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Fallout Deposition in the Marshall Islands from Bikini and Enewetak Nuclear Weapons Tests

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Abstract

Deposition densities (Bq m^{-2}) of all important dose-contributing radionuclides occurring in nuclear weapons testing fallout from tests conducted at Bikini and Enewetak Atolls (1946-1958) have been estimated on a test-specific basis for all the 31 atolls and separate reef islands of the Marshall Islands. A complete review of various historical and contemporary data, as well as meteorological analysis, was used to make judgments regarding which tests deposited fallout in the Marshall Islands and to estimate fallout deposition density. Our analysis suggested that only 20 of the 66 nuclear tests conducted in or near the Marshall Islands resulted in substantial fallout deposition on any of the 25 inhabited atolls. This analysis was confirmed by the fact that the sum of our estimates of ^{137}Cs deposition from these 20 tests at each atoll is in good agreement with the total ^{137}Cs deposited as estimated from contemporary soil sample analyses. The monitoring data and meteorological analyses were used to quantitatively estimate the deposition density of 63 activation and fission products for each nuclear test, plus the cumulative deposition of $^{239+240}$Pu at each atoll. Estimates of the degree of fractionation of fallout from each test at each atoll, as well as of the fallout transit times from the test sites to the atolls were used in this analysis. The estimates of radionuclide deposition density, fractionation, and transit times reported here are the most complete available anywhere and are suitable for estimations of both external and internal dose to representative persons as described in companion papers.

Keywords

fallout; ^{137}Cs; nuclear weapons; Marshall Islands

Introduction

From 1946 through 1958, 66 nuclear weapons tests were conducted in or near the Marshall Islands, including 23 at Bikini Islands, 42 at Enewetak, and one at a nearby open-ocean site (DNA 1979;Simon and Robison 1997;Simon, Beck et al. 2009). Of special significance was the test BRAVO, a 15-Mt test conducted on 1 March (local time) 1954 on Bikini Atoll, which, as a result of an unexpected wind shear condition, resulted in heavy fallout on atolls east of the site and high radiation doses to the populations of those atolls. Numerous studies have been conducted to monitor the islands and people, to develop land remediation strategies, and to assess contemporary and possible future doses that might be received by inhabitants of certain

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atolls in the Marshall Islands. Particular emphasis has been given to the northern Marshall Islands. Many of these studies were chronicled in a July, 1997 special edition of Health Physics [73(1), 1997].

However, while there have been numerous measurements made over the decades of radioactivity in soil collected from many of the atolls (particularly the northern atolls and primarily for ^{137}Cs), no assessment of the deposition of all of the many radionuclides contributing to radiation exposure from each test has ever been made for all of the atolls of the Marshall Islands

Some of the difficulties in estimating deposition of the many fallout nuclides at the more than 30 atolls and separate reef islands has been the absence of measurement data of nuclides other than ^{137}Cs, lack of a reliable model for predicting the relative deposition of deposited nuclides for both thermonuclear and non-thermonuclear tests, and absence of good data on time of transit for fallout from each test to reach the atolls from the test sites. There has also been little information regarding the effects of weathering that reduced residual radioactivity, particularly soil ^{137}Cs levels, more quickly than would be expected by radioactive decay alone.

In this work, all available historical and contemporary measurement data was collected and reviewed, including data not previously published in the open literature. An analysis of these measurements along with model calculations of relative nuclide activity and estimated fractionation, supplemented by meteorological modeling, have allowed us to make deposition estimates of all important fallout radionuclides at all atolls from each test.

In a 2004 report to Congress (NCI 2004), simplistic estimates of deposition across the Marshall Islands were made and used to estimate the maximum doses to people living on the inhabited atolls. In this work, the estimates of fallout that were used in the 2004 report have been re-analyzed and improved. In the 2004 report to Congress, the total activity of ^{137}Cs deposited per unit area of ground on each atoll of the Marshall Islands was inferred from soil samples collected in 1991-1994 by Simon and Graham (1997). The calculations assumed that that all of the total ^{137}Cs present after correcting for global fallout was deposited in 1954 as the result of the BRAVO test. The BRAVO deposition was then estimated by subtracting an estimated contribution from “global fallout” from the maximum ^{137}Cs measured in soil sampled in 1991-1994 at each atoll and then decay-correcting back to the time of deposition. The activity ratio of other radionuclides was then estimated from published values of activity ratios for BRAVO debris (Hicks 1982) and the estimated fallout transit time (h), called time-of-arrival (TOA), for BRAVO fallout to reach each atoll.

The methodology used in 2004 for deriving deposition estimates was not completely realistic for several reasons:

1. The estimation of ^{137}Cs in Marshall Islands soils at the time of deposition was based on back-correcting contemporary measurements only for radioactive decay and did not take into account the continual loss of ^{137}Cs from the upper layers of soil due to downward migration (Robison et al. 2003). That gradual loss is primarily a result of heavy tropical rainfall in the Marshall Islands, which was more abundant in the southern atolls than in the northern atolls, as well as the absence of clay in the soil which might otherwise retain ^{137}Cs. Therefore, the true amounts of ^{137}Cs deposited at the times of the tests were greater than those derived for the 2004 report with a larger relative correction made for the southern atolls compared to the northern atolls because of higher annual precipitation there.
2. For the more southern atolls, global fallout ^{137}Cs comprises a substantial fraction (as much as 50%) of the total measured ^{137}Cs inventory. In a 2004 report (NCI 2004),

the contribution of global fallout ^{137}Cs, which is relatively constant over the Marshall Islands, to the total measured ^{137}Cs, was also overestimated. The extent of the overestimation was such that negative values were derived for some southern atolls for the local fallout.

3. Because the 2004 calculations were conservatively based on the maximum ^{137}Cs observed in soils collected at each atoll in 1994-96, rather than the best estimate of the average inventory at undisturbed sites, some, but not all, of the underestimation due to losses from the expected deposition was compensated for.

4. The assumption that all the fallout was a result of the BRAVO test biased some of the 2004 deposition estimates toward higher values than were likely because some tests had substantially longer fallout times-of-arrival (as great as 6-8 days compared to 5-40 h). Also, as shown later in this paper, a substantial fraction of the fallout in some of the northern atolls was from tests other than BRAVO and in the most southern atolls, most of the fallout was from tests other than BRAVO.

Our new estimates of fallout are much more detailed as they take into consideration all radionuclides that contributed substantially to either external or internal radiation exposure from each of the most important weapons tests conducted at Bikini and Enewetak. The present analysis also attempts to correct for all the potential sources of error noted above. Based on available environmental measurement data, ground deposition density for ^{137}Cs, $^{239+240}$Pu, and 62 other radionuclides listed in Simon, Beck et al. (2009, Table 4), have been estimated for each nuclear test that we believe deposited substantial levels of fallout at each atoll or reef island. All together, fallout deposition has been estimated for 20 tests, listed in Simon, Beck et al. (2009, Table 1), at 31 atolls or separate reef islands. The complete list of the 34 atolls and separate reef islands of the Marshall Islands is given in Simon, Beck et al. (2009, Table 2); deposition estimates are given for all of them, with the exception of Bikini and Enewetak, where the tests were conducted.

Materials and Methods

Overview

Based on evaluations of several types of data, we have estimated the deposition density (Bq m^{-2}) of 63 individual radionuclides by atoll or island as well as by nuclear test, plus the cumulative ^{239,240}Pu from all tests. The various types of data reviewed included environmental measurements of ^{137}Cs and other radionuclides (both historical and contemporary), historical measurements of exposure-rate following individual tests derived from aerial surveys, ground surveys, and continuous monitors, historical measurement data of beta activity collected on gummed film during the years of nuclear testing, and recent results from meteorological analyses. Findings from the analysis of these data were coupled with information on the predicted mixture of radionuclides from specific nuclear tests as a function of time after detonation by Hicks (1981; 1982b) to predict deposition densities as a function of fallout time-of-arrival.

The estimates of deposition density of ^{137}Cs in this work are our best estimates inferred from all available monitoring data, much of it historical and available only in the gray literature, e.g., government laboratory reports and internal memoranda from laboratories. As discussed, in a previous assessment of radiation dose to residents of the Marshall Islands from regional fallout by the National Cancer Institute (NCI 2004), emphasis was placed on estimating the fallout deposition in the 1950's from contemporary analyses of cumulative ^{137}Cs deposition at each atoll. Furthermore, in that work it was assumed for purposes of simplicity that the total deposition was a result of a single nuclear test (the 1954 BRAVO test). In this paper, we use the various historical measurement data to make estimates of ^{137}Cs deposition density and

fallout time-of-arrival (TOA) for each atoll and for each of 20 individual tests. The resultant total ^{137}Cs deposited by all tests from this analysis, after appropriate decay to account for the actual effective decay rate in the Marshall Islands discussed previously, were then compared with the retrospective ^{137}Cs remaining in the soil as measured at various by different investigators in 1978 and in 1991-93. This comparison was used to demonstrate the validity and relative accuracy of our individual test ^{137}Cs deposition density estimates.

The test- and atoll-specific estimates of ^{137}Cs fallout were then used to estimate the deposition densities (Bq m^{-2}) of all other radionuclides considered in this study, taking into account the estimated fallout transit times. A nuclide mixture was assumed for all thermonuclear tests identical to that for the Castle BRAVO nuclear test (Hicks 1982b), adjusted for estimated fractionation effects, while a nuclide mixture of a typical plutonium-fueled fission device that was detonated at the Nevada Test Site was used for the non-thermonuclear tests (Hicks 1981).

The measurements we used for the estimation and validation of the ground deposition of ^{137}Cs and other radionuclides included:

- measurements in soil of ^{137}Cs and other long-lived radionuclides including ^{90}Sr, and $^{239+240}$Pu and ^{241}Am, carried out at least a decade after the last test;
- historical measurements of exposure rates, made soon after tests, from which the ground deposition of ^{137}Cs can be derived using calculated ratios of ^{137}Cs deposition per unit exposure rate and information on the time-of-arrival of fallout;
- historical measurements of total beta activity deposited on gummed film (GF) that were collected daily at several locations after a number of tests; and
- meteorological data on wind speed and direction, that were used as input to an atmospheric transport model to predict the geographic pattern of dispersion and to make crude estimates of the ground deposition of ^{137}Cs on any atoll after any test.

These data as well as the models used to estimate deposition density are described in detail in the following paragraphs.

Contemporary measurements of ^{137}Cs and other long-lived radionuclides

Considerable amounts of data are available on the total inventories of long-lived radionuclides at various times as a result of soil sampling carried out by the Department of Energy-sponsored Northern Marshall Islands Radiological Survey (Robison et al. 1982; 1997) and soil sampling and *in situ* gamma spectrometry surveys by the Republic of the Marshall Islands Nationwide Radiological Study (NWRS) (Simon and Graham 1997;Simon et al. 1999). Some data in other years are also available in reports from University of Washington (Nelson 1977;1979) and the Atomic Energy Commission's Health and Safety Laboratory (HASL 1956). These various soil measurement data were used to validate the estimates of total ^{137}Cs deposition density inferred from measured exposure rates and other data by comparing the estimated inventories (from summing individual test depositions, decay corrected to the year of soil analysis) with the contemporary measurements of inventory. The decay correction used for this comparison relied on an estimate of the effective half-life, accounting for both physical decay and loss of activity due to weathering and downward migration.

