ALIMENTARY TRACT ABSORPTION (f_1 VALUES) FOR RADIONUCLIDES IN LOCAL AND REGIONAL FALLOUT FROM NUCLEAR TESTS

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Abstract—**This paper presents gastrointestinal absorption fractions (f_1 values) for estimating internal doses from local and regional fallout radionuclides due to nuclear tests. The choice of f_1 values are based on specific circumstances of weapons test conditions and a review of reported f_1 values for elements in different physical and chemical states. Special attention is given to fallout from nuclear tests conducted at the Marshall Islands. We make a distinction between the f_1 values for intakes of radioactive materials immediately after deposition (acute intakes) and intakes that occur in the course of months and years after deposition, following incorporation into terrestrial and aquatic foodstuffs (chronic intakes). Multiple f_1 values for different circumstances where persons are exposed to radioactive fallout (e.g., local vs. regional fallout and coral vs. continental tests) are presented when supportive information is available. In some cases, our selected f_1 values are similar to those adopted by the International Commission on Radiological Protection (ICRP) (e.g., iodine and most actinides). However, f_1 values for cesium and strontium derived from urine bioassay data of the Marshallese population are notably lower than the generic f_1 values recommended by ICRP, particularly for acute intakes from local fallout (0.4 and 0.05 for Cs and Sr, respectively). The f_1 values presented here form the first complete set of values relevant to realistic dose assessments for exposure to local or regional radioactive fallout.**

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Key words: fallout; Marshall Islands; radionuclide; nuclear weapons

INTRODUCTION

FROM 1946 through 1958, 66 nuclear weapons tests were conducted by the U.S. in or near the Marshall Islands located in equatorial waters in the Pacific. During roughly the same period (1951–1958), the U.S. also conducted 119 nuclear tests at the Nevada Test Site (NTS) (U.S. DOE 1988). All together, over 500 atmospheric nuclear weapons tests took place at various sites around the world from 1945 to 1980 (Beck and Bennett 2002). These tests resulted in the deposition of significant quantities of radioactive debris to the local, regional and global environments resulting in human health consequences that have ensued over the decades since (Simon 1997; Simon et al. 2006). Since nuclear testing ceased, a number of dose reconstruction assessments have been conducted for specific populations (Beck et al. 2006). Depending on the level of detail considered and the degree of realism sought in assessment calculations, dose estimation parameters may require evaluation and interpretation based on the available historical data for each assessment.

The assessment of internal dose from the ingestion of radionuclides for the general public requires knowledge on the fraction of the radioactive material absorbed from the alimentary tract to blood, using a model parameter usually termed the "f_1 value." Literature information for f_1 values vary significantly depending on the origin of the measurements or assumptions made about the chemical and physical properties of the ingested form in addition to other biological and environmental factors. Metabolic investigations on radionuclide uptake using soluble compounds used in laboratories and some industries often produced higher f_1 values compared to other less soluble forms in both animal and human studies. Typically, the International Commission on Radiological Protection (ICRP) and other radiation protection organizations tend to adopt conservative f_1 values to ensure a margin of safety for protection of humans exposed to a wide range of radionuclide forms in the workplace and to members of the public. The use of generic f_1 values, however, may not be desirable in dose reconstructions, particularly where "best

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estimate" values are needed to calculate realistic population-average or typical individual doses.

The purpose of this paper is to (a) review and evaluate historical data on the physical and chemical characteristics of radionuclides associated with fallout debris including fractionation, solubility, and other factors which may affect their absorption by humans; (b) review and evaluate f_1 values for key fallout radionuclides from the available literature as they pertain to fallout in general and to the unique coral environment of the Marshall Islands, in particular; and (c) provide an evaluation and present our "best estimates" for f_1 values for use in reconstruction of realistic internal doses from acute intake of local and regional fallout from nuclear tests, and for chronic intake of radioactive materials following their incorporation into terrestrial and aquatic foodstuffs. In this paper, local fallout is assumed to have occurred at close-in locations that are typically associated with large particle sizes (≥ 50 μm) and extend roughly up to 300 km from ground zero, whereas regional fallout that occurs at distant locations is composed of only smaller particle sizes and is assumed to extend from 300 to 3,000 km from ground zero.

Justifications for the choice of f_1 values for fallout radionuclides were made taking into account the particular circumstances of various nuclear test conditions and the expected nuclides' solubility and biological availability, whenever such information was available. Information presented in this paper resulted from a thorough review of the available fallout literature, declassified government documents, and various reports from national and international organizations. Although emphasis was given to nuclear weapons testing conducted at the Marshall Islands, information provided here may be applicable to fallout from tests or detonations conducted elsewhere.

A large number of fission and activation products are formed in nuclear weapons tests (Hicks 1981). Most of them are either produced in low yield or have short half-lives. Key fallout radionuclides that contribute substantially to the internal dose to humans resulting from nuclear tests conducted in the Marshall Islands are isotopes of iodine (I), tellurium (Te), cesium (Cs), strontium (Sr), yttrium (Y), molybdenum (Mo) barium (Ba), lanthanum (La), cerium (Ce), praseodymium (Pr), iron (Fe), cobalt (Co), zinc (Zn), and neptunium (Np) (Simon et al. 2010). Plutonium (Pu), americium (Am), uranium (U), zirconium (Zr), ruthenium (Ru), rubidium (Rb), and bismuth (Bi) are among other elements of interest present in radioactive fallout. Review and evaluation of f_1 values for the above elements from exposure to fallout are addressed in this document.

BACKGROUND AND METHODS

Unique characteristics of the Pacific Proving Ground

The basic components of topsoil at coral atolls like the Marshall Islands are coral-based sand, hard and weathered coral, and marine shells. Coral-based soil is typically low in organic matter, potassium, clay, and many other nutrients, but enriched in lime ($CaCO_3$) content (Simon et al. 2002).

In the case of detonations over coral or coral-based soils, calcium carbonate in the ground is heated to very high temperatures or completely vaporized, resulting in the formation of relatively fine particles. When such particles mix with bomb debris in the cooling fireball, they adsorb fission radionuclides within and on the surface of particles (Norman 1973). Fresh coral debris is largely CaO, $CaCO_3$, and $Ca(OH)_2$ (Heidt et al. 1953; Maxwell et al. 1955) and, hence, raises the pH considerably (pH 10–11) when it dissolves in water (Crocker et al. 1965). The maximum soil adsorption and retention of Sr, Cs, Y, Ce, and Pu also occurs at this high pH range. In addition, lime rich soil may further increase the retention of Sr and Cs (NAS 1963). In fallout particles from water-surface tests, radioactivity is carried in water droplets which form a cloud of mist after detonation. In the presence of sea water, some fission products may be fused in sodium oxide particles or precipitated as sulfate on particle surfaces (discussed in detail later). For tests conducted on barges or towers, iron from the steel structures can incorporate in fallout particles to form mixed iron, calcium, and fission-products oxides (Adams et al. 1958). Small fallout particles from coral islands were found to be 10–50% water soluble and almost completely soluble in acids (Cohn et al. 1954; Heidt et al. 1953; and personal experience).

Fractionation, solubility, and bioavailability of fallout debris

Fractionation and solubility of fallout debris are important phenomena because of their relationship to bioavailability and the absorption of various radioactive species by humans. The fractionation occurs due to both physical and chemical separation of fallout particles and/or fission products. The degree of fractionation is a function of many interacting factors including the physical and chemical properties of nuclides formed in the fission process as well as those of progeny elements in a given decay chain, particle size and composition, distance from ground zero, the specific conditions of nuclear detonation (weapon composition, yield, burst height, meteorological conditions, etc.), and the properties of the surrounding environment after the deposition of fallout particles (Tompkins and Werner 1955; Beck

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and Bennett 2002). In general, fractionation decreases with the magnitude of the device yield and increases with proximity of the detonation to the ground surface.

Radionuclides will fractionate to varying degrees due to differences in fission products' volatilities. In the fireball, bomb material (FeO) and earth/water-surface components form molten droplets in which refractory elements dissolve and distribute uniformly through the particles. As the fireball cools (below 1,500°C) and the particles form and solidify, the more volatile elements and their progeny condense and distribute according to the available particle surface, where the amount of activity varies with r^2 (Adams et al. 1958; Crocker et al. 1965; Hicks 1982). The degree of fractionation can then be defined as the ratio of refractory (R) to volatile (V) nuclides, where an *R/V* value of 1 represents the relative ratio of unfractionated debris (Beck et al. 2010). Various particles in the debris can also agglomerate to form a complex heterogeneous mix of particles of different shapes and sizes (Norman and Winchell 1970; Vajda 2001). ^{89}Sr, ^{137}Cs, and ^{90}Sr are examples of radionuclides that, having gaseous precursors, are subject to additional fractionation and, thus, are partially depleted in local fallout occurring at close-in locations compared to other volatile nuclides. ^{95}Zr and ^{144}Ce are refractory in fallout and are frequently used as reference radionuclides to determine the degree of fractionation for other nuclides. In general, volatile radionuclides exhibit a high degree of solubility relative to the refractory radionuclides.

