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Mass balance approach to estimating radionuclide loads and concentrations in edible fish tissues using stable analogues

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ABSTRACT

Humans can consume a number of types of biota tissues, which have varying propensities to accumulate radionuclides. As a result, depending upon the biota species, the radionuclide and the tissue under consideration, it may be necessary to estimate the percent radionuclide load in specific edible tissues, and in cases where whole organisms are consumed, to estimate the radionuclide load in the whole body of an organism, based on data that have been collected for individual tissues. To accomplish this, data were compiled that can be used to estimate the partitioning patterns and percent loads of various groups of elements in edible tissues of freshwater fishes. General trends in partitioning, such as those provided in this paper, can be used to predict radionuclide transfer to humans and the corresponding potential radiological dose to humans via dietary pathways, in this case following the consumption of fish.

1. Introduction

Humans can consume a number of different types of biota tissues, which have varying tendencies to accumulate radionuclides (Livingston and Livingston, 1993; Yankovich and Beaton, 2000). For example, in the case of fishes, which can represent an important part of human's diets, depending upon the species of fish and the cultural habits of the individuals consuming the fish, muscle tissue or whole fish may be eaten (Zach et al., 1996). As a result, the propensity of a given radionuclide to accumulate in a given edible tissue can dictate the potential radiological dose received by humans through dietary pathways. A notable example is the partitioning of strontium-90 (^{90}Sr) and other bone-seeking calcium analogues in hard tissues (Ophel and Judd, 1967; Ophel et al., 1971; Yankovich, 2002). Under such circumstances, consumption of whole fishes from areas receiving ^{90}Sr inputs could lead to substantially higher internal doses to humans than would be expected if only fish muscle were being consumed. However, many routine environmental monitoring programs (REMPs) have been designed to measure radionuclides only in muscle tissue of edible species, and as a result, literature reviews of radionuclide transfer to edible fishes have often focused on transfer from water to fish muscle (e.g., IAEA, 1994).

By comparison, from the perspective of radiological protection of non-human species, concentration ratios (CRs) are typically compiled on the basis of whole-body activity concentrations relative to those in surface waters (e.g., Hosseini et al., 2008), with focus on reference organisms. Additional literature data may also be available for other tissues that have been collected for a different purpose, but that could be used to estimate concentrations in edible tissues. Therefore, by developing of a standardized approach to estimate radionuclide levels in edible tissues based on measurements taken for other tissue types, such data could be used in the assessment of radionuclide transfer to human dietary items. Such an approach was documented in the revised International Atomic Energy Agency (IAEA) Technical Report Series (TRS) 364 document on radionuclide transfer to humans in terrestrial and freshwater environments, and the corresponding technical document (TECDOC) (IAEA, in press-a, in press-b). In doing so, literature data relating tissue biomass to organism fresh weight were compiled for edible fishes along with data reflecting partitioning patterns of nuclides (both radionuclides and their stable analogues)

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Table 1
Predictive relationships between whole body weight (in kg fresh weight), *X*, and tissue weight (in g fresh weight) for teleost fishes, *Y* (Crile and Quiring, 1940; Muir and Hughes, 1969; Quiring, 1950; Reynolds and Karlotski, 1977; Yankovich, 2002).