Historical measurements of exposure rates

Data on exposure rates after various tests in the 1952 IVY and 1954 CASTLE series were obtained from Joint Task Force 7 memoranda (JTF 1954a,b,c), Breslin and Cassidy (1955), Klein (1952), Eisenbud (1953), Steton et al. (1956), Martin and Roland (1954), Heidt et al. (1952), DNA (1979), Graves, 1954, Graveson et al. (1956), SAIC (1981), and Dunning

(1957). During the years of testing, islands were generally surveyed soon after individual detonations by aerial reconnaissance using fixed-wing aircraft as a monitoring platform, as well by ground-level measurements with gamma survey meters. Automatic gamma-ray monitors, designed by HASL, were also located at Rongerik, Kwajalein, Majuro and Ujelang during Operation CASTLE (Breslin and Cassidy 1955). Additionally, the U.S. Public Health Service (PHS) monitored fallout on Ujelang, Utrik, Wotho and Rongerik during the 1956 tests and Ujelang, Utrik, Wotho, and Rongelap during the 1958 tests (PHS 1956; 1958), reporting the maximum daily exposure rates measured with survey meters. Except for test YOKE at Kwajalein, we could not locate any exposure rate data for tests conducted before 1952.

All reported exposure rate survey data for the CASTLE series were multiplied by a factor of 1.3 to correct for deficiencies in the energy and angular response of the survey meters used at that time that caused readings to be low by 20%-30% (Sondhaus and Bond 1959).

The correction factor to convert measurements made at altitude to ground level used by Breslin and Cassidy (1955) was found to be in reasonable agreement with model calculations made later of the variation of exposure rate with height above the ground for surface deposits of radioactivity (Beck and dePlanque 1968). In some cases, however, the exposure rate estimates made in the 1950s at altitude may not have accurately reflected the true amount fallout deposited. This was likely true for small islands because the relatively large field of view at the flight altitude would include some open water. Thus, estimated exposure rates at atolls from the test sites where the net signal was on the order of the average background signal (0.05 to 0.1 mR h^{-1}) are very uncertain and assumed to have been generally biased low. This relatively high background signal, determined by measurements over open ocean in the vicinity of the atoll, and due primarily to airplane and detector background, limited the ability to accurately detect small amounts of fallout on the atolls.

The exposure rates, corrected for all known deficiencies, were normalized to the same time post-detonation (12 h afterwards, termed H+12) using the temporal variation reported by Hicks (1982) for a number of nuclear tests. The variation with time reported for BRAVO was used for all thermonuclear (TN) tests, while the variation with time reported for TESLA was used for the non-thermonuclear tests. In a companion paper (Bouville et al. 2009), we show that the decay rate as a function of time after detonation does not vary substantially from one thermonuclear test to another and, for that reason, we assumed the BRAVO decay rate for all thermonuclear tests and the TESLA decay rate for non-thermonuclear tests to be an acceptable approximation. We also show in Bouville et al. (2009) that the decay rate during the first few weeks after the test does not vary substantially with the degree of fractionation of the fallout or the likely degree of weathering. In both cases, sums of 10 exponential terms were used to accurately fit the reported time-dependence.

In order to estimate ^{137}Cs deposition density from exposure rate measurements we used the model calculations reported by Hicks (1981; 1982; 1984). Besides the variation in exposure rate as a function of time, Hicks also reported the values of relative deposition density for a large number of radionuclides, normalized to 1 mR h^{-1} at H+12, for various degrees of fractionation and for several nuclear tests. The ratios of ^{137}Cs to exposure rate at H+12 were used to estimate ^{137}Cs deposition density from exposure rate data. The ratio of ^{137}Cs to exposure rate at H+12 depends strongly on the degree of fractionation [(estimated as a ratio of refractory (R) to volatile (V) nuclides relative to no fractionation, (R/V=1)] as shown in Table 1. Our estimates of fractionation at each atoll are discussed later in this section. For example, for distant atolls (where R/V=0.5), the ^{137}Cs deposition density at the time of fallout was taken to be equal to 43.7 Bq m^{-2} per mR h^{-1} at H+12 for all non-TN tests and to equal 31.8 Bq m^{-2} per mR h^{-1} at H+12 for all TN tests.

Historical measurements of daily deposition density

The daily deposition density could be estimated, in some cases, from historical measurements of beta activity collected on gummed film used to monitor fallout (Bouville and Beck 2000; Harley et al. 1960). Gummed film was a passive collector of atmospheric aerosols that was mounted horizontally above ground at 1 m height and that would collect particles through their adherence to its sticky surface. Gummed film was usually exposed for 24 hours, after which the total beta activity of the radioactive material collected on the film was measured. In the Marshall Islands, routine gummed film sampling was carried out only on Kwajalein (1952-1959) and Majuro (1952 and 1954 only) atolls, and further away but still relatively close-by at two locations in Micronesia, Pohnpei and Kosiae as indicated by archival gummed film data (List 1955; Heidt et al. 1952).

However, some additional gummed film data for other atolls (unpublished) were also found in the archives of the Department of Energy's Health and Safety Laboratory (HASL) in NYC. Those gummed film data were used to help infer fallout patterns and confirm ground and aerial survey measurements. In order to estimate the ^{137}Cs deposition or the exposure rate at H+12 from the beta activity measured on the film, the gummed film data were corrected for collection efficiency, loss of volatiles, and decayed in a manner similar to that used to analyze the gummed film data from the nuclear tests that were conducted at the Nevada Test Site (NTS) (Beck et al.1990).

The gummed film data were occasionally not consistent with other types of measurements. Some of the difficulties in interpreting the gummed film data were the lack of information on the exact date of counting and any counting efficiency corrections that might have been made to the original data. Because much of the fallout at the sites of the gummed film stations was associated with very heavy precipitation, the ability of gummed film to retain fallout particles was likely to have been very low at times. These various factors resulted in high uncertainty in our interpretations of the gummed film data, particularly for the purpose of making quantitative interpretations.

Despite various limitations, the gummed film data were good indicators of the specific calendar days on which fallout occurred at the sampling sites and unequivocally demonstrated the continuation of fallout for several days after a major test. The gummed-film data and the HASL automatic gamma-ray monitors were both indicative that the HASL air surveys were often conducted prior to the end of fallout deposition and occasionally, even conducted prior to the arrival of fallout. Thus, some earlier reported deposition estimates based only on the air survey data were too low, particularly in the southern Marshall Islands where much of the fallout occurred several days after the detonations.

Use of an atmospheric transport model

Because there is no known fallout monitoring data for the Marshall Islands prior to the 1952 IVY series (except at the test site atolls), it was necessary to use a meteorological model and archival meteorological data to estimate fallout deposition at the atolls for tests conducted in 1946, 1948, and 1951. The model used here, HYSPLIT (Draxler and Hess 1997; Draxler et al. 2007), was developed and is maintained by the National Ocean and Atmospheric Administration (NOAA) and under certain conditions, can be applied to estimating dispersion of fallout from nuclear testing. Our application of the HYSPLIT model for estimating Marshal Island fallout is discussed in a companion paper in this issue (Moroz et al. 2009). The HYSPLIT code models the fallout deposition downwind from available meteorological data at the time of the test, using reanalysis data sets of historical meteorological measurements extrapolated to a sparse grid. The model simulates the transport and deposition of particles of different sizes originating at different altitudes at the location of the test. The HYSPLIT model does not

simulate the weapons debris cloud itself or the radioactivity associated with particles of a given size.

Modeling the transport and deposition of particles released from a nuclear weapons test is a complex and highly uncertain exercise, even if perfect information on the spatial variations in wind speeds and directions over the entire region is available. The actual activity-particle size distribution in a nuclear debris cloud varies with the particular type and yield of the tests, the height of the burst and the local topography. No data of these types are available for the Pacific tests. Thus, the amount of ^{137}Cs attached to particles of a particular size released from a particular altitude could only crudely be estimated for the tests of interest based on limited data from Nevada tests.

In order to relate the geographic pattern of deposition and number of deposited particles estimated by HYSPLIT and ^{137}Cs deposition as a function of yield, debris cloud size and altitude, we developed a crude model to describe the relative numbers of particles released from the cap of the stabilized mushroom cloud at various altitudes as well as from the stem (Moroz et al. 2009). An activity-particle size distribution was also estimated based on data of beta-activity as a function of particle size from measurements following NTS tests, modified slightly to reflect the fact that ^{137}Cs tends to be depleted on the larger particles due to a predisposition for the larger particles to be deposited prior to the formation of ^{137}Cs from its gaseous precursor, ^{137}Xe. The total amount of ^{137}Cs released was estimated from the estimated fission yield of each test (UNSCEAR 2000; Hicks 1981; 1982a; 1982b). Note, however, that even the fission yields for U.S. thermonuclear tests are only an estimate (UNSCEAR 2000), since those data are still classified.

Comparisons were made of the geographic deposition patterns predicted by HYSPLIT with historical ground or aircraft radiological monitoring data. Based on comparisons for tests where monitoring data were available, it appears that when HYSPLIT predicted fallout in the areas where fallout was actually observed, the estimated deposition generally agrees with measured ^{137}Cs to within a factor of 10 (Moroz et al. 2009). Thus, the meteorological model could be used to only provide crude estimates of fallout where no measurements were made. A more reliable use for HYSPLIT was to support our interpolations of deposition at atolls from real measurements of deposition at nearby atolls.

Although the HYSPLIT model estimates are very uncertain, in some cases they were the only source of any information on fallout deposition and, in general, supported numerous anecdotal reports of substantial fallout prior to 1952 at some atolls, in particular at Ujelang from two 1951 tests, DOG and ITEM. The HYSPLIT predictions were also used to complete the fallout deposition pattern for 1956 and 1958 tests for which only a limited number of atolls were monitored for radioactive debris. The 1956 and 1958 tests, however, deposited relatively little fallout and, thus, any uncertainty introduced by these simulations has very little impact on our estimates of total fallout deposition.

^{137}Cs ground deposition due to global fallout

Because the fallout estimates of most radionuclides were derived from measured activities of ^{137}Cs in soil, careful attention was paid to the separation of the ^{137}Cs activity due to fallout during the first week or so after the test - when the radioactive cloud was passing over the Marshall Islands for the first time (so-called “local fallout”) – compared to the ^{137}Cs activity resulting from fallout after the radioactive debris from that test of other tests had circled the globe once or more (producing “global fallout”).

The estimated concentrations of ^{137}Cs in soil samples collected in 1978 by Robison et al. (1982; 1997) and in 1991-1993 by Simon and Graham (1997) had two sources: (1) the local fallout

due to tests conducted at Bikini and Enewetak, and (2) the global fallout due to nuclear weapons tests conducted months or years earlier, in the Pacific as well in other sites of the northern hemisphere.