In terms of particle size distribution, the fallout process has the effect of sorting the debris by diminishing particle size along the forward path of the cloud. Consequently, the refractory and volatile phases are also partially separated over the initial period following detonation. It has been observed that fallout particle size and total radioactivity are correlated and both decreased with distance after the 1954 Bravo test at Bikini Atoll (Lessard 1986). About 20–30% of the total fallout activity is deposited at close-in locations and is mostly associated with large particles dominated by the refractory phase. About 90% of the more volatile nuclides deposit on small particles at more distant locations. This is particularly evident in high-yield explosions with a high cloud top, often moving at high velocity due to upper atmosphere winds. For low-yield explosions, at low cloud height and low wind velocity, a significant fraction of small particle size can deposit near ground zero. As with distance, time of arrival of fallout may also be a surrogate for change in particle size (Simon 1990; Anspaugh 2004).

Detonations at altitudes sufficiently high to eliminate incorporation of ground soil into the fireball tend to produce small, spherical and highly radioactive particles with activity distributed more evenly throughout particle volume. These particles tend to be more soluble than for ground surface detonations (Crocker et al. 1965). If soil and other on-site materials are incorporated in the fireball, the resulting particles tend to be large in size, of irregular shape, and of low specific activity.

Solubility and leachability of radionuclides from fallout particles are governed by several factors including the following: a) the physical and chemical characteristics of the host particle (size, surface area, surface chemistry, surface orientation of radionuclides, age of fallout material, etc.); b) radionuclide speciation and oxidation states; c) conditions associated with detonation including the degree of fractionation during particle formation; and d) environmental factors including soil/water chemistry. The most extensively investigated solvents used in leaching studies have been water, sea water, and 0.1 N HCl (Crocker et al. 1965). Dilute HCl is generally considered as a simulant for stomach fluids. Data on leaching of fallout debris have usually been reported in terms of the fraction of gross beta or gross gamma activity found in the soluble phase. Results in terms of individual radionuclide solubility are much more useful, but these data are very scarce (Anspaugh 2004).

Solubility of fallout particles in HCl at pH 1.0 from the NTS was greater for smaller particles (LeRoy et al. 1966). Reducing particle size by approximately one-half resulted in an increase of the fraction leached activity by a factor of two (Table 1). Also, acid leaching was much more effective in dissolving fallout particles than was water or alkaline solutions and solubility increased with multiple extractions and the duration of contact. Measurements of total gamma activity leached by water from coral samples under different test conditions were reported by Crocker et al. (1965) and the information is summarized in Table 2. In surface-water tests, fallout particles were much more water soluble than from ground (coral)-surface tests by approximately an order of magnitude. Data from the NTS (Baurmash et al. 1958) on fallout particle solubility in 0.1 N HCl showed that, in tower tests, when the fireball interacted with surface material, the soluble fraction of the formed material was much lower (0.20–0.30) than for higher altitude air

Table 1. Solubility of total radioactivity from local fallout as a function of particle size; data from the Nevada Test Site (NTS) on particles collected six days after detonation (from LeRoy et al. 1966).

Particle size (μm)	Fraction of activity leached in HCl at pH 1.0
~500	~0.06
180	0.19
90	0.42

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Table 2. Total gamma activity leached by water from local fallout particles as a function of test conditions and leaching time (Crocker et al. 1965).

Test condition	Leaching time in days	Fraction dissolved
Coral-surface tests	4–8	0.08
Coral-surface tests	20	0.23
Surface-water (barge) tests	2–3	0.60–0.70

Table 3. Solubility of various radionuclides from simulated fallout particles (from LeRoy et al. 1966).

Particle characteristics[a] used in the study	Soluble fraction leached out in 0.1 N HCl (or 0.1 N NaOH) ^{85}Sr	^{134}Cs	^{133}Ba
Local fallout simulated particles:			
Microspheres of fabricated glass particles containing carrier-free tracers, where 10% of activity could be leached out in dilute HCl	0.06 (0.08)	0.01 (0.05)	0.05 (0.07)
Distant location fallout simulated particles:			
Fabricated particles containing carrier tracers, Sr and Ba in oxide form and Cs as silicate. The mix was calcined at 1,000°C, 50–100% of activity could be leached out in dilute HCl	0.72 (0.15)	0.38 (0.12)	0.91 (0.23)

[a] Particle size ranged from 30–40 μm and particle specific activity was about 1.85 MBq per gram.

balloon tests (0.65–0.85), where the fireball did not interact with the ground surface. Particle solubility in neutral, alkaline and acid solutions from continental detonations ranged from less than 0.01 to 0.5 of total radioactivity depending on particle size and location with respect to the detonation site (LeRoy et al. 1966). Russian and U.S. studies, following ground-surface tests, determined that the soluble fraction of fallout debris in the close-in zones within 25 km from point of detonation generally ranged from 0.005–0.10 of total radioactivity (Cohn et al. 1954; Gordeev 1999). Surface orientation of individual nuclides in fallout particles plays a dominant role in the degree of leaching (Norman and Winchell 1970). Therefore, volatile nuclides absorbed preferentially on particle surfaces are expected to be more soluble than refractory nuclides. In summary, as the distance from ground zero increases, the degree of fractionation (*R/V*) decreases, the particle size of deposited fallout decreases, and the solubility increases.

One of the few radionuclide-specific solubility studies was conducted using simulated fallout particles spiked with ^{85}Sr, ^{134}Cs, and ^{133}Ba tracers (LeRoy et al. 1966). For simulated local fallout (close-in) particles, the fraction leached by 0.1 N HCl or 0.1 N NaOH was very small, ranging from 0.01 to 0.08 for those tracers. For simulated particles of distant fallout, 0.38, 0.72, and 0.91 of the total activity was leached by acidic solution for Cs, Sr, and Ba tracers, respectively. Much less activity was leached by alkaline solution, however (Table 3). In the environment, ^{239,240}Pu was measured in seawater and lagoon sediment from the Bikini and Enewetak Atolls where the nuclear tests in the Marshall Islands were conducted (Noshkin et al. 1987). The fraction of plutonium mobilized by seawater at any given time was estimated at about 0.001 of the total sediment inventory.

Following contact of fallout debris with the surroundings, additional nuclide separation can occur. For example, soluble radionuclides may be preferentially leached out by sea water in deep and surface water detonations. It was found that the adsorption of ^{90}Sr ions onto soil decreases with increasing soil acidity (Maxwell et al. 1955; Adams et al. 1958). The presence of phosphate ions in the soil resulted in markedly increased adsorption of strontium onto soil particles. Mild corrosion of the metallic surfaces of fallout particles could, over time, increase surface area, creating more favorable conditions for radionuclide leaching. In general terms, the interaction of radionuclides with various environmental conditions can either decrease or increase their solubility/mobility and biological availability over time.

The ratio of biologically available activity to total radioactivity in local fallout is reported to be generally small. LeRoy et al. (1966) fed 10 healthy human volunteers 1-wk-old particulate fallout collected about 1.5 km following the land-surface detonation of the 1962 Small Boy at the NTS. An average fraction of only 0.032 of the total gamma radioactivity was absorbed by the alimentary tract; the range was 0.003 to 0.088. Some of the observed variability in alimentary tract absorption in that study was due to several factors including large variation in fallout particle sizes and radionuclide content, uncertain whole-body counting geometry, as well as variation among individuals in terms of age, sex, diet, and lifestyle.

Absorption of radionuclides by humans, either through ingestion or inhalation, is likely controlled by dissolution of the fallout particle matrix. Therefore, radionuclide absorption in humans from fallout particles may be different from that generally associated with simple compounds of a given element as often used in most animal uptake studies (ICRP 1996). It should be recognized, however, that the biological availability of fallout debris as expressed by various transfer parameters (e.g., f_1 values) are element-specific and cannot be simply predicted on the basis of solubility tests alone

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using various leaching solvents (in vitro). It should be also noted that the gastrointestinal absorption of radionuclides incorporated in terrestrial and aquatic foodstuffs (ingested during chronic intakes) are not necessarily correlated with the properties of the same radionuclides immediately after deposition (ingested during acute intakes).

Selection of f_1 values

In this report, the literature information on alimentary tract absorption (f_1) for key fallout radionuclides was carefully examined. Although f_1 values assigned by ICRP for workers or members of the public are convenient to use in dose assessments for radiation protection purposes, radionuclide absorption in man may be profoundly modified by such factors as (a) the physical and chemical characteristics of fallout particles; (b) absorbability of the chemical compounds formed in the alimentary tract; and (c) the transit time of fallout material through the human digestive system (LeRoy et al. 1966). The ICRP (1986) stated that where specific information for a given material is available, this should be used to derive separate f_1 values for specific compounds or circumstances.

