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Review

Revision and meta-analysis of selected biosphere parameter values for chlorine, iodine, neptunium, radium, radon and uranium

S.C. Sheppard*, M.I. Sheppard, J.C. Tait, B.L. Sanipelli

*ECOMatters Inc., 24 Aberdeen Avenue, P.O. Box 430, Pinawa,
Manitoba, Canada R0E 1L0*

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Abstract

There is a continual supply of new experimental data that are relevant to the assessment of the potential impacts of nuclear fuel waste disposal. In the biosphere, the traditional assessment models are data intensive, and values are needed for several thousand parameters. This is augmented further when measures of central tendency, statistical dispersion, correlations and truncations are required for each parameter to allow probabilistic risk assessment. Recent reviews proposed values for 10–15 key element-specific parameters relevant to ^{36}Cl , ^{129}I , ^{222}Rn , ^{226}Ra , ^{237}Np and ^{238}U , and some highlights from this data update are summarized here. Several parameters for Np are revised downward by more than 10-fold, as is the fish/water concentration ratio for U. Soil solid/liquid partition coefficients, K_d , are revised downward by 10–770-fold for Ra. Specific parameters are discussed in detail, including degassing of I from soil; sorption of Cl in soil; categorization of plant/soil concentration ratios for U, Ra and Np; Rn transfer from soil to indoor air; Rn degassing from surface water; and the Ca dependence of Ra transfers.
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Keywords: Chlorine; Iodine; Neptunium; Radium; Radon; Uranium; Pathways; Foodchain; Dose

* Corresponding author. ECOMatters Inc., WB Business Centre, 24 Aberdeen Avenue, Suite 105, Pinawa, Manitoba, Canada R0E 1L0. Tel.: +1 204 753 2747; fax: +1 204 753 8478.
E-mail address: sheppards@ecomatters.com (S.C. Sheppard).

1. Introduction

The computational models used to facilitate the environmental safety assessment of nuclear waste disposal and the environmental impact of naturally-occurring and technologically-enhanced radionuclides are data intense. Several hundred parameters must be assigned values if the assessment is at all comprehensive. There are isotope-specific dose conversion factors, element-specific environmental transfer factors, and ecosystem-specific descriptive or state parameters. In general, the dose conversion factors for humans (dose coefficients as Sv Bq⁻¹) are well known, and the ecosystem-specific descriptive parameters are measurable and also well known. As a group, the least well known are the element-specific transfer factors. This arises because these parameters, in addition to being element-specific, are often also specific to certain ecosystems. This implied factorial, of dozens of parameters, 30 or more elements and an indefinite but large number of ecosystem categories results in the need for a large database.

Not all element-specific parameters are deficient in data: there is a massive literature dealing with cesium (Cs) in the environment, and several other elements are well studied. However, there are elements and environmental circumstances where the parameter values are poorly known. In addition, there is a continued international effort to increase the amount of data available for all the elements of concern. As a result, there is a periodic need to update the database of parameter values used in any nuclear environmental safety assessment.

Selecting values for use in safety assessments is not trivial. Attention must be given to issues such as temporal and spatial averaging. For example, parameters related to soil properties must represent (be averaged over) the spatial scale used in the model, whether it be local, regional or continental. Parameters also have specific ecological contexts. For example, a water-to-fish transfer parameter may be intended to encompass a complete food web leading to the organism in question, and so values measured with that organism but in the absence of appropriate contaminated food (for example, using contaminated water only) would be inappropriate. Similarly, they may be intended to compute dose to humans consuming fish, or dose to the fish themselves. Finally, if the assessment is to be probabilistic, many parameter values must be described with a best estimate, a measure of statistical dispersion, one or more truncations and possibly correlations to other parameters.

The objective of this paper is to report an update of selected biosphere parameter values suitable for probabilistic assessment. The elements included were chlorine (Cl), iodine (I), neptunium (Np), radium (Ra), radon (Rn) and uranium (U). The assessment context is generic to locations on the Canadian Precambrian Shield and for times many millennia in the future. Abbreviated definitions of the parameters are given in Table 1. The revised parameter values are presented here in tabular form, whereas the discussion deals with aspects of the data that were different from expected, or otherwise of scientific interest.

2. Methods

2.1. Search methods and data acceptance

The intent of the reviews was to emphasize peer-reviewed sources and grey literature sources that met similar quality standards. Computerized searches of key databases including Science Direct and STN International were completed. Both keyword and citation searches were done. In addition, personal collections were perused.

Table 1
Symbol and definitions of the parameters reviewed

	Definition
λ_{vol}	Surface water degassing rate, a first-order rate constant for loss from water column into the atmosphere by processes such as gas formation, and excluding aerosol formation.
AIML	Aquatic iodine mass loading, a volume ratio to represent the volume of water that would contain the aquatic-source iodine found in the a unit volume of atmosphere, where the mass loading of iodine into the atmosphere may be a gas or as suspended aerosols from processes such as bubble and wave break.
λ_v	Soil degassing rate, a first-order rate constant for loss from soil rooting zone into the atmosphere by processes such as gas formation, and excluding resuspension of dust.
C_r	Plant/soil concentration ratio, where soil concentration is for the rooting depth of soil on a dry-weight basis. Both fresh-weight and dry-weight concentrations are reported for plant tissues, and so it must be indicated.
B	Fish/water or aquatic concentration ratio, where fish concentration is usually for the edible flesh and is always on a fresh-weight basis.
α_{sed}	Lake-water to bottom-sediment removal rate, a first-order rate constant for net loss from water column into the sediment. This is a net loss term, so that re-release from sediment to the water column is encompassed.
K_d	Soil solid/liquid partition coefficient, effectively a concentration ratio with concentration on soil solids on a dry-weight basis.
f^j	Transfer factor from ingestion to edible animal product, where the superscript j refers to meat, poultry, eggs and milk. It is the fraction of radionuclide ingested by the animal daily that is transferred to the edible product, so that when multiplied by the daily ingestion rate of radionuclide the product is the concentration in the edible product.

See Table 4 for parameters reviewed that are specific to ^{222}Rn .

Ideal data were from measurements in realistic and relevant settings, with low contaminant concentrations, and where measurements were of the biotic compartment most relevant to human or biota exposure. Although our emphasis was on settings typical of boreal Canada, species from more temperate regions were considered potentially representative of future climates in northern Canada. Data acceptance criteria did vary somewhat among the elements. There is a voluminous literature on U, and so relatively ideal criteria could be realized. With Np, as a contrasting example, there are scant data and it was necessary to consider less ideal data and use expert judgment to account for their relevance. Considerable detail is given in the underlying references (Sheppard et al., 2002, 2004a,b, 2005a,b) as to how values were included or excluded.

Where a decision was made to summarize the data in a reference (coalesce multiple data to a single value to represent the study), geometric means (GMs) were used on the default assumption that ratio parameters are lognormally distributed. This is based on the Central Limit Theorem which, when applied to ratio data, indicates a tendency for lognormal distributions. This tendency is almost always confirmed by empirical evidence. As a result, summaries were usually the GM of observations within a reference, with geometric standard deviations (GSD) reported where appropriate. Our convention with summaries prepared for this paper is to report GSD to two significant digits, and to report GM with two significant digits only if the corresponding $\text{GSD} < 5$, otherwise we report only one significant digit. When citing values from references, we report the number of significant digits used by the original authors.

