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Reference Gastrointestinal Absorption Fractions for Radionuclides Ingested in Soil

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OAK RIDGE National Laboratory

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REFERENCE GASTROINTESTINAL ABSORPTION FRACTIONS FOR RADIONUCLIDES INGESTED IN SOIL

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ABSTRACT

Oak Ridge National Laboratory is conducting a project for the Environmental Protection Agency (EPA) Office of Superfund Remediation and Technology Innovation involving derivation of cancer risk coefficients for ingestion of radionuclides in soil. EPA provides guidance on quantifying gastrointestinal (GI) absorption of a chemical in a given medium for use in risk evaluations including assessments for Superfund sites. Essentially, a medium-specific GI absorption fraction may be applied if and only if there is sound information to support that value. Otherwise, the applied GI absorption fraction should be the best estimate of fractional absorption of the chemical when ingested in highly soluble form. Based on our review of the literature on GI absorption of elements in soil, we have concluded that for many elements there is convincing evidence of substantially reduced absorption in ingested soil compared with ingestion in readily dissolved form. This report summarizes the reviewed information and proposes a comprehensive set of GI absorption fractions for ingestion of elements in soil. For most elements, the proposed GI absorption fractions are lower than the default absorption fractions recommended by the International Commission on Radiological Protection (ICRP) for radionuclides ingested in highly soluble form, even though considerable conservatism has been incorporated into the proposed soil-specific GI absorption fractions.

1. INTRODUCTION

ORNL is conducting a project for the Environmental Protection Agency (EPA) Office of Superfund Remediation and Technology Innovation involving derivation of cancer risk coefficients for ingestion of radionuclides in soil. This requires estimates of gastrointestinal (GI) absorption fractions for radionuclides contained in the soil. The default GI absorption values used in radiation protection for intake of radionuclides in diet generally are based on data for tracers ingested in material that is readily dissolved in the GI tract. Depending on the element and the soil type, a substantial portion of a radionuclide ingested in soil may be contained in particles that are insoluble in the GI tract, resulting in lower fractional absorption to blood than when ingested in readily soluble form.

EPA provides guidance on quantifying GI absorption of elements in soil for use in risk evaluations including assessments for Superfund sites (EPA, 2007b, 2022). The report "Guidance for Evaluating the Oral Bioavailability of Metals in Soils for Use in Human Health Risk Assessment" (EPA, 2007b) addresses potential uses of medium-specific bioavailability (fractional absorption) of the metals. Section 6, pp. 4-5, states that "in the absence of data to the contrary, the bioavailability of a chemical should be assumed to be equal in soil, diet, and water… [but] cases may exist where sufficient data are available for a chemical to support development of medium-specific and chemical-specific default values to increase the accuracy of exposure and risk calculations…" Ingestion of lead (Pb) in soil is used in that report as an example of medium-specific adjustment of GI absorption in risk assessments. In EPA's Integrated Exposure Uptake Biokinetic Model for Lead in Children (SRC, Inc., 2021), it is assumed that fractional absorption for Pb in soil or dust is 60% of its fractional absorption in diet or water as a rounded central value based on Pb ingestion studies on laboratory animals. The absorption fraction for Pb in diet or water is estimated by EPA as 0.5. The estimated absorption fraction for Pb in soil is $0.6 \times 0.5 = 0.3$, or 30% when ingested in soil compared with 50% when ingested in diet or water.

Based on our review of the literature, we have concluded that: (1) for many elements there is evidence of substantially reduced absorption in ingested soil compared with ingestion in readily dissolved forms, and (2) there is sufficient information in the literature to develop a system of GI absorption fractions for ingestion of elements in soil that is more realistic than blanket assignment of absorption fractions commonly applied to intake in diet or water. This report provides an overview of soil bioavailability data for elements, proposes GI absorption fractions for elements ingested in soil, and discusses the basis for the assigned absorption fractions.

2. METHODS

2.1 DEFINITIONS

As defined by the EPA reports (EPA, 2007b, 2012), the bioavailability of an element ingested in a medium is the fraction or percentage of the ingested amount that is absorbed into the systemic circulation (EPA, 2007b, 2012). The bioaccessibility of an element ingested in a medium refers to the physiological solubility of the element in the gastrointestinal tract and is expressed in terms of the fraction or percentage dissolved in a specified region of the GI tract (EPA, 2007b). The bioaccessibility of an element ingested in soil is commonly estimated from in vitro measurements, with in vitro conditions designed to simulate in vivo conditions in the human alimentary tract.

2.2 GENERAL CONSIDERATIONS

Soil is a system of solids and liquids characterized by processes that vary with soil type, e.g., loam, clay, sand, or soils rich in organic material ("organic soils"). The bioavailability of an element in a specific soil depends on the chemical and physical characteristics of that element as related to characteristics of the

soil such as pH (perhaps the most important single characteristic), texture, quantity and type of organic matter, and mineral content (Alloway, 2012; Iurian et al., 2015; Petruzzelli et al., 2020). The bioaccessible portion of an element in soil is expected to exist in one of the following conditions: in soluble form in the liquid phase of soil, adsorbed to minerals, or adsorbed or complexed by organic matter. The inaccessible portion of the element is likely to exist as oxides, carbonates, phosphates, or other strongly bound mineral compounds, or contained in crystal lattices of hydroxides, clay, or minerals (Petruzzelli et al., 2020). Studies of the bioavailability, bioaccessibility, or mobility of many different elements in soil provide a database with which to construct a system of GI absorption fractions for radionuclides in soil. The mobility of an element in soil refers to its capacity to move between different components of soil such as the solid and liquid phases, and to its related capacity to transfer vertically or horizontally over time. The mobility of an element is one of the primary factors determining its bioavailability in soil (Garrett, 2016).