The global fallout of ^{137}Cs in the Marshall Islands was estimated in this work based on a comparison of the deposition of ^{90}Sr, measured in steel pots at Majuro, Enewetak, and Ponape Micronesia (alternate spelling: Pohnpei) during 1960-1970 with that collected in New York City (NYC) during the same period (Larsen 1983), and the total ^{137}Cs inventory from global fallout in NYC. In NYC, 2.6 kBq m^{-2} of ^{90}Sr was deposited between 1960 and 1970, compared to 0.43 kBq m^{-2} at Majuro for the same period, a ratio of 17%. Similar ^{90}Sr deposition ratios for the same period are obtained for Enewetak (15%), Kosrae Micronesia (11%) and Ponape Micronesia (15%), even though Majuro has a much different annual rainfall, about 340 cm compared to 200 cm for Enewetak and 500 cm for Ponape. Thus, it appears that all atolls of the RMI received on average about 16% as much global fallout as did NYC. The similarity of the ratios for locations at different latitudes and with different precipitation levels agrees with the findings of Simon and Graham (1997) who noted that the decrease in global fallout deposition with decreasing latitude (UNSCEAR 2000) is offset in the Marshall Island by an increase in deposition at lower latitudes because of their greater annual rainfall rates.

The inventory of ^{137}Cs from global fallout in NYC [inferred from measurements of ^{90}Sr through 1981 reported by Larsen (1983)] was about 5.6 kBq m^{-2} in the early 1960s at the end of the period of heavy fallout. Hence, the global fallout deposition density of ^{137}Cs in the Marshall Islands in the early 1960s was estimated to be about 5.6 kBq m^{-2} × 0.16 = 0.9 kBq m^{-2}. This is similar to the estimate of 0.7 kBq m^{-2} derived by Whitcomb (2002) on the basis of a literature survey.

Using the estimate of 0.9 kBq m^{-2} for total global-fallout-^{137}Cs deposited in Marshall Islands, we estimated the residual global fallout component in 1978 and 1994 when soil activity measurements were made. For the purposes of our calculations, we have assumed that all of the global fallout was deposited in 1961.

Effective decay rate of ^{137}Cs in the Marshall Islands

We accounted for the fact that ^{137}Cs was lost from the upper horizons of soil in the Marshall Islands at a rate higher than due to radioactive decay alone, as suggested by Robison et al. (2003). Failure to account for this weathering effect resulted in previous estimates of the deposition density at the time of fallout, based on contemporary measurements of ^{137}Cs inventory assuming only simple radioactive decay being significantly underestimated (by as much as a factor of three to four for some southern atolls). Contemporary ^{137}Cs to ^{90}Sr ratios in soils collected by Robison et al. (1982) in 1978, as well as a comparison of ^{137}Cs in soils of atolls sampled by both Robison et al. (1982) in 1978 and Simon and Graham (1997) in 1991-93, confirm the Robison et al. (2003) finding that ^{137}Cs is lost from the soil profile to ground water in the Marshall Islands. Because the soils of the coral atolls have virtually no clay content, and because the soil is pure $CaCO_3$ below the soil horizon where the organic matter is located (at most about 50 cm), there is a continual downward migration of ^{137}Cs due to the low binding capacity of the upper soil horizons. Our estimates of effective half-lives, taking into account the combination of the environmental loss and radioactive decay, are given in Table 2 for all atolls of the Marshall Islands, ordered by increasing average annual precipitation.

The estimated half-lives for environmental loss ranged from about 15-60 y corresponding to effective half-lives ranging from 10 to 20 years. Our estimated environmental loss rates are somewhat less than those estimated for Bikini and Enewetak by Robison et al (2003). However, the calculations of Robison et al. (2003) did not account for fractionation. At relatively close-in distances to ground zero for a near surface explosion, fractionation can be very high and

thus the ratio of ^{137}Cs to ^{90}Sr in fallout much lower than the ratio expected based on the atom ratio of these nuclides from nuclear fission (see next section). When corrected for fractionation, the effective half-life at Bikini, reported by Robison et al. (2003) would thus probably be closer to 12 y rather than the 8-10 y they reported. Although an effective half-life of 12y is still somewhat smaller than the values we estimate for the northern atolls, the Robison et al. (2003) estimates are only for a few sites on Bikini and Enewetak atolls. Our analyses, based not only on ^{137}Cs to ^{90}Sr ratios, but also on a comparison of ^{137}Cs measured in soils years apart, indicated that the degree of environmental loss varied considerably from island to island on each atoll, as well as from atoll to atoll, depending on the depth of the soil layer and type of vegetation.

The effective half-life generally decreases with increasing average annual rainfall rates, as is to be expected because of precipitation-driven downward migration of the radioactivity in the soil. We estimate that the uncertainty in our estimates of effective half-life range from ±1 to ±2 y in the northern Marshall Islands, where the estimated half-lives are based on ^{137}Cs to ^{90}Sr soil measurements and comparisons of ^{137}Cs inventories in different years, to ±2 to ±3 y in the southern Marshall Islands, where the estimates are based primarily on relative precipitation rates. While the environmental half-life strongly depends on long-term precipitation rates, it is not solely a function of average annual rainfall since the depth of the soil layer varies among islands, depending on their size and stage of vegetative development. Our estimated uncertainty in the effective half-life for ^{137}Cs in soils of the northern Marshall Islands is based on the variations in the loss rates calculated from ^{137}Cs to ^{90}Sr ratios at atolls with similar rainfall. We assigned a larger uncertainty to the effective half-life at southern atolls where^{137}Cs to ^{90}Sr ratios were not measured and the estimate of effective half-life was based on extrapolation. Average precipitation rates in the Marshall Islands are known to increase from north to south with some references suggesting they level off just south of Majuro, consistent with the observed remaining inventories of ^{137}Cs in soil at atolls south of Majuro.

Influence of fractionation

The radionuclides created during the explosion of a nuclear device are usually classified as either refractory (R) or volatile (V) according to whether their melting point is higher or lower than 1,500 °C (Hicks 1982). For example, isotopes of iodine and cesium are volatile and isotopes of zirconium and cerium are refractory.

The mixture of radionuclides in the fallout cloud can change with time after the test (due to factors other than radioactive decay) as part of a phenomenon termed fractionation. A radionuclide mix is fractionated when the activity ratio of the refractory to the volatile radionuclides in deposited fallout differs from what would be expected from the composition in the initial debris cloud. Fractionation is due to the tendency of refractory radionuclides to be distributed throughout fallout particles and volatile nuclides to be distributed preferentially on the surface of particles. This phenomenon gives the ratio of the refractory to the volatile radionuclides a dependence on particle size, with smaller particles, because of their relatively larger surface to volume ratio, typically enriched in volatile nuclides and larger particles typically enriched in refractory nuclides. Large particles, i.e., those greater than ~50 μm in diameter, deposit quickly because of their higher gravitational settling velocity. They also tend to be enriched in refractory nuclides because those nuclides have higher volatilization temperatures and condense more quickly as the fireball cools. Conversely, the volatile nuclides have lower volatilization temperatures and tend to remain gaseous longer and are adsorbed onto smaller particles that remain aloft longer.

The so-called “unfractionated” radionuclide composition varies only slightly from test to test according to the fissionable material and construction characteristics of the nuclear device. Fractionation is very sensitive to the explosive yield, type of soil, height of burst and other

factors (Freiling 1961; 1962; 1963; Freiling et al. 1965). On average, the unfractionated activity ratio of the refractory to the volatile radionuclides in fallout is about 1.6 at H+12.

In this analysis, it is only of interest to compare the level of fractionation at each location of interest to the unfractionated radionuclide mix; therefore, the degree of fractionation is simply denoted as R/V where a ratio of R/V=1.0 represents unfractionated fallout, while a ratio of R/V=2.0 represents fallout where there is twice as much activity of each refractory nuclide in the cloud relative to that in an unfractionated cloud, and, conversely, a degree of fractionation, R/V=0.5 represents fallout where one half of the atoms of each refractory radionuclide has been removed, typical of fallout at long distances from the detonation site (Hicks 1982).

Because of the dependence of fractionation on particle size and because the time varies for deposition of particles of different sizes, we define a *critical time, T_{cr}* to be the length of time since detonation for all particles greater than 50 μm diameter to be deposited. At distances where the TOA is greater than T_{cr}, refractory nuclides are assumed to be depleted relative to volatile nuclides as described in Hicks (1982). Conversely, at increasingly closer distances to the point of detonation, volatile nuclides including ^{137}Cs are assumed to be increasingly depleted in the deposited particles relative to refractory nuclides. Cesium-137 and ^{90}Sr, because their precursors (^{137}Xe, ^{90}Kr) are noble gases, are assumed to be even further depleted compared to other volatile nuclides such as ^{131}I, i.e.^{137}Cs/V < 1. The calculated ratios of ^{137}Cs to the exposure rate at H+12 (termed Ė12) for several values of the degree of fractionation were given in Table 1. Hicks (1982b) reported the radionuclide composition for BRAVO fallout as a function of time for both R/V=0.5 and R/V=1.0. Using Hicks' unfractionated nuclide mixture, we extended his calculations to other values of R/V up to R/V=3.0. The calculated ratios of ^{137}Cs to ^{131}I, ^{137}Cs/^{90}Sr and ^{137}Cs/V as a function of R/V and TOA/T_{cr} are given in Table 3. The approximate relationship between R/V and TOA/T_{cr} was inferred from comparisons of measured ratios of ^{137}Cs in soils downwind from the NTS to corresponding post-test measurements of Ė12 (McArthur and Miller 1989; Thompson et al. 1994) with the calculated ^{137}Cs to Ė12 ratios as a function of R/V in Table 1.

As was shown in Table 1, the degree of fractionation has a substantial impact on the ratio of ^{137}Cs to Ė12. From Table 3, it can be seen that the deposition of ^{137}Cs per unit of ^{131}I or per unit of ^{90}Sr deposited, is 30% to 40% less when the TOA is small compared to T_{cr}, i.e., at locations with a greater degree of fractionation.

Based on estimated TOAs and estimated T_{cr} which are based on reported debris cloud top heights (DNA, 1979), we believe, as shown later, that only fallout from BRAVO was enriched in refractory nuclides to any significant degree and only at a few of the northern atolls in the Marshall Islands. This phenomenon was primarily a consequence of the relatively high speed at which the cloud traveled, resulting in a greater than usual downwind transport of both large and small particles and a greater than usual degree of fractionation. This is consistent with the estimate of T_{cr} for BRAVO which was about 48 h. At the locations of the southernmost atolls, almost all the fallout occurred at times greater than T_{cr} and, thus, the deposition calculations there are based on a relative activity ratio of refractory to volatile radionuclides (R/V) of 0.5, implying a deficit of one-half of the refractory nuclides when compared to a radionuclide mix without fractionation.