2.2. Statistical handling

The accepted data were gathered in a spreadsheet. Frequency distributions were prepared and statistical analyses describing distributions and other characteristics of the data such as

correlations were computed. Categorization of the data was an important consideration, and is discussed in more detail with the results. By categories, we mean differentiation of parameter values among categories of environmental factors. For example, should a generic plant/soil concentration ratio be applied to all plants, or should there be multiple values categorized with one value for each of a few or of many categories of plant or soil types. Categorization was driven by two factors. One was the number of categories used in the present model. For example, the present system model Gierszewski et al. (2004) now uses four soil types and two plant types. The other factor was the differences found among the data. In some cases, the data available indicated that refinement of the model might be justified or even required to account for the notable differences in parameter values observed. In other cases, the data were sufficiently homogenous that categorization would have no statistical value.

3. Results

3.1. Summary of findings

The reviews summarized in this paper update certain parameter values used in the assessments described by Goodwin et al. (1994), Wikjord et al. (1996) and Gierszewski et al. (2004). Some of these parameter values were from summaries completed in the 1970s or early 1980s. Many of these older summaries are still used throughout the world, because a comprehensive update is a major undertaking. One of the last major compilations was the IAEA Handbook (IAEA, 1994), and there is a revision underway at present through the IAEA EMRAS Program. Our revised parameter values are given in Tables 2–4. The detailed underlying reports are given in Sheppard et al. (2003, 2004a,b, 2005a,b).

In many cases, the present reviews markedly increased the amount of data underlying the selected values, sometimes by as much as a 100-fold. Interestingly, many of the previous values were shown to still be very representative of the more recent data. This is quite encouraging; it implies an asymptote in the process of derivation of new data and suggests that other parameters need the ongoing attention. In many cases, the value selected was a relatively minor change from the previous, less than an order of magnitude, which is usually not statistically different given the typical variation observed. In a few cases, there were large changes (Table 5). The large changes could often be traced to a situation where the previous value was largely expert judgment, and this is now appropriately replaced with data-based values. In some cases, the older values were found to be based on suspect data or data not well supported with ancillary information, and this underlying data could now be rejected and replaced with new.

The following sections deal with a few specific parameters and elements. These were chosen because they were judged to be of most scientific interest.

3.2. Degassing of iodine from soil

Iodine is nearly unique among the elements assessed for nuclear fuel waste disposal because it is biologically essential, relatively mobile, and potentially volatile. Its volatile nature is not usually considered an important exposure pathway, but rather is potentially important as a loss mechanism. Once volatilized from a contaminated environment, ^{129}I would be broadly dispersed and no longer a significant source of local or global exposure. Since ^{129}I has such a long half-life, volatilization may be the only effective local loss mechanism. Volatilization occurs from both aquatic and terrestrial environments, terrestrial volatilization is discussed

Table 2

Observed geometric means (*n*, geometric standard deviation) and recommended geometric means (geometric standard deviations) for the parameters that have been relatively well investigated

Parameter	Units	Chlorine		Iodine		Neptunium		Radium		Uranium	
		Obs.	Rec.	Obs.	Rec.	Obs.	Rec.	Obs.	Rec.	Obs.	Rec.
Plant/soil C_r fully generic plant type	FW ^a	5.5 (67, 4.1)		0.008 (80, 9.3)	0.008 (10)	0.0038 (135, 11)	0.003 (5.7)	0.009 (740, 10)	0.01 (11)	0.0017 (502, 9.0)	0.002 (10)
Plant/soil C_r for human foods	FW	3.7 (43, 5.0)	3.7 (5.7)	0.005 (61, 8.)		0.0006 (40, 6.7)		0.0041 (315, 7.5)		8×10^{-4} (231, 6.3)	
Plant/soil C_r for native browse and forage	FW	4 (24, 5.1)	4 (5.7)	0.03 (27, 8.3)		0.005 (124, 10)		0.02 (432, 11)		0.003 (287, 8.0)	
Soil Kd for sand	L kg ⁻¹	0.1 (7, na ^b)	0.1 (10)	8 (41, 7.2)	8 (10)	3 (26, 7.7)	3 (10)	40 (11, 24)	47 (4.9)	40 (30, 28)	40 (20)
Soil Kd for loam	L kg ⁻¹	0.1 (7, na)	0.1 (10)	20 (38, 9.7)	20 (10)	10 (21, 16)	10 (10)	30 (17, 10)	47 (4.9)	200 (63, 22)	200 (20)
Soil Kd for clay	L kg ⁻¹	0.1 (7, na)	0.1 (10)	10 (15, 8.5)	10 (10)	20 (11, 6.3)	20 (10)	30 (8, 8.0)	47 (4.9)	200 (20, 13)	200 (20)
Soil Kd for organic	L kg ⁻¹	0.1 (7, na)	2000 (10)	80 (20, 22)	80 (22)	500 (5, 5.2)	500 (10)	200 (1, -)	47 (4.9)	2000 (21, 6.7)	2000 (20)
Fish/water <i>B</i>	L kg ⁻¹ FW		50 (12)	30 (21, 14)	6 (12)		200 (12)	20 (63, 6.4)	20 (6.4)	3 (25, 10)	3 (10)
Lake-water-to-sediment removal rate constant	a ⁻¹	0.005 (13, 12)	0.005 (12)	3.9 (4, -)	4 (7.8)	0.5 (12, 12)	0.02 (6.9)	0.3 (94, 6.1)	0.3 (6.1)	0.2 (73, 15)	0.2 (14)

^a FW – refers to tissue concentrations on a fresh-weight basis.^b na – Geometric standard deviation not applicable to the data because there are zero values.

Table 3

Recommended geometric mean (geometric standard deviations) for parameters that are not well investigated

Parameter	Chlorine	Iodine	Neptunium	Uranium	Radium
Degassing from water (a^{-1})	0	—	0	0	0
Aquatic iodine mass loading	—	2.6×10^{-6} (3.0)	—	—	—
Degassing from soil (s^{-1})	3×10^{-11} (10)	7×10^{-10} (10)	0	0	0
Ingestion transfer coefficient from plants to terrestrial animal human-food products					
Cow milk ($d L^{-1}$)	0.015 (2.2)	0.0076 (2.9)	5.0×10^{-6} (3.2)	3.7×10^{-4} (3.2)	6.2×10^{-4} (3.2)
Beef meat ($d kg^{-1}$)	0.020 (2.2)	0.012 (3.2)	2.0×10^{-4} (3.2)	4.0×10^{-4} (3.2)	9.0×10^{-4} (3.2)
Poultry/eggs ($d kg^{-1}$)	2.0 (2.2)	7.5 (3.2)	2.0×10^{-2} (3.2)	1.2 (3.2)	0.13 (7)

here because it is less well studied. One of the difficulties in measuring this parameter is that the loss rate is slow enough, with a half-time of a decade or more, that the loss is analytically difficult to detect.

The soil degassing rate describes the fraction of a nuclide in the root zone of the soil that is lost as a gas to the atmosphere per unit time. ICRU (2001) recommends the notation k in units s^{-1} . The soil degassing rates used in Canadian assessments originally were lognormally distributed with a GM of $1 \times 10^{-9} s^{-1}$ ($0.032 a^{-1}$) and a GSD of 10 (Sheppard, 1992), independent of the soil type. In a subsequent study (Zach et al., 1996), this value was reduced to a GM of $6.7 \times 10^{-10} s^{-1}$ ($0.021 a^{-1}$) with a much smaller GSD of 3.0 based on the paper by Sheppard et al. (1994). This GSD of 3.0 was a subjective estimate, taken as intermediate between a GSD of 1.9 from research done in the field on Canadian soils (Sheppard et al., 1994) and a GSD of 11 that included field values from the arid Hanford site in the US (Garland et al., 1987 cited by Sheppard et al., 1994). Sheppard et al. (1994) summarized much of the earlier literature, and the range of iodine loss rate constants was 6×10^{-12} to $1 \times 10^{-7} s^{-1}$ with a GM of $6.7 \times 10^{-10} s^{-1}$, the value used by Zach et al. (1996).

