Paper

URINARY EXCRETION OF RADIONUCLIDES FROM MARSHALLESE EXPOSED TO FALLOUT FROM THE 1954 BRAVO NUCLEAR TEST

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Abstract—**Soon after the Bravo nuclear test at Bikini Atoll in the Marshall Islands on 1 March 1954, urine samples were collected for analysis of excreted radioactivity from native residents exposed to radioactive fallout on two atolls as well as from U.S. military personnel on a third atoll. The earliest acquired samples, obtained by the Los Alamos Scientific Laboratory (LASL), were assayed for various radionuclides and provided the first known measurements of ^{131}I in urine following exposure to fallout from a nuclear test. Over the course of 1954, many additional samples were collected by the LASL, as well as by the Atomic Energy Commission New York Operations Office's Health and Safety Laboratory and the Naval Radiological Defense Laboratory. Collectively, the groups sampled included Marshallese exposed on Rongelap and Ailinginae Atolls, American military weather observers temporarily resident on Rongerik Atoll, and sailors from the Japanese fishing vessel, the *Lucky Dragon*. While the bioassay measurement data and individual urine volumes have been crucial to various attempts to assess intakes of radioactivity and the related internal radiation doses among the Marshallese, those data have never been published in any peer-reviewed journal, but have been restricted to agency memoranda, laboratory reports, and summaries in some publications and book chapters. Reconstructions of internal doses to Marshallese in 1954 and in later years have depended on these data and, hence, they have considerable historical importance as well as importance to ongoing health risk projections for Marshallese. This paper presents much of the original data on urine volumes and radioactivity from the various assays of urine for radionuclides, and compares estimates of ^{131}I intakes made in 1954, 1985, 1987, and 2008.**

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INTRODUCTION

The Bravo nuclear test on 1 March 1954 (local Pacific time) resulted in early fallout on neighboring atolls in the Marshall Islands lying primarily to the east of the test site on Bikini Atoll (Fig. 1, Simon et al. 2010a). Exposure to the fallout resulted in moderate to high radiation exposures to small groups of native Marshallese and Americans living or staying on these nearby atolls (64 Marshallese on Rongelap Island of Rongelap Atoll, 18 members of the Rongelap community on Sifo Island, Ailinginae Atoll, 159 on Utrik Atoll, and 28 American military men on Rongerik Atoll). In addition, but not considered here, were 23 sailors on the Japanese fishing vessel, the *Lucky Dragon.*

The Marshallese exposed to Bravo fallout on the atolls directly downwind were, collectively, one of the first populations to be exposed to both high levels of internal and external radiation from radioactive fallout, the first population to provide information regarding late-effects of acute to moderately-protracted environmental exposures, and the first population to give information about the health effects of exposure to radioiodines. In companion papers, Bouville et al. (2010) and Simon et al. (2010b) provide an assessment of external and internal doses received by natives of all atolls of the Marshall Islands from all tests conducted in the Marshall Islands, including those highly exposed from Bravo fallout. The estimated internal doses of Simon et al. (2010b) are based, to a large degree, on the assay data reported here.

The primary pattern of high deposition from Bravo as reported by numerous references (e.g., Conard 1975; DNA 1979; Martin and Rowland 1982) was cigar-shaped, 32–64 km wide, and extended eastward towards Utrik Atoll (located 570 km east of Bikini) with a slight northward curvature (also see Lessard et al. 1985). The atolls of Rongelap, Rongerik, Ailinginae all lay at intermediate distances to Utrik and had either permanent or temporary resident populations at the time of the Bravo test. Much lower depositions were also received at atolls

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lying at more southern latitudes, e.g., Kwajalein, Majuro and others (Breslin and Cassidy 1955; Beck et al. 2010).

Prior to the Bravo test, there was little experience with assessing internal contamination from exposure to fallout or predicting the related health consequences. One central reason that investigators from the Los Alamos Scientific Laboratory (LASL) conducted the urine sampling was to correlate data collected during Operation Greenhouse (Enewetak 1951) and in the UK on determining the relative hazards of external and internal exposure to radioactive debris from atomic weapons.[§] The data obtained from bioassay were also believed to be potentially useful for radiation safety purposes in the future, particularly since the weapons testing program was envisioned to continue for years to come.

Soon after the exposures took place from the deposition of Bravo fallout, investigators at LASL realized that urinary excretion of radionuclides by the Marshallese could be used as an index of internal contamination,[§] and plans were made to obtain 24-h urine samples from the islanders and the military weather observers stationed on Rongerik.[§] On the 15th day after fallout, the first urine collection was conducted by LASL. Other institutions, in particular, the Naval Radiological Defense Laboratory (NRDL), and later Brookhaven National Laboratory (BNL), also collected urine samples in 1954; however, their purposes were primarily to document the effects and responses to, at that time, levels of environmental exposure without precedent (Bond et al. 1955; Conard et al. 1956).