Tissue type	Relationship between whole body weight and tissue biomass	Tissue-to-body weight (%) geomean (Min.–Max.) [*n*]
Bone	$Y = 40.68X^{1.03}$ ($r^2 = 0.992$)	4.71 (2.34–9.1) [17]
Gills	n.a.	1.3 (0.7–1.8) [4]
Scales	n.a.	7.0 [1]
Brain	$Y = 0.960X^{0.504}$ ($r^2 = 0.747$)	0.087 (7.02×10^{-5}–2.29) [183]
Eyes	$Y = 5.36X^{0.76}$ ($r^2 = 0.727$)	0.504 (0.034–1.65) [174]
Gizzard	n.a.	1.80 (1.8, 1.8) [2]
Gizzard Contents	n.a.	0.242 (0.03–0.7) [5]
Gonads (female)	$Y = 3.67X^{0.729}$ ($r^2 = 0.340$)	1.53 (0.040–6.41) [39]
Gonads (male)	$Y = 2.03X^{1.13}$ ($r^2 = 0.421$)	0.860 (0.034–1.8) [35]
Heart	$Y = 1.92X^{1.00}$ ($r^2 = 0.915$)	0.192 (0.077–2.71) [180]
Kidney	$Y = 5.16X^{1.03}$ ($r^2 = 0.891$)	0.518 (0.155–1.44) [137]
Liver	$Y = 13.42X^{1.08}$ ($r^2 = 0.899$)	1.43 (0.222–6.23) [216]
Muscle	n.a.	64.3 (55.3–76.7) [5]
Skin	n.a.	7.1 [1]
Skin and scales	n.a.	12.0 (9.3–14.1) [5]
Spleen	$Y = 1.12X^{0.98}$ ($r^2 = 0.856$)	0.112 (0.031–0.413) [77]
Stomach/ intestine	$Y = 39.61X + 36.76$ ($r^2 = 0.894$)	5.06 (0.200–12.3) [157]
Thyroid	$Y = 0.0131X + 8 \times 10^{-5}$ ($r^2 = 0.628$)	1.42×10^{-3} (2.03×10^{-6}–0.162) [170]
Viscera	n.a.	10.4 (6.5–16.1) [3]

in edible tissues. This information was then used to tabulate nuclide loads in edible fish tissues with respect to tissue compartment size (or tissue biomass).

The objective of this paper is to summarize an approach to estimate concentrations of nuclides in edible freshwater fish tissues, based on data for other tissues and analogous elements, using a mass balance approach that is conceptually similar to that of Reference Man (ICRP, 1975). In addition, generic parameter values describing the distribution of nuclides in freshwater teleost fish tissues, which have been derived using this approach, have been provided.

2. Methods

Information on the amount or load of nuclides in the tissue compartments of organisms, such as freshwater fishes, can be used to estimate concentrations in edible tissues using a mass balance approach when data on edible tissues are lacking (Yankovich and Beaton, 2000). As a first step, literature data have been compiled for teleost fishes of varying body sizes to quantify the biomass of each tissue compartment, and predictive relationships were developed to relate tissue biomass to organism fresh weight. In doing so, focus was placed on edible freshwater fish tissues, which included whole-body for forage fishes; muscle, liver and eggs for benthivorous fishes; and muscle and eggs for piscivorous fishes (Livingston and Livingston, 1993). Freshwater fishes were considered because of their importance in the human diet and the relatively large amount of data available for this type of organism.

Biomass estimates for fishes and their internal compartments were made, and corresponding concentration measurements were taken for each tissue. These were then utilized to estimate the expected loads of nuclides for all groups of metals (alkalis, alkaline earths, basic metals, lanthanides and actinides, and transition metals), as well as metalloids and non-metals, in fish tissues for use in estimation of radionuclide uptake by humans via the dietary pathway, as follows:

$$L_n^t = \frac{C_t m_t}{C_{wh} m_{wh}} 100\% \quad (1)$$

where L_n^t is the load of nuclide, n, in tissue, t, relative to the load in the whole body; C_t is the nuclide concentration in a given tissue (in mg/kg fresh weight or Bq/kg fresh weight); m_t is the biomass of that tissue (in kg fresh weight); C_{wh} is the element concentration in the whole organism (in mg/kg fresh weight or Bq/kg fresh weight); and m_{wh} is biomass of the whole organism (in kg fresh weight).

Therefore, the fraction of a nuclide in a tissue, t, relative to the whole body, wh, can be determined using Equation (1), which is based on measured concentrations.

In cases where concentration data are not available for a tissue, it is possible to use a generic tissue-specific concentration ratio, CR_t, to estimate missing values for a given nuclide. CR_t can be defined as the ratio between the nuclide concentration in the tissue of interest and the concentration in a Reference Tissue for which a large amount of data are available. For example, for teleost fishes, muscle can be used as a Reference Tissue (Yankovich and Beaton, 2000). By taking the product of CR_t and the concentration of the nuclide in the Reference Tissue (C_{RT}), it is possible to estimate the concentration in the tissue of interest. $L_n^{t:ref}$ can then be estimated based on the concentration in tissue, t, relative to the concentration in the Reference Tissue (muscle) by re-arranging Equation (1), as follows:

$$L_n^{t:ref} = \frac{CR_t C_{RT} m_t}{C_{wh} m_{wh}} 100\% \quad (2)$$

where $L_n^{t:ref}$ is the expected load of nuclide, n, in tissue, t, relative to the load in the whole body; CR_t is the tissue-specific concentration ratio of a tissue of interest (based on literature data); and C_{RT} is the concentration of the element of interest measured in the Reference Tissue (i.e., muscle) (in mg/kg fresh weight or Bq/kg fresh weight).