Measurements done by Sheppard et al. (1994) over three years in a field setting, using a vegetated mineral soil, indicated a loss rate by volatilization of $7 \times 10^{-9} s^{-1}$ (GSD = 1.4). A shorter-term laboratory study with an organic soil gave a volatilization rate of $2 \times 10^{-10} s^{-1}$

Table 4

Radon specific parameter values

Parameter		Results
Surface water degassing transfer coefficient ($m s^{-1}$)	AM ^a (SD)	6.7×10^{-6} (3.3×10^{-6}) — correlated to wind speed or lake area
Soil relative emission rate ($mol_{Rn} m^{-2} s^{-1} (mol_{Ra})^{-1} kg_{soil}$)	GM ^a (GSD)	2.7×10^{-9} (2.2)
Soil-to-indoor air transfer factor ($mol^{222}Rn m^{-3} air / (mol^{226}Ra kg^{-1} dry soil)$)	GM (GSD)	1×10^{-5} (2.6)
Fraction released from domestic water into indoor air	GM (GSD)	0.52 (1.3)

^a AM (SD) indicates arithmetic mean (standard deviation) implying a normal distribution, GM (GSD) indicates geometric mean (geometric standard deviation) implying a lognormal distribution.

Table 5

Geometric mean (geometric standard deviation) of parameter values where recommendations differed from previous values (Davis et al., 1993) by more than 10-fold

Parameter	Previous	Recommended
Soil solid/liquid partition coefficient (L kg^{-1}) – Ra	500–36,000 (10)	47 (4.9)
Fish/water concentration ratio (L kg^{-1}) – Np	2500 (12)	200 (12)
Fish/water concentration ratio (L kg^{-1}) – U	50 (12)	3 (10)
Lake water to sediment transfer rate (a^{-1}) – Np	0.5 (3)	0.02 (6.9)

(Sheppard et al., 1994). Less volatilization from organic soils would be expected because they often more effectively sorb iodine (Bostock et al., 2003).

Muramatsu and Yoshida (1995) reported experimental work on volatilization of iodine from the soil–plant system, using ^{125}I . They assessed the volatilization from both rice growing in flooded soil and oat in the same soil drained. They concluded that iodine emissions were highly stimulated by the presence of plants and they observed seasonal patterns for iodine emission for both plant types. The volatilized species was methyl iodide (CH_3I) and was emitted primarily from the plant shoot rather than the soil. For rice grown in flooded soils, the emission rate varied from $4.9 \times 10^{-9} \text{ s}^{-1}$ to a high of $2.9 \times 10^{-8} \text{ s}^{-1}$ at tillering. The average over the 60-d growing season was about $9 \times 10^{-9} \text{ s}^{-1}$. For the oat plants, the average over the 60-d growing season was $3 \times 10^{-10} \text{ s}^{-1}$. The most interesting information is that the field-moist soil alone gave an initial emission rate of $2 \times 10^{-10} \text{ s}^{-1}$, very similar to that of the planted pot, then its emission rate dropped as low as $3 \times 10^{-11} \text{ s}^{-1}$. The flooded soil alone started much lower ($8 \times 10^{-10} \text{ s}^{-1}$) than the planted flooded soil.

More recently, Bostock et al. (2003) carried out iodine volatilization experiments with a coniferous forest and a grassland soil for varying time periods (48 h and 22 d). The soils were acidic in nature and the forest soil was rich in organic matter (up to 9.7%). The emission rates varied between the soils, a range of $0\text{--}2 \times 10^{-9} \text{ s}^{-1}$ was measured from the 48-h grassland study and 6×10^{-12} to $6 \times 10^{-10} \text{ s}^{-1}$ from the 22-d forest soil study. Both short-term processes and longer-term processes are apparently occurring. Some of this effect of time was attributed to sorption of iodine onto organic substances in the soil.

The work of Muramatsu and Yoshida (1995) and Bostock et al. (2003) confirm the GM of $6.7 \times 10^{-10} \text{ s}^{-1}$ used for the second Canadian assessment (Zach et al., 1996), and we recommend this value for future use as well. However, the GSD should be increased to 10, recognising the large variability in these data. This GM value is within about an order of magnitude of the loss rate expected from leaching (assuming a K_d of 10 L kg^{-1}), but 5×10^5 -fold faster than from radioactive decay of ^{129}I . Thus, it is clearly an important parameter and perhaps deserves more research attention. There are no truncations or correlations proposed, although there may be evidence to support a negative correlation with the soil solid/liquid partition coefficient, K_d , especially in high organic matter mineral and organic soils. When more I is sorbed (high K_d), apparently, and presumably, less is available to volatilize.

3.3. Non-zero soil solid/liquid partition coefficient (K_d) for chlorine

There is an almost implicit assumption in many sectors of the literature that Cl, specifically the Cl^- anion, does not sorb to earth materials. For this reason, it is commonly used as a tracer of water flow in porous media. However, in the context of ^{36}Cl from nuclear fuel waste, even

a very modest degree of sorption in soil could markedly change the dose estimates. For example, compared to nil sorption, a very low K_d of 0.1 L kg^{-1} could result in a near doubling of the steady-state soil ^{36}Cl concentrations (the increase in steady-state concentration would be approximately $1 + K_d(\rho/\theta)$, where ρ is soil bulk density (kg L^{-1}) and θ is the volumetric soil moisture content).

In addition to the limited scientific interest in K_d values for Cl, measuring K_d values as low as this is problematic because many methods compute sorption by difference in pore water concentration before and after contact with soil, and for Cl these differences are small. However, there are a few data (Table 6).

The major compendia documenting K_d values for several nuclides (Coughtrey et al., 1985; Sheppard and Thibault, 1990; IAEA, 1994) all excluded Cl. In earlier Canadian assessments (Zach et al., 1996; Sheppard et al., 1996), the K_d values for Cl were derived from a regression across many elements, with plant/soil concentration ratios as the independent variable. For sand, loam, clay and organic soils, the GM K_d values were 0.8, 0.25, 4.4 and $11 \text{ L water kg}^{-1}$ dry soil. The GSD for all soil types was 10. The correlation between K_d and plant/soil concentration ratio was at -0.7 , as it was for all radionuclides. For a Swedish nuclear fuel disposal safety assessment, Bergström et al. (1999) set the median soil K_d value at 1 L kg^{-1} , with a range of $0.1\text{--}10 \text{ L kg}^{-1}$. The RESRAD model used to predict risks of radionuclide releases from contaminated soil has a default soil K_d for Cl^- of 0.1 L kg^{-1} (RESRAD). Koch-Steindl and Pröl (2001) concluded that Cl^- is the only species of Cl to be considered within the stability field of water; it is only weakly bound to soil, is not sorbed, not accumulated in soil and is quantitatively lost from soil by leaching.