2.3 MODIFICATION OF REFERENCE GI ABSORPTION FRACTIONS FOR APPLICATION TO INGESTION IN SOIL

The International Commission on Radiological Protection (ICRP) is updating its biokinetic and dosimetric models and dose coefficients for workers and members of the public. Updated models for workers were published in a series of ICRP reports called the OIR (Occupational Intake of Radionuclides) series, completed in 2022. The updated models for members of the public will be published in a series of reports called the Dose Coefficients for Intakes of Radionuclides by Members of the Public series, colloquially referred to as the EIR (Environmental Intake of Radionuclides) series. The elements considered in this report are those addressed in the OIR and EIR series of reports as parent radionuclides taken into the body.

For each element, the EIR series recommends default age-specific GI absorption fractions, called f_A values, representing absorption of the element when ingested in a form that is readily soluble in the GI tract. For some elements, additional f_A values are recommended for ingestion of specific chemical forms of the element. The f_A values are specified for each of six ages at intake: infancy (100 d), 1 y, 5 y, 10 y, 15 y, and adult. For example, the default f_A values for barium are 0.6 for infants, 0.3 for ages 1-15 y, and 0.2 for adults. Much lower f_A values are recommended by the ICRP for application to barium in highly insoluble forms such as barium sulfate.

The ICRP's default f_A values applied in the EIR series are used here as the starting place for developing a system of GI absorption fractions, in this report referred to as "f_S values", for elements ingested in soil. The elements addressed in this project are divided into five groups representing different levels of bioavailability in soil as assessed from a review of the literature. Elements in a group are assigned a common bioaccessibility factor, BF, applicable to all ages at intake. For an element in Group N (N = 1, 2, 3, 4, or 5), the absorption fraction $f_S(j)$ for the element ingested in soil at age j is derived as $BF_N \times f_A(j)$, where $f_A(j)$ is the ICRP's default f_A value for that element for intake at age j. Groups 1-5 are assigned the following BF values: $BF_1 = 1.0$, $BF_2 = 0.8$, $BF_3 = 0.6$, $BF_4 = 0.4$; $BF_5 = 0.2$. For example, for an element assigned to Group 3, the value f_S for age j is derived as $0.6 \times f_A(j)$. Group 1 consists of elements whose default absorption fractions applied by the ICRP were not changed for application to ingestion in soil, due either to evidence that bioavailability in soil approximated that in food or water, or as a cautious measure due to a paucity of data on bioavailability of the element in soil.

The assignment of BF values to an element follows the ICRP's general principles in assignment of f_A values to an element. That is, the assigned absorption fraction is ideally a central value for the population subgroup of interest but often is a cautiously high value that accounts for limitations in available data.

2.4 TYPES OF DATA USED TO ASSESS BIOAVAILABILITY OF ELEMENTS INGESTED IN SOIL

The preferred type of data for assessment of bioavailability of an element in soil are GI absorption fractions determined in human subjects or, more commonly, in laboratory animals following ingestion or oral administration of element tracers in soil. This type of data may come from: (1) studies that compare GI absorption of a tracer ingested in soil with absorption of the tracer administered in a form expected to be completely available for absorption to blood; or (2) studies that provide only an estimate of GI absorption of a tracer ingested in soil. In the second case, the bioavailability of the element is based on comparison of observed GI uptake with the ICRP's default f_A. The laboratory animals most often used in bioavailability studies are swine, monkeys, rats, mice, and rabbits, with swine considered by many investigators as a particularly good laboratory model for humans regarding digestive processes (Weis and Lavelle, 1991; Casteel et al., 1997; Heinritz et al., 2013).

Much of the data found in the literature related to bioavailability of elements in soil was developed in *in vitro* studies of the bioaccessibility of elements in soil. The goal of a bioaccessibility study is to determine the fraction of an element ingested in soil that is dissolvable in alimentary tract fluids and hence available for absorption to blood. In *in vitro* bioaccessibility studies, conditions are created to simulate *in vivo* conditions in the human alimentary tract such as pH, residence time, and enzymatic action in the mouth, stomach, and intestines. If an f_S value for an element is based largely on *in vitro* measurements of its bioaccessibility, some conservatism should be applied to account for limitations in *in vitro* modeling of the chemical environment of the human alimentary tract.

The mobility of an element in the aqueous surface environment (e.g., soil, streams, lakes) is considered an indicator of its bioavailability (Garrett, 2016). Discussions and tabulations of the mobility of elements in the surface environment address different measures of mobility including exchangeability between the solid and liquid phases of soil and rates of transfer through the larger surface environment including rates of vertical or horizontal relocation. Table 1 categorizes the relative mobility of groups of elements in the surface environment at different pH and redox conditions, based on a review by Garrett (2016).

A widely used measure of the mobility of an element in soil is the solid-liquid distribution coefficient, K_d, as determined for a specific element and soil. K_d is calculated as the concentration of the element in dry solids (mg kg^{-1} dry soil) divided by the concentration in pore water (mg L^{-1}), giving units of L kg^{-1}. The K_d value of an element is inversely related to its mobility. For example, K_d values of 0.5, 15, and 1000 for a given element and soil indicate high, moderate, and low mobility, respectively, of the element in that soil. As illustrated in Table 2, K_d for an element can vary considerably from one soil type to another. The variability results from differences in characteristics of different soils such as pH, clay content, and organic content, among several potentially important variables. Table 3 lists central K_d values for 48 elements as determined in a variety of soils, based on a review by Vidal et al. (2009). The review addressed 67 elements, but Table 3 addresses only those elements for which K_d values were reported for at least three soil types among loam, clay, sand, and highly organic soils.

