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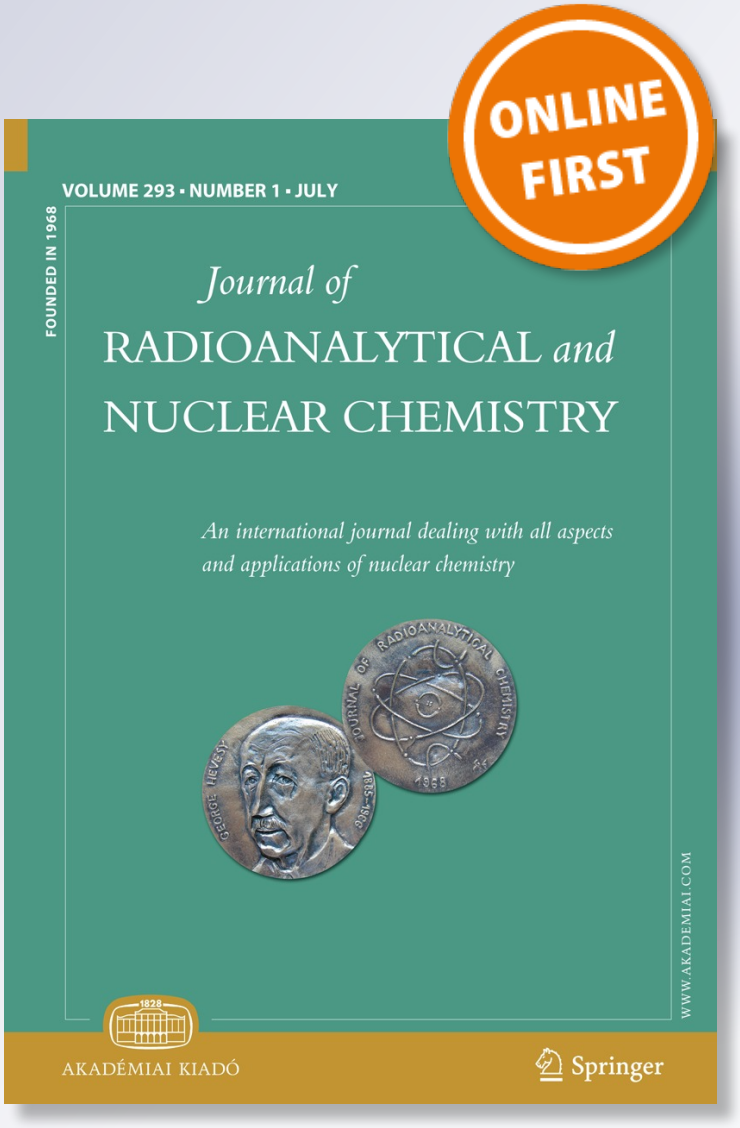

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Abstract Default values for the solubility of various compounds in the lung are provided in publications of the International Commission on Radiological Protection as absorption types to characterizes the potential uptake of radionuclides to blood. The default assignments are conservative and reflect compounds likely to be encountered in the workplace. In practice, solubility profiles for many compounds, both natural and man-made, are complex, with a fraction of the compound in each absorption type, denoted as F, M, or S. Only soluble compounds of tritium and iodine can be reasonably assumed to be of one absorption type. The assumption of a single absorption type for airborne distributions of solid particulate matter can introduce order of magnitude errors in internal dosimetry calculations. The problem is particularly acute for isotopes with dual toxicity (e.g. uranium which is both nephrotoxic and radiotoxic), and when a dose estimate must be derived with only a single bioassay measurement. For inhalation exposures during an accident, treatment decisions frequently must be made quickly to be effective. While much work has been done to develop rapid bioassay methods that will provide data in a clinically useable timeframe, little consideration has been given to the magnitude of the error in the dose estimate resulting from the assumption of the default solubility profiles.

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Introduction

Clearance from the lung of radionuclides attached to inhaled particulate matter occurs through mechanical transport of the deposited particles to the gastrointestinal tract, to lymph nodes and absorption to blood following dissociation of the radionuclide from the particles. The clearance processes act in a competing manner, however absorption of the soluble fraction of the aerosol dominate the early kinetics and accounts for the bulk of the activity of the radionuclide appearing in an early urine samples [1].