At a broader scale, some elements, such as calcium analogues, tend to preferentially and predictably partition into hard tissues, as opposed to soft tissues. As a result, it is reasonable to sub-divide a fish into soft and hard tissue compartments (Yankovich, 2002). In doing so, the sum of the loads in all the hard tissues should be taken, so they can then be compared to the sum of the loads in all the soft tissues. A load ratio, LR_n, for nuclide, n, can then be calculated to compare the tendency to partition between the hard and soft tissue compartments, as follows:

$$LR_n = \frac{L_n^{hard}}{L_n^{soft}} \quad (3)$$

where L_n^{hard} is the sum of the loads of nuclide, n, in all hard tissues (i.e., bone, gills and scales); and L_n^{soft} is the sum of the loads of nuclide, n, in all soft tissues (e.g., muscle, liver, eggs, etc.).

When comparing the load of a nuclide in the hard tissues versus the soft tissues for different fish species, differences in the sizes of these two compartments can add variability, making it more difficult to produce predictive patterns in partitioning between species. To address this, LR_n can be weighted for the biomass of soft and hard tissues in a given fish species, using the following equation, as past work has indicated that it is reasonable to compartmentalize fishes into soft and hard tissues for such purposes (e.g., Yankovich and Beaton, 2000; Yankovich, 2002):

$$BR_n = \frac{L_n^{hard} m_{hard}}{L_n^{soft} m_{soft}} \quad (4)$$

where BR_n is the hard-to-soft tissue load ratio, LR_n, weighted for the biomasses of hard, m_{hard}, and soft tissues, m_{soft}.

Literature data on nuclide partitioning between fish tissues and the fresh biomass of each tissue were supplemented with measured data for brown bullheads (*Ameirus nebulosis*), a benthic fish species, and northern pike (*Esox lucius*), a piscivorous species, that were collected from Perch Lake (Chalk River, Ontario) between 1995 and 1999 (Yankovich and Cornett, 2001; Yankovich, 2002). Individual fishes were dissected to remove blood, boneless hypaxial muscle, bone, liver, kidney and gonadal tissues for stable element analysis. In doing so, care was taken to avoid contamination of soft tissues with bone or other hard tissues, such as scales, particularly when dissecting northern pike, a scaled species which has Y-bones passing through the hypaxial muscle. Concentrations of stable analogues of radionuclides (referred to here as 'nuclides') were then measured in tissues using Inductively-Coupled Plasma Mass Spectroscopy (ICP-MS) and ICP-Atomic Emission Spectroscopy (ICP-AES).

3. Results and discussion

3.1. Tissue-to-whole body biomass ratios

Relationships between body sizes of fishes and tissue biomasses, as well as the percent biomasses of each tissue, have been estimated based on both literature data and measurements taken for Perch Lake fishes (Table 1). As expected, with the exception of gonadal tissue (which is known to vary with maturity, season, temperature, sampling location, nutritional status and other factors), predictive relationships between fresh biomasses of whole fishes and their edible tissues are fairly strong (Calder, 1996; Peters, 1983; Yankovich and Cornett, 2001; Table 1). In general, the strongest relationships were found when comparing whole body fresh biomasses to those of bone ($r^2 = 0.992$), heart ($r^2 = 0.915$), liver ($r^2 = 0.899$), stomach/intestine ($r^2 = 0.894$), kidney ($r^2 = 0.891$) and spleen ($r^2 = 0.856$), with slightly lower r^2-values for brain (0.747), eyes (0.727) and thyroid (0.628). By comparison, r^2 values of only 0.340 and 0.421