A K_d value can be used to estimate the fraction of the element in the liquid phase of a soil based on typical fractions of solids and liquids in that soil type. The fraction of the element in the liquid phase is expected to be bioaccessible but usually is a considerable underestimate of the bioaccessible fraction. This is because a large portion of the element contained in the solid phase that is not readily exchangeable with water under normal soil conditions may be extractable from soil particles in the chemical environment of the alimentary tract. Thus, a low K_d value (e.g., <1) is a strong indication of high bioaccessibility. Overall, there appears to be an inverse relation between K_d and bioaccessibility, but a high K_d value alone does not ensure low bioaccessibility. For example, Pb typically has higher bioaccessibility than its K_d values would indicate. This may result from low solubility of common forms

of Pb (oxides, sulfates, carbonates) in soil water but relatively high solubility in acidic environments such as portions of the human alimentary tract (Traina and Laperche, 1999).

Table 1. Relative mobility of selected elements in the surface environment

Mobility	Oxidizing[a] (pH 5-8)	Oxidizing (pH < 4)	Reducing[a]
High	I, Cl, Br, S, C, Mo, Se, Te	I, Cl, Br, S, C	I, Cl, Br
Moderate	Ca, Na, Mg, F, V, Zn, Ag, As, Sb, Sr, Hg, U	Ca, Na, Mg, Sr, Li, F, Zn, Cd, Hg, Cu, Ag, Co, Ni, V, As, Mn, P, U	Ca, Na, Mg, Li, Sr, Ba, Ra, F, Mn
Low	K, Rb, Ba, Mn, Si, Ge, P, Pb, Cu, Ni, Co, Cd, In, Ra, Be, W	K, Rb, Ba, Si, Ge, Ra	K, Rb, Si, P, Fe
Very low	Fe, Al, Ga, Sc, Ti, Zr, Hf, Th, Sn, REEs[b], Pt metals[c], Au, Cr, Nb, Ta, Bi, Cs	Fe, Al, Ga, Sc, Ti, Zr, Hf, Th, Sn, REEs, Pt metals, Au, As, Mo, Se	Fe, Al, Ga, Sc, Ti, Zr, Hf, Th, Sn, Fe, REEs, Au, Cu, Pt metals, Ag, Pb, Zn, Cd, Hg, Ni, Co, As, Sb, Bi, Se, Te, Mo, In, Cr, Nb, Ta, U

Source: Garrett, 2016

[a.] Oxidizing and reducing refer to gain and loss of oxygen, respectively.

[b.] REEs = rare earth elements (Y, Sc, and the lanthanide family La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu)

[c.] Pt metals = platinum group metals (Ru, Rh, Pd, Os, Ir, Pt)

Table 2. Central estimates of K_d values for selected elements in different types of soil

	Element (number of samples)				
	Cesium (469)	Strontium (255)	Uranium (178)	Zinc (92)	Cadmium (61)
All soils	1200	52	200	950	150
Sand	530	22	110	110	110
Loam	3500	57	310	2400	100
Clay	5500	95	28	2400	130
Organic	270	110	1200	560	650

Source: Vidal et al., 2009

Table 3. Central estimates[a] of K_d values for elements studied in multiple soil types[b]

Element	K_d	Element	K_d
Tc	0.23	Zr	410
Cl	0.3	Bi	480
Na	3.4[c]	Co	480
Mg	3.8[c]	Cu	530
I	6.9	Pu	740
Ca	8	Ta	780
K	13	Fe	880
Np	36	Ho	930[c]
Mo	38	Sm	930[c]
Cr	40	Zn	950
Sr	52	Be	990
Br	55[c]	Ce	1200
Sb	62	Cs	1200
P[c]	87	Mn	1200
Si[c]	130	Nb	1500
Cd	150	Sn	1600
Pd	180	Ac	1700[c]
Se	200	Th	1900
U	200	Pa	2000[c]
Rb	210[c]	Pb	2000
Po	210	Hf	2500
Ru	270	Ra	2500
Ni	280	Am	2600
Ag	380	Cm	9300

Source: Vidal et al., 2009

[a.] Values for Sb, Pd, Po, Ta, Sn, Hf, and Am are arithmetic means. All other values are geometric means.

[b.] Elements were studied in loam, sand, clay, and organic soils except Na and Mg were not studied in organic soil.

[c.] Based on only one study.

3. SUMMARY OF REVIEWED DATA ON BIOAVAILABILITY OF ELEMENTS INGESTED IN SOIL

3.1 ALKALI METALS

The alkali metals addressed here are sodium (Na), potassium (K), rubidium (Rb), cesium (Cs), and francium (Fr). Cesium is the most frequently studied alkali metal regarding bioavailability in soil. Estimates of bioavailability range from about 3% to about 20%. Cooke et al. (1997) estimated mean GI absorption of ^{137}Cs of 19% following its ingestion by sheep in clay and 3% following ingestion in a soil with high organic content. These values indicate low bioaccessibility, as Cs typically is nearly completely absorbed from food and water (ICRP, 2017). Gastrointestinal absorption of ^{134}Cs from artificially contaminated soil administered to rats was estimated as 21% (Iwata et al., 2019). Oughton et al. (1992) found that over 80% of Chernobyl-derived ^{137}Cs in Norwegian and Soviet soils was strongly attached to solids. Takahara et al. (2017) estimated that bioaccessibility of ^{137}Cs in tested soils was at most 16%. Ellickson et al. (2002) estimated bioaccessibility of 15.8 ± 8.6% (mean ± S.D.) for ^{137}Cs in soil samples from the Savannah River Site. K_d values for Cs typically are very high in all studied soil types, suggesting low bioavailability. The K_d value for Cs was the highest among 54 elements in Canadian soils studied by Sheppard et al. (2007) and fourth from highest among 52 elements in Swedish soils studied by Sheppard et al. (2009).

Reported K_d values suggest that mobility of alkali metals in soil increases with decreasing mass and ionic radius: Cs < Rb < K < Na (Sheppard et al., 2007, 2009; Vidal et al., 2009). Data on mobility of alkali metals in the surface environment (Table 1) suggest a similar ordering except that Rb and K are both categorized as having low mobility under all conditions. Overall, K_d values for K indicate moderate mobility, and comparative K_d values for K and Rb indicate greater mobility of K than Rb. K_d values for Na generally are among the lowest of the frequently studied elements, suggesting relatively high mobility in soil. Based on the above considerations, the following BF values for the alkali metals are assigned: 0.2 for Cs (and for Fr by analogy with Cs), 0.4 for Rb, 0.6 for K, and 0.8 for Na.