Thus, the approach used in the following sections was to evaluate the available information in order to arrive at the most realistic f_1 values for fallout radionuclides from various nuclear tests, with particular focus on local and regional fallout tests conducted at the Marshall Islands for use in internal dose assessments (Simon et al. 2010). Furthermore, f_1 values for Cs and Sr derived from urine bioassay data of the Rongelap residents following the Bravo test (presented next) were given considerable weight in the analysis presented here for these radionuclides. Our approach represents what may be considered a "best estimate," since the f_1 value(s) is selected to represent the average feature of a given nuclide metabolic behavior and not an exceptionally high or low value of the parameter. During the process of evaluation and selection of f_1 values for the particular situation, adequate amounts of objective information were not always available and some judgment, based on our experience were applied. In some cases, selected f_1 values presented in this document were similar to the values adopted by the ICRP. In other cases, our proposed values were different after considering the specifics of the nuclear tests, the nature and radiochemical properties of fallout particles, expected solubility, distance from ground zero, and other considerations. Sometimes, more than one f_1 value was recommended for the different exposure circumstances. This approach was taken when sufficient information was available to justify any changes from the ICRP recommendations.

In this paper, we present specific f_1 values for deriving acute intakes occurring immediately after fallout under different test conditions (coral vs. continental tests) and for various distances from ground zero (close-in vs. distant locations, that is, for local fallout vs. regional fallout), as well as for chronic intakes occurring months to years after fallout resulting from ingestion of fallout nuclides incorporated in foodstuffs or ingested by inadvertent intake of soil. The local fallout area, which corresponds to close-in locations, is defined here as the area where all particles with diameters greater than 50 μm are deposited. For high-yield tests conducted in the Marshall Islands, the local fallout area roughly extended to about 300 km downwind from ground zero. Regional or tropospheric fallout consists of particles with diameters smaller than 50 μm, which are more soluble than local fallout particles. In the regional fallout area, the degree of fractionation (*R/V*) is assumed to be constant and equal to 0.5. In this paper, regional fallout is considered to extend to a distance of about 3,000 km (representing the size of the territory of the Marshall Islands), although, technically, it covers the entire world as it is due to fallout occurring in the tropospheric part of the radioactive cloud, which circles the Earth for the first time in about 2 to 3 wk. Because of the very small size of land area at the detonation sites of the Pacific Proving Ground (Bikini and Enewetak), the relative fractions of coral soil and sea water incorporated into fallout debris were likely to be similar for most ground and water (sub and surface) tests. Therefore, no distinction was made to our selection for f_1 between land and water tests.

There is indirect and limited evidence from some animal studies that fasting conditions prior to ingestion could increase the absorption of certain radionuclides through the alimentary tract. Therefore, fasting f_1 values were proposed here for some nuclides when such information was available in the literature. In terms of age-specific f_1 values, we suggest complying with the recommendations given in recent ICRP documents (ICRP 1993, 1996).

Uptake of fallout radionuclides ingested by the Marshallese

Pooled urine samples from adults were collected from residents of Rongelap within three weeks after intake of fallout from the test Bravo and in the several months following for radiochemical analysis to determine the degree of internal contamination (Harris et al. 2010). The earliest samples of urine in which ^{131}I and other nuclides were measured were obtained by Harris of Los Alamos Scientific Laboratory (Harris 1954; Harris et al. 2010). We examined these data sets, referred to here as the (1) Walter Reed data (Woodward et al. 1959), (2)

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the U.S. NRDL data (acronym for U.S. Naval Radiological Defense Laboratory, cited as U.S. AEC 1956), and (3) the Harris data (Harris 1954), for purposes of determining the availability and alimentary tract absorption of fallout radionuclides at close-in distance (the sites of exposure being between 150 km and 210 km from the detonation point). Only intake by ingestion is considered, as it is the primary pathway of acute internal exposure due to the dominance of larger particles in local fallout (Simon et al. 2010).

Among the three data sets, there were six radionuclides assayed though no single data set reported all of them. Our evaluation of the validity and quality of the three sets of urine measurements suggested that ^{131}I, ^{137}Cs, and ^{90}Sr from Walter Reed and from Harris were of good quality and had reasonably low uncertainties. Measurements of ^{89}Sr by U.S. NRDL and Harris were satisfactory but had significantly larger relative uncertainties. Both ^{140}Ba and ^{103}Ru data were likely not reliable due to the lack of specific radiochemical separations and the detection methods used in the bioassay.

We selected the measurement of ^{131}I in urine as likely to have been the most precise because of reliable measurement techniques which verified the half-life of the activity (Harris 1954; Harris et al. 2010). Moreover, ^{131}I measurements were selected because all literature supports the notion of complete absorption of radioiodines in the alimentary tract (presented later) for all commonly occurring forms [i.e., $f_1(^{131}\text{I}) = 1$], thus giving us a basis for interpreting the bioassay measurements of other radionuclides. As indicated in Harris et al. (2010) and Simon et al. (2010), the bioassay measurements of ^{131}I in pooled urine samples were used to derive a population average acute intake, $\bar{Q}$ (Bq), of ^{131}I at Rongelap as follows:

$$\bar{Q}(^{131}\text{I}) = \frac{A(^{131}\text{I}, t_s)}{EF(^{131}\text{I}, t_s)}, \tag{1}$$

where $A(^{131}\text{I}, t_s)$, in Bq, is the activity of ^{131}I in urine in a 24-h urine sample at the time of sampling, t_s, in days, and $EF(^{131}\text{I}, t_s)$, unitless, is the fraction of ingested ^{131}I excreted in urine on day of sampling, t_s, following the intake of fallout particles, and is inferred from the metabolic model of iodine described in Simon et al. (2010).

For radionuclides other than ^{131}I, denoted as i in eqns (2), (3), and (4), the f_1 values cannot be assumed to be equal to unity. In that case,

$$EF(i, t_s) = f_1(i) \times ET(i, t_s), \tag{2}$$

where $ET(i, t_s)$ is the fraction of ingested radionuclide i that is totally absorbed in the alimentary tract and that is excreted in urine on day of sampling, t_s, following intake.

Combining eqns (1) and (2) allows us to estimate f_1 for any radionuclide i that was assayed in the urine samples:

$$f_1(i) = \frac{A(i, t_s)}{\bar{Q}(i) \times ET(i, t_s)}, \tag{3}$$

where the intake, $\bar{Q}(i)$, for any radionuclide i, was calculated as:

$$\bar{Q}(i) = \bar{Q}(^{131}\text{I})\left[\frac{Dep(i)}{Dep(^{131}\text{I})}\right], \tag{4}$$

and $Dep(i)$ is the deposition density (Bq m^{-2}) of radionuclide i from fallout, calculated according to Beck et al. (2010), while $ET(i, t_s)$ was derived from ICRP models [ICRP Report 56 (1989) for ^{137}Cs and Report 67 (1993) for ^{90}Sr, ^{131}I, ^{140}Ba, and ^{103}Ru].

Using the ICRP models to estimate the excretion fractions for total absorption of ^{137}Cs, ^{90}Sr, ^{140}Ba, and ^{103}Ru on the days of sampling, the bioassay data from Woodward et al. (1959), U.S. Atomic Energy Commission (U.S. AEC 1956), and Harris (1954), as well as the assumption that f_1 for ^{131}I is equal to unity (even when ingested as fallout particles), we estimated the f_1 values for ^{137}Cs, ^{90}Sr, ^{89}Sr, ^{140}Ba, and ^{103}Ru ingested in the form of fallout particles at close-in distances from ground zero. The bioassay data are discussed in Harris et al. (2010) and the deposition data are presented in Beck et al. (2010). The results of calculations to estimate f_1 for those four radionuclides are presented in Table 4.

As shown in Table 4, we estimated an f_1 value for ^{137}Cs of 0.44 and an average f_1 value for both ^{89}Sr and ^{90}Sr equal to 0.02. In addition, we estimated an f_1 of 0.031 for ^{140}Ba similar to that for Sr and a value of $\sim 1 \times 10^{-4}$ for ^{103}Ru. The f_1 value estimated for ^{103}Ru is much smaller than literature values (presented later). However, as discussed earlier, the reliability of the ^{140}Ba and ^{103}Ru

Table 4. Estimated f_1 values based on urine bioassay measurements of the Marshallese population on Rongelap Atoll following the Bravo test.

	Data set (see text)		
Radionuclide	USNRDL	Walter Reed	Harris
^{137}Cs	—	0.440	—
^{89}Sr	0.020	—	0.020
^{90}Sr	—	0.021	—
^{131}I	—	—	1.0[a]
^{140}Ba	0.031[b]	—	0.031[b]
^{103}Ru	—	—	~0.0001[b]

[a] Assumed based on literature information.

[b] Estimates may not be reliable since bioassay measurements may not be reliable (see text).

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measurements is in doubt. The opportunity afforded by the historical bioassay data to estimate f_1 for close-in fallout, particularly for ^{137}Cs, and ^{89}Sr and ^{90}Sr, is unique. For this reason, the derived cesium and strontium f_1 values are given considerable weight in the analysis presented here.