In the aftermath of a radiological event resulting in exposure to dispersing radioactive aerosol, medical treatment decisions must be made quickly for maximum efficacy of treatment protocols seeking to reduce the radiation dose to exposed individuals. This time constraint limits the type and number of bioassay samples obtained upon which to base an estimate of the intake and projected dose to the exposed individual. Clinical decisions regarding treatment are generally based on the projected dose estimate for the exposed individuals [2, 3].

Internal dosimetry calculations must be performed to estimate the intake and projected dose for the radionuclide(s) of the aerosol based on their measurements within the exposed individual or in excreta. The routes of intake, distribution within the body and subsequent excretion of a radionuclide are shown in Fig. 1. DCAL [4] and IMBA [5] are two popular software products that implement the International Commission on Radiological Protection (ICRP) Publication 66 lung model [6], see Fig. 2, models describing the distribution among the systemic organs, and

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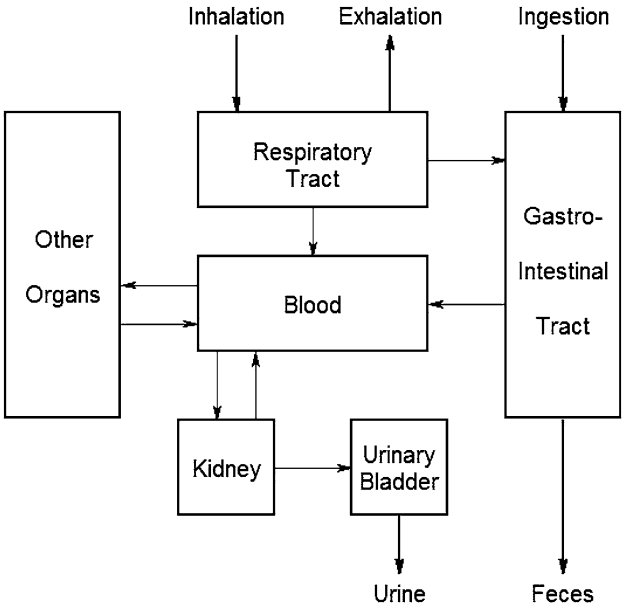

Fig. 1 Schematic of the routes of intake, internal distribution, and elimination. The respiratory and gastrointestinal tracts are the routes of entry from which uptake to blood with subsequent distribution among organs and tissues prior to elimination. The complexity of the various kinetic models varies greatly; e.g., tritiated water is rather uniformly distributed in the body while radioiodine is mainly associated with the thyroid gland

the dosimetric models of ICRP Publication 68 [1]. The computational procedures require that assumptions be made regarding the absorption of the inhaled aerosol. The ICRP lung model [6] defines three absorption types to characterize the absorption to blood of the inhaled radionuclide from particulates deposited in the lung. The transfer coefficient define the fractional rate of transfer of the radionuclide from a lung region to blood is expressed as a half-time. Mechanical clearance is an additional clearance mechanism however it is independent of the solubility of the inhaled particulates; e.g., in the ciliated regions of the tracheobronchiolar region it is related to the rate of mucus clearance. The ICRP lung model assigns the absorption process of the particulate matter to one of three default absorption types: Type F (fast dissolution and a high level of absorption to blood), Type M (a moderate rate of dissolution and level of absorption to blood), and Type S (a slow dissolution and low level of absorption to blood). Absorption is viewed as a two stage process where the radionuclide is freed (referred to as dissolution) from the particulate matrix and taken up by the blood. For particulates characterized as Type F the absorption rate corresponding to a half-time of 10 min; Type M the absorption is biphasic, 10 % absorbed with a 10 min half-time and 90 % with a 140 day half-time; and Type S absorption is also biphasic, 0.1 % absorbed with a 10 min half-time and 99.9 % with a 7,000 day half-time.