3.2 ALKALINE EARTH ELEMENTS

The alkaline earth elements are beryllium (Be), magnesium (Mg), calcium (Ca), strontium (Sr), barium (Ba), and radium (Ra). Strontium, Ba, and Ra are physiological analogues of Ca, especially regarding bone physiology. Strontium has chemical and physiological properties particularly close to those of Ca and strongly competes with Ca for uptake by plants and animals. Measurements by Sysoeva et al. (2005) indicate high mobility of ^{90}Sr in different soil types, with readily exchangeable portions in the range 70-90%. Oughton et al. (1992) examined the bioaccessibility of Chernobyl-derived ^{90}Sr in Norwegian and Soviet soils and found up to 70% in the extractable fractions. Ellickson et al. (2002) studied bioaccessibility of ^{90}Sr in soil samples from the Savannah River Site. The mean and median extraction levels were about 72% and 78%, respectively, for gastric extraction and 45% and 47%, respectively for intestinal extraction. Shock et al. (2007) assessed the bioaccessibility of Ba in soils near mining operations and found high variation in different soil samples, ranging from virtually zero to 66%. Calcium, Sr, and Mg are categorized in Table 1 as having moderate mobility in the surface environment; Ba and Ra are categorized as having low mobility for oxidizing conditions and moderate mobility for reducing conditions; and Be is categorized as having low mobility in oxidizing conditions at relatively high pH but is not addressed elsewhere in the table. The affinity of Ra, Ba, Sr, Ca, and Mg for ion exchange and presumably for binding to the solid phase of soil decreases in the order Ra > Ba > Sr > Ca > Mg (Iurian et al., 2015). Willett et al. (1994) investigated chemical extractability of ^{226}Ra from uranium mine tailings alone or mixed with acidic soils near the mine. Mean extractability from the tailings was 11% and in tailings mixed with acidic soils was 2-7% except for one soil in which extractability was 44%. Based on the above considerations, the following conservatively high BF values for the alkaline elements are assigned: 0.4 for Be and Ra; 0.6 for Ba; and 0.8 for Ca, Sr, and Mg.

3.3 TRANSITION METALS

3.3.1 Technetium and Rhenium

Technetium (Tc) and rhenium (Re) are unusually mobile transition metals in soil, at least under aerobic conditions. The most stable form of technetium is pertechnetate (TcO_4), which is highly mobile in aerobic soil environments such as cultivated soils (Echevarria et al., 1997). K_d values for Tc are near zero for aerobic soils (Sheppard et al., 2007). The most stable form of rhenium is perrhenate (ReO_4), a close chemical and physiological analogue of pertechnetate (ICRP, 2022). The behavior of perrhenate in soils appears to follow that of pertechnetate. In a study of soils in Canada, Re had the lowest K_d value of the 54 studied elements (Sheppard et al. 2007). K_d values for Tc are higher in anaerobic soils (e.g., wetlands and floodplains) than aerobic soils (Sheppard et al., 2007), but exposures to Tc and Re in soil seem more likely for aerobic than anerobic soils. A BF value of 1.0 is conservatively assigned to Tc and Re.

3.3.2 Cadmium and Zinc

Cadmium (Cd) and zinc (Zn) are adjacent elements in Group 2B of the periodic table. Cadmium is a toxic element but bears a physiological resemblance to the essential element Zn and can replace Zn in several

biological processes. The toxic effects of Cd may result largely from competition with Zn in biological functions (Cotzias et al., 1961; Brzoska and Moniuszko-Jakoniuk, 2001). Based on data collected from the literature, typical bioaccessibility of Cd may be around 60%. Reported measurements of bioaccessibility of Zn vary considerably and on average are lower than estimates for Cd. Padoan et al. (2021) measured the bioaccessibility of Cd, Zn, Pb, Cr, Co, Cu, and Ni in soils near two former mining sites in Italy. Average bioaccessibility was higher for Cd and Zn than for the other elements. For relatively small particles (< 10 μm), bioaccessibility of Cd varied from about 75% to nearly 100%, and bioaccessibility of Zn varied from 8% to 99%. For particle sizes 10-200 μm, bioaccessibility varied from about 68% to about 82% for Cd and from about 50% to about 99% for Zn. In a naturally contaminated reference soil from North Lincolnshire in the UK, the measured bioaccessibility was 60.3 ± 9.9% (SD) for Cd and 22.3 ± 3.7% for Zn. Plotted data of Morman et al. (2009) summarizing bioaccessibility of Cd in a collection of soils from the U.S. and Canada indicate values of about 55-70%. Schroder et al. (2003) compared the *in vitro* bioaccessibility of Cd with bioaccessibility of Cd in juvenile swine. The bioaccessibility measurements for swine, expressed as the ratio of the absorption fractions for Cd ingestion in soil and ingestion in soluble form, averaged about 63%. Results of the bioaccessibility studies of Morman et al. depended on the specific method applied but indicated a maximum bioaccessibility of 63%. Babatunde et al. (2014) estimated bioaccessibility of about 48% for Zn and 64% for Cd in soils in a region of Nigeria. Petruzzelli et al. (2020) estimated mean bioaccessibility of about 21% for Zn in soil samples collected near a cement plant. Geometric means of K_d values for Cd were similar to values for Zn in sand and organic soils (Table 3) but much lower than values for Zn in all soils combined, which is consistent with higher average bioavailability of Cd than Zn in soil. A BF value of 0.6 is assigned to Cd and Zn. This may overstate typical bioavailability of Zn but takes into consideration the high reported bioaccessibility for some soils.