LITERATURE REVIEW AND SELECTED f_1 VALUES

In the following sections, we discuss individual elements and the available data on each from which our "best estimate" of the f_1 values for radionuclides in particulate fallout were made.

Iodine (I)

Iodine is a highly fractionating element in fallout debris due to its volatility and, hence, its tendency is to deposit on the surface of particles rather than throughout them (Norman 1973). Exposure to direct environmental conditions, including sunlight, greatly accelerates iodine release from fallout particles (Norman 1973). After detonation, its isotopes are probably present in free form in the gaseous cloud or condensed onto the surface of small fallout particles. Over time, iodine is expected to deposit on the ground and become attached to solid and liquid particulates. Iodine isotopes are readily soluble and mobile in the environment and exhibit high accumulation ratios through the food chain, including milk (Whicker and Schultz 1982). Iodine is an essential nutrient and, when assimilated by mammals, it concentrates primarily in the thyroid gland.

^{131}I was readily detected in urine of people residing at the Marshall Islands in urine samples collected 16 d after their exposure to Bravo fallout debris (Harris 1954; Harris et al. 2010). Measurable concentrations of ^{131}I have also been reported in human and animal thyroids from the U.S. and other countries as a result of global fallout during the years of worldwide atmospheric nuclear weapons testing (Van Middlesworth 1963).

In humans and animals, absorption of I isotopes in the alimentary tract is almost complete when ingested in a wide range of forms including aqueous solutions, milk, and incorporated in foodstuffs. Studies of ingestion of local fallout particles from land-surface detonations indicate that most ^{131}I is available for alimentary tract absorption by humans (LeRoy et al. 1966). Literature review by Hamilton (1944, 1948) also supports a complete oral absorption of iodine in the rat. Lessard (1986) reported that the ingestion of Bravo debris resulted in an efficient release of radioiodine trapped in fallout particles due to the acid environment of the human stomach. An f_1 value of 1.0 for radioiodines has been recommended by various organizations for both occupational and environmental exposures for all ages (ICRP 1979, 1989, 1996; Kendall et al. 1988; U.S. EPA 1988).

Selected f_1 values for iodine. All literature information support complete absorption for all commonly occurring forms of iodine. An f_1 value of 1.0 should apply well to most fallout conditions including tests conducted at the Marshall Islands. We, therefore, chose f_1 to be unity for most types of nuclear tests and at all distances from ground zero for both acute and chronic intakes of I. This f_1 value is also chosen for all ages and for direct ingestion of coral particles.

The exception to complete alimentary tract absorption of I might be for local fallout from land tests involving silicate soil. Fractional release of radioiodine from fallout particles is a function of the particle diameter, diffusion coefficient, and time. It has been proposed that iodine fraction released from silicate particles resides within 0.1 μm of the surface (Norman and Winchell 1970). For ^{131}I, about 30% may be locked inside large particles (Norman and Winchell 1970) and therefore unavailable either for vapor release or water solubility. Based on this information, it is appropriate to set a lower f_1 value of 0.7 for local fallout resulting from ground bursts on silicaceous soil, such as at the NTS and other similar sites.

Tellurium (Te)

^{132}Te is an important radionuclide in fallout and is the precursor for ^{132}I. In the fireball, tellurium is a volatile and moderately fractionating element (Freiling 1961). Therefore, it is likely to become condensed onto fallout particle surfaces as the fireball cools following the detonation. In fallout, Te is expected to be in the oxide form (TeO_2 and/or TeO_3); both are water soluble. The metabolism and alimentary tract uptake of Te has been reported only in a few animal species. In a review by Hamilton (1948), the f_1 for tellurium was reported to be about 0.25 in rats. Mullen and Stanley (1974) reported that 0.25 of tellurate fraction was absorbed from the alimentary tract of dairy cows. The ICRP (1979) adopted an f_1 value of 0.2 for acute intakes by workers and 0.3 for chronic intakes by the general public (ICRP 1993, 1996).

Selected f_1 value for tellurium. Due to the lack of information on Te absorption in humans, we suggest using the f_1 values adopted by ICRP for both acute and chronic intakes (0.20 and 0.30, respectively). It is possible that a small fraction of Te could be trapped in large fallout particles, particularly if refractory silicates and oxides are incorporated, as in the case of some ground bursts, which may reduce its solubility and alimentary

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tract absorption. It is unlikely, however, that the bound fraction of Te is significant enough to impact its absorption from coral particles.

Cesium (Cs)

Because of its relatively high fission yield and long half-life (~30 y), ^{137}Cs is one of the most abundant radionuclides in fallout debris. It is a highly fractionating isotope in fallout due to its gaseous precursor ^{137}Xe and adsorbs onto particle surfaces to a large extent. It is readily mobile in the environment and tends to accumulate in biota. Cesium is a chemical analog of potassium, which is a major nutrient element. Absorbed Cs is distributed uniformly in the human body and its retention tends to increase with the intake of potassium (Leggett 1986).

The absorption fraction of Cs from the alimentary tract to blood varies greatly with the physical and chemical forms, potassium ingestion rate, and animal species, among other factors. Human and animal data indicate that soluble salts (i.e., CsCl) are efficiently absorbed from the alimentary tract (ICRP 1979; NCRP 2006). However, Cs can bind to certain clay minerals, where its environmental mobility may be significantly reduced. Lime was found to increase Cs retention in soil and thus may reduce its solubility and bioavailability in the environment (NAS 1963).

A sample of f_1 values for Cs from the literature is provided in the Appendix. Reported f_1 values for various ingested forms ranged from 0.2 to 1.0 in humans and animal species. While the ICRP (1979, 1989, 1996) adopted an f_1 value of 1.0 for both acute and chronic intakes for workers and the general public, the literature data indicate that the uptake of Cs may not always be complete. The wide range of f_1 values reported could be attributed to the wide range of the ingested Cs forms used in the various studies. The reported f_1 value of 0.9 for ^{137}Cs from the ingestion of contaminated food items by a U.S. native (Hardy et al. 1965) is based on measurements of the urinary to fecal excretion ratio to estimate the uptake in a single individual, many years after the nuclear tests were carried out at the Marshall Islands and, therefore, uncertain.

Selected f_1 values for cesium and rubidium. Our "best estimate" of f_1 values for Cs and Rb under various types of intake conditions are presented in Table 5. For acute intakes from local fallout, we chose an f_1 of 0.4 based on the value estimated for ^{137}Cs from the Marshallese urine bioassay data following exposure to local fallout from the Bravo test (Table 4). This estimate is also consistent with the f_1 value reported by LeRoy et al. (1966) on the absorption of Cs from ingested local fallout simulated particles in humans (Appendix). The same f_1 value (0.4) is chosen for direct intake when contaminated soil is inadvertently ingested by humans. For acute intakes from regional fallout and for chronic intakes, an f_1 value of 0.8 is selected based on the mid-range of literature values for Cs incorporated in various food items and the ingestion of simulated distant fallout particles (LeRoy et al. 1966). The above selected f_1 values reflect the relative absorption of Cs associated with different particle sizes and exposure conditions for the reasons discussed.

Table 5. Selected f_1 values for cesium and rubidium.

	Acute intakes		
Exposure conditions	Local fallout	Regional fallout	Chronic intakes All locations
Coral tests, all types	0.40	0.80	0.80
Land tests, continental soil	0.40	0.80	0.80
Direct soil ingestion	—	—	0.40

The internationally adopted f_1 value for Rb is 1.0 for both occupational and environmental exposures (ICRP 1980, 1996; U.S. EPA 1988). Rb is very similar in its chemical and metabolic behavior to cesium. We, therefore, chose f_1 values for Rb equal to those for cesium as discussed above.

Strontium (Sr) and barium (Ba)

Several Sr isotopes are produced in high yield in the fission process. The short-lived ^{89}Sr (53 d half-life) is a dominant component in fresh fallout material and the long-lived ^{90}Sr (~28 y half-life) is abundant in aged fallout (Cohn et al. 1954). Both ^{89}Sr and ^{90}Sr are fractionating "volatile" radionuclides in typical fallout. However, the term "volatile" is a characteristic of their noble gases precursors and not for strontium itself.

Radiochemical analysis of urine samples from a group of Marshallese indicated that ^{89}Sr alone contributed 42% of the total beta activity at 45 d post-detonation following exposure to fallout from the Bravo test (Cohn et al. 1956). Several studies on plant and animal uptake concluded that more Sr was assimilated at distant locations than for local fallout (close-in from ground zero) due to particle size and solubility differences along the forward path of fallout (Maxwell et al. 1955).

Pathways of intake of strontium by humans from fallout are through the direct ingestion of fallout particles, ingestion of contaminated food and milk, and to a lesser extent, inhalation. Increased calcium intake has been shown to reduce strontium absorption and retention in man (Spencer et al. 1963). Fasting was also found to

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increase strontium absorption in humans (Hamilton 1944).

