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Whole-organism concentration ratios for plutonium in wildlife from past US nuclear research data

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ABSTRACT

Whole-organism concentration ratios ($CR_{wo\text{-}media}$) for plutonium (Pu) in wildlife were calculated using data from the broad range of organism types and environmental settings of the US nuclear research program. Original sources included site-specific reports and scientific journal articles typically from 1960s to 80s research. Most of the calculated $CR_{wo\text{-}media}$ values are new to existing data sets, and, for some wildlife categories, serve to fill gaps or add to sparse data including those for terrestrial reptile; freshwater bird, crustacean and zooplankton; and marine crustacean and zooplankton. Ratios of Pu concentration in the whole-organism to that in specific tissues and organs are provided here for a range of freshwater and marine fish. The $CR_{wo\text{-}media}$ values in fish living in liquid discharge ponds were two orders of magnitude higher than those for similar species living in lakes receiving Pu from atmospheric fallout, suggesting the physico-chemical form of the source Pu can dominate over other factors related to transfer, such as organism size and feeding behavior. Small rodent data indicated one to two order of magnitude increases when carcass, pelt, and gastrointestinal tract were included together in the whole-organism calculation compared to that for carcass alone. Only 4% of Pu resided in the carcass of small rodents compared to 75% in the gastrointestinal tract and 21% in the pelt.

1. Introduction

Plutonium (Pu) can be important in dose calculation due to the relatively high energy (~5 MeV) of the alpha radiation emitted from most Pu isotopes. However, because of the short range of the alpha particle, valid estimation of dose to living organisms from Pu requires quantification of the amount of Pu that is physically internalized within an organism's body. For wildlife that live in Pu-contaminated environments, the amount of internalized Pu can be estimated from concentrations in soil, water, and, in some cases, air, through the concept of concentration ratios (CRs) defined as the concentration in the organism compared with the concentration in a host media (Beresford, 2010; Hosseini et al., 2008). Current biota dose assessment models utilize whole-organism (fresh mass basis) concentration ratios (Beresford et al., 2008), designated as $CR_{wo\text{-}media}$ ($CR_{wo\text{-}soil}$ relative to soil and $CR_{wo\text{-}water}$ relative to water).

Currently available compilations of $CR_{wo\text{-}media}$ values indicate uneven coverage for Pu-uptake data in various categories of wildlife. For example, $CR_{wo\text{-}media}$ values for terrestrial mammals and invertebrates are numerous, while those for freshwater bird, crustacean, and reptile are sparse or non-existent (Beresford et al., 2008; Wood et al., 2010). In general, for those categories where data exist, there can be disproportionate representation of some species, and of some environments, particularly where studies in the past were focused on the human food-chain (Howard et al., 2012).

A number of approaches have been suggested to address these data limitations, including new empirical studies, and mechanistic model-based approaches such as the kinetic-allometric approach in Higley (2010). The calculation of $CR_{wo\text{-}media}$ values can also be accomplished utilizing past studies which contain valid data on radionuclide uptake in biota, but that have not been widely accessed for biota dose purposes. For example, Fesenko et al. (2010) have published transfer data derived from former Soviet Union sources that had previously been difficult to access by the international community.

One source yet to be fully utilized are the data from research and testing reports associated with past United States nuclear programs which included activities involving Pu production, research, and weapons testing dating from the 1940s. The US nuclear program was extensive and was conducted in a variety of ecosystem types including semi-arid desert (e.g., Nevada Test Site), humid sub-tropical (e.g., Savannah River Site) and tropical marine (e.g., Marshall Islands). It therefore contains Pu data sets covering

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a broad range of wildlife in a variety of environmental conditions. Many of these data sets have been reported in scientific literature (e.g., Whicker and Shultz, 1982). However, many data have remained in site-specific reports, or rarely-referenced journal articles, and have not yet been used in to development of the whole-organism concentration ratios that have been widely published. This appears to be in part due to a tendency toward focusing on human-dose assessment in the past, which presented data in a form not directly useable in studies on non-human biota. Also, the site-specific reports containing the data were previously difficult to access in the paper-based systems of the past, but are now more readily available electronically.

The main objective of this paper is to report on the process and results of calculating $CR_{wo\text{-}media}$ values for Pu in wildlife using data from the US nuclear research program. These data sources have not been widely accessed for the purposes of assessing doses to biota. As they represent a broad range of organism types and environmental settings, these sources are expected to address gaps in key current publications and databases, and bolster the range of parameters available to current biota dose assessment software such as ERICA-Tool (Brown et al., 2008; Beresford et al., 2008; http://www.erica-tool.com) and RESRAD-BIOTA (USDOE, 2004; http://web.anl.gov/resrad/home2/biota.cfm). We present and discuss the process used for calculating the $CR_{wo\text{-}media}$ values from disparate data types, and, as a secondary objective, draw comparisons among values from differing study locations and conditions.