3.3.3 Mercury

The bioaccessibility of mercury (Hg) in soil has been studied in a variety of soils (Davis et al., 1997; Barnett and Turner, 2001; Neculita et al., 2005; Zagury et al., 2009; Welfringer and Zagury, 2009; Gray et al., 2010; Rodrigues et al., 2014; Safruk et al., 2015; Jiang et al., 2020). Most estimates are well below 10%. The highest estimated mean bioaccessibility found in the studies cited above was 26.2% for a site polluted by chlor-alkali production, which can involve sizable releases of Hg to the environment. A conservative BF of 0.4 is assigned to Hg.

3.3.4 Yttrium and Scandium

Yttrium (Y) and scandium (Sc) are rare earth elements (REEs), a chemical family that also includes the 15 lanthanides addressed separately in this paper. The REEs are classified in Table 1 as having very low mobility in the surface environment. Loell et al. (2011) measured bioaccessibility of REEs including Y and Sc in 232 soil samples from 63 soil profiles in central Germany. The estimated mean and median estimates were 11.9% and 12.1% for Sc and 31.0% and 32.3% for Y. Jeske and Gworek (2013) studied bioaccessibility of Y and Sc in different types of soil in forest areas in Poland. The extractable portions were in the range 4-28% for Y and 3-14% for Sc. Hamilton et al. (2015) determined mean bioaccessibility of Y of 46.3% in a naturally contaminated reference soil. Based on these estimates, the assigned BF values are 0.4 for Y and 0.2 for Sc.

3.3.5 Platinum Group Metals (PGMs)

The PGMs are ruthenium (Ru), rhodium (Rh), palladium (Pd), osmium (Os), iridium (Ir), and platinum (Pt), which show similar chemical and physiological characteristics. PGMs are characterized by Garrett (2016) as having low mobility in the surface environment. Anionic forms of Ru, the most frequently studied PGM, tend to be more mobile than cationic forms due to strong adsorption of cationic forms to

soil solids (Sanzharova et al., 2009). Some PGMs may be more mobile in soil than others as indicated by comparative studies of Pt, Pd, and Rh, which are used in catalytic converters and over time are released to the environment due to deterioration of the catalyst surface (Puls et al., 2012). Rh was found to be adsorbed to soil solids to a lesser extent than Pt or Pd (Herincs et al., 2013), and Pd compounds appear to be more soluble and mobile in the environment than Pt compounds (Feichtmeier and Leopold, 2015). In a study of bioaccessibility of inhaled Pd and Pt, Puls et al. (2012) studied their extractability from simulated GI fluids assuming complete transfer of the inhaled metals to the GI tract via mucociliary clearance. The metals were collected by air sampling in an urban area polluted by traffic. The bioaccessibility of Pd and Pt was estimated as 41% and 27% for Pd and Pt, respectively. Studies by Turner and Price (2008) indicate that extractability of Rh, Pd, and Pt in simulated stomach and intestinal fluids generally was only a few percent and was far less than that of Pb (discussed in Section 3.4.1) in stomach fluids. Based on the limited available quantitative information on PGMs, it seems unlikely that they would show high bioavailability in soils. A BF value of 0.4 is assigned to PGMs.

3.3.6 Some Transition Metals with Extremely Low Mobility

Titanium (Ti), zirconium (Zr), hafnium (Hf), niobium (Nb), tantalum (Ta), and iron (Fe) are characterized in Table 1 as having very low mobility in the surface environment. This characterization is supported by K_d values published by Vidal et al. (2009) and Sheppard et al. (2007, 2009) for Zr, Hf, Nb, Ta, and Fe (no K_d value was provided for Ti). The estimated mean bioaccessibility was <1% for Ti, Nb, Zr, Hf, and Fe in a naturally contaminated reference soil (Hamilton et al., 2015). The estimated mean bioaccessibility of Ta in that soil was 16.1%, but the standard deviation was >100%. We assign a BF value of 0.2 to Ti. Zr, Hf, Nb, and Fe and, as a cautious measure, 0.4 to Ta.

3.3.7 Remaining Transition Metals

The remaining transition metals are vanadium (V), chromium (Cr), manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), molybdenum (Mo), silver (Ag), tungsten (W), and gold (Au). The reviewed bioaccessibility values for these elements generally were in the range 2-40%. For example, Izquierdo et al. (2015) estimated mean bioaccessibility of 6.3% for Cr, 28% for Ni, 32% for Cu, 34% for Co, and 40% for Mn in 48 soil samples from urban gardens in Madrid. Hamilton et al. (2015) estimated mean bioaccessibility of 1.8% for V, 16% for Ni, 17% for Cr, 17.8% for Mo, 21% for Co, 27.2% for W, 33% for Cu, and 41% for Mn in a naturally contaminated reference soil. Padoan et al. (2021) measured the bioaccessibility of Cr, Co, Cu, and Ni as a function of particle size in soils near two former mining sites in Italy. For sizes <10 µm, mean bioaccessibility (read from box plots) was roughly 30% for Cu,19% for Co, 9% for Ni, and 4% for Cr. For 10-200 µm, bioaccessibility was roughly 18% for Cu,17% for Co, and 2-3% for Ni and Cr. Ljung et al. (2007) estimated maximum mean bioaccessibility of 4.5% for Cr and 7.1% for Ni in urban playground soils in Sweden. Babatunde et al. (2014) estimated bioaccessibility of 48.2% for Cu and 37.9% for Cr in soils near an oil depot in Nigeria. Ratte (1999) concluded from a review of the literature that Ag compounds generally have low bioaccumulation by plants or animals from soils or marine environments. Dang et al. (2018) estimated bioaccessibility of Ag nanoparticles in soil to range from 0.5 to 10.9%, compared with 4.7-14.4% for $AgNO_3$. No information regarding bioavailability in soil was found for Au, but it is considered to have very low mobility under either oxidizing or reducing conditions. A cautiously high BF value of 0.4 is assigned to V, Cr, Co, Ni, Cu, W, Ag, and Au. A cautiously high value of 0.6 is assigned to Mn because of its small data set and the fact that reported bioaccessibility was never below 0.4. The value of 0.6 is also applied to Mo as a cautious measure because of its limited data and the fact that it is considered highly mobile in the surface environment at pH 5-8, which is a common pH level for soil.