A sample of literature values of f_1 for Sr is provided in the Appendix. In general, Sr is absorbed more efficiently from the alimentary tract when incorporated in dietary components (chronic intakes) compared to other ingested forms. Human data on the absorption fraction of Sr in soluble forms, dietary sources, and from simulated fallout particles give values ranging from about 0.10 to about 0.50. An f_1 value of 0.3 was reported by Hardy et al. (1965) for ^{90}Sr absorption. Their calculated value was based on a single U.S. human subject who consumed food products obtained from the Marshall Islands and may be relevant to chronic intakes. The ICRP (1979, 1989, 1993, 1996) and Nuclear Energy Agency (Kendall et al. 1988) adopted an f_1 value of 0.3 for both acute and chronic intakes based on the upper limit of absorption for various ingested forms. For occupational exposure, the U.S. Environmental Protection Agency (U.S. EPA 1988) and ICRP (1979) list two different ingestion solubility classes for strontium with f_1 values of 0.3 and 0.01 for classes D and Y, respectively, or Type M and Type S according to ICRP Publication 66 (1994).

In general, isotopes of strontium generated from nuclear tests on coral islands are expected to be in the oxide or the hydroxide form in fresh fallout debris and in the carbonate form in aged material. However, in most tests conducted at the Marshall Islands (land and water), fallout debris was exposed to seawater to various degrees. Chemical analysis of fallout particles from the Mike test indicated the presence of hydrated calcium sulfate (gypsum). Apparently, when calcium hydroxide formed in the fireball is exposed to the sea-water environment, which is rich in sulfate and magnesium ions, calcium can precipitate as sulfate on particle surface to form a shell of gypsum and magnesium hydroxide around its inner core (Heidt et al. 1953; Adams et al. 1958). Strontium and Ba are likely to react with the sea-water sulfate in the same manner. It is well known that Sr and Ba sulfates are very poorly soluble in water; therefore, their biological availability and absorption by humans may also be diminished. Additionally, Libby (1957) hypothesized that in high-yield (megaton) weapons, the fireball remains sufficiently hot, so that ^{90}Sr will have been produced before the condensation of particles causing ^{90}Sr to incorporate in the center of fallout particles. In that case, ^{90}Sr would be less biologically available than in low magnitude (kiloton) weapons where ^{90}Sr would be condensed more on fallout particle surfaces. As discussed earlier, the solubility of Sr and Ba from close-in fallout simulated particles was much lower than for distant locations (Table 3). Therefore, it is possible that a significant proportion of the total Sr and Ba from tests conducted at the Marshall Islands might have been trapped in insoluble particles at close-in locations and, thus, less biologically available. No information on the alimentary tract absorption of Sr sulfate could be found in the literature, however.

As for Ba, the alimentary tract absorption fraction of ^{133}Ba tracer in the oxide form in simulated fallout particles averaged 0.01 and 0.06 for close-in and distant locations, respectively (LeRoy et al. 1966). Barium sulfate is also very poorly absorbed (less than 1%) from the alimentary tract of adult humans (Boender and Verloop 1969; Figueroa et al. 1968). For workers, the ICRP recommendation (1980) for f_1 was 0.10 based on literature values for all compounds of Ba. In more recent ICRP reports (1993, 1996), an f_1 value of 0.20 was adopted for members of the public equal to Ra (in view of the chemical similarities of Ba and Ra). Several metabolic similarities between Sr, Ba, and Ca were also noted by Leggett (1992). Hamilton (1948) reported that the metabolic behavior of Sr and Ba are essentially indistinguishable in the rat.

Selected f_1 values for strontium and barium. Our "best estimates" for f_1 for Sr under various intake conditions are presented in Table 6. For acute intakes from local fallout, we estimated an f_1 of about 0.02 for ^{89}Sr and ^{90}Sr from the Marshallese urine bioassay data (Table 4). However, analytical methods and counting technology that were available in the 1950's for the measurement of beta-emitting Sr isotopes were minimal. Therefore, several sources of uncertainties associated with the urine bioassay data and the derivation of f_1 for Sr (presented earlier) should be expected. For the above reasons, it was prudent to choose a higher f_1 value of 0.05 for the acute intakes of Sr isotopes from local fallout. This f_1 value reflects the likely chemical forms and associated solubility of Sr in coral fallout exposed to sea water (discussed earlier). For chronic intakes, an f_1 value of 0.3 is selected based on reported literature values for Sr incorporated in food and milk (Appendix). This value is currently adopted by the ICRP (1996) for Sr intakes by members of the public. Our selected f_1 value of 0.2 for regional fallout represents the mid-range of

Table 6. Selected f_1 values for strontium.

	Acute intakes		
Exposure conditions	Local fallout	Regional fallout	Chronic intakes All locations
Coral tests, all types	0.05	0.20	0.30
Land tests, continental soil	0.15	0.20	0.30
Direct soil ingestion	—	—	0.15

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reported data on acute intake from other ingested forms and Sr absorption from simulated distant fallout particles. For local fallout of Sr incorporated in continental soil, an f_1 value of 0.15 is selected based on few data on the absorption of its silicate form (Hamilton 1944) and simulated local fallout in the oxide form (LeRoy et al. 1966). The same f_1 value (0.15) is also chosen for direct soil ingestion from chronic intakes. Literature information suggests that overnight fasting of human subjects resulted in increased Sr absorption of about 30%, on average (ICRP 1993). The Bravo detonation occurred early in the morning when the nearby populations, having had their last meal about 12 h earlier, were likely in the equivalent of a fasting condition. If fasting conditions can be assessed, f_1 values may be increased by the same ratio (30%) for acute intakes.

For barium intakes from local fallout, an f_1 value of 0.03 was estimated from the Marshallese urine bioassay data (Table 4). Although ^{140}Ba measurements used to derive this f_1 value may not be reliable, it is similar to that estimated for Sr from the bioassay data. Given the reported chemical and metabolic similarities between Ba and Sr, we selected an f_1 value for Ba equal to that for Sr (0.05) for acute intakes from local fallout. For chronic intakes, the f_1 value of 0.2 adopted by ICRP (1996) appears to be appropriate. For regional fallout, the f_1 value of 0.1 adopted by ICRP for acute intakes was selected.

The actinide elements (Pu, Np, Am, U)

Under high temperatures in nuclear explosions, the actinide elements are expected to form metal oxides. If continental-type soil is incorporated in the fireball, fallout particles will include significant amounts of silicate and oxide species. In nuclear tests on coral islands, the actinide elements are expected to exist in oxide forms, incorporated in calcium oxide particles. The metal oxide forms for most actinides are refractory and typically insoluble under normal environmental conditions. In water tests (sub or surface), some of the actinide elements may fractionate slightly. In general, most actinides are characterized by high soil adsorption (K_d), low plant and animal uptake and low environmental mobility. Inhalation is generally the major intake pathway to humans since alimentary tract absorption is very low for most actinides. However, alimentary tract uptake can contribute to exposure when food with surficial contamination is ingested or direct soil ingestion is appreciable. Direct soil intake could be a significant mode of exposure in developing countries (Simon 1998).

Plutonium (Pu). Typical literature values for Pu f_1 values are presented in Appendix. Reported values ranged from 2×10^{-1} to 10^{-5} depending on the Pu form ingested. The level of alimentary tract absorption for Pu is influenced by several factors including concentration (mass), oxidation states and binding to complexing agents in water, soil, plant, and animal tissues (Harrison et al. 1981). Durbin (1975) stated that Pu absorption from the alimentary tract is affected by the chemical form ingested and that f_1 values are higher for soluble compounds relative to the hydrolysable salts and insoluble compounds. At low ingested mass of Pu, as may be encountered under environmental conditions, absorption may be significantly greater than at high mass intake as in some animal studies (ICRP 1989). However, Harrison et al. (1981) did not observe such effect in hamsters or rabbits. Additionally, when the ingested mass is small, the influence of chemical form on Pu absorption seems largely to diminish (ICRP 1989). Some studies indicated that incorporation of the actinides into plant or animal tissues, as in dietary sources, resulted in a two to four fold increase in ingested dose as compared with absorption of Pu salt solutions (Sullivan 1981). In all species studied (rats, mice, guinea pigs, dogs and swine), neonates absorbed more actinides than did the adults by two to three orders of magnitude (Sullivan 1981). Also, animal studies suggest that fasting can increase Pu absorption by as much as an order of magnitude (ICRP 1986, 2006).

The solubility and absorption of Pu oxides also vary with the temperature at which the oxide species was produced. For example, Pu systemic absorption, following inhalation, is higher for low-temperature fired oxides than for high-temperature oxides (ICRP 1986) and it would not be unreasonable to assume that the same is valid for ingested Pu oxides. On the other hand, a significant part of the ^{239}Pu deposited on the Earth's surface from fallout was produced from volatilized ^{238}U via a (n, γ) reaction and subsequent decay of the ^{239}U through ^{239}Np and ^{239}Pu (Watters et al. 1980). This material is formed as single atoms and was not subjected to the high temperatures of nuclear explosions. This type of Pu oxide appears to be more soluble than would be expected if the oxide had been produced at high temperatures. Additionally, Sullivan (1981) concluded that the ingestion of smaller particle sizes resulted in higher f_1 values due to higher solubility in the alimentary tract.

