especially the method of oxide formation. High-fired plutonium oxide results in insoluble material form with a slower lung absorption rate than plutonium oxide that results from air oxidation at ambient temperatures.[8]

General Discussion

Solubility and Bodily Absorption

Chemical form of inhaled material determines solubility and subsequent transport behavior within the body. Three broad solubility categories have been defined for use with ICRP 68/72 inhalation DCFs and are expressed in terms of lung absorption types categorized as fast (F), moderate (M), and slow (S). These lung absorption types relate to the rate of absorption from the respiratory tract to body fluids as defined below:

- Lung Absorption Type F: Deposited materials that are readily absorbed into body fluids from the respiratory tract (Fast absorption).
- Lung Absorption Type M: Deposited materials that have intermediate rates of absorption into body fluids from the respiratory tract (Moderate absorption).
- Lung Absorption Type S: Deposited materials that are relatively insoluble in the respiratory tract (Slow absorption).

Association of Chemical Compound Forms and Lung Absorption Type

The assignment of lung absorption type assignments to the various chemical compound forms in which radionuclides can exist is given in Annexe F of ICRP Publication 68[3] (Table 3 of FGR 11 serves a similar function for the FGR-11 system[2]). Some of the common chemical compound forms explicitly addressed include oxides, hydroxides, nitrates, halides, carbides, sulphides, sulphates, and fluorides.

There are no apparent universal rules that govern the assignment of a specific chemical compound form (e.g., oxides) to a solubility category, so each radionuclide must be treated individually. For example in the ICRP-68/72 system, thorium oxide is assigned to lung absorption type S, while technetium oxide is assigned to lung absorption type M.[3] Furthermore,

a single lung absorption type may cover all chemical compound forms for a particular radionuclide (e.g., americium), while other radionuclides have chemical compound forms that span two lung absorption types (e.g., plutonium) or three (e.g., uranium) as shown by the examples below.[3]

- Americium compounds are all lung absorption type M.
- Plutonium compounds can be lung absorption type S (insoluble oxide) or lung absorption type M (all other plutonium compounds).
- Uranium compounds can be lung absorption type S (UO_2 and U_3O_8), lung absorption type M (UO_3, UF_4, and UCl_4), or lung absorption type F (UF_6, UO_2F_2, and $UO_2[NO_3]_2$).

Particle Size Effects

The particle size also affects material transport in the body and thus the inhalation DCF value. The ICRP Publication 68 gives inhalation DCFs that are based on both 1-μm and 5-μm activity median aerodynamic diameter (AMAD).[3] The AMAD signifies that fifty percent of the activity in the aerosol is associated with particles of aerodynamic diameter greater than the AMAD. The 1-μm and 5-μm AMAD particle sizes addressed in ICRP Publication 68 are broadly consistent with the particle-size range generally considered in accident analysis. Recommended airborne release fractions given in DOE-HDBK-3010-94 are based on 10-μm aerodynamic equivalent diameter (AED) and less.[9]

The ICRP 68/72 models predict greater deposition of larger particles in the upper respiratory tract, where the rate of absorption to the blood is relatively high.[10] As a result for highly soluble particles (i.e., lung absorption type F), the total predicted absorption to the blood tends to greater for the 5-μm particles in comparison with the 1-μm particles.[10] As an example, consider cesium for which all compound forms are categorized under lung absorption type F. The 5-μm AMAD inhalation DCF for ^{137}Cs is approximately 40% higher than that corresponding to 1 μm (i.e., 6.7E-09 Sv/Bq versus 4.8E-09 Sv/Bq).[3]

In contrast for particles that dissolve more slowly (lung absorption types M and S), the models predict that "a substantial portion of the deposited particles is cleared to the gastrointestinal tract by mechanical transport before there is sufficient dissolution to allow absorption of the radionuclide into blood".[10] The net effect tends to be lower inhalation DCFs for 5-μm particles in comparison with the 1-μm particles. Take for example ^{238}Pu and consider lung absorption type M. The 5-μm AMAD inhalation DCF is approximately 30% lower than that corresponding to 1 μm (i.e., 3.0E-05 Sv/Bq versus 4.3E-05 Sv/Bq).[3] Similar results are observed with other plutonium isotopes and thorium, americium, curium, and californium radionuclides that are characterized as lung absorption type M or S and which tend to have relatively high inhalation DCFs.

Practical Applications

Well Characterized Source Term Material

For a well characterized source term, specifying the appropriate inhalation DCFs is relatively straightforward following the recommendations given in Annexe F of ICRP Publication 68 for relating chemical compound forms to a lung absorption type for each radionuclide. For some radionuclides, a specific lung absorption type will be identified for the specific chemical form of interest, and the inhalation DCF associated with that lung absorption type is applicable. For other radionuclides, Annexe F of ICRP Publication 68 will not explicitly associate the chemical form of interest to a lung absorption type. In these cases, ICRP Publication 68 identifies a lung absorption type to be used for either *unspecified compounds* or *all compounds* that is appropriate.[3]

The ICRP Publication 72 gives inhalation DCFs based on particles of 1-μm AMAD.[4] For radiological exposures to members of the general public, ICRP Publication 71 recommends a particle size of 1-μm AMAD as a default value.[8] The older set of inhalation DCFs that are documented in FGR 11 are also based on the 1-μm AMAD particle size.[2]

The ICRP Publication 68 gives inhalation DCFs that are based on both 1-μm and 5-μm AMAD particle sizes.[3] The ICRP Publication 68 recommends a particle size of 5-μm AMAD as a default value for occupational exposures stating that 5-μm AMAD is generally more representative of workplace aerosols.[3] The inhalation DCFs based on 1-μm AMAD are also given since this smaller particle size has been shown from field measurements to be more appropriate "in some situations".[3] Thus, a well-characterized inventory of powder material will include information on particle size to guide the analyst in selection on the more appropriate set of inhalation DCFs to use for the worker consequence analysis. Note that the 1-μm AMAD DCFs from ICRP Publications 68 and 72 do not always exactly agree with one another based on differences in the models used for the worker and general public, respectively. The differences though are generally not that significant (e.g., 10% or less).