3.4 BASIC METALS

3.4.1 Lead

The US EPA (2007a) reviewed and analyzed bioavailability data for Pb including several animal studies involving comparisons of the level of absorption of Pb following ingestion in soil and ingestion in a highly soluble reference material (see Casteel et al., 1997; Ruby et al., 1999; EPA, 2020; and their bibliographies). The EPA derived a rounded median bioavailability of Pb in soil of 0.6. This value is used in the EPA's Integrated Exposure Uptake Biokinetic Model for Lead in Children (IEUBK model) (SRC, Inc., 2021) and is adopted here as the BF for Pb ingested in soil.

3.4.2 Other Basic Metals

The basic metals other than Pb are aluminum (Al), gallium (Ga), indium (In), tin (Sn), thallium (Tl), and bismuth (Bi). Central K_d values found for these elements are in the range 300-31,000 (Sheppard et al., 2007, 2009; Vidal et al., 2009). Al, Ga, Sn, and Bi are characterized in Table 1 as having very low mobility in the surface environment, and In is characterized as having low mobility under oxidizing conditions (pH 5-8) and very low mobility under reducing conditions. The following estimates of bioaccessibility (mean ± standard deviation) were determined by Hamilton et al. (2015) for a naturally contaminated reference soil: Bi, 1.7 ± 0.87%; Sn, 2.6 ± 2.4%; Al, 6.7 ± 0.66%; Ga, 7.6 ± 13.8%; Tl, 14.6 ± 9.0%. Shock et al. (2007) determined bioaccessibility of Al in the range 0.31-4.0% in soils near mining operations. The estimated mean bioaccessibility of Sn was in the range 0.4%-2.75% in surface sediments from the Bernam River in Malaysia (Kadhum et al., 2017). The BF value 0.2 is assigned here to Al and Sn. The more conservative value 0.4 is assigned to Ga, In, Bi, and Tl in view of their limited data.

3.5 METALLOIDS

3.5.1 Arsenic

Arsenic (As) is the most extensively studied metalloids regarding bioavailability. The US EPA reviewed data on the comparative absorption of arsenic (As) ingested in soil or in a highly soluble reference material by human subjects, swine, monkeys, and mice (EPA, 2012). The relative bioavailability of As ingested in soil was 34.5% (95% confidence interval of 30.2–38.8, n=64) for swine, 19.2% (15.8–22.6, n=24) for monkeys, and 33.5% (27.1–39.8, n=15) for mice. EPA (2012) cited a human balance study by Stanek et al. (2010) suggesting higher relative bioavailability (~54%) of As ingested in soil (compared with bioavailability in food and beverages), but that estimate was not considered reliable by the authors of EPA (2012) due to limitations in the human study. In the present paper we adopt a BF of 0.4 for As ingested in soil as a rounded mean of the results of the studies on human subjects, swine, monkeys, and mice.

3.5.2 Other Metalloids

Metalloids other than As with potentially important radioisotopes are silicon (Si), germanium (Ge), antimony (Sb), tellurium (Te), and polonium (Po). Bioaccessibility of Sb in different soils appears to be broadly similar to that of As (Pascaud et al., 2013; Li et al., 2014; Hamilton et al., 2015). In Table 1, Sb is characterized as having moderate mobility under oxidizing conditions at pH 5-8 and very low mobility under reducing conditions; Te is characterized as having high mobility in the surface environment under oxidizing (pH 5-8) conditions and very low mobility under reducing conditions, and Si and Ge are characterized as having low mobility under all addressed conditions. K_d values for Po suggest low mobility in sand, loam, and clay, and very low mobility in soils with high organic content (Vidal et al., 2009). The following BF values are assigned: 0.6 for Te and 0.4 for Si, Ge, Sb, and Po.

3.6 LANTHANIDES

The lanthanide family consists of 15 elements: lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), and lutetium (Lu). Available K_d values for the lanthanides indicate a high percentage of adsorption to solids, with at most a few tenths of 1% typically found in the liquid phase of the soil. Loell et al. (2011) studied the bioaccessibility of all lanthanides except Pm and Ho in 232 soil samples from 63 soil profiles in central Germany. The median estimated bioaccessibility of individual elements ranged from 9.6% to 29.1% and averaged 19.1%. Hamilton et al. (2015) determined an overall mean bioaccessibility of 38.2% (range for individual elements, 34.6-42.1%) for 14 lanthanide elements (all lanthanides except Pm) in a naturally contaminated reference soil. A BF value of 0.4 is assigned to all lanthanide elements.

3.7 ACTINIDES

The actinide elements to be addressed in the ICRP's EIR series include actinium (Ac), thorium (Th), protactinium (Pa), uranium (U), neptunium (Np), plutonium (Pu), americium (Am), curium (Cm), berkelium (Bk), californium (Cf), einsteinium (Es), and fermium (Fm). Data for the actinides indicate low mobility in soil except for U, which is considered moderately mobile under oxidizing conditions, and Np, which is considered relatively mobile in soils other than highly organic soils.

K_d values for actinides addressed in Table 3 are 36 for Np, 200 for U, and in the range 740-9,300 for other listed actinides (Pu, Ac, Th, Pa, Am, Cm). The relatively high mobility of Np in soil compared with the other actinides may result from weaker bonding of its most stable oxidation state, Np (+5), to soil solids than occurs for the most stable oxidation states of the other actinides (+4 for Pu and Th, +6 for U, and +3 for the remaining actinides) (Cataldo et al., 1988; Sanzharova et al., 2009).