The fractionation ratio (R/V) for BRAVO fallout at each atoll was estimated from various ratios of nuclides obtained from radiological analysis of soil samples, including ^{137}Cs to $^{239+240}$Pu, ^{137}Cs to ^{90}Sr, ^{90}Sr to $^{239+240}$Pu, as well as the ratio of TOA to T_{cr}. As explained above, greater degrees of fractionation (i.e., the larger R/V value) result in smaller ratios of ^{137}Cs or ^{90}Sr to refractory nuclides such as $^{239+240}$Pu in soil samples, as well as smaller ratios of ^{137}Cs to ^{90}Sr.

Our best estimates of R/V for BRAVO fallout at a range of atolls, are presented in Table 4. The measured ^{137}Cs to $^{239+240}$Pu ratios in soils collected in 1978 (Robison et al., 1978) are shown to illustrate the clear variations in the ratio of a refractory nuclide activity ($^{239+240}$Pu) to a volatile nuclide (^{137}Cs) activity in the soil sample data as the level of fractionation changes with distance. The ratio varies from about 4 at atolls close to the test site to 15-20 at atolls far away. Because the various ratios used to estimate R/V did not always provide consistent results (for example, the ^{137}Cs to $^{239+240}$Pu ratio measured at Rongerik Island, as shown in Table 4) and also because the soil samples also contained various amounts of unfractionated fallout from tests other than BRAVO, expert judgment was used to correct for the fraction of ^{137}Cs from other tests, to evaluate all the soil data activity ratios, and to make a best estimate of the R/V for BRAVO fallout. These estimated R/V are also consistent with the approximate ratios expected based on the ratio of TOA to T_{cr}. Based on the estimated TOA to T_{cr} ratios for BRAVO, we expect an R/V at Rongelap (where TOA/T_{cr} ~0.12) on the order of 2, about 1.5 at Rongerik (where TOA/T_{cr} ~ 0.16), and an R/V of about 1.0-1.3 for Utrik (TOA/T_{cr} ~0.44). Smaller R/V would be expected for Likiep and Mejit Island.

Although we believe our estimates of R/V for BRAVO to be reasonable, the uncertainty can be large. Furthermore, the relative degree of fractionation assumed for ^{137}Cs as a function of other volatile nuclides (Table 3) is a relatively crude estimate based on a model for fractionation for surface tests that is very sensitive to the particular test conditions and type of soil (Freiling 1961;1962;1963;Freiling et al., 1965). Because most of our estimates of ^{137}Cs deposition are from measurements of exposure rate, and because the value of R/V has a substantial influence on the temporal variation of the exposure rate, any error in the estimated R/V for BRAVO fallout and the estimated activity ratios as a function of R/V (given in Tables 1 and 3) will amplify the error in the estimated ^{137}Cs deposited by the BRAVO test. Thus, if the actual BRAVO R/V at Rongelap Island is actually 1.0 or 1.5 as opposed to our current estimate of 1.3, the ^{137}Cs deposited could be in error by as much as 50%. Even if the R/V estimates are valid, the estimated ratio of ^{137}Cs to other volatile nuclides is based on a general fractionation model and is likely to vary from test to test and, thus, may not reflect the actual ratios for BRAVO. Hence, even if our estimate of fractionation is correct, the error in the ^{137}Cs to Ė12 ratio could still be on the order of 10-15% just from the uncertainty in the estimated ^{137}Cs to V ratio. Although the R/V estimates for BRAVO fallout are very uncertain, the good agreement between the ^{137}Cs deposition estimates (which incorporate our R/V estimates) and the soil inventory measurements discussed below, suggest that our fractionation estimates are indeed reasonably based.

Variations in fallout nuclide composition with time

The atom ratios of various nuclides released in different nuclear weapons test is known to vary slightly due to differences in fissile material and device construction (Hicks, 1981). As discussed above, the radionuclide ratios in fallout at any particular location will also depend on the assumed degree of fractionation. However, for a given value of R/V, the differences from test to test are small compared to the uncertainty in the measured or estimated deposition, as illustrated in Table 5 for selected nuclide ratios. Because only fallout from BRAVO was fractionated at Marshall Island sites, the variations from test to test for fractionated fallout are not relevant here.

Note that the ^{131}I to ^{137}Cs ratio is quite insensitive to the particular test, even for non-thermonuclear compared to thermonuclear tests. However, the ratios for other nuclides contributing to either external or internal dose differ between thermonuclear tests and non-thermonuclear tests for some nuclides, reflecting the different fission yields for ^{239}Pu fission as a function of ^{238}U fast fission. Therefore, the radionuclide mix for the BRAVO test was used for deposition-density estimates for all thermonuclear tests, while for non-thermonuclear

tests, TESLA, which was a typical ^{239}Pu-fueled weapon that was detonated at the Nevada Test Site (Hicks 1981), was taken to be representative of the non-thermonuclear tests conducted in the Marshall Islands.

Results

Tests depositing fallout in the Marshall Islands

The 20 tests we estimate deposited substantial fallout on any of the inhabited atolls of the Marshall Islands are presented in Simon, Beck et al. (2009, Table 1). The list includes one test in 1948, two in 1951, two in 1952, six in 1954, three in 1956, and six in 1958. Though there were 45 tests conducted at Bikini and Enewetak (DNA 1979; Simon et al 2009a) in addition to those shown in Table 1 of Simon, Beck et al (2009), we believe those 45 tests deposited local or regional fallout primarily only on the test site atolls of Bikini and Enewetak and on the open ocean outside of the area of the Marshall Islands. This conclusion is based on our analysis of available monitoring and meteorological data, as well as corroboration by data in unpublished reports.

Figure 1 shows an example of calculated directions of travel of air masses as they moved away from the nuclear test site, clearly illustrating the fallout moving away from the Marshall Islands. The air-mass trajectories were reconstructed the NOAA-HYSPLIT model and archival meteorological data. Using the HYSPLIT model one can determine whether air masses likely moved towards or away from the inhabited atolls of the Marshall Islands. In the absence of actual measurements, this type of analysis helped confirm our conclusions on which tests likely deposited fallout on the inhabited atolls.

Fallout time-of-arrival (TOA)

Estimates were made of the TOA of fallout for each test at each atoll. These estimates, expressed in hours post-detonation, are presented in Table 6. Times of arrival varied considerably depending on the distance of each atoll from the test site, wind speed, and wind direction. For example, at Rongelap in the northern Marshall Island, estimated TOAs varied from as short as six hours for BRAVO to about 80 hours for NECTAR and Zuni. Conversely, at Majuro in the southern Marshall Islands, TOA varied from about 36 hours for King to about 140 hours for YOKE. The shortest TOA was about four hours for BRAVO fallout at Ailinginae and the longest TOA was about 170 hours (>1 week) for UNION fallout at Kili Island.

The TOA estimates tend to be conservative in that, as illustrated by both auto-monitor and GF data, the fallout at some atolls often continued over many hours or days. Since gummed film data represented the deposition for a 24-hour interval, our estimates of the time of initial TOA are uncertain to about ±12 h if only based on those data. Although the initial time of arrival can be determined more precisely from the auto monitor data which recorded data every 6 hours, in many cases, the fallout arrived over a period of many hours to many days with no specific peak. For other tests, the TOA could only be crudely estimated based on whether fallout occurred prior to an airplane survey or between repeated surveys. For tests with little actual data, the fallout and TOA was estimated from the HYSPLIT model. As discussed in a companion paper in this issue (Moroz et al. 2009), the HYSPLIT model tended to predict a larger TOA than indicated by actual data, particularly at large distances, possibly a result of small errors in the wind speed input data, although errors in wind direction vectors and in the debris cloud model could also be partly responsible The actual TOA may also have been shorter than predicted due to rainout from clouds that otherwise would not have reached ground level (Moroz et al. 2009).

Estimated ^{137}Cs deposition at each atoll from each test

Using the various types of measurement data discussed earlier, supplemented by calculations from the NOAA-HYSPLIT model, deposition estimates have been made for each atoll for each of the tests listed in Simon et al. (2009a, Table 2). A best estimate of ^{137}Cs deposition was made from all available data for each test and each atoll. Results, expressed in Bq m^{-2}, are presented in Table 7. If no actual data were available for a particular atoll, estimates were based on interpolation based on the observed pattern of fallout measurements made at nearby atolls. When there was reason to suspect the quality of any measurement data, again based on the pattern of fallout at nearby atolls, some initial deposition estimates were subsequently modified to achieve better agreement of the total estimated fallout with the measured soil inventory data.

A major problem in making a best estimate of fallout on a given atoll from the available data was deciding which data were more reliable when estimates based on different measurements did not agree. One problem was that no information was available on the exact locations of most of the various measurements. Some of the atolls such as Kwajalein are very large and fallout could have varied over the atoll area. Ground survey measurements were generally few in number and, thus, might not be representative of the average fallout. The airplane survey measurements were usually the maximum recorded reading during the flyover of the entire atoll. However, because of the flight altitude, the measured value, even after correction for altitude, may often be lower than the actual highest ground level value, especially for the smaller islands.

It is also difficult to interpret data from measurement made at times of several weeks or more after the initial fallout occurred. In such cases, heavy local precipitation between the time of deposition and measurement would have resulted in a reduction in the exposure rate as discussed in Bouville et al. (2009). The possibility of weathering was considered in our assessment of Ė12 for a particular test or atoll, particularly when the data were obtained over a several day period or when the measurements were inconsistent.

Because the estimates of ^{137}Cs deposition were uncertain, a key test of their overall validity was the high degree of agreement between the cumulative estimated ^{137}Cs deposited at each atoll with the total inventories measured many years later. This agreement is discussed below under “Estimates of total ^{137}Cs deposition from all tests”.

Uncertainty in ^{137}Cs deposition estimates

As discussed, a best estimate of the Ė12 exposure rate from each test and the resultant ^{137}Cs deposited was made based on available data, HYSPLIT modeling, or interpolation. An uncertainty estimate was assigned to each of these estimates based on the quality and amount of available data and the uncertainty in the ratio of ^{137}Cs to Ė12. In general, if the estimates were based on multiple sources of consistent measurements, a geometric standard deviation (GSD) of 1.3 was assigned, if the data were sparser or somewhat inconsistent, a GSD of 1.5 was assigned, and if the estimate was based on a questionable measurement, a GSD of 1.8 was assigned. The GSDs assigned to the estimated ^{137}Cs deposition were identical to those for Ė12 unless there was fractionation, in which case the GSDs were increased to reflect the large uncertainty in the estimated ^{137}Cs to Ė12 ratio. If no data were available, and an estimate was based on interpolation of data at nearby atolls, the uncertainty estimate was based on the quality of the data at those atolls and the apparent variability as a function of distance. For a few tests, particularly those prior to 1952, no actual monitoring data were available, and all fallout estimates were based on the meteorological modeling. For those estimates, a GSD of 3.0 was assigned to reflect the very high uncertainty. The net uncertainty in the total ^{137}Cs deposited from all tests at an atoll was calculated assuming all individual test estimates were uncorrelated.