In general, the range of f_1 values in various animal species that ingested soluble Pu forms (citrate, chloride, etc.) lie between 10^{-3} to 10^{-4}, and for the oxide forms, the average f_1 value is about 10^{-5} (ICRP 1989). The majority of f_1 values for low mass intake of Pu from global fallout are greater than 10^{-4} (ICRP 1996). Based on a urine bioassay study, Sun and Meinhold (1997) estimated an f_1 value of 4×10^{-4} for Pu incorporated in

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a typical Marshallese diet (chronic intakes). In that study, direct soil ingestion was assumed to be 500 mg d^{-1}, though the value was not based on empirical evidence and, hence, the predicted f_1 value is uncertain.

The earlier ICRP (1979) recommended f_1 value of 1×10^{-5} for Pu in the oxide and hydroxide forms and 1×10^{-4} for all other compounds for occupational exposure. The recent ICRP (1996) adopted an f_1 value of 5×10^{-4} for adults and 5×10^{-3} for children during the first year of life for chronic intakes from environmental exposures.

Selected f_1 values for plutonium. Based on literature values (Appendix) and information presented earlier on Pu characteristics in fallout, we chose the f_1 values presented in Table 7. For acute intakes from local fallout for both coral and continental tests, we select an f_1 value of 1×10^{-5}. This value is supported by ICRP (1979) and U.S. EPA (1988) recommendations for acute intakes of the oxide and hydroxide forms of Pu and reflects the expected low solubility of Pu associated with large particles. Our selected f_1 value of 5×10^{-4} for chronic intakes represents the mid-range of literature values for Pu incorporated in foodstuffs and is supported by rough estimates for the Marshallese population (Sun and Meinhold 1997) and is equal to the recent ICRP (1996) recommendation for members of the public. For acute intakes of regional fallout we chose $f_1 = 1 \times 10^{-4}$, which is assumed to be an order of magnitude greater than for local fallout to reflect the increased Pu solubility and bioavailability with distance as discussed earlier. Although there is no direct evidence to show whether fasting would increase Pu absorption in humans, the fact that a marked increase in Pu absorption was observed after fasting in some animal species suggests that it would be prudent to assume that similar effects may occur in man (ICRP 1986). Therefore, f_1 values for acute intakes may be increased by an order of magnitude, if fasting conditions can be assessed. Safety tests (non-nuclear detonations of nuclear fuel material) typically produce large particles dominated by the refractory Pu oxide forms. The resulting debris is much more insoluble than in some fallout. For this reason, the lower f_1 (10^{-5}) value is selected in this case. The same f_1 value (10^{-5}) is also appropriate for direct soil ingestion.

Table 7. Selected f_1 values for plutonium.

	Acute intakes		
Exposure conditions	Local fallout	Regional fallout	Chronic intakes All locations
Coral tests, all types	1×10^{-5}	1×10^{-4}	5×10^{-4}
Land tests, continental soil	1×10^{-5}	1×10^{-4}	5×10^{-4}
Direct soil ingestion	—	—	1×10^{-5}

Neptunium (Np). Neptunium and plutonium are much alike in their chemical and metabolic characteristics. In ICRP (1980), an f_1 of 0.01 was recommended for acute intakes by workers based on the absorption of Np nitrate in rats (Appendix). In ICRP (1986, 1989), an f_1 value of 1×10^{-3} was recommended for environmental exposures to low Np mass. The most recent ICRP recommendation (1996) reduced the Np f_1 value to 5×10^{-4} for chronic intakes by members of the public, equal to that for Pu. Based on strong similarities, we chose the same f_1 values for Np as for Pu, for all types of intakes (Table 7).

Americium (Am). The chemical and biological behavior of Am is similar to the lanthanide rare earths (Hamilton 1948). Reported f_1 values for Am in various ingested forms are within similar range to Pu (Appendix). In ICRP (1979), an f_1 value of 5×10^{-4} was recommended for all Am compounds for acute intakes in the work place. The most recent recommendation for the Am f_1 value is 5×10^{-4} (ICRP 1996) for chronic exposure of the general public, equal to that for Pu. Based on the state of knowledge at this time, we chose the same f_1 values for Am as for Pu for all types of intakes (Table 7).

Uranium (U). The metabolic behavior of U is different from most actinides. In the oxide form, U is slightly more soluble in water than Pu and Am. Its absorption from the alimentary tract is also higher than most other actinides. This is likely due to its smaller ionic size in the +6 (UO_2^{+2}) oxidation state, the most commonly encountered form in the environment. Uranium uptake and retention in the skeleton is less than most actinides and exhibits higher accumulation in the kidney.

Some f_1 literature values for uranium are presented in the Appendix. In general, U absorption from the alimentary tract increases with increasing solubility of the ingested compound. Literature values indicate fractional absorption in the range of 5×10^{-3}–2×10^{-2}. Uranium uptake was found to be substantially greater in animals under fasting condition (Bhattacharyya et al. 1989).

Selected f_1 values for uranium. Based on the preceding information, our chosen f_1 values for uranium are presented in Table 8. For acute intakes from local fallout (coral and continental tests), we chose an f_1 value of 2×10^{-3}, consistent with that adopted for workers

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Table 8. Selected f_1 values for uranium.

Exposure conditions	Acute intakes Local fallout	 Regional fallout	Chronic intakes All locations
Coral tests, all types	2×10^{-3}	2×10^{-2}	2×10^{-2}
Land tests, continental soil	2×10^{-3}	2×10^{-2}	2×10^{-2}
Direct soil ingestion	—	—	2×10^{-3}

ingesting U oxides (U.S. EPA 1988). For regional fallout and chronic intakes, the recent ICRP recommendation of $f_1 = 2 \times 10^{-2}$ seems appropriate and is representative of literature values for U in diet and drinking water. For direct soil ingestion, the lower f_1 value (2×10^{-3}) is selected, which reflects the expected low U solubility for this type of intake. For fasting conditions, the f_1 value is increased to 5×10^{-2} corresponding with results from animal studies.

The rare earth elements (Ce, Y, La, Pr)

The principal fallout elements in this group include isotopes of Ce, Y, La, and Pr. Most radionuclides of these elements are highly refractory and virtually insoluble in fallout. These elements have no essential function in biological systems and they exhibit low mobility and biological uptake through the food chain. As a group, the rare earths are very poorly absorbed from the alimentary tract. The ICRP (1980, 1981, 1996) adopted f_1 values of these elements are in the range of 10^{-3} to 10^{-4} based on limited literature data for various ingested forms (Appendix).

Selected f_1 values for rare earth elements. The ICRP recommendations (1980, 1981) for f_1 for most of the rare earths applied to acute intakes by workers are based on very limited literature data, some using soluble chloride and nitrate forms of the elements. Due to the same limitations of the available information, most ICRP recommendations (1989, 1996) for chronic intakes applied to the general public are set similar to some of the actinide elements. Due to the lack of fallout-relevant data and similarity of the physico-chemical properties of most rare earths and some actinides, we chose f_1 values for these elements equal to those selected for Pu and other actinide elements, for all types of intakes (Table 7).

The heavy metals (Fe, Co, Zn, Zr, Ru)

The fallout radionuclides giving the most significant dose among the heavy metals group are Fe, Co, Zn, Zr, and Ru. Radionuclides ^{55}Fe, ^{60}Co, and ^{65}Zn are present in fallout from neutron activation of bomb construction material. Some ^{65}Zn may also be due to activation of Zn in seawater being entrained into the fireball. Most heavy metals are expected to occur as refractory oxides in fallout. Literature values for the alimentary tract absorption of these elements are summarized in the Appendix.

Iron (Fe). Iron absorption from the alimentary tract is a function of several factors including: the chemical form ingested, age, gender, and the amount of reserve iron in the body. Due to the lack of fallout-relevant information for Fe, we chose an f_1 value of 0.1 for all types of intakes as adopted by ICRP (1980, 1995).

Cobalt (Co). The available data on the absorption of Co are variable and are affected by the mass and chemical form ingested (Appendix). Due to the lack of fallout-relevant data on Co absorption, the ICRP (1979) adopted f_1 value of 0.05 for acute intakes of the oxide and hydroxide forms are applied here for local fallout. For regional fallout and chronic intakes, an f_1 value of 0.10 is chosen equal to the ICRP (1993, 1996) adopted value for exposure of the public to environmental levels of Co.