Rashid et al. (2022) measured GI absorption of U in rats exposed orally to U-contaminated soil. Estimated absorption to blood was in the range 0.17-0.65% of the ingested amount. Träber (2020) estimated mean absorption of U of 0.53% in 10 healthy human subjects who ingested a known mass of U in soil mixed in water, compared with the ICRP's reference f_A of 2% for relatively soluble forms of U. Yusuf et al. (2023) performed in vitro studies of bioaccessibility of U in various chemical forms (e.g., UF_6, UO_2, U_3O_8) in aerosols collected in workshops in a nuclear fuel fabrication plant. The estimated bioaccessibility was about 20% for moderately soluble compounds and 1% for relatively insoluble compounds.

In vivo studies of absorption of Pu or Am isotopes following ingestion in soil or dust by laboratory animals indicate absorption levels much lower than the ICRP reference values of 5×10^{-4} for ages 1 y or greater (and 5×10^{-3} for infants). Harrison et al. (1994) measured fractional absorption of Pu and Am ingested by rats or guinea pigs in dusts from three nuclear weapons test sites in Australia. Estimated absorption of Pu ranged from 2×10^{-6} to 1×10^{-5}. Estimated absorption of Am ranged from 3×10^{-6} to 5×10^{-5}. Cooke et al. (1997) measured GI absorption of Pu and Am in sheep following ingestion of two contaminated soils. Mean estimates were 7×10^{-5} for Pu and 4×10^{-5} for Am. Animal studies reviewed by Howard et al. (2009) also indicate extremely low absorption of Pu and Am from ingested soil.

Based on the above information, a BF value of 0.2 is assigned to Pu and Am; 0.4 is assigned to U; and 0.6 is assigned to Np. By analogy with Pu, the value 0.2 is applied to Th, which has the same frequently occurring oxidation state as Pu (+4). By analogy with Am, the value 0.2 is also applied to Ac, Cm, Bk, Cf, Es, and Fm, all of which commonly occur in the same oxidation state as Am (+3).

3.8 NONMETALS

3.8.1 Hydrogen

Hydrogen (H) as ^{3}H presumably exists primarily in the liquid phase of soil, an assumption supported by the fact that reported K_d values for ^{3}H are near zero (Serne, 2007; Vidal et al., 2009). A BF value of 1.0 is assigned to ^{3}H.

3.8.2 Carbon, Phosphorus, Sulfur, Selenium

There is little information on the bioaccessibility of carbon (C), phosphorus (P), sulfur (S), and selenium (Se). Inorganic forms of S generally are more mobile than organic forms, with sulfate being the most mobile form (Edwards, 1998). As classified in Table 1, C and S are highly mobile in the surface environment under oxidizing conditions, Se is highly mobile under oxidizing conditions at pH 5-8, and P is moderately mobile under oxidizing conditions with pH < 4 and has low mobility under other conditions. Hamilton et al. (2015) estimated bioaccessibility of Se as 57.6 ± 55.7 in a naturally contaminated reference soil. In view of the limited quantitative information on these elements, the following conservative BF values were assigned: 1.0 for C, S, and Se; and 0.8 for P.

3.8.3 Halogens

The halogens are fluorine (F), chlorine (Cl), bromine (Br), iodine (I), and astatine (At). Some reported K_d values for Cl are near zero (Serne, 2007; Vidal et al.,2009), indicating high bioavailability. Br is a close chemical and physiological analogue of Cl and is thought to be highly mobile in the surface environment (Table 1), but has higher reported K_d values (Table 3) than Cl. Fluorine is characterized in Table 1 as only moderately mobile in the surface environment. Iodine typically has low K_d values and is considered highly mobile under most conditions, including oxidizing, reducing, acidic, and alkaline environments (Plant et al., 2001). No information was found regarding At, but it is presumed to have relatively high bioavailability in view of its broad chemical and physiological similarities to I. A BF value of 1.0 is assigned to all five halogens F, Cl, Br, I, and At. This value appears to be well supported for Cl and I and is assigned as a cautious measure for their analogues F, Br, and At.

3.8.4 Noble Gases

Rn is the only noble gas addressed in the EIR series as a parent radionuclide. Rn is virtually inert and expected to escape readily from ingested soil. A BF value of 1.0 is assigned to Rn.

4. SUMMARY AND CONCLUSIONS

The default GI absorption (f_A) values used in radiation protection for ingested radionuclides generally are based on data for tracers ingested in highly soluble form. With some exceptions, these f_A values are expected to overestimate absorption of radionuclides ingested in soil because some portion of the activity in soil is bound to particles that are relatively insoluble in the GI tract. Presumably, solubilization of an ingested form of an element in the GI tract is required before the element can be absorbed through the gut wall.

This report proposes a set of element-specific multipliers, called bioaccessibility factors or BF values, for modifying the ICRP's age-specific GI absorption fractions for food and water to derive absorption fractions suitable for application to radionuclides ingested in soil. These reduction factors are based on a review of the literature on the bioavailability, bioaccessibility, and mobility of elements in soil. Considerable conservatism has been applied in many cases in view of limitations in the data including

application of *in vitro* data in lieu of measurements of bioavailability in humans or laboratory animals, reliance on sparse element-specific data in many cases, and reliance on element analogies in some cases.

The BF value assigned to each element addressed in this report is shown in Table 4. The rationale for assignment of a BF value to a given element is provided in the preceding section, in the subsection that addresses the portion of the periodic table in which that element appears.

To illustrate the application of BF values, we consider GI uptake of Ra-226. The ICRP's age-specific f_A values for radium are 0.6 for infant (age 100 d or less), 0.3 for ages 1-15 y, and 0.2 for age 25 y and up. BF for radium is 0.4 (Table 4). The f_S values for radium (GI absorption values for radium ingested in soil) are $0.4 \times 0.6 = 0.24$ for age 100 d or less, $0.4 \times 0.3 = 0.12$ for ages 1-15 y, and $0.4 \times 0.2 = 0.08$ for age 25 y and up. As in the ICRP methodology, absorption fractions for intermediate ages (e.g., an age between 15 y and 25 y) are determined by linear interpolation.