2. Materials and methods

2.1. Source locations and ecosystem types

Source documents were accessed through Argonne and Savannah River National Laboratories, through document services provided by the US Department of Energy (USDOE, Information Bridge) and through scientific journals. Many of the documents were not readily accessible through typical means of locating scientific research documents. Primary sources, including site-specific reports, were used which served to provide information on data provenance, laboratory methods, and quality assurance methods, and were examined even in cases where the data were summarized in subsequent publications.

Few data were in directly-usable formats. All data required review regarding provenance and utility. Most data required conversion, or further calculation, to obtain the $CR_{wo\text{-}soil}$ (terrestrial) and $CR_{wo\text{-}water}$ (aquatic) values presented here. In calculating $CR_{wo\text{-}media}$ values on a fresh mass basis from the disparate data sources, typical issues encountered were tissue concentrations/CRs provided in dry mass instead of fresh mass, $CR_{wo\text{-}media}$ values provided for tissues/organs instead of on a whole-organism basis, and ambiguously-related organism and media concentrations. In these cases, the whole-organism $CR_{wo\text{-}media}$ values were calculated using the following process.

When data were reported on a dry-, or ash- basis, instead of fresh-mass basis, the necessary conversions were made using the following (abbreviated codes used in the tables of this paper):

FW:DW(a) fresh:dry mass ratios as reported in original source;

FW:DW(b) fresh:dry mass ratios for terrestrial systems (e.g., 0.3-small mammal whole-body, 0.8-mammal bone, 0.25 detritivorous invertebrate) as reported in Beresford et al. (2008), or ratios for aquatic systems (e.g., 0.21 mollusc and crustacean, 0.18 marine biota) as reported in Hosseini et al. (2008);

FW:DW(c) fresh:dry mass ratios from the other available publications as noted;

FW:ASH fresh:ash mass ratios for small mammals (e.g., 0.013-muscle, 0.017-organs, 0.029-GI tract, 0.14-bone, 0.03-skin/pelt) derived from data on 19 rabbits (*Lepus californicus*) reported by Smith et al. (1978).

When data were reported for specific tissues or organs, instead of whole-organism, a conversion was made to a whole-organism basis using whole-organism:tissue factors (e.g., ratio of whole-organism concentrations to tissue-specific concentrations as described in Yankovich et al., 2010) in the following decreasing order of priority:

WO:T(a) whole-organism:tissue factors from reference study data were used when reported;

WO:T(b) whole-organism:tissue factors from Yankovich et al. (2010) or Hosseini et al. (2008);

WO:T(c) whole-organism:tissue factors reported in other publications, or author's unpublished work as noted.

The *n* values used for summary statistics represent the number of calculated $CR_{wo\text{-}media}$ values. In some instances the *n* values were reported for pooled samples (multiple specimens in one analysis sample), or dissected samples (multiple tissue samples analyzed for one specimen sample).

Original data reported for different Pu isotopes (^{238}Pu, $^{239,240}Pu$, etc.) were assumed to have sufficiently similar physico-chemical qualities to be considered as being within the same population when deriving summary statistics. For example, if both ^{238}Pu and $^{239,240}Pu$ data were available, values were calculated for each then averaged to arrive at a $CR_{wo\text{-}media}$ value for a given specimen.

Data from separate studies were not combined in this analysis to allow each study data set to be examined and reported on its own merit and with respect to its particular environmental conditions. The resulting $CR_{wo\text{-}media}$ values were grouped according to wildlife group categories in the Wildlife Transfer Parameter Database (www.wildlifetransferdatabase.org). Geometric means (GM) and geometric standard deviation (GMSD) were calculated using the approximation equations in Howard et al. (2012), and GM is not reported where $n < 3$.

2.2. QA screening process

Original data were included in calculating $CR_{wo\text{-}media}$ values when the following criteria were met:

- Documentation of sample collection, preparation, and analysis method were described and conformed to generally accepted practice.
- Wildlife organisms were not constrained to laboratory enclosures or mesocosms (e.g., mesocosm studies at a pond at Hanford, Washington State, were not included).
- Sampling of host media (water, soil) was representative of home range of the sample organism (several studies were rejected for using generic concentration factors from the US Environmental Protection Agency)
- The period of time of organism exposure to Pu contamination was generally consistent with other wildlife CR studies that make use of the equilibrium assumption (e.g., studies on fish and waterfowl with short exposure periods were rejected).
- Pooled samples were for the same or closely-related species (e.g., data were rejected when tissues from lizards + rodents were combined). However, zooplankton was considered without species identification.

3. Results and discussion

3.1. Calculated $CR_{wo\text{-}media}$ values

Values of $CR_{wo\text{-}media}$ for Pu uptake were determined in seventy-two wildlife types, primarily at the species level, from twenty-one