The uptake to blood of a radionuclide inhaled as a particulate depends on the chemical form of the matrix in which the radionuclide is bound. With the exception of a few radionuclide; e.g., tritium, the chemical form is rarely known. Consequently default absorption Types (F, M, or S) are normally assigned in a conservative manner when deriving radiation protection guidelines. However these assignments, while yielding conservative estimates of the dose per unit inhaled activity, underestimate the

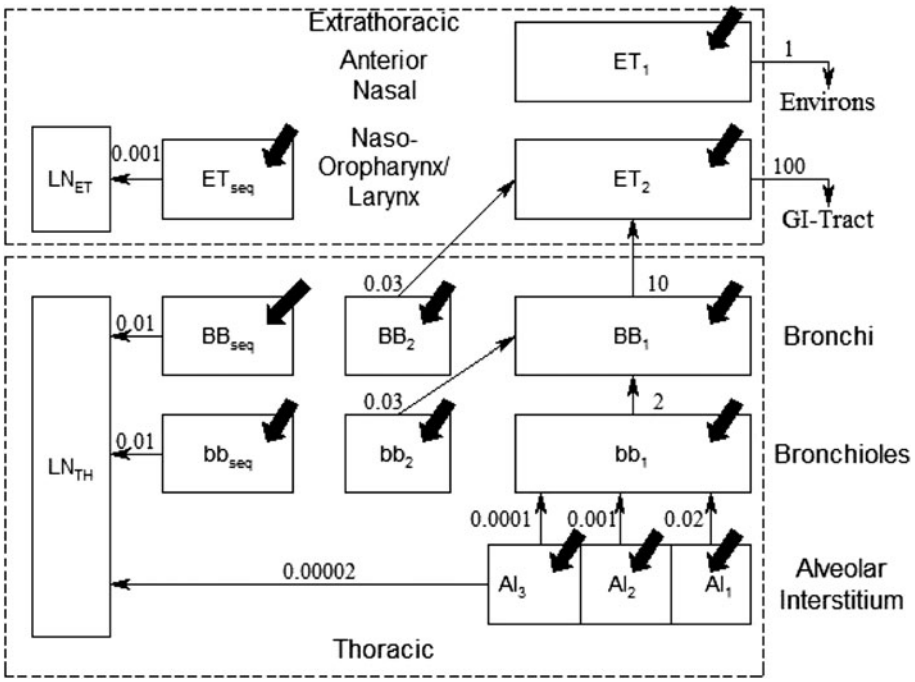

Fig. 2 Compartment model representing the time-dependent transport from each region of the ICRP Publication 66 respiratory tract model. The *thick arrows* denote the regions of initial aerosol deposition and the *thin arrows* denote the mechanical transfer; the numerical value being the rate (d^{-1}). The compartments with a label footnoted by *seq* are regions of prolonged retention within the airways. Uptake by blood (absorption) is assumed to occur from all compartments except ET_1

Table 1 Default and measured solubility profiles for various compounds

Product	Default solubility type	Actual solubility type (%)		
		Type F	Type M	Type S
Uranium metal—recycling smelting	100 % S	2	0	98
DU munitions	100 % S	23	0	77
DU munitions after heat treatment	100 % S	5	0	95
UO_2 from in situ mine	100 % S	76	24	0
PuO_2 (1,750 °C)	100 % S	4.6	0	95.4
AmO_2 (1,750 °C)	100 % S	6.7	0	93.3
U_3O_8 (High fired—Exxon)	100 % S	25.5	0	74.5

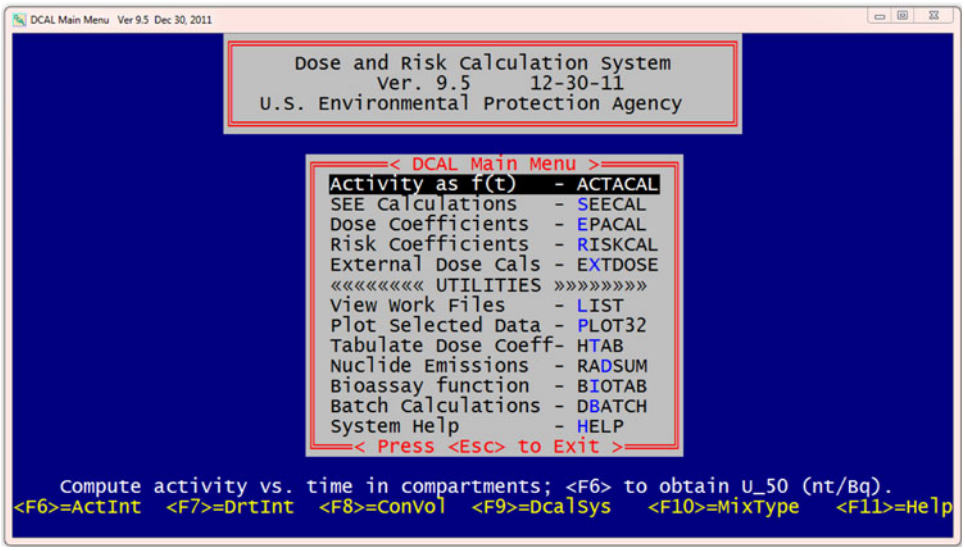

Fig. 3 The main menu of the DCAL software used in the calculations. The MixType module invoked by the F10 key enables calculations for an inhaled aerosol based on a mixing of the absorption Types F, M, and S