There have been a few direct measurements of K_d for Cl. Sheppard et al. (1993) measured soil K_d for Cl in soil at about field capacity; initially the K_d was 0.63 L kg^{-1} , then became 0 L kg^{-1} over a period of 21 d (their detection level for K_d was not reported). The soil had a pH of 7.5, clay content of 18%, organic matter content of 18.4%. Sheppard et al. (1993)

Table 6
Soil solid/liquid partition coefficients, K_d , for Cl as reported in the literature

Source	Soil description	Soil texture	K_d (L kg^{-1})
<i>Experimental</i>			
Sheppard et al. (1993)	Garden soil	Loam	0
Takebe (2000)	Na-type bentonite	Clay	1
	Ca-type bentonite	Clay	0
	JAERI Tokai, pH 8.5	Sand	0.1
	JAERI Tokai, pH 8.7	Silt	0
	Ibaraki Kuroboku, pH 6.9		1
Sheppard et al. (2004a) inferred from Milton et al. (2003)	Litter	Organic	2200
<i>Assumed values</i>			
Zach et al. (1996),	Clay	Clay	4.4
Sheppard et al. (1996)	Loam	Loam	0.25
	Sand	Sand	0.8
	Organic	Organic	11
Bergström et al. (1999)	Generic soil		1
Klos et al. (1996)	Generic soil		0
Predicted from C_r value of 16 reported by Sheppard et al. (1999)	Garden soil		0.92
Bright and Addison (2002)	Generic soil		0.6

also noted a soil residence half-time for Cl in the field of 140 d, slightly longer than for Br in the same experiment, which implies Cl was not fully mobile. Takebe (2000) reported Kd values from batch-type experiments for several materials, including soils, and the values for soils ranged from 0 for sands and silts to 1 L kg^{-1} for sodium bentonite and for a soil formed on volcanic ash that may have had an organic matter content in the order of 10%. Milton et al. (2003) recently suggested that the increase in ^{36}Cl concentration measured at depths $<2 \text{ m}$ on soils of the Canadian Shield probably indicates organic sequestering of Cl. Unfortunately, few soil Kd values are possible from their data. Sheppard et al. (2004a) inferred a range, using ancillary data from Milton et al. (2003), of $830\text{--}5700 \text{ L kg}^{-1}$. These are remarkably high values, but may be appropriate for organic soils. Cornett et al. (1997) measured ^{36}Cl in environmental samples and concluded that it was moving through the biosphere at slower rates than would a non-reactive anion or water, but they did not provide any retention or Kd data.

One of the explanations for the observations by Milton et al. (2003) of Cl accumulation in upper soil layers is a combination of organic association of Cl in tandem with efficient recycling by vegetation. Recent experiments in the Chernobyl 10-km zone have demonstrated that ^{36}Cl , added as a tracer to the soil, was rapidly incorporated into the standing biomass, where it was efficiently recycled and thereby prevented from being lost by leaching (Kashparov, 2004).

There are a number of studies that offer inferential evidence of non-zero Kd values in groundwaters (Ogard et al., 1988; Phillips et al., 1988). Chlorine-36 was among the suite of radionuclides spiked into the groundwater below a 70-cm soil column in a lysimeter experiment in the UK (Butler and Wheeler, 1999). The results showed that the soil Kd for ^{36}Cl was definitely non-zero. Lee et al. (2001) investigated further and showed that ^{36}Cl attached to humic substances and associated preferentially with low-molecular-weight fractions. They felt they had clear evidence for the existence of chlorinated humic substances that retarded the migration and reduced the bioavailability of ^{36}Cl .

The median value for all soils in Table 6, including the zero values, is 0.1 L kg^{-1} . The GM is not very meaningful because there are as many zero values as non-zero values (a GM cannot be computed including zero values). There are insufficient data to differentiate Kd among the mineral soil types, so the median of 0.1 L kg^{-1} is recommended as the GM. The value of 2000 L kg^{-1} (from Milton et al., 2003) is tentatively recommended as a GM for organic soils. Because GSD cannot be computed where there are zero values, a generic estimate is used. In Thibault et al. (1990), the median GSD for 26 radionuclides and four soil texture classes was 5.0 (range 1–100). Here, the recommended GSD for ^{36}Cl is 10, consistent with the default used by Zach et al. (1996) and still appropriate because of the scarcity of data and the inconsistent evidence of apparent retardation for Cl. No truncations are recommended, and a negative correlation with the plant/soil concentration ratio probably remains appropriate.

3.4. Effect of soil pH on soil Kd for uranium

Several authors have pointed to the relationships between soil Kd and selected physical or chemical properties. Soil pH, clay and organic matter contents as well as mineralogy have all been shown to be important for determining soil Kd for a variety of elements. Soil pH and organic matter content can vary significantly with time, depending on soil management and fertilisation practices.

For U, a study that investigated the effects of these soil characteristics was Echevarria et al. (2001). They found no significant effect of clay or organic matter content; however, they did

find a significant relationship between soil Kd (as L kg^{-1}) and pH. For 21 soils with pH from 5.5 to 8.8, the relationship was:

$$\log K_d = a \text{ pH} + b \quad (1)$$

where $a = -1.29 \pm 0.17$ (standard error) and $b = 11 \pm 1.2$ (standard error).

The r^2 value was 0.76. This significant relationship with soil pH is probably due to the existence of different uranium complexes as a function of soil pH. Echevarria et al. (2001) stated that soil pH should be the focus variable for reduction of uncertainty associated with soil Kd for risk assessments, at least for U.

The present updated compilation of soil Kd values includes 134 data for mineral soils with a pH range of 4.0–8.8. However, the negative relationship between Kd for U and pH is undoubtedly related to the formation of soluble U-carbonate complexes at higher pH. It follows then that above pH 5.5, where HCO_3^- becomes an important species (Lindsay, 1979), is the domain where pH would be a useful predictor. In Fig. 1, there is a lot of scatter, but above about pH 5.5, the regression relationship (Eq. (1)) has the coefficients $a = -1.07 \pm 0.133$ (standard error) and $b = 9.80 \pm 0.930$ (standard error), neither of which are significantly different ($P > 0.05$) than those of Echevarria et al. (2001). The r^2 is 0.41, less than that of Echevarria et al. because this compilation contains values from many different protocols for obtaining sorption measurements, whereas Echevarria et al. used one consistent protocol. Recommended Kd values for U would utilize the regression equation for soils above pH 5.5, and a constant value of 300 L kg^{-1} below pH 5.5. In the absence of information about soil pH, the recommendations (L kg^{-1}) as GM of the reviewed values are sand: 40; loam: 200; clay: 200; and organic: 2000.

3.5. Plant type categories for plant/soil concentration ratios

Plant/soil concentration ratios CrS are perhaps the most thoroughly investigated of the parameters used in environmental safety assessment models. For example, in this project there were 535 relatively recent data for U and 624 for Ra. This amount of data suggests that there is potential for discrimination of values among various plant and soil types. Obviously, there are several drivers for this. From the perspective of a dose assessment model, there is a limit to the number of different plant and soil types that can be reasonably accounted for, unless there is

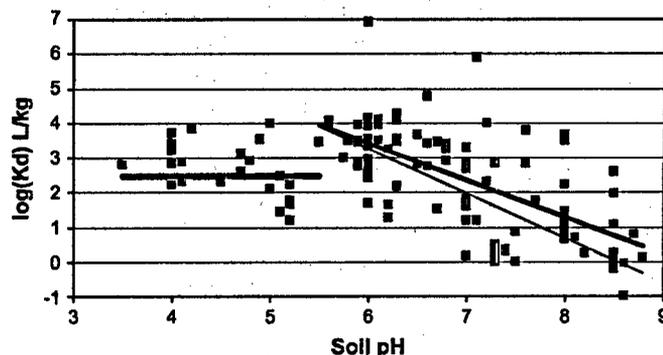


Fig. 1. Soil Kd (log of Kd in L kg^{-1}) for U versus pH. Below pH 5.5, there was no significant effect of pH (GM Kd = 300 L kg^{-1}). Above pH 5.5, the bold line is the best-fit regression, and the fine line is that of Echevarria et al. (2001).

detailed information about the diet and location of the critical group. Certainly, dose assessment for waste management requires projections into the future where diet, in particular, cannot be well specified. There is also a limitation imposed by the data: reporting CrS for different plant and soil types is not justified if they are not significantly different.