Zinc (Zn). All ICRP reports since 1980 (1980, 1993, 1996) adopted an f_1 value of 0.50 for Zn for both acute and chronic intakes, although it may be an overestimation when incorporated in some foods (ICRP 1993). As discussed in Simon et al. (2010), ^{65}Zn was an important contributor to chronic exposure from the ingestion of contaminated seafood. Due to the lack of fallout-relevant information, the mid-range of 0.35 for dietary Zn (Honstead and Brady 1967) was selected of all fallout exposure conditions.

Zirconium (Zr). ^{95}Zr is highly refractory and fairly insoluble in fallout debris, particularly when incorporated into large particles that deposit locally. It is expected to be present in the oxide form after detonation (Freiling 1961). The limited literature data presented in the Appendix are based on the alimentary tract absorption of soluble chemical forms of Zr in rats. No studies on the uptake of the oxide form could be found. For acute intakes the adopted ICRP (1979) f_1 value for all compounds of Zr is 2×10^{-3}. For chronic intakes, ICRP recommendation (1989, 1996) increased f_1 to 1×10^{-2} to allow for the possibility that Zr uptake might be greater for biologically incorporated forms in food. Due to the lack of fallout-relevant information, we select the ICRP f_1 value of 2×10^{-3} for local and regional fallout and 1×10^{-2} for chronic intakes.

Ruthenium (Ru). For Ru intake from local fallout, an f_1 value of about 1×10^{-4} was estimated from the Marshallese urine bioassay data (Table 4). However, this

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estimate is low and may not be reliable for the reasons stated earlier. The ICRP (1980, 1989, 1996) adopted an f_1 value of 5×10^{-2} for both acute and chronic intakes based on the absorption for soluble compounds from animal studies. The f_1 value for the oxide form from one animal study is about 1×10^{-3} (Runkle et al. 1980) and is appropriate for local fallout. The limited data on the absorption of Ru incorporated in diet suggest an f_1 value of 1×10^{-2} (Yamagata et al. 1969), and is chosen for all other intakes (Table 9).

Other radionuclides

Molybdenum (Mo). ^{99}Mo is a moderately fractionating radionuclide in fallout debris, particularly in water-surface bursts (Freiling 1961; Crocker 1966). In ICRP (1979), an f_1 value of 0.8 was adopted for workers for all compounds other than the sulfide form, for which an f_1 value of 0.05 was adopted (Appendix). For the public, the f_1 value was increased to 1.0 in the recent ICRP (1993, 1996) recommendation for chronic intakes without much justification. Based on the above information, we believe that complete absorption ($f_1 = 1.0$) for Mo is unrealistically high when applied to fallout particulates. Furthermore, it is possible that a fraction of the Mo in local fallout debris exposed to seawater may be present as sulfate or sulfide. Due to the lack of fallout-relevant data on Mo, we selected a middle range value between the sulfide and all other forms ($f_1 = 0.2$) for acute intakes. For chronic intakes of Mo incorporated in food, a higher f_1 (0.5) was selected. Dose estimates for Mo based on the above f_1 values will, however, have a high degree of associated uncertainty.

Table 9. Summary of selected f_1 (adult values) for key fallout radionuclides for various intake categories[a] used in internal dose assessments for the Marshallese populations.

	Acute intakes		
Nuclide	Local fallout	Regional fallout	Chronic intakes All locations
Iodine (I)	1.0	1.0	1.0
Tellurium (Te)	0.2	0.2	0.3
Cesium (Cs), Rubidium (Rb)	0.4	0.8	0.8
Strontium (Sr)	0.05	0.2	0.3
Barium (Ba)	0.05	0.1	0.2
Plutonium (Pu), Neptunium (Np), Americium (Am), Cerium (Ce), Yttrium (Y), Lanthanum (L), Praseodymium (Pr)	1×10^{-5}	1×10^{-4}	5×10^{-4}
Uranium (U)	2×10^{-3}	2×10^{-2}	2×10^{-2}
Iron (Fe)	0.1	0.1	0.1
Cobalt (Co)	0.05	0.1	0.1
Zinc (Zn)	0.35	0.35	0.35
Zirconium (Zr)	2×10^{-3}	2×10^{-3}	1×10^{-2}
Ruthenium (Ru)	1×10^{-3}	1×10^{-2}	1×10^{-2}
Molybdenum (Mo)	0.2	0.2	0.5
Bismuth (Bi)	0.05	0.05	0.05

[a] Recommendations for f_1 values for other exposure conditions are presented in the text and in Tables 5–8.

Bismuth (Bi). Based on a limited amount of experimental information, ICRP (1980, 1996) adopted an f_1 value of 0.05 for Bi for both acute and chronic intakes. This value was thought to be appropriate for most common Bi compounds. Given the limited data available, we have no reason to suggest any other value.

SUMMARY

The purpose of this review was to evaluate the available data and to determine the most appropriate f_1 values for key fallout radionuclides for use in realistic reconstructions of internal dose from the intake of fallout from nuclear tests conducted at the Marshall Islands in the 1940's and 1950's. Currently used f_1 values adopted by ICRP and other radiation protection organizations are sometimes conservative and based on data related to radionuclides present in food or as specific forms; some are soluble compounds. Such generic f_1 values may not be suitable for the specific conditions of exposure to particulate fallout from nuclear weapons tests. Presented f_1 values are our "best estimates" and were generally based on the mid-range of the available literature values. Specific circumstances of weapons tests conditions were also considered (e.g., radionuclide fractionation and solubility) in our evaluation and used to propose appropriate modifications to currently adopted f_1 values. Our estimated f_1 values derived from the urine bioassay data of the Marshallese population were given considerable weight in the selected f_1 values for Cs, Sr, and Ba presented here. The opportunity to derive empirically-based absorption parameter values is unique and proved to be significant in our evaluation of f_1. Multiple f_1 values for various circumstances where persons are exposed to fallout (e.g., close-in vs. distant locations and acute vs. chronic intakes) were presented when supportive information was available (Tables 5–8). Although special attention was given to tests conducted at the Pacific Proving Ground, the approach taken here provides values that should be useful for assessments of exposure to fallout from other regional nuclear tests or events. In the process of evaluation and selection of f_1 values for the various circumstances of intake, adequate amounts of objective information were not always available and some judgment based on the authors' experience was applied. For these and other reasons, some degree of uncertainty associated with the f_1 values presented here is expected. The above examination led to the selection of a consistent and complete set of f_1 values

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(presented in Table 9) deemed to be appropriate for use in realistic internal dose reconstruction for exposures to fallout from nuclear tests conducted in the Marshall Islands.

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APPENDIX

Table A1. Summary of literature values for alimentary tract absorption fraction (f_1) of key fallout radionuclides in various ingested forms and experimental conditions.

Ingested form	f_1	Comments	References
Iodine (I)			
Literature information, based on ingestion from aqueous solution, milk and incorporated in foodstuffs	1.0	Adopted value for workers (acute intakes) and the general public (chronic intakes)	ICRP 1979, 1989, 1996; U.S. EPA 1988; Kendall et al. 1988 (NEA)
Cesium (Cs)			
Contaminated food items from Rongelap, Marshall Islands	0.90	Only one human subject investigated, chronic intakes	Hardy et al. 1965
^{134}Cs + carrier in silicate form, simulated distant fallout particles (see Table 3)	0.82 (0.73–0.84)[a]	5 human subjects investigated	LeRoy et al. 1966
^{134}Cs carrier free in microspheres, simulated local fallout particles (see Table 3)	0.31 (0.29–0.36)[a]	3 human subjects investigated	
^{134}Cs in chloride solution	0.89 (0.87–0.90)[a]	4 human subjects investigated	
^{137}Cs and ^{132}Cs, carrier free	0.99	2 human subjects investigated	Iinuma et al. 1971
^{137}Cs incorporated in venison meat	0.78 (0.1–0.4)[a]	10 human subjects investigated, chronic intakes	Henrichs et al. 1989
^{137}Cs incorporated in shellfish from the Irish Sea	0.23 (0.1–0.4)[a]	6 human subjects investigated, chronic intakes	Hunt et al. 1998
^{137}Cs incorporated in food and ^{137}Cs in chloride form	0.98	3 human subjects investigated	Raaf et al. 2004
Typical literature information for various animal species	0.57	Reindeer	NCRP 2006
	0.78	Mule deer	
	0.27	Lab mouse	
Literature information on various physico-chemical forms	1.0	Adopted value by various organizations for workers and members of the public	ICRP 1979, 1989, 1996; U.S. EPA 1988; Kendall et al. 1988 (NEA)
Strontium (Sr)			
Sr in bicarbonate form	0.10	Animal studies	Hamilton 1944
Sr in silicate form	0.15		
^{90}Sr in diet and milk	0.26 (0.09–0.48)[a]	4 human subjects investigated, chronic intakes	Fujita 1965
Contaminated food items from the Marshall Islands (Rongelap)	0.30	1 U.S. subject investigated, f_1 was calculated from intake and excretion, chronic intakes	Hardy et al. 1965
^{85}Sr + carrier in oxide form incorporated in particles to simulate distant locations (see Table 3)	0.23 (0.07–0.47)[a]	9 human volunteers in two groups fed different levels of activity	LeRoy et al. 1966
^{85}Sr carrier free in oxide form incorporated in glass micro-spheres to simulate local fallout (see Table 3)	0.16 (0.06–0.25)[a]	5 human subjects investigated	
^{85}Sr in chloride solution	0.17 (0.08–0.34)[a]	11 human subjects investigated	
Contaminated food	0.17	10 adult humans on high Ca diet, chronic intakes	Spencer et al. 1963
Literature information on various ingested forms	0.3	For all compounds	ICRP 1979; U.S. EPA 1988
	0.01	For $SrTiO_3$, acute intakes (workers)	
Literature information on various ingested forms	0.30	Based on upper limit of soluble forms of Sr, chronic intakes (general public)	ICRP 1989, 1993, 1996; Kendall et al. 1988 (NEA)
Barium (Ba)			
^{133}Ba in local fallout simulated particles	0.01	Ingested by 4 humans subjects	LeRoy et al. 1966
^{133}Ba in distant locations simulated particles	0.06		
Literature information on diet	0.10	Adopted for workers (acute intakes)	ICRP 1980
Literature data on Ra	0.20	Based on Ra absorption, adopted for the public (chronic intakes)	ICRP 1993, 1996