elimination of the radionuclide from the body. Use of the default absorption type when estimating inhaled activity based on a urine bioassay sample can result in a systematic bias which is not desirable as decisions regarding treatment options themselves may entail risk. A realistic, rather than conservative estimate of dose and risk are desirable.

Experimental

Measured solubility profiles for seven compounds were drawn from the literature [7–9] and used to estimate the inhaled activity intake and projected dose indicated by a single urine bioassay sample containing 1 Bq/mL of the radionuclide in urine at 24 h after an acute exposure. The aerosol was taken to have a lognormal particle size distribution corresponding to an activity median aerodynamic diameter (AMAD) of 5 μ (geometric standard deviation of 2.5). The fractional solubility profiles from the literature, see Table 1, were input into the DCAL software package [4] to derive an estimate of intake and dose corresponding to the assumed urine measurement. The software was modified to include an additional module enabling calculations involving solubility profiles expressed as a linear combination of the ICRP absorption types. DCAL's main menu is shown in Fig. 3. To illustrate the computational procedure, consider the first product listed in Table 1; i.e., *Uranium Metal-Recycling Smelting*:

1. DCAL's ACTACAL module is invoked for each absorption type of the solubility profile, for this product Type F and S, to derive the time dependent content in each compartment of the mathematical models describing the fate following a unit activity intake.
2. Invoke the MixType module by pressing the <F10> key and enter the fraction assigned to each absorption type as requested; i.e., for this product 0.02 is of Type F and 0.98 is Type S. This module creates a file of the time-dependent content of the model compartment as the weighted sum of the content of each absorption type. The resultant file is named following, DCAL's

Table 2 Comparison of estimated intake for default and actual solubility profiles based on a spot urine bioassay sample at 24 h of 1 Bq/mL

Product	Estimated intake (Bq)		Intake ratio Default:actual
	Default profile	Actual profile	
Uranium metal—recycling smelting	8.70E + 06	3.17E + 06	2.74
DU munitions	8.70E + 06	4.17E + 05	20.9
DU munitions after heat treatment	8.70E + 06	1.64E + 06	5.30
UO_2 from in situ mine	8.70E + 06	1.25E + 07	69.6
PuO_2 (1,750 °C)	6.76E + 08	1.50E + 07	44.9
AmO_2 (1,750 °C)	1.63E + 08	5.41E + 06	30.1
U_3O_8 (High fired—Exxon)	8.70E + 06	3.77E + 05	23.0

Table 3 Comparison of estimated dose for default and actual solubility profiles based on a spot urine bioassay sample at 24 h of 1 Bq/mL

Product	Dose (Sv)		Dose ratio Default:actual
	Default profile	Actual profile	
Uranium metal—recycling smelting	49.8	17.9	1.79
DU munitions	49.8	1.9	26.3
DU munitions after heat treatment	49.8	8.97	5.56
UO_2 from in situ mine	49.8	0.105	472
PuO_2 (1,750 °C)	5700.	220	26.0
AmO_2 (1,750 °C)	1400	93.8	16.7
U_3O_8 (High fired—Exxon)	49.8	1.67	29.9

naming convention, with *Z* designating the absorption type.

3. All DCAL modules, other than ACTCAL, can be invoked to complete calculations for the solubility profile; e.g., invoke in sequence SEECAL, EPACAL and HTAB to derive organ-specific dose coefficients and BIOTAB to tabulate the expected urinary and fecal excretion and total body retention as a function of time.

For illustrative purposes we assume a spot-urine sample of 200 mL obtained 24 h post an acute inhalation intake reflecting urine production over the preceding 4 h period. Intake and dose estimates for the default absorption type and for the solubility profile are given in Tables 2 and 3, respectively.