Sheppard and Evenden (1997) and Sheppard (2005) suggest that for all parameters, there is an inherent level of variation that reflects sources such as natural variation and inaccuracy of the ratio model. For Cr this coincides with a GSD in the order of 3–6, which in turn implies that Cr values that differ by less than about 10-fold are probably not significantly different. As a result, based on statistical differences, there are generally only a few categories of plant and soil types that warrant different Cr values.

Table 7 shows the results of categorizing Cr for Np, Ra and U into nine plant types. These elements are highlighted here because there were more data than for Cl or I. Note that the overall GSDs were 12, 6.4 and 8.4, respectively. After analysis of variance that accounted for the nine plant types, the residual GSDs (that variation not explained by the plant type categories) were much lower at 6.7, 3.4 and 5.7, respectively. Clearly, these plant type categories were useful in explaining some of the overall variation (Sheppard, 1995a).

For each element, there were significant differences among the plant types. Lichens, mosses and heather had substantially higher CrS than other plants, probably reflecting plant stature and the specific ability of these plants to retain dust – dust is a major source of nutrients for lichens and moss because they do not have roots. Dust and soil splash are also important contributors to radionuclides in forages consumed by animals (Sheppard, 1995b). Among the other plant types, there was a general tendency for the human consumption plants to have lower CrS than the animal forage, native browse, shrub and trees. However, the differences among the plant types were not consistent from element to element. For example, Cr values for trees were especially low for U, but were third highest for Ra. This highlights that categorization among plant types will be different among elements, which of course is not surprising given that the plants have some metabolic discrimination in the uptake and redistribution of contaminant elements. From

Table 7
Geometric mean Cr (dry-weight basis), in brackets are (n, GSD)

Plant type	Neptunium	Radium	Uranium
Vegetables	0.0059 (1, –)abc	0.02 (96, 5.3)d	0.0045 (81, 5.0)cd
Root crops	0.0046 (2, 1.2)abc	0.018 (78, 4.8)d	0.006 (64, 6.2)bcd
Cereal grains	0.003 (43, 7.8)c	0.03 (79, 6.1)cd	0.003 (55, 8.7)d
Fruits, berries, nuts	0.0028 (7, 3.4)bc	0.04 (27, 5.9)bcd	0.0021 (36, 4.1)d
Forages	0.03 (78, 8.9)ab	0.1 (170, 5.6)ab	0.01 (104, 5.8)b
Native browse	0.6 (2, 9.6)a	0.1 (80, 6.5)ab	0.01 (83, 8.7)bc
Shrubs	–	0.078 (25, 3.6)abc	0.007 (33, 5.2)bcd
Trees	8.6 (1, –)a	0.10 (58, 4.1)ab	0.004 (68, 16.3)d
Lichens, moss, heather	–	0.3 (11, 6.3)a	0.3 (9, 11)a
Overall	0.01 (134, 12)	0.05 (624, 6.4)	0.006 (535, 8.4)
Residual GSD	6.7	3.4	5.7
Concluding comments	Cereal grain and forages not the same	Lichens, moss and heather most unique, human consumption plants not the same as forages and browse	

Values followed by the same letter within a column are not significantly different ($P < 0.05$). Residual GSD is after analysis of variance accounting for the effect of plant type.

the perspective of dose assessment models, this means that although there may be a generic categorization to variation in plant types across all elements in the model, the data may not support different values. To illustrate, the model may be specified to use different Cr values for vegetables and root crops, but based on Table 7, there is no justification to use different values for these crops. Although not investigated here, it is also relevant to consider correlations to soil elemental concentration for U and perhaps all elements (Sheppard and Evenden, 1988).

In summary, the model of Gierszewski et al. (2004) uses only two categories of plants – human consumption versus animal consumption, and the present reviews support this in general. Further categorization would be supported by the data, but the categories would not be consistently different across the many elements in an assessment model.

3.6. Radon degassing from surface water

Radon-222 concentration in the atmosphere as a result of degassing from surface water is modeled by estimating the flux of ^{222}Rn out of the water body, followed by a treatment of atmospheric dispersion to give the air concentration (Table 4). The degassing parameter is a velocity (m s^{-1}), so when multiplied by a concentration in water ($\text{Bq }^{222}\text{Rn s}^{-1}$) yields a flux density ($\text{Bq }^{222}\text{Rn m}^{-2} \text{ s}^{-1}$). This parameter is often described as a transfer coefficient.

A number of studies have measured this parameter in ideal settings. Emerson et al. (1973) measured the mass transfer coefficient for ^{222}Rn for Lake 227 in the Experimental Lakes Area (ELA, northwest Ontario, Canada), and suggested that lower values they observed for lakes, compared to open ocean studies, were due to lower wind speed. Lower wind speed would increase the boundary layer thickness and decrease the gas exchange rate. Hesslein et al. (1980) measured the ^{222}Rn mass transfer coefficient for Lake 224 at the ELA in late July to early September. The measured transfer coefficients in a whole-lake experiment ranged from 4.1×10^{-6} to $10 \times 10^{-6} \text{ m s}^{-1}$ with a mean of $7 \times 10^{-6} \text{ m s}^{-1}$. The data were taken at weekly intervals and thus would average the effects of wind speed on the transfer coefficient. Experiments in limnocorrals with a diameter of 5–10 m gave similar transfer coefficients. Again, the higher transfer coefficient values measured for Lake 224 compared to other lakes (Emerson et al., 1973, 1975) were attributed to “higher wind stress” on Lake 224; although, wind speeds were not specified by Hesslein et al. (1980).

Wind speed may be the main cause of this separation of the values obtained on lakes versus those obtained on oceans, although other processes such as the occurrence of surface films may also have an effect. To more fully examine the effect of wind speed, the results of several studies were combined (Fig. 2). These results suggest that the transfer coefficient is constant at $2.1 \times 10^{-6} \text{ m s}^{-1}$ for wind speeds below about 2.5 m s^{-1} , and then is a linear function at higher wind speeds (transfer coefficient = $-2.1 \times 10^{-5} + 8.5 \times 10^{-6}$ (wind speed), $r^2 = 92\%$, both regression coefficients $P < 0.05$).

Alternatively, a value specific to lakes could be surmised on the basis of the ELA data. The median of the means of the ELA data $6.7 \times 10^{-6} \text{ m s}^{-1}$ with a standard deviation of $3.3 \times 10^{-6} \text{ m s}^{-1}$. The distribution function should be truncated at 0 m s^{-1} since negative values are impossible. Because wind speed is likely to exhibit a normal distribution over a year, it is reasonable to expect that the distribution of transfer coefficients would also be normal. Although the transfer coefficient is nearly independent of wind speed below $\sim 2.5 \text{ m s}^{-1}$ (Fig. 2), a positive correlation with wind speed or lake size would be appropriate. A correlation of $r = 0.7$ is recommended, based solely on the rationale that a correlation is expected, and a value less than 0.7 will have little impact in stochastic analysis (Sheppard and Sheppard, 1989).

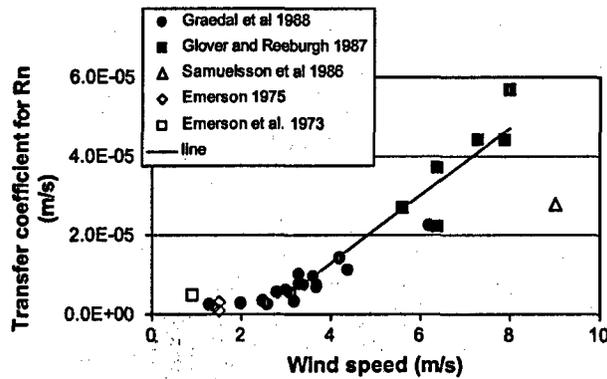


Fig. 2. Relationship between gas exchange (transfer) coefficient and wind speed, with data from several references and including both lake and ocean. The line is the best-fit line for wind speeds above 2.5 m s^{-1} .