(Continued)

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Table A1. (*Continued*)

Ingested form	f_1	Comments	References
The Actinides			
Plutonium (Pu)			
Oral administration of soluble salts	7×10^{-3}	Review of various studies on rats	Hamilton 1948
Bicarbonate and citrate forms	$(2 \times 10^{-3}–4 \times 10^{-3})^a$	Fasting mice and rats	Larsen et al. 1981
Pu in chlorinated drinking water	$(1 \times 10^{-3}–2 \times 10^{-1})^a$	Range of values based drinking water intakes	Larsen et al. 1983
Fallout Pu incorporated in reindeer meat	8×10^{-4}	Estimated in Finnish Lappa population, chronic intakes	Mussalo-Rauhamaa et al. 1984
Incorporated in plants via roots	1×10^{-3}	Suggested f_1 values for nuclear fuel facility	Pinder et al. 1990
Surface contamination on plants	1×10^{-5}		
Pu (+4) fed to fasting baboons	1×10^{-4}	f_1 values were similar in mice and humans	Bhattacharyya et al. 1989
Pu (+4)	$(2 \times 10^{-4}–9 \times 10^{-4})^a$	Range of values in human volunteers	Popplewell et al. 1994
$^{239,\,240}$Pu incorporated in typical Marshallese diet	4×10^{-4}	34 human subjects investigated, chronic intakes	Sun and Meinhold 1997
$^{239,\,240}$Pu incorporated in shellfish	3×10^{-4} $(2 \times 10^{-5}–7 \times 10^{-4})^a$	6 human subjects investigated, from Sellafield plant discharge in the Irish Sea, chronic intakes	Hunt 1998
Oxides and hydroxide forms	1×10^{-5}	Adopted values, based on limited literature for workers	ICRP 1979; U.S. EPA 1988
All other compounds	1×10^{-4}		
Literature information for various ingested forms	1×10^{-3}	Adopted value for the public	ICRP 1989
Literature information for various ingested forms	5×10^{-4}	Revised value for adults (chronic intakes)	ICRP 1996
	5×10^{-3}	Adopted value for children (chronic intakes)	
Incorporated into food and drinking water	1×10^{-3}	Recommended values based on literature information (chronic intakes)	Kendall et al. 1988 (NEA)
Neptunium (Np)			
Literature information, nitrate form	1×10^{-2}	Based on the absorption of Np in rats, adopted for workers	ICRP 1980
Literature information	1×10^{-3}	Recommended for environmental exposures (low mass), chronic intakes	ICRP 1986, 1989
	5×10^{-4}	Revised value, chronic intakes	ICRP 1996
Americium (Am)			
^{241}Am incorporated in shellfish from the discharge of Sellafield plant	1×10^{-4}	6 human subjects investigated, chronic intakes	Hunt 1998
Literature information on various ingested forms	5×10^{-4}	For all Am compounds, acute intakes for workers	ICRP 1979
	1×10^{-4}	For occupational exposure	U.S. EPA 1988
	1×10^{-3}	Adopted value for the public	ICRP 1989
	5×10^{-4}	Most recent recommendation, chronic intakes for the public	ICRP 1996
Uranium (U)			
Soluble uranyl nitrate	$(5 \times 10^{-3}–5 \times 10^{-2})^a$	4 hospital patients investigated	Hursh et al. 1969
Incorporated in diet	1×10^{-3}	Review of literature data	Wrenn et al. 1985, 1989
Drinking water	2×10^{-2}		
U (+6)	5×10^{-2}	Fed to fasting baboons	Bhattacharyya et al. 1989
	5×10^{-3}	Non-fasting baboons	
Drinking water	2×10^{-2}	Chronic intakes	Harduin et al. 1994
Central values for different studies	$(1 \times 10^{-2}–2 \times 10^{-2})^a$	Review and re-analysis of several studies	Leggett and Harrison 1995
Central estimates for environmental studies	$(3 \times 10^{-3}–3 \times 10^{-2})^a$	Chronic intakes	
Overall range from food intake	$(1 \times 10^{-2}–1.5 \times 10^{-2})^a$	Chronic intakes	
Soluble inorganic forms	5×10^{-2}	Adopted value for workers	ICRP 1979
Soluble forms	5×10^{-2}	Recommended for occupational exposure	U.S. EPA 1988
Oxide (insoluble) forms	2×10^{-3}	Acute intakes	
Dietary forms	2×10^{-2}	Adopted for the general public (chronic intakes)	ICRP 1995, 1996

(*Continued*)

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Table A1. (*Continued*)

Ingested form	f_1	Comments	References
The Rare Earths			
Cerium (Ce)			
Various ingested form	3×10^{-4}	Adopted value for workers	ICRP 1979
	1×10^{-3}	Value set equal to the actinide elements, general public	ICRP 1989
	5×10^{-4}	Revised value for the general public (chronic intakes)	ICRP 1996
Yttrium (Y)			
Various forms	1×10^{-4}	Adopted for both acute and chronic intakes	ICRP 1980, 1996; U.S. EPA 1988
Lanthanum (L)			
Chloride form, animal studies	1×10^{-3}	Adopted for workers	ICRP 1981
Various forms, animal studies	5×10^{-4}	Revised value for the general public (chronic intakes)	ICRP 1996
Praseodymium (Pr)			
Soluble forms	3×10^{-4}	Adopted for workers, value set equal to Ce	ICRP 1981
	5×10^{-4}	Revised value for the general public	ICRP 1996
The Heavy Metals			
Iron (Fe)			
Incorporated in diet	0.10	Adopted for acute and chronic intakes	ICRP 1980, 1995
$^{59}FeSO_4$	(0.1–0.3)[a]	In normal male subjects	Boender and Verloop 1969
Various forms	0.30	"Cautious value" based on man and animal studies, chronic intakes	Kendall et al. 1988 (NEA)
Cobalt (Co)			
Organic and inorganic forms	0.3	Adopted value for workers (acute intakes)	ICRP 1979
Oxide and hydroxide forms	0.05		
Environmental (trace) levels	0.10	Adopted for the public (chronic intakes)	ICRP 1993, 1996
^{60}Co incorporated in shellfish from Sellafield plant discharge in the Irish Sea	0.2	Human study, chronic intakes	Hunt 1998
Zinc (Zn)			
Inorganic forms and organic incorporated in diet	0.50	Adopted for acute and chronic intakes	ICRP 1980, 1993, 1996
Incorporated in fish from the Columbia River	0.35 (0.25–0.45)[a]	7 human volunteers investigated, chronic intakes	Honstead and Brady 1967
Zirconium (Zr)			
Oxalate form	1×10^{-3}	Absorption in the rat	Shiraishi and Ichikawa 1972
Various forms, animal studies	2×10^{-3}	Adopted for workers (acute intakes)	ICRP 1979
Soluble compounds, animal studies	1×10^{-2}	Adopted for the public (chronic intakes)	ICRP 1989, 1996
Ruthenium (Ru)			
Contaminated clams	1×10^{-2}	One male volunteer	Yamagata et al. 1969
Oxide form	$(3 \times 10^{-3}–1 \times 10^{-2})$[a]	Animal study	Runkle et al. 1980
Soluble chloride, animal studies	5×10^{-2}	Adopted for acute and chronic intakes	ICRP 1980, 1989, 1996
Other Radionuclides			
Molybdenum (Mo)			
Sulfide form (MoS_2)	0.05	Adopted for acute intakes by workers	ICRP 1979; U.S. EPA 1988
All other compounds	0.80		
No specific information provided	1.0	Adopted for chronic intakes by the general public	ICRP 1993, 1996
Bismuth (Bi)	0.05	Adopted for acute and chronic intakes	ICRP 1980, 1996

[a] Range of values.

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