Discussion

With the exception of tritium and cesium, which can generally be safely assumed to be 100 % Type F, the compounds of most other radionuclides have complex solubility profiles with a fraction in more than one absorption type. This is true even of chemical forms that are normally considered to be highly insoluble (e.g. PuO_2). To illustrate this point, the retention of high-fired PuO_2 in the lung with the default solubility profile of 100 % Class S, and the more realistic value of 4.6 % Class F and the remainder Class S, is shown in Fig. 4. The daily fraction excreted for these same compounds is shown in Fig. 5. In

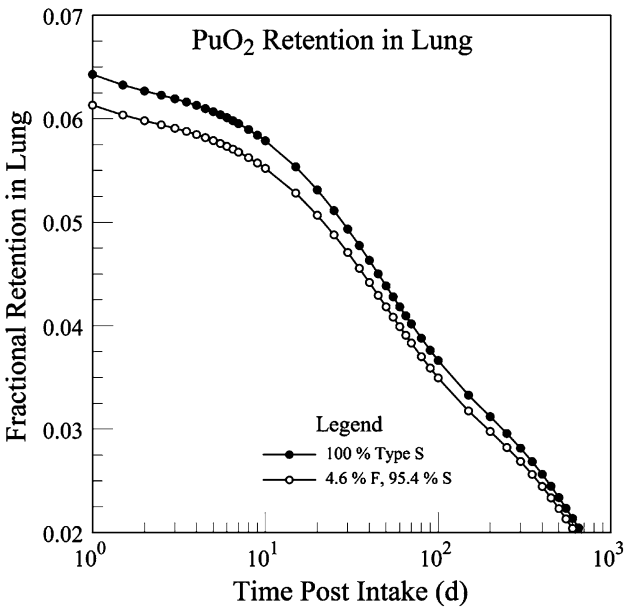

Fig. 4 The retention in the lung (thorax) of inhaled PuO_2 as a Type S aerosol and as an aerosol of mixed solubility profile (4.6 % Type F and 95.4 % Type S). The inhaled activity particle size distribution characterized by an AMAD of 5 μ

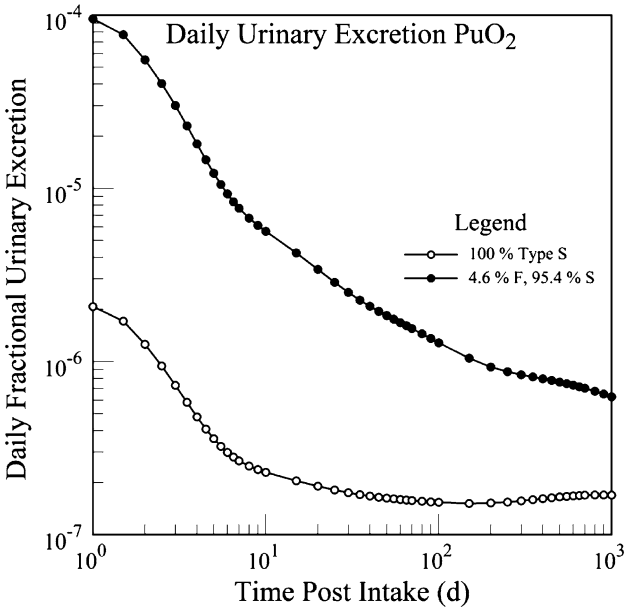

Fig. 5 The daily fraction of the inhaled Pu excreted in urine assuming a Type S aerosol and an aerosol of mixed solubility profile (4.6 % Type F and 95.4 % Type S)

these profiles, the fraction in Type F is the main determinant of the activity in the early urine bioassay samples. Failure to estimate the fraction of the compound in this absorption type can lead to order-of-magnitude errors in the intake and dose assessment (see Tables 2, 3) when only early bioassay data is available. The dose estimate based on a single urine sample while uncertain will normally bias high if the default absorption type is used in the analysis. For uranium compounds, which are nephrotoxic in their soluble forms, but radiotoxic in the insoluble forms, the error will inevitably overestimate one risk and underestimate the other. Note that the lung model plays a role twice in the analysis; once to determine the intake, and a second time to estimate in the dose projection. Thus errors in the lung model compound.

Conclusion

Fractional solubility profiles should be used for all compounds of the inhaled radionuclide not known to be readily soluble in the lung. Where no measured solubility profiles exist for the compound or no information is available on the chemical nature of the aerosol, efforts should be made to at least estimate the fraction of dust might be Type F, as this is one of the key factors in the accurate analysis of early urine bioassay data.

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