The selected transfer coefficient would overestimate the annual transfer of ^{222}Rn to air from water bodies that may be frozen for a significant portion of the year and thus releasing little or no ^{222}Rn over this period. Similarly, solubility will vary with temperature, probably resulting in higher coefficients in late summer than other seasons. The transfer coefficient may be different between thermally stratified lakes (summer conditions in deep lakes, coincidentally the conditions where many of the data were collected) and mixed lakes (spring and fall or in shallow lakes). Overall, the use of the data from ELA should be conservative with respect to these seasonal effects (i.e., lead to higher dose estimates from atmospheric pathways) because they probably represent the seasons where the transfer coefficients are highest.

3.7. Radon transfer from soil to indoor air

A large portion of background radiation exposure to humans is attributed to ^{222}Rn (and its radioactive progeny), and ^{222}Rn in indoor air is a major vector. Against this background, it is still necessary to compute the incremental dose resulting from ^{222}Rn that arises because of nuclear fuel waste. Several models (Gierszewski et al., 2004; Azlina et al., 2003) account for ^{222}Rn in indoor air from both contaminated domestic water and direct transfer of ^{222}Rn gas from contaminated soil to indoor air. One approach is a simple ratio, where the soil-to-indoor air transfer coefficient is the ^{222}Rn concentration in indoor air divided by the ^{226}Ra concentration in soil.

$$K_{iRn} = \frac{C_{air}^{Rn}}{C_{soil}^{Ra}} \quad (2)$$

where:

K_{iRn} is the transfer coefficient from soil to indoor air $[(\text{mol } ^{222}\text{Rn m}^{-3} \text{ air})/(\text{mol } ^{226}\text{Ra kg}^{-1} \text{ dry soil})]$,

C_{air}^{Rn} is the indoor air ^{222}Rn concentration $(\text{mol m}^{-3} \text{ air})$, and

C_{soil}^{Ra} is the soil ^{226}Ra concentration $(\text{mol kg}^{-1} \text{ dry soil})$.

Although simple in concept, this parameter is very difficult to measure because, in many buildings, the building materials are an important source of the ^{222}Rn measured indoors, not the surrounding soil. Additionally, there is variation in the process caused by:

- indoor ventilation rates, which vary from winter to summer,
- building design (sub-grade rooms always have higher ^{222}Rn concentrations in air than grade-level or higher floors),
- the emanation efficiency from soil (the fraction of the ^{222}Rn produced in solids that is available to migrate),
- fraction of indoor air that previously passed through the contaminated soil,
- porosity and moisture content along the flow path from soil to indoors,
- the relative contribution from subsoil and underlying rocks, and
- thermal and barometric gradients that control convective migration.

Nason and Cohen (1987) measured ^{226}Ra in soil and ^{222}Rn in soil gas and indoor ^{222}Rn concentrations in several areas throughout the U.S. to determine if there was any correlation between measurements. Correlations between ^{222}Rn and ^{226}Ra concentrations were weak for all cases:

Rn(indoor air):Ra(soil), $r = -0.26-0.45$;
 Rn(indoor air):Rn(soil gas), $r = 0.0-0.37$; and
 Rn(soil gas):Ra(soil), $r = 0.30-0.46$.

The denominator of the parameter, the soil ^{226}Ra concentration, varies little, with a value of $3 \times 10^{-12} \text{ mol kg}^{-1}$ dry soil (25 Bq kg^{-1}) being representative of Canadian Shield conditions (Davis et al., 1993; Amiro, 1992) and 35 Bq kg^{-1} worldwide (UNSCEAR, 2000). The relatively small variability probably reflects the fact that ^{226}Ra is part of a primordial decay series that is ubiquitous. However, a corollary of this is that few studies that report indoor ^{222}Rn also report soil ^{226}Ra concentrations, limiting the amount of data available to determine $K_{i\text{Rn}}$.

Indoor ^{222}Rn concentrations are lognormally distributed and vary more widely. Studies on the Canadian Shield exhibit values ranging from 1.6×10^{-17} to $1.3 \times 10^{-16} \text{ mol m}^{-3}$ air ($20-166 \text{ Bq m}^{-3}$) and GSD values exceeding 3 (Amiro, 1992).

The literature found useful to compute $K_{i\text{Rn}}$ is summarized in Table 8. Preference was given to large surveys of areas with normal levels of soil ^{226}Ra . Where the literature reference did not provide direct measures of ^{226}Ra , then country-wide or global average values from UNSCEAR (2000) were used. Although correlations between indoor ^{222}Rn and soil ^{226}Ra are not commonly reported, meta-analysis such as compiled here is perhaps the best prospect to examine the correlation. The log values of indoor ^{222}Rn and soil ^{226}Ra concentration in Table 8 are significantly correlated ($r = 0.63$, $P < 0.001$). Notably, this resulted from the high values of both reported by Stranden and Strand (1988) and Voutilainen et al. (1988), all the other data were generally clustered together. With no ^{226}Ra there would be no ^{222}Rn , which implies a correlation would be an appropriate a priori assumption. However, with housing, quite often the construction materials contain ^{226}Ra . Because of proximity, ^{222}Rn from construction materials can often dominate over ^{222}Rn from the surrounding soil.

Values of $K_{i\text{Rn}}$ as calculated in Table 8 ranged from 0.1 to 9 ($\text{Bq } ^{222}\text{Rn m}^{-3} / (\text{Bq } ^{226}\text{Ra kg}^{-1} \text{ dry soil})$), with a GM of 1.5 ($\text{Bq } ^{222}\text{Rn m}^{-3} / (\text{Bq } ^{226}\text{Ra kg}^{-1} \text{ dry soil})$) and GSD

Table 8
Summary data for indoor air ^{222}Rn concentrations, soil ^{226}Ra concentrations and corresponding $K_{i\text{Rn}}$ values.

^{222}Rn (Bq m^{-3})	$^{226}\text{Ra}^a$ (Bq kg^{-1})	$K_{i\text{Rn}}$ ($^{222}\text{Rn}/^{226}\text{Ra}$) ($\text{Bq Rn m}^{-3}/\text{Bq Ra kg}^{-1}$)	Reference
90	46	2	Castren et al. (1985)
33	40	0.8	Nero et al. (1985)
43	60	0.7	McAulay and McLaughlin (1985)
41	26	2	Poffijn et al. (1985)
25	35	0.7	Sciocchetti et al. (1985)
19–88	17	1–5	Sorensen et al. (1985)
10–1000	10–60	4	Nason and Cohen (1987)
6–1800	5–200	3	Keller and Schütz (1988)
170–840	890–5600	0.2	Stranden and Strand (1988)
1100	1000	1	Voutilainen et al. (1988)
63, 34	32	2, 1	Baixeras et al. (1996)
60–330	35	2–9	Friedmann et al. (1996)
5–57	35	0.1–2	Harley (1996)
242	42	6	Hubbard et al. (1996)
22	20–70	0.3–1	Jin et al. (1996)
44	35	1	Keller et al. (1996)
60, 150	35	2–4	Kies et al. (1996)
26–48	40	0.7–1	Price and Nero (1996)
78	35	2	Siniscalchi et al. (1996)
116–179	35	3–5	Steck et al. (1996)
21	33	0.6	Fujimoto et al. (1997)
30	33	0.9	UNSCEAR (2000)
128	30	4.2	Dumitrescu et al. (2001)
28–64	29	1–2	Choubey et al. (2003)
130	25	5	Louizi et al. (2003)
70, 36	29	2, 1	Ramola et al. (2003)

^a Average ^{226}Ra concentration of 35 Bq kg^{-1} (UNSCEAR, 2000) assumed where no data were available.

of 2.6. The median GSD among those reported in the underlying studies (Table 8) was 2.5, suggesting this is a reasonable value.