The estimated SD in the effective half life was used to estimate the resultant SD in decay to either 1978 or 1991-93. The latter was then combined with the estimated uncertainty in the sum of the measured fallout ^{137}Cs to estimate the overall uncertainty in the expected inventory in 1978 or 1991-93.

Estimates of total ^{137}Cs deposition from all tests

The total ^{137}Cs deposition at each atoll from all tests was summed and the uncertainty estimate in this sum calculated by combining the individual uncertainty estimates. The estimated total ^{137}Cs deposited at each atoll from all tests is shown in Fig. 2 of Simon, Beck et al. (2009), and a grouping of the atolls into four categories, based on similar magnitudes of cumulative ^{137}Cs deposition, is shown in Table 8. While these categories are relatively distinct in terms of the magnitude of the mean deposition density, the deposition on individual islands could vary considerably.

As a means to verify our estimates of ^{137}Cs deposited from each test at each atoll, the deposition estimates at each atoll from each test were each decay-corrected to later years when the inventory of ^{137}Cs was measured using the effective decay rates discussed earlier. Thus, in making the decay correction, we considered not only radiological decay but, also, the environmental loss, i.e. the gradual loss of ^{137}Cs from the upper layers of soil due to downward migration as a result of long-term precipitation. The loss of ^{137}Cs due to both decay and migration are described by the effective decay rate described previously. The sum of the decay-corrected ^{137}Cs depositions from each of the tests was compared with the inventory (corrected for global fallout ^{137}Cs) at the same atoll measured years after the testing had ceased. A ratio of the estimated total inventory (from summing depositions from each test) to the measured inventory (also reflecting all tests) that was reasonably close to unity was considered as evidence or corroboration that the individual test-specific depositions were reasonable. Where the ratio deviated from unity by more than our estimate of the uncertainty on our sum of the cumulative deposition from all tests, we considered the possibility that some of the measurement data were problematic or that an important source of additional fallout was unaccounted for. To resolve those inconsistencies, we used plots of the geographic pattern of the estimated fallout deposition from each test, as well as from all tests together, supplemented with the patterns of fallout deposition predicted by a meteorological model (NOAA-HYSPLIT), to identify where our interpretations of data might have been faulty. These steps were implemented to assist us reformulating individual deposition density estimates with the overall purpose of improving parity between the sum of our test-specific deposition estimates and measurements of total inventory.

It is important to note that for many tests and atolls, only sparse historical measurement data were available. In addition, contemporary measurements, which generally are of high precision, can have limitations in their usefulness since soil disturbance over the intervening years sometimes render them unrepresentative of the original deposition. The iterative technique of comparing fallout measurements with fallout estimates, followed by adjustments based on geographically-based patterns, was used to achieve an improved concordance between the total measured soil ^{137}Cs inventory and the total estimated ^{137}Cs deposition. We believe that more reliable estimates of the deposition from individual tests could be made this way compared to relying only on sparse historical measurement data.

The calculated inventory of ^{137}Cs for each atoll for 1978 and 1991-93 was compared with the average soil inventory measured by Robison et al. (1982) at 12 atolls in 1978 and by Simon and Graham (1997) at 30 atolls in 1993 (see Table 9). Since the depositions occurred at different times, each individual test deposition estimate was decayed (using the effective decay rates given in Table 2) to the time of a relevant soil measurement before summing to make this comparison. The uncertainty in the ratio was calculated from the estimated uncertainty in the

individual deposition estimates, in the atoll soil inventory estimates, and in the effective decay. Considering the large uncertainties in each component of the ratios, the agreement is quite good. For almost all atolls, the final ratio of total estimated ^{137}Cs deposited (from summing test specific estimates) to the contemporary measurements of soil inventory were within the range 0.7 to 1.3, well within the range of uncertainty for the cumulative estimate (Table 9). While most of the individual atolls ratios are within the range 0.7-1.3, the overall weighted average ratio for both data sets is close to unity. This suggests that our estimated deposition values can be used to reliably estimate external and internal doses for all atolls even though the precision for each atoll individually is not the same. It also suggests that we have not neglected any substantial fallout on any atoll.

The ^{137}Cs deposition densities (kBq m$^{-2)}$ from the CASTLE (1954) tests BRAVO, ROMEO, and YANKEE, on four atolls, Majuro, Kwajalein, Utrik, and Rongelap, are compared in Fig. 2. We believe that the deposition densities at these four atolls represent the overall range of deposition from regional nuclear testing on inhabited atolls. The cumulative deposition density of ^{137}Cs in the southernmost atolls, which includes the capital of Majuro, was between 1 and 3 kBq m^{-2}. The cumulative deposition at many of the mid-latitude atolls (~10° N) ranged between 3 and 8 kBq m^{-2}.

Here it should be recalled that global fallout ^{137}Cs, which is not included in our estimates, was about 0.9 kBq m^{-2} across the Marshall Islands implying that the deposition of ^{137}Cs from regional fallout ranged from about the same value as global fallout on the most southern atolls, to about 8 times that from global fallout on the mid-latitude atolls (Kwajalein and nearby atolls). Although global fallout deposition was responsible for a large fraction of the ^{137}Cs measured in soils at some of the atolls, this does not imply the doses that were received locally from Marshall Island tests were comparable to the doses received from global fallout since, as discussed in the companion papers by Bouville et al. (2009) and Simon et al. (2009), most of the dose from Marshall Island tests to residents was from short-lived radionuclides.

For purposes of comparison only, mid-latitude continental locations (30°-40° N) received much higher ^{137}Cs deposition from global fallout than did the Marshall Islands. Based on extrapolation from measurements of ^{90}Sr in fallout collectors (Larsen, 1983) at numerous locations worldwide, ^{137}Cs deposition density from global fallout in the Marshall Islands was only about 15-20% of that in the mid-latitudes.

A considerably greater amount of ^{137}Cs from regional nuclear testing was deposited at Utrik and at the small uninhabited neighboring atoll of Taka, about 20-30 kBq m^{-2}, or about 20 to 30 times the global fallout deposition of ^{137}Cs. The ^{137}Cs deposition on a northern group of atolls and islands that included Ailinginae Atoll (normally uninhabited), Rongelap Island (home to the Rongelap community), Rongerik Atoll (usually uninhabited) and Bikar Atoll (never inhabited) were considerably higher, ranging from 50 to 200 kBq m^{-2}. Those atolls clearly received fallout deposition from the BRAVO test.

The results of our study indicate that most of the total fallout deposition in the Marshall Islands occurred in 1954 from the CASTLE series tests. However, contrary to most every previous report, the CASTLE–BRAVO test was not the single test that contributed the largest fraction of fallout at every atoll. As a means to emphasize this point, we graphically compare deposition density of ^{137}Cs in Fig. 3. In the southern atolls of the Marshall Islands, epitomized by Majuro, the CASTLE-ROMEO test contributed the largest fraction of the total ^{137}Cs deposited. Conversely, in the northern Marshall Islands including Utrik and Rongelap, the BRAVO test was easily the largest contributor. At atolls at mid latitudes (Kwajalein and others), CASTLE-YANKEE contributed most of the fallout.

Estimates of deposition of radionuclides other than ^{137}Cs

Out of the several hundreds of fission products and activation products produced in a nuclear weapons test, the 63 most important radionuclides, including ^{137}Cs, representing over 99% of the potential ingestion dose, have been selected. Deposition densities of those 63 radionuclides have been estimated for each of the 20 tests that resulted in any fallout deposition in the Marshal Islands (other than at the test site atolls) and for each of the 31 atolls and separate islands that are considered. As discussed earlier, the dependence of activity ratios on fractionation was calculated by appropriately adjusting Hicks' (1981) activity ratios for BRAVO unfractionated fallout. Because of the widespread interest in the ground contamination by plutonium, deposition densities of $^{239+240}Pu$ also have been estimated, but only for the total activity deposited from all tests. A representative sample of the estimated deposition densities is shown in Table 10.

In general, the uncertainty in these deposition density estimates is driven by the uncertainty in the ^{137}Cs deposition density estimates. The uncertainties in the Hicks' (1981) activity composition as a function of R/V are small compared to the uncertainties in the estimates of R/V used to estimate ^{137}Cs deposition density from exposure-rate measurements. However, as discussed earlier, the good agreement between the sum of the various ^{137}Cs deposition density estimates with retrospective soil sample data, even for atolls with fractionated fallout, suggest that the deposition density estimates for all radionuclides of interest are sufficiently accurate to allow reasonable estimates to be made of radiation doses from external and internal exposure at all of the populated atolls of the Marshall Island.

Summary and Conclusions

While there have been numerous measurements made over the decades, of radioactivity in soil collected from many of the atolls (particularly the northern atolls and primarily of ^{137}Cs), no assessment of the deposition of all of the major radionuclides contributing to radiation exposure from each test has ever previously been made for all of the atolls of the Marshall Islands. In this paper, for the first time, the deposition densities of all the major radionuclides in fallout from individual nuclear weapons tests at Bikini and Enewetak Atolls have been estimated at each atoll of the Marshall Islands. These new deposition estimates are based on a thorough analysis of all the available data. All together, depositions at 31 atolls have been estimated for 63 radionuclides originating from the 20 nuclear tests which were likely to have deposited substantial amounts of fallout anywhere in the Marshall Islands. The agreement of the total estimated fallout at each atoll with measurements of soil radioactivity inventory from two different studies conducted years apart provides strong confirmation of the validity of the deposition estimates and that all incidences of substantial fallout are accounted for.

Previous estimates of fallout deposition in the Marshall Islands based only on contemporary measurements of ^{137}Cs in the soil generally underestimated fallout deposition due to considering only physical decay and neglecting the loss of ^{137} Cs from the soil horizon over time.

Deposition of ^{137}Cs, one of the more important of the moderately long-lived nuclides, and representative of the total fallout deposited, varied considerably over the Marshall Islands, but the atolls can be grouped into four groups of similar levels of contamination. Atolls south of Kwajalein, but including Ujae received cumulative deposition of only few times that of global fallout, in the range of one to three kBq m^{-2}. Seven mid-latitude atolls (Kwajalein and others) received slightly higher cumulative deposition, about three to eight times the global fallout deposition. Utrik received considerably higher deposition, about 30 kBq m^{-2}, while Rongelap Island received about 175 kBq m^{-2}, close to 200 times that of global fallout.

In the southern and mid-latitude atolls of the Marshall Islands (inclusive and south of Kwajalein), the ROMEO test contributed the largest fraction of the total deposition density of ^{137}Cs. At atolls north of Kwajalein, the BRAVO test contributed the largest fraction of the total deposition density of ^{137}Cs. In the mid latitude region (Kwajalein), YANKEE contributed the largest fraction.

Times of transit for fallout to reach atolls varied from as short as four hours for BRAVO fallout to reach Ailinginae to about 170 hours for UNION fallout to reach Kili Island. All important radionuclides in fallout were estimated based on atom ratios derived for a thermonuclear or non-thermonuclear test as a function of fractionation and the estimates of fallout transit time.