Table 5 lists the ICRP's default f_A value for workers (ICRP, 2016, 2017, 2019, 2022) for each of the 91 elements listed in Table 4 and the corresponding f_S (GI absorption) values proposed in this report for ingestion of the element in soil by an adult worker or member of the public. As illustrated above, the proposed f_S value for an element is calculated as the ICRP's default f_A value for that element multiplied by the bioaccessibility factor for the element listed in Table 4.

Table 4. Summary of assigned BF values

Group	Bioaccessibility factor (BF)	Element
1	1.0[a]	At, Br, C, Cl, F, H, I, Re, Rn, S, Se, Tc
2	0.8	Ca, Mg, Na, P, Sr
3	0.6	Ba, Cd, K, Mn, Mo, Np, Pb, Te, Zn
4	0.4	Ag, As, Au, Be, Bi, Ce, Co, Cr, Cu, Dy, Er, Eu, Ga, Gd, Ge, Hg, Ho, In, Ir, La, Lu, Nd, Ni, Os, Pd, Pm, Po, Pr, Pt, Ra, Rb, Rh, Ru, Sb, Si, Sm, Ta, Tb, Tl, Tm, U, V, W, Y, Yb
5	0.2	Ac, Al, Am, Bk, Cf, Cm, Cs, Es, Fe, Fm, Fr, Hf, Nb, Pa, Pu, Sc, Sn, Th, Ti, Zr

[a] The ICRP's f_A value for ingestion of the element in highly soluble form is left unchanged for application to its ingestion in soil.

Table 5. ICRP's default f_A values for ingestion of radionuclides by a worker compared with proposed absorption fractions for radionuclides ingested in soil by an adult

Element	ICRP	Soil	Element	ICRP	Soil
Actinium (Ac)	0.0005	0.0001	Molybdenum (Mo)	0.9	0.54
Aluminum (Al)	0.003	0.0006	Neodymium (Nd)	0.0005	0.0002
Americium (Am)	0.0005	0.0001	Neptunium (Np)	0.0005	0.0003
Antimony (Sb)	0.05	0.02	Nickel (Ni)	0.05	0.02
Arsenic (As)	1	0.4	Niobium (Nb)	0.01	0.002
Astatine (At)	1	1	Osmium (Os)	0.01	0.004
Barium (Ba)	0.2	0.12	Palladium (Pd)	0.005	0.002
Berkelium (Bk)	0.0005	0.0001	Phosphorus (P)	0.8	0.64
Beryllium (Be)	0.005	0.002	Platinum (Pt)	0.01	0.004
Bismuth (Bi)	0.05	0.02	Plutonium (Pu)	0.0005	0.0001
Bromine (Br)	1	1	Polonium (Po)	0.1	0.04
Cadmium (Cd)	0.05	0.03	Potassium (K)	1	0.6
Calcium (Ca)	0.4	0.32	Praseodymium (Pr)	0.0005	0.0002
Californium (Cf)	0.0005	0.0001	Promethium (Pm)	0.0005	0.0002
Carbon (C)	1	1	Protactinium (Pa)	0.0005	0.0001
Cerium (Ce)	0.0005	0.0002	Radium (Ra)	0.2	0.08
Cesium (Cs)	1	0.2	Radon (Rn)	1	1
Chlorine (Cl)	1	1	Rhenium (Re)	0.9	0.9
Chromium (Cr)	0.01	0.004	Rhodium (Rh)	0.05	0.02
Cobalt (Co)	0.1	0.04	Rubidium (Rb)	1	0.4
Copper (Cu)	0.5	0.2	Ruthenium (Ru)	0.05	0.02
Curium (Cm)	0.0005	0.0001	Samarium (Sm)	0.0005	0.0002
Dysprosium (Dy)	0.0005	0.0002	Scandium (Sc)	0.001	0.0002
Einsteinium (Es)	0.0005	0.0001	Selenium (Se)	0.8	0.8
Erbium (Er)	0.0005	0.0002	Silicon (Si)	0.5	0.2
Europium (Eu)	0.0005	0.0002	Silver (Ag)	0.05	0.02
Fermium (Fm)	0.0005	0.0001	Sodium (Na)	1	0.8
Fluorine (F)	1	1	Strontium (Sr)	0.25	0.2
Francium (Fr)	1	0.2	Sulfur (S)	1	1
Gadolinium (Gd)	0.0005	0.0002	Tantalum (Ta)	0.001	0.0004
Gallium (Ga)	0.001	0.0004	Technetium (Tc)	0.9	0.9
Germanium (Ge)	1	0.4	Tellurium (Te)	0.3	0.18
Gold (Au)	0.1	0.04	Terbium (Tb)	0.0005	0.0002
Hafnium (Hf)	0.002	0.0004	Thallium (Tl)	1	0.4
Holmium (Ho)	0.0005	0.0002	Thorium (Th)	0.0005	0.0001
Hydrogen (H)	1	1	Thulium (Tm)	0.0005	0.0002
Indium (In)	0.005	0.002	Tin (Sn)	0.02	0.004
Iodine (I)	1	1	Titanium (Ti)	0.001	0.0002
Iridium (Ir)	0.01	0.004	Tungsten (W)	0.5	0.2
Iron (Fe)	0.1	0.02	Uranium (U)	0.02	0.008
Lanthanum (La)	0.0005	0.0002	Vanadium (V)	0.2	0.08
Lead (Pb)	0.2	0.12	Ytterbium (Yb)	0.0005	0.0002
Lutetium (Lu)	0.0005	0.0002	Yttrium (Y)	0.0001	0.00004
Magnesium (Mg)	0.5	0.4	Zinc (Zn)	0.5	0.3
Manganese (Mn)	0.05	0.03	Zirconium (Zr)	0.002	0.0004
Mercury (Hg)	0.1	0.04			

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