A stochastic analysis similar to that of Amiro (1992) was undertaken, using a Monte-Carlo risk assessment tool (Crystal Ball 2000 – Decisioneering Inc.). Lognormal probability density functions (pdf) were chosen for the indoor ^{222}Rn and the soil ^{226}Ra concentration with the GM and GSD values presented in Table 9. The pdf for ^{222}Rn was the GM ($5 \times 10^{-17} \text{ mol m}^{-3}$ air or 60 Bq m^{-3}), with a GSD of 4.3. The pdf used for ^{226}Ra had a GM of 35 Bq kg^{-1} (world average from UNSCEAR, 2000) with a GSD of 3. The GM for $K_{i\text{Rn}}$ was calculated as 1.7 ($\text{Bq }^{222}\text{Rn m}^{-3}/(\text{Bq }^{226}\text{Ra kg}^{-1} \text{ dry soil})$). This is remarkably consistent with the GM from Table 8. In addition, the indoor ^{222}Rn and soil ^{226}Ra concentrations were also correlated in the Monte-Carlo analysis with correlation factors of 0, 0.3 and 1, to investigate the effect of correlation on the GSD for $K_{i\text{Rn}}$. As expected, the GSD decreased with increasing correlation (Table 9). Since there appears to be little or no observed correlation between indoor ^{222}Rn and soil ^{226}Ra concentrations, the results for correlations of $r = 0$ and $r = 0.3$ are probably most appropriate to estimate the GSD for $K_{i\text{Rn}}$, and these bracket the median GSD of 2.5 from Table 8.

Table 9

Geometric mean (GM), geometric standard deviation (GSD) for ^{222}Rn and ^{226}Ra used in the stochastic calculation of $K_{i\text{Rn}}$, also showing the calculated GM and GSD of the $K_{i\text{Rn}}$ parameter with three levels of correlation between ^{222}Rn and ^{226}Ra

	^{222}Rn (Bq m^{-3})	^{226}Ra (Bq kg^{-1})	$K_{i\text{Rn}}$ ($\text{Bq m}^{-3} \text{ }^{222}\text{Rn}/(\text{Bq kg}^{-1} \text{ }^{226}\text{Ra})$)	$K_{i\text{Rn}}$ ($\text{mol m}^{-3} \text{ }^{222}\text{Rn}/(\text{mol kg}^{-1} \text{ }^{226}\text{Ra})$)
Pearson correlation (Rn:Ra)			0	0.3
			0.3	1
			0.3	0.3
GM	60	35	1.7	1.7
GSD	4.3	3	6.5	4.6
			1.4	1.4

This survey indicated that the $K_{i\text{Rn}}$ value derived by Amiro (1992) of a GM of 4.3×10^{-5} ($\text{mol } ^{222}\text{Rn m}^{-3} \text{ air}/(\text{mol } ^{226}\text{Ra kg}^{-1} \text{ dry soil})$) and GSD of 2.6 was probably slightly high, intended to be conservative. The recommended value based on this review for $K_{i\text{Rn}}$ is a log-normal distribution with a GM of 1×10^{-5} ($\text{mol } ^{222}\text{Rn m}^{-3} \text{ air}/(\text{mol } ^{226}\text{Ra kg}^{-1} \text{ dry soil})$) or $1.5 (\text{Bq } ^{222}\text{Rn m}^{-3})/(\text{Bq } ^{226}\text{Ra kg}^{-1} \text{ dry soil})$ and a GSD of 2.6. No truncations are required, and no correlations to other parameters are justified.

3.8. Calcium dependence of radium transfer factors

It is very probable that all of the contaminant elements in nuclear waste interact to some degree with stable elements in the biosphere. For some, such as ^{129}I and ^{36}Cl , the key interaction is with stable isotopes of the same element (Sheppard et al., 1996). For some, and the same two elements serve as an example, there is some coincidental interaction: like elements (anions for this example) will behave similarly. Most interesting are the interactions that result from chemical interferences. Calcium is the best example, it is such a common cation in soil and water that it interferes with sorption of elements onto mineral surfaces and competes with elements for uptake at the organism membrane surface (Paquin et al., 2000). In general, these interactions are ignored, or are accounted for by selecting data from experiments where the media had similar characteristics to the media actually assessed. In the present review, the effect of Ca on Ra was evident for a number of transfer parameters.

The relationship between plant/soil concentration ratio (C_r) for Ra and the plant tissue Ca concentration is shown in Fig. 3. The best-fit relationship is $\log(C_r) = -2.07 + 0.616 (\log[\text{Ca}_{\text{tissue}}])$, where $[\text{Ca}_{\text{tissue}}]$ is the plant tissue Ca concentration (g kg^{-1}). Although apparently well described, this may be of only marginal importance because a 600-fold change in tissue Ca concentration results in only a 50-fold change in C_r . Values of C_r normally have variation in the order of 30-fold, although there was less variation for Ra than observed for other elements (Table 7). A 600-fold range in tissue Ca concentrations can be reasonably expected, especially if there is a range of soil conditions. Interestingly, one might anticipate a competitive relationship between Ra and Ca in plants, and the positive relationship shown here reflects that soil and plant factors are enhancing uptake of both elements (perhaps a coincidental interaction, as described above). In an assessment context, this effect may be indirectly modeled if soil Kd values are dependent on soil properties such as texture and pH.

Another example is the effect of Ca in water on the fish/water transfer factor (B). Combining data from three studies (Fig. 4), the relationship $\log(B) = 2.72 - 0.048 [\text{Ca}_{\text{water}}]$, where B is the fish/water concentration ratio (L kg^{-1}) and $[\text{Ca}_{\text{water}}]$ is the water Ca concentration (mg L^{-1}), seems to describe the data reasonably well. This implies that a 4-fold increase in Ca

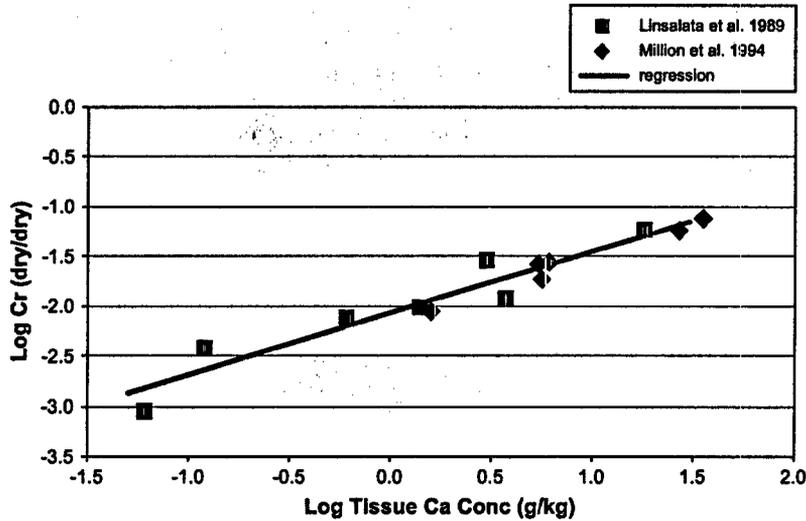


Fig. 3. Relationship between the dry-weight plant/soil C_r values for Ra and the plant tissue Ca concentration (regression is $\log(C_r) = -2.1 + 0.62 \log(\text{Ca})$, correlation of $\log(C_r)$ with $\log(\text{Ca})$ concentration is $r = 0.96$).