Two companion papers present estimates of radiation doses to Marshallese from external irradiation (Bouville et al. 2009) and from internal irradiation (Simon et al. 2009) that are based on the deposition densities reported here. Estimates of radiation-induced cancers derived from the dose estimates are presented in a fourth companion paper (Land et al. 2009).

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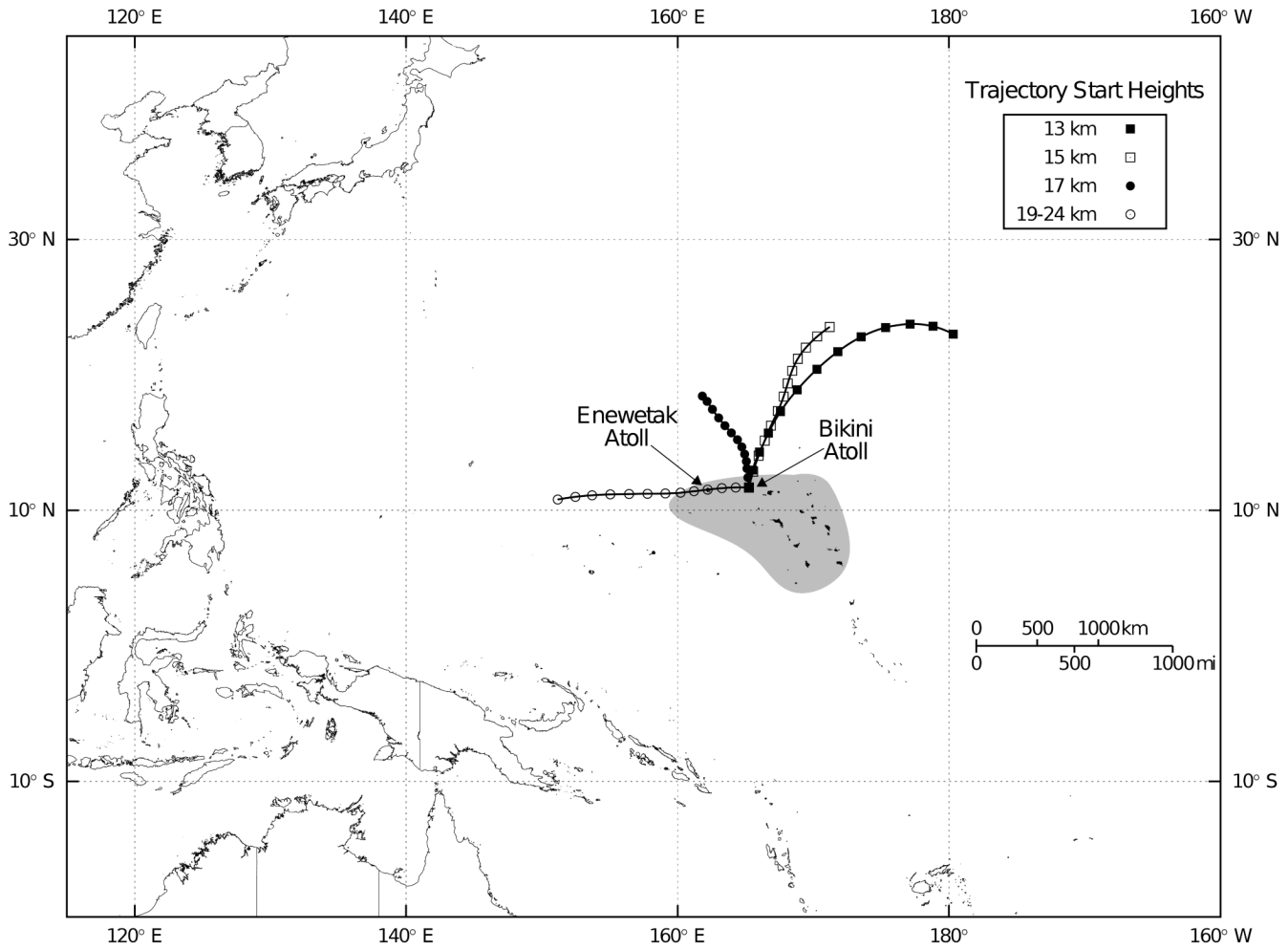

Fig. 1.

Upper level air mass trajectories moving W and NE away from Bikini Atoll (i.e., away from Marshall Islands, shown as gray shaded area) during the first 39 h following the POPLAR detonation as derived from archival meteorological data and simulated with the NOAA-HYSPLIT model for the day of the Hardtack I POPLAR test (7/12/58, 9.3 MT explosive yield). Note: trajectory symbols represent three h intervals.

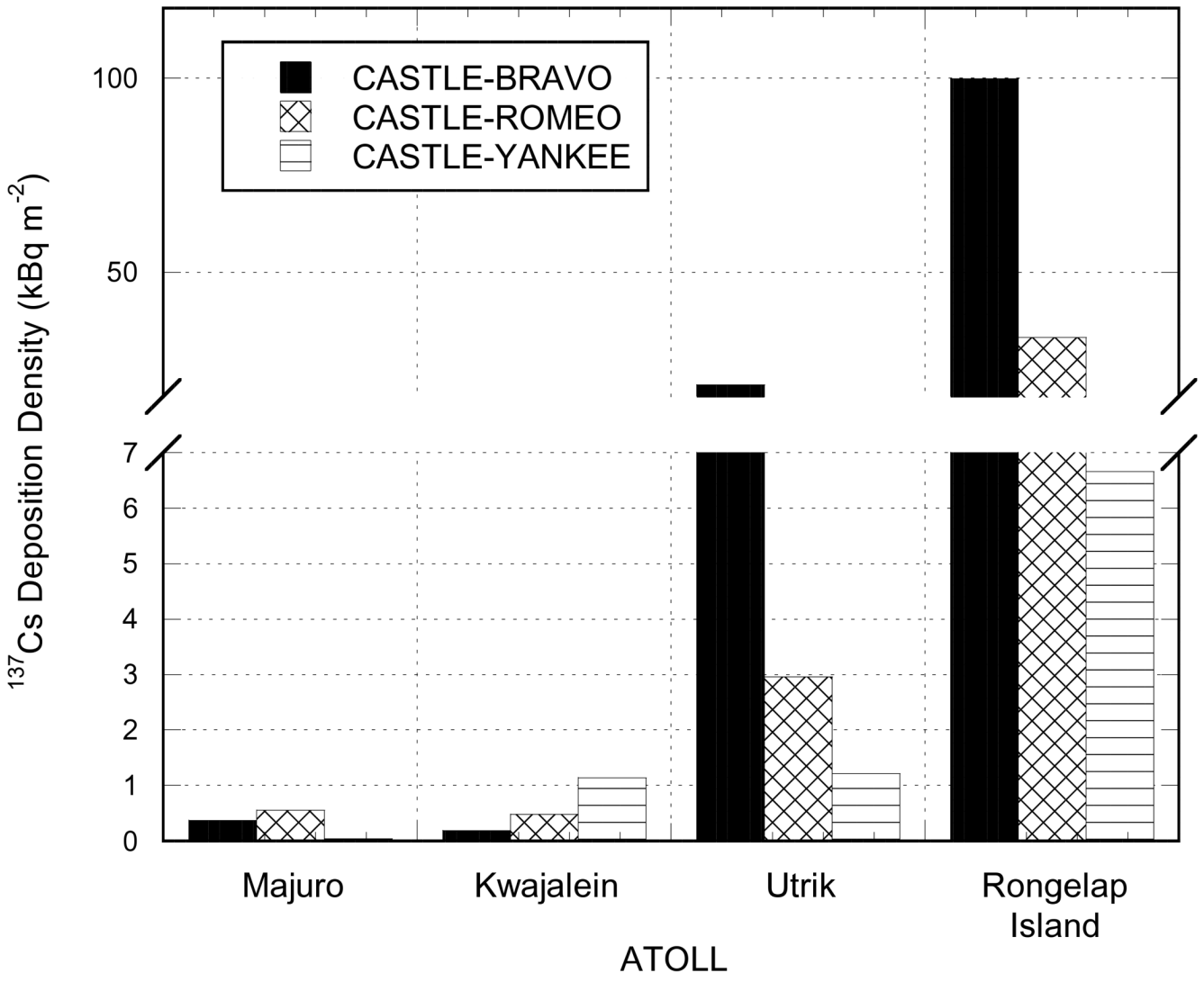

Fig 2.
Comparison of BRAVO, ROMEO, and YANKEE deposition densities (kBq m^{-2}) of ^{137}Cs at Majuro, Kwajalein, Utrik and Rongelap. Note break in Y-axis between 7 and 18 kBq m^{-2}.

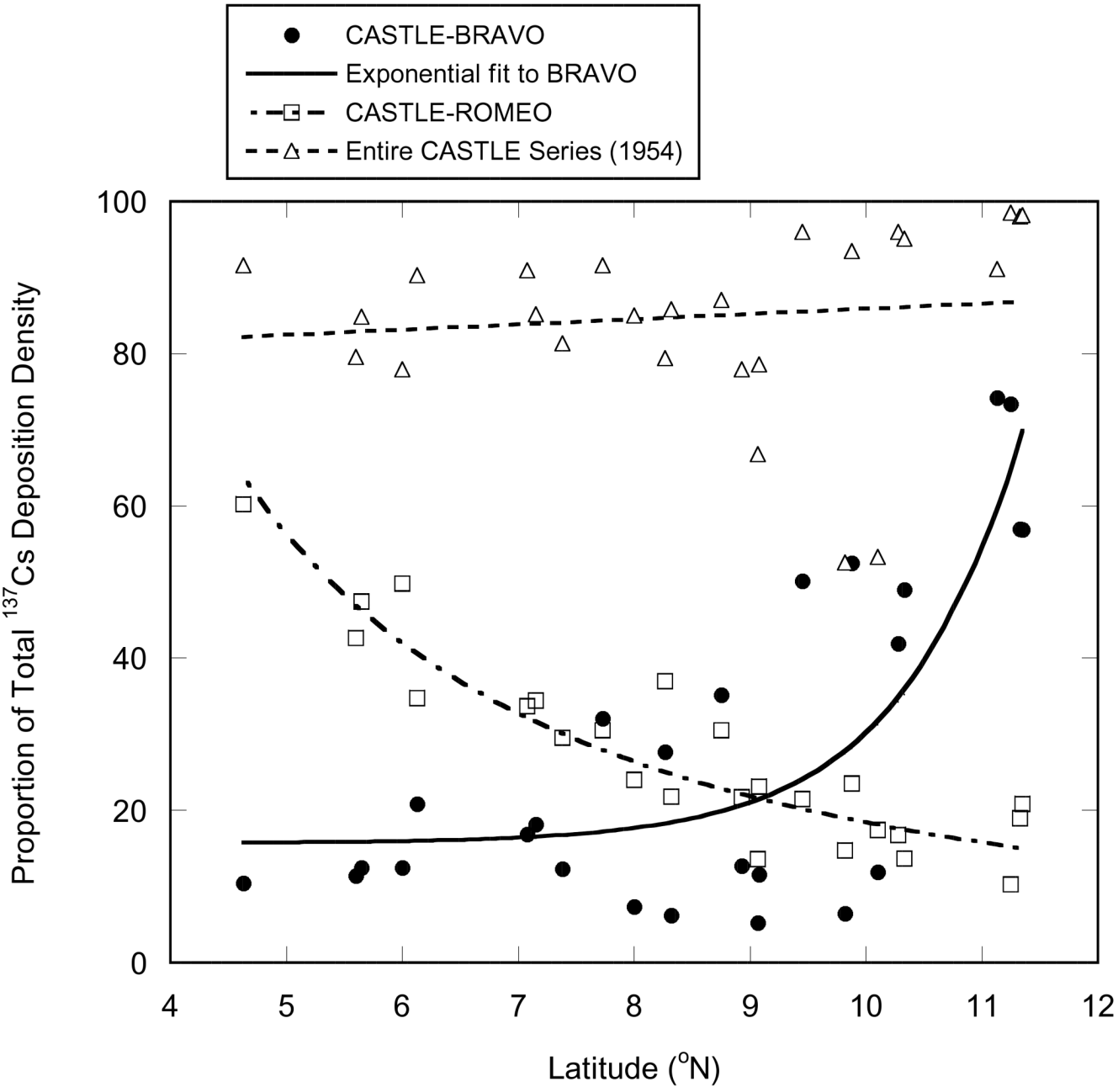

Fig. 3.