Particles of 10-μm AED and less particles are generally assumed to be released in unmitigated accident analysis,[9] for which either the 1-μm or 5-μm AMAD DCFs would be applicable. In a mitigated accident scenario in which active ventilation systems are credited, filters not only reduce the magnitude of the source term but also reduce the characteristic particle size that is released to the atmosphere. For these scenarios, inhalation DCFs that are based on a characteristic particle size much less than 1-μm (e.g., 0.1 μm) may be more appropriate. The extended database given in the compact disc issued by the ICRP gives inhalation DCFs for ten aerosol sizes (0.001 μm to 10 μm AMAD) using the ICRP 68/72 models.[7]

Typical SRS Application

Most radiological inventories at SRS exist in oxide powder, oxide metal or nitrate solution forms. For inventories that are well characterized, the approach summarized above generally applies. Even for well-characterized inventories, however, potential environmental effects associated with the some accident scenarios (e.g., fire, explosion, criticality) may introduce

uncertainty in the chemical form of the released material. Also, liquid solutions in some SRS facilities may change chemical form during processing so the consequence analysis needs to cover both material forms. A common situation in consequence analyses performed at SRS is to consider the possibility of both nitrate and oxide chemical compound forms in order to address the uncertainties in chemical form of the released material during postulated accidents.

For each radionuclide, a reasonably conservative DCF is selected for use based on considering the various lung absorption types that are represented by the nitrate and oxide chemical compound forms and selecting the maximum DCF from among the lung absorption types representing these forms. In order to implement this approach, five distinct cases must be addressed. These five cases and the particular strategy for handling each case are summarized in Table 1.

Table 1. Strategy for Source Terms that May be Either Nitrate or Oxide Chemical Forms.

Case	Case Description	Strategy
1	All chemical compounds of the radionuclide are assigned to a single lung absorption type.	The inhalation DCF is based on the single lung absorption type that is identified for all chemical compounds.
2	The oxide form and nitrate form are explicitly assigned to the same lung absorption type.	The inhalation DCF is based on the lung absorption type that is identified for the nitrate and oxide forms.
3	The oxide form is explicitly assigned to one lung absorption type and the nitrate form is explicitly assigned to a different lung absorption type.	The inhalation DCF is specified on the basis of the higher of the two inhalation DCFs considering the lung absorption type for the oxide and nitrate forms.
4	Either the oxide form or the nitrate form but not both are explicitly assigned to a lung absorption type and a different lung absorption type covers "unspecified compounds."	The inhalation DCF is specified on the basis of the higher of the two inhalation DCFs considering the lung absorption type for the oxide or nitrate forms and the lung absorption type for "unspecified compounds."
5	Neither the oxide form nor the nitrate form is explicitly assigned to a lung absorption type and more than one lung absorption type option is provided.	In absence of other information, the inhalation DCF is specified on the basis of the highest inhalation DCF among those associated with a listed lung absorption type in Annexe F of ICRP Publication 68.

Ill Characterized Inventory

When little information is available on the chemical form of the inventory, a bounding set of default inhalation DCFs is appropriate. The method for specifying the inhalation DCF for each radionuclide is the same as the strategy listed for Case 5 in Table 1. Specifically, the inhalation DCF is specified on the basis of the highest inhalation DCF among those associated with a listed lung absorption type in Annexe F of ICRP Publication 68 for the given radionuclide.

Plutonium

Since plutonium in the source term is often the main contributor to dose, specifying the appropriate inhalation DCFs for plutonium radionuclides is especially important and thus given particular emphasis. In previous ICRP guidance, all plutonium oxide forms were assigned to the same solubility category (least soluble category).[8] Annexe F of ICRP Publication 68 makes a subtle, but important, distinction in the assignment of plutonium oxide material to a lung absorption type. Recent guidance from ICRP publications on plutonium lung absorption has focused on the various oxide forms of plutonium and the importance of the material's history, especially the method of oxide formation. High-fired plutonium oxide results in insoluble material form with a slower lung absorption rate than plutonium oxide that results from air oxidation at ambient temperatures.[8] Thus, only *insoluble* oxide is assigned to lung absorption type S, and all other plutonium compounds are assigned to lung absorption type M.[3] Incomplete oxidation seems to be the contributing factor for the plutonium oxide that forms at ambient temperatures being more soluble and having absorption characteristics representative of lung absorption type M.[8] Complete oxidation occurs at high temperatures (~ 1000 °C and above for studies cited in ICRP 71) and results in lung absorption S[8] with a lower inhalation DCF[3,4].

When specific knowledge of the origin of the plutonium oxide in the source term is lacking, or it is not clear how environmental effects (e.g., fire, explosion, criticality) from the assumed accident condition will affect the material-at-risk, the analyst should conservatively assume the soluble oxide form and use the higher inhalation DCFs that are associated with lung absorption type M.

Concluding Remarks

The methodology at the SRS for specifying inhalation DCFs is summarized in this paper. Uncertainties in chemical form that result from either processing uncertainties or accident-related transformations are considered. For each radionuclide, a reasonably conservative DCF is selected for use based on considering the various lung absorption types that are represented by possible chemical compound forms and selecting the maximum DCF from among the lung absorption types representing these forms. Following recent ICRP guidance, the assignment of plutonium oxide material to a lung absorption type considers material's history as it relates to the method of oxide formation.

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