concentration causes a 700-fold decrease in B . This is a realistic range of water Ca concentrations, so that the effect on B is of practical importance. In this case, the effect would appear to be competitive, and it is well known that Ca competes with other cations for uptake at the media/membrane interface (Paquin et al., 2000). It is anticipated that because Ra and Ca have

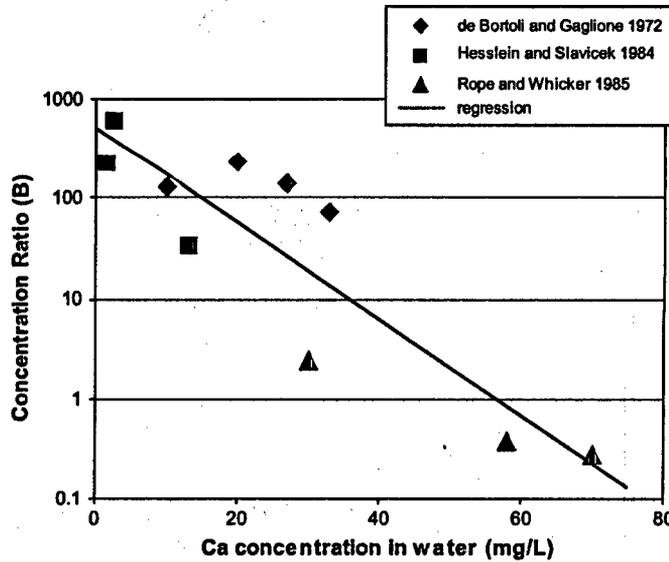


Fig. 4. Correlation between the fish/water concentration ratio, B , of ^{226}Ra in fish and the calcium concentration in the water (regression is $\log(B) = 2.7 - 0.048(\text{Ca})$, correlation of $\log(B)$ with Ca concentration is $r = -0.89$).

similar ionic properties, the much higher molar concentrations of Ca will have a profound effect on Ra uptake.

This relationship to Ca has been known for many years, and led to the concept of observed ratio (OR), where the concentration ratio for, in this case Ra, is normalized to the concentration ratio for Ca. Hesslein and Slavicek (1984) investigated the concept of the OR in organisms versus water and food to explain the differences in bioaccumulation. They found that Ra is discriminated against versus Ca by fish, but favored relative to Ca by macrophytes and crayfish.

This dependence of transfer factors on elements other than those directly related to waste materials has important implications. The simplest resolution is to select the transfer parameter values to represent the element composition of the media at the site being assessed. However, for long-term assessments, this requires an assumption that the soil and surface water chemistries will remain unchanged. Of all elements, this is perhaps least tenable for Ca. Calcium is an important and often dominant base cation, so that Ca concentrations are markedly affected by the weathering of soils and acidification of surface waters. Decreasing Ca concentrations over millennia may be a reasonable expectation. This implies that wherever Ca is a competing cation, as shown for Ra in fish, the transfer factors may increase with time as the Ca concentrations decrease.

3.9. Radium concentration ratios – whole fish versus flesh

The fish/water concentration ratio, B , was originally intended to model transfer to human food sources, but it may also be used to compute doses to non-human biota. Radium tends to accumulate in the skeleton – a further implication of the relationship between Ra and Ca. Thus, for humans the emphasis needs to be on Ra in flesh (muscle), whereas for non-human biota the Ra in bones should be included. For any pathway where the whole fish including bones may be ingested, it is probable that the Ra in the bones will be released by acidic gastric processes. This suggests a requirement for different parameter values for human versus non-human dose estimates. Of course, humans do ingest bones of small freshwater species such as smelt (*Osmerus mordax*).

Not many studies report both flesh and whole-fish data. Justýn and Havlík (1990) reported B values for Ra in *Ophiocephalus*, showing values for flesh of 5–15 L kg⁻¹, whereas for bone the B values were 40–77 L kg⁻¹. Blaylock (1982) reported B values for Ra in whole fish (less viscera) that were 63-fold higher than those they reported for flesh. Justýn and Havlík (1990) also pointed out relationships between Ra uptake and metabolism rate, so that the rate of accumulation of Ra in younger fish was twice as fast as in older fish. Anderson et al. (1963) reported Ra concentrations in several fish species, including whole fish and flesh. The B values calculated from these data varied from 0.72 to 44 L kg⁻¹ for whole fish (juveniles) and from 2.8 to 4.2 L kg⁻¹ for flesh (older fish). It is more probable that younger fish would be ingested whole.

The overall review identified 63 values of B for Ra in flesh, with a GM of 20 L kg⁻¹ and a GSD of 6.4. However, there were also 98 data for whole fish, with a GM of 71 L kg⁻¹ and a GSD of 4.8. Taken all together, the GM was 40 L kg⁻¹ with a GSD of 6.2. Although this clearly supports the expected trend, the values are not statistically different.

Similar to other parameters, this raises an interesting dilemma. The theory and considerable evidence suggests that different B values should apply to flesh-only than to whole fish. However, the overall variability arising from uncertainty in species, water chemistry and other

environmental variables (including food preparation) overshadow the variability controlled by differentiating flesh versus whole fish. Although this variability is known, the general desire to have simple assessment models probably does not allow for incorporation of known, but statistically insignificant, effects.

4. Summary/recommendations

A predominant observation from this review is that the research to support and derive new transfer parameters is extremely biased toward certain elements and certain parameters. Very little has been done with feed-to-animal-product transfers, probably because controlled studies are very expensive, especially if they involve radionuclides. Relatedly, these parameters are often found to be of lesser importance for dose estimates to humans. Biokinetic studies have been used to derive these transfer factors (Thorne, 2003), but these also require data that are not commonly available. There is a prospect for improvement for all parameter values resulting from the greater availability of extremely sensitive analytical methods. As an example, inductively coupled plasma mass spectroscopy (ICP-MS) typically provides data for over 50 elements in every sample, to very low concentrations. This means that analysis of stable-element isotopes and analogues to the elements of concern can be measured in everyday food production samples. In addition, stable element data facilitates the development of specific activity models particularly relevant for C, Cl and I (Sheppard et al., 1996).

In contrast to animal transfer parameters, for some elements such as U, there is a vast literature on soil-to-plant transfers. The arguments presented by Sheppard and Evenden (1997) and Sheppard (2005) suggest there is a finite level of precision that is possible with the simplistic concentration ratio model (a GSD of about 3–6). It could be argued that for elements such as U, further measurement of C_r is not going to advance the precision and accuracy of assessment. This may be true even for site-specific measurements (Sheppard, 2005), and is surely true of more generic assessments. For advancement of the prediction of soil-to-plant transfers of U, it is clearly time to adopt a more mechanistic model, or redirect the focus to less-well-researched elements.

There is at least one aspect of experimentation to obtain parameter values that is, in general, very inadequate. The science has proceeded, for many years, in a rather reductionist manner: parameters specific to one element and process are measured in isolation. The predominant example is the reporting of plant/soil concentration ratios without ancillary reporting of soil properties, and especially Kd. However, the same applies to other parameters, other environmental variables, and perhaps also from element to element. The science may well be advanced if more studies reported findings for a suite of elements, so that correlations and commonalities can be utilized. In the context of probabilistic risk assessment, the similar (correlated) behaviour of co-contaminants in a given realization should be investigated. For example, if the probabilistic case is one with higher-than-average soil Kd for one contaminant element, then this should imply (but in practice usually does not) that co-contaminants may also have higher-than-average Kd.

An indirect conclusion from this review is that transfer parameters do vary with environmental conditions, and obviously environmental conditions will vary over the time scales relevant to nuclear waste management. The increased bioavailability of Ra as Ca concentrations decrease is a good example. This argues for the modeling of an evolving environment, and not simply physical changes, but also chemical changes.

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