Percentage of total ground deposition density from BRAVO, ROMEO, and all CASTLE Series (1954) tests at 25 inhabited atolls and islands. The CASTLE series data points include the contributions from BRAVO and ROMEO as well as all other tests in that series. The abscissa represents the latitude (°N) of separate reef islands or the centroid location of atolls (except location for Kwajalein which represents the southern half of atoll at about 9.1 °N).

Table 1

Estimated ratios of ^{137}Cs/E12 (Bq m^{-2} per mR h^{-1} at H+12) as a function of R/V.

R/V	^{137}Cs/E12
0.5	31.8
1.0	20.7
1.5	7.8
2.0	5.2

Table 2

Estimated effective half-lives (y) of ^{137}Cs ordered (top to bottom) by increasing annual average rainfall.

Atoll or island	Estimated effective half-life (y)
Utirik	16
Rongelap Atoll: Rongelap Island	16
Rongerik Atoll: Eniwetak Island	16
Ailinginae	20
Taka	15
Ailuk	15
Mejit	14
Wotho	15
Jemo	14
Erikub	14
Ujelang	20
Wotje	14
Likiep	15
Kwajalein (northern half of atoll)	14
Ujae	14
Kwajalein (southern half of atoll)	14
Lae	14
Maloelap	14
Lib	13
Aur	14
Namu	13
Alinglapalap	13
Majuro	12
Arno	12
Jaluit	12
Knox	11
Namorik	11
Kili	12
Ebon	12
Mili	11

Table 3

Estimated activity ratios of $^{137}Cs/V^{a}$, $^{137}Cs/^{131}I$, $^{137}Cs/^{90}Sr$, as a function of R/V

R/V	TOA/T_{cr}	($^{137}Cs/V$) *a*	($^{137}Cs/^{131}I$) *b*	($^{137}Cs/^{90}Sr$)
0.5	1	1.1	1.32×10^{-3}	1.1
1.0	0.5 to <1.0	1.0	1.20×10^{-3}	1.0
1.5	0.25 to >0.5	0.7	0.84×10^{-3}	0.85
2.0	0.1 to <0.25	0.6	0.72×10^{-3}	0.75
3.0	<0.1	0.5	0.6×10^{-3}	0.63

a Cs-137/V represents the activity of ^{137}Cs to any other volatile nuclide relative to the ratio for unfractionated debris.

b At H+12. Does not include ^{131}I that will grow in from ^{131}Te and ^{131m}Te precursors.

Table 4

$^{137}Cs/^{239+240}Pu$ activity ratios measured in 1978 soil samples (Robison et al., 1981) and estimated fractionation (R/V) for BRAVO fallout. The estimated uncertainty (1 Std. Dev.) of the fractionation estimate is shown in parenthesis.

Atoll or island	$^{137}Cs/^{239+240}Pu$	R/V (Est. Std. Dev.)
Northern Rongelap Atoll	4	2.0 (0.5)
Rongelap Atoll: Rongelap Island	6	1.4 (0.2)
Alinginae Atoll	8	1.3 (0.2)
Rongerik Atoll: Rongerik Island	14	1.0 (0.2)
Rongerik Atoll: Eniwetak Island	3	1.5 (0.2)
Taka Atoll	13	1.0 (0.2)
Utrik Atoll	6	1.0 (0.2)
Ailuk Atoll	9	1.0 (0.1)
Mejit Island	8	0.9 (0.1)
Likiep Atoll	12	0.7 (0.2)
Ujelang Atoll	12	0.5 (0.1)
Wotho Atoll	17	0.5 (0.1)

Table 5

Variation in selected nuclide activity ratios and in ^{137}Cs/E12 quotients (Bq m^{-2} per mR h^{-1}) for various thermonuclear tests resulting in significant fallout in the Marshall Islands. Values for a typical NTS fission test (TESLA) are also given for comparison. All ratios are for R/V=0.5 and H+12.

TEST	$^{131}I/^{137}Cs$	$^{89}Sr/^{137}Cs$	^{137}Cs/E12	$^{140}Ba/^{137}Cs$
MIKE	823	158	31	695
BRAVO	838	124	32	674
ROMEO	840	145	31	679
YANKEE	840	124	31	682
ZUNI	873	129	33	685
TEWA	899	123	32	684
TESLA	866	70	44	528

Table 6

Estimated times-of-arrival of fallout (TOA) in hours after each detonation at each atoll considered.

	Yoke	Dog	Item	Mike	King	Bravo	Romeo	Koon	Union	Yankee	Nectar	Zuni	Flat-head	Tewa	Cactus	Fir	Koa	Maple	Red-wood	Cedar
Ailinginae	24	42	-	~40	24	4	30	12	24	30	80	70	31	-	-	52	-	-	72	-
Ailingaplap	148	-	-	~40	24	52	110	24	85	40	120	-	51	-	-	128	-	-	-	-
Ailuk	140	56	-	~40	24	27	90	24	30	35	-	70	-	-	-	60	-	-	-	-
Arno	135	-	-	~40	24	48	120	72	120	48	-	-	65	-	-	115	-	-	-	-
Aur	136	134	-	~40	24	42	115	65	80	40	-	-	60	-	-	113	-	-	-	-
Bikar	40	58	-	~40	26	16	60	24	53	23	-	-	127	-	-	37	-	-	-	-
Ebon	156	-	-	~40	-	54	150	100	125	90	130	-	-	-	-	135	-	-	-	-
Erikub	141	-	-	~40	28	40	110	60	60	45	-	-	64	-	-	70	-	-	-	-
Jabat	143	-	-	~40	24	50	115	30	85	40	120	-	18	-	-	135	-	-	-	-
Jaluit	150	-	-	~40	36	54	140	90	170	110	130	-	60	-	-	126	-	-	-	-
Jemo Island	142	56	-	~40	24	25	90	24	40	35	-	70	-	-	-	60	-	-	-	-
Kili Island	153	-	-	70	-	56	144	85	170	110	130	-	59	-	-	131	-	-	-	-
Knox	148	-	-	~40	36	50	120	80	140	120	-	-	90	-	-	118	-	-	-	-
Kwajalein (N)	33	65	-	~40	-	35	100	16	52	30	110	50	42	-	-	80	-	-	-	-
Kwajalein (S)	36	65	-	~40	20	40	100	18	54	30	110	54	50	-	-	135	-	-	-	-
Lae	161	66	-	~40	20	40	90	24	54	40	90	60	43	-	-	141	-	-	-	-
Lib Island	155	73	-	~40	24	60	110	22	60	40	90	-	45	-	-	137	-	-	-	-
Likiep	144	-	-	~40	24	26	80	24	28	35	-	90	-	-	-	124	-	-	-	-
Majuro	137	-	-	~40	36	48	100	72	120	48	-	-	74	-	-	114	-	-	-	-
Maloelap	132	127	-	~40	24	42	110	60	90	40	-	-	65	-	-	112	-	-	-	-
Mejit Island	125	56	-	~40	28	30	110	30	35	40	-	-	70	-	-	60	-	-	-	-
Mili	148	-	-	~40	36	50	120	80	140	120	-	-	90	-	-	118	-	-	-	-
Namorik	152	-	-	~40	-	56	140	100	105	130	130	-	70	-	-	134	-	-	-	-
Namu	152	-	-	~40	24	50	110	24	60	40	110	-	48	-	-	132	-	-	-	-
Rongelap (N)	26	45	-	~40	-	6	32	18	24	30	80	80	32	-	-	54	-	-	73	-
Rongelap	26	45	-	~40	-	6	32	18	24	30	80	80	32	-	-	54	-	-	73	12
Rongerik	28	47	-	~40	-	8	35	18	25	30	-	90	54	-	-	56	-	-	72	-
Taka	145	66	-	~40	24	20	50	24	30	33	-	-	-	-	-	54	-	-	-	-

	Yoke	Dog	Item	Mike	King	Bravo	Romeo	Koon	Union	Yankee	Nectar	Zuni	Flat-head	Tewa	Cactus	Fir	Koa	Maple	Red-wood	Cedar
Taongi	43	-	-	~40	-	40	60	40	24	24	140	-	-	-	-	21	28	-	70	23
Ujae	162	68	-	~40	24	37	90	24	24	40	85	60	43	-	-	-		-	-	-
Ujelang	12	17	27	12	12	18	42	50	24	22	80	50	22	42	18	54	75	45	65	30
Utrik	50	58	-	48	-	22	60	24	31	35	-	90	100	-	-	54	-	-	-	-
Wotho	30	63	-	~40	-	13	90	24	45	35	80	50	66	-	-	66	-	-	-	12
Wotje	136	61	-	~40	28	39	100	50	50	40	-	-	68	-	-	70	-	-	-	-

Table 7

Estimates of ^{137}Cs deposition density (Bq m^{-2}) on atolls of the Marshall Islands from each of the nuclear tests with evidence of substantial fallout.

Atoll or Island	Yoke	Dog	Item	Mike	King	Bravo	Romeo	Koon	Union	Yankee	Nectar	Zuni	Flat-head	Tewa	Cactus	Fir	Koa	Maple	Red-wood	Cedar
Ailinginae	360	180	0	74	0	32,000	11,000	5,000	150	2,100	37	2,800	260	0	0	41	0	0	110	0
Ailingaplap	4	0	0	74	37	190	440	220	37	37	300	0	150	0	0	17	0	0	0	0
Ailuk	3	52	0	74	37	370	1,000	440	330	170	0	110	0	0	0	93	0	0	0	0
Arno	7	0	0	74	37	370	740	740	37	110	0	0	74	0	0	6	0	0	0	0
Aur	6	<1	0	74	37	560	740	110	37	150	0	0	260	0	0	37	0	0	0	0
Bikar	4	23	0	74	0	48,000	6,300	4,400	2,000	7,400	0	0	2	0	0	44	0	0	0	0
Ebon	4	0	0	74	0	150	860	150	37	74	37	0	0	0	0	41	0	0	0	0
Erikub	5	2	0	74	37	780	740	480	74	150	0	0	37	0	0	48	0	0	0	0
Jabat	5	2	0	74	37	180	440	370	37	300	0	0	150	0	0	21	0	0	0	0
Jaluit	5	0	0	74	37	190	740	150	37	37	11	0	190	0	0	27	0	0	0	0
Jemo Island	2	56	0	74	74	2,600	1,100	260	74	740	0	110	0	0	0	89	0	0	0	0
Kili Island	2	0	0	74	0	190	700	190	37	37	110	0	150	0	0	37	0	0	0	0
Knox	5	0	0	74	37	330	560	440	74	37	0	0	37	0	0	2	0	0	0	0
Kwajalein (N)	740	9	0	74	0	370	480	480	110	1,500	22	190	260	0	0	56	0	0	0	0
Kwajalein (S)	740	9	0	74	37	190	480	410	110	1,100	37	37	220	0	0	56	0	0	0	0
Lae	2	3	0	74	74	260	440	37	37	260	560	110	190	0	0	2	0	0	0	0
Lib Island	2	0	0	74	37	150	520	370	74	37	560	0	190	0	0	37	0	0	0	0
Likiep	3	0	0	74	74	2,900	1,300	150	74	740	0	150	0	0	0	59	0	0	0	0
Majuro	6	0	0	74	37	370	700	590	37	37	0	0	150	0	0	37	0	0	0	0
Maloelap	6	<1	0	74	37	850	740	410	37	74	0	0	150	0	0	48	0	0	0	0
Mejit Island	4	36	0	74	37	2,800	1,100	440	370	1,700	0	0	7	0	0	110	0	0	0	0
Mili	5	0	0	74	37	330	560	440	74	37	0	0	37	0	0	2	0	0	0	0
Namorik	6	0	0	74	0	150	560	220	7	37	74	0	150	0	0	37	0	0	0	0
Namu	3	<1	0	74	37	150	480	480	37	74	480	0	150	0	0	37	0	0	0	0
Rongelap (N)	350	140	0	74	0	480,000	33,000	22,000	10,000	6,700	11	2,200	220	0	0	220	0	0	110	0
Rongelap (S)	350	140	0	74	0	100,000	33,000	22,000	10,000	6,700	11	2,200	220	0	0	220	0	0	110	15
Rongerik	210	120	0	74	0	67,000	24,000	14,000	4,100	5,400	0	1,300	190	0	0	220	0	0	4	0
Taka	2	89	0	74	0	13,000	3,000	1,900	370	1,300	0	0	0	0	0	190	0	0	0	0

Atoll or Island	Yoke	Dog	Item	Mike	King	Bravo	Romeo	Koon	Union	Yankee	Nectar	Zuni	Flat-head	Tewa	Cactus	Fir	Koa	Maple	Red-wood	Cedar
Taongi	21	2	0	74	0	410	190	37	26	37	4	0	0	0	0	370	22	0	4	7
Ujae	1	8	0	74	37	190	370	37	26	150	480	37	190	0	0	0	0	0	0	0
Ujelang	13	220	890	37	37	260	590	190	330	330	410	37	37	410	22	37	7	74	74	11
Utrik	3	85	0	74	0	21,000	3,000	2,100	890	1,200	0	74	11	0	0	190	0	0	0	0
Wotho	440	67	0	74	0	480	700	260	150	330	220	930	220	0	0	110	0	0	0	37
Wotje	4	56	0	74	37	2,600	1,100	480	37	740	0	0	11	0	0	74	0	0	0	0

Table 8

Four groups of atolls and islands based on similar cumulative ^{137}Cs deposition densities (kBq m^{-2}) from regional fallout. Atolls are listed alphabetically within each group.

Atoll group	Atolls	Mean Cumulative Deposition Density of ^{137}Cs (kBq m^{-2})	Range of ^{137}Cs Deposition Density (kBq m^{-2})
Southern atolls	Ailinglaplap, Arno, Aur, Ebon, Erikub, Jabat, Jaluit, Kili Island, Knox, Lae, Lib Island, Majuro, Maloelap, Mili, Namorik, Namu, Ujae	1.9	1.3 – 2.4
Mid-latitude atolls	Ailuk, Jemo Island, Kwajalein, Likiep, Mejit Island, Ujelang, Wotho, Wotje	5.1	3.5 – 7.5
Utrik/Taka	Utrik, Taka	24	20 – 29
Northern atolls	Ailinginae, Bikar, Rongelap Island, Rongerik	82	54 – 117

Table 9

Ratio of ^{137}Cs inventory in 1991-93 and 1978 estimated from all available data to measured inventories by NWRS (Simon and Graham 1997) and LLNL (Robison et al 1978). The soil sample data obtained for Bikar and Knox are highly uncertain, resulting in the large standard deviation (S.D.) shown; the data for Bikar and Knox are not included in the weighted mean calculations.

ATOLL	NWRS data for 1991-93 (1 S.D.)	LLNL data for 1978 (1 S.D.)
Rongelap Island (south part of Rongelap Atoll)	0.9 (0.6)	0.8 (0.2)
Rongerik: Enewetak Island	1.0 (0.8)	1.2 (0.6)
Bikar	1.0 (0.1)	2.7 (1.4)
Utrik	1.0 (0.7)	0.8 (0.5)
Taka	0.8 (0.7)	1.1 (0.4)
Ailuk	1.1 (0.8)	1.1 (0.5)
Mejit Island	0.8 (0.4)	1.0 (0.5)
Jemo Island	0.8 (0.5)	-
Likiep	1.1 (0.7)	0.7 (0.3)
Wotje	0.8 (0.5)	-
Erikub	0.6 (0.4)	-
Maloelap	1.5 (0.7)	-
Aur	1.3 (0.6)	-
Wotho	1.1 (0.7)	0.9 (0.5)
Ujelang	0.8 (0.5)	0.7 (0.3)
Kwajalein (north part of atoll)	0.8 (0.5)	-
Lae	1.4 (0.8)	-
Ujae	1.1 (0.7)	-
Kwajalein (south part of atoll)	1.5 (0.7)	1.3 (na)
Namu	1.1 (0.7)	-
Lib Island	1.2 (0.7)	-
Alinglapalap	1.1 (0.7)	-
Majuro	1.2 (0.9)	1.5 (0.8)
Arno	1.2 (0.9)	-
Mili	0.8 (0.5)	-
Knox	0.8 (0.6)	-
Namorik	0.9 (0.5)	-
Jaluit	0.7 (0.6)	-
Kili Island	0.8 (0.6)	-
Ebon	1.2 (0.7)	-
Jabat	0.9 (0.6)	-
Arithmetic mean (1 S.E.M.)	1.01 (0.042)	1.00 (0.072)
Weighted mean[a] (weighted error on the mean)[b]	0.94 (0.012)	0.86 (0.012)

[a] $\mu' = \frac{\sum(x_i/\sigma_i^2)}{\sum(1/\sigma_i^2)}$ (Bevington 1969)

$$_b\sigma_\mu = \sqrt{\frac{1}{\sum(1/\sigma_i^2)}}$$

Table 10

Sum of deposition densities (kBq m^{-2}) from all tests for 24 selected radionuclides at four atolls. All are fission products unless otherwise noted.

		ATOLL			
Nuclide	**Half-life**	**Majuro**	**Kwajalein (S)**	**Utrik**	**Rongelap Island**
^{55}Fe*	2.7 a	1.8×10^{-1}	3.1×10^{-1}	4.7×10^{0}	4.2×10^{1}
^{89}Sr	51 d	2.2×10^{2}	3.5×10^{2}	3.7×10^{3}	2.8×10^{4}
^{90}Sr	29 a	6.9×10^{-1}	1.2×10^{0}	1.1×10^{1}	7.5×10^{1}
^{92}Y	3.5 h	5.8×10^{0}	8.3×10^{2}	3.3×10^{4}	2.7×10^{6}
^{93}Y	10 h	2.1×10^{2}	2.7×10^{3}	8.1×10^{4}	2.1×10^{6}
^{95}Zr	64 d	1.5×10^{2}	2.5×10^{2}	4.2×10^{3}	3.9×10^{4}
^{99}Mo	66 h	1.6×10^{3}	4.0×10^{3}	7.3×10^{4}	7.6×10^{5}
^{103}Ru	39 d	1.8×10^{0}	2.0×10^{2}	3.0×10^{1}	1.8×10^{2}
^{106}Ru	370 d	4.0×10^{1}	7.0×10^{1}	6.1×10^{2}	4.3×10^{3}
^{131m}Te	30 h	2.4×10^{2}	9.4×10^{2}	1.1×10^{4}	1.1×10^{5}
^{131}I	8.0 d	1.3×10^{3}	2.5×10^{3}	2.3×10^{4}	1.7×10^{5}
^{132}Te	78 h	2.4×10^{3}	5.7×10^{3}	5.7×10^{4}	4.6×10^{5}
^{132}I	2.3 h	2.5×10^{3}	5.9×10^{3}	5.8×10^{4}	4.7×10^{5}
^{133}I	21 h	2.5×10^{3}	1.3×10^{4}	1.7×10^{5}	1.9×10^{6}
^{135}I	6.6 h	1.7×10^{2}	4.6×10^{3}	8.5×10^{4}	2.7×10^{6}
^{137}Cs	30 a	2.0×10^{0}	3.5×10^{0}	2.9×10^{1}	1.8×10^{2}
^{140}La	1.7 d	9.0×10^{2}	1.1×10^{3}	8.2×10^{3}	3.2×10^{4}
^{141}La	3.9 h	7.0×10^{0}	8.5×10^{2}	2.5×10^{4}	3.3×10^{6}
^{141}Ce	33 d	3.8×10^{2}	6.4×10^{2}	7.8×10^{3}	4.6×10^{4}
^{143}Ce	33 h	1.1×10^{3}	3.9×10^{3}	8.1×10^{4}	1.0×10^{6}
^{144}Ce	280 d	2.0×10^{1}	3.5×10^{1}	5.3×10^{2}	4.7×10^{3}
^{145}Pr	6.0 h	3.0×10^{1}	1.0×10^{3}	3.8×10^{4}	2.1×10^{6}
^{239}Np*	2.4 d	9.1×10^{3}	2.4×10^{4}	4.4×10^{5}	4.8×10^{6}
$^{239+240}Pu$	24000/6600a	7.2×10^{-2}	6.6×10^{-2}	3.5×10^{0}	1.6×10^{1}

* Activation product