The earliest report on radioactivity assays in urine samples collected from Marshallese was from one of us in the form of a laboratory memo referred to here as Harris (1954)**. In that document, summary results of assays (count rates and activity concentrations) of ^{131}I in urine from the exposed adult population on Rongelap Island and on nearby Sifo Island on Ailinginae were reported and simple estimates of thyroid doses from ^{131}I estimated. As will be discussed, the LASL urine samples, while obtained from individual adults, were mixed to form "pooled" samples from which aliquots were assayed for ^{131}I activity and other radionuclides. At a later date, a modest change in detector efficiency for ^{131}I measurements was determined.[††] Neither the modifications, nor the original data, however, were ever published.

In addition to collections by LASL, urine samples were also collected by the Atomic Energy Commission (AEC) New York Operations Office's (NYOO) Health and Safety Laboratory (HASL) (Bouville and Beck 2000) and the NRDL (Cronkite et al. 1956; Woodward et al. 1959). There does not appear, however, to be any single source of information on all urine samples obtained by the different laboratories. From the sparse historical record, it appears that some of the sampling efforts were partially coordinated. For example, urine samples collected by Navy personnel on 24 and 25 March 1954, under the NRDL auspices, were distributed to HASL, LASL, and NRDL.[‡‡] In 1958, the Walter Reed Army Institute of Research (Department of Nuclear Medicine), in conjunction with the Department of Pathology of BNL reported on the determination of internally deposited radioisotopes from urine samples collected in 1954. Those data appear to be counting data from which the radionuclide excretions (in terms of activity per day) were reported in 1959 by Walter Reed Army Institute of Research.

Possibly because of the poor documentation available on the various urine samplings conducted by different laboratories, James (1964), for an estimation of doses to Rongelap children, mistakenly assumed and reported that the pooled urine samples collected by LASL were 20.1% urine (by volume) from ages 5–16 y and 4.8% from ages <5 y. Those volumes described the Walter Reed samples reported by Woodward et al. (1959). The LASL samples, in fact, only included urine from adults.

In later years, dose assessments for the Marshallese on Rongelap, Ailinginae and Utrik (Lessard et al. 1985) and the American military on Rongerik (Goetz et al. 1987) were conducted, though none were published in peer-reviewed journals. Lessard et al. (1985) estimated external and internal thyroid doses to the Marshallese on Ailinginae, Rongelap, and Utrik from the Bravo test using data of Harris** and other kinds of information. Other authors later cited Lessard et al. (1985) as that analysis was easily the most thorough and best documented analysis at that time. However, all of the above reports incorrectly reported that the pooled urine sample collected by LASL included urine from children.

It is unusual, given the significance of the primary bioassay data, that after 55 years, it is so poorly documented in the open literature. However, one of us

[§] Telex communication (memorandum). Los Alamos Scientific Laboratory to USAEC, Washington, DC. 10 March 1954. Available at: http://www.hss.energy.gov/healthsafety/ihs/marshall/collection/data/ihp1d/400045e.pdf.

** Harris PS. A summary of the results of urine analysis on Rongelap natives Americans and Japanese fishermen to date. Memo to AEC. Los Alamos, NM: Los Alamos Scientific Laboratory; 1954.

[††] Harris PS. Bravo fallout. Written circa 1980, unpublished manuscript.

[‡‡] Memorandum from E.P. Cronkite (NRDL) to Merril Eisenbud (NYOO), 4 April 1954.

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(P.S.H.) conducted the very first urine sampling and the measurements of ^{131}I and has personal knowledge that explains those unusual circumstances. Within the first few days to weeks after the exposure took place, there was an immediate need for estimates of intake and radiation dose so that the medical community could plan for the appropriate care of the exposed Marshallese, based on their expectations of the late effects that might occur as a consequence of the exposures (Bond et al. 1955). To assist in understanding the extent of the contamination and for estimating exposures, urine samples were collected and radionuclide analyses made. However, the initial report by Harris** was marked for "official use only" making publication of the data at that time impossible. That classification was maintained until the mid-1980's. Other data useful to reconstructing doses, e.g., the urine volumes of the NYOO samples,[‡‡] were contained in memoranda not declassified until the mid-1990's. While all the memoranda referred to in this publication are now unclassified, the 1954 ^{131}I bioassay data are difficult to locate, even from document archives.

The importance of the LASL urine sample measurements is underscored by the fact that no estimates of intakes have been published that are independent of those data. More recently, Musolino et al. (1997) and Takahashi et al. (2001) discussed simple strategies to estimate intakes from other tests and at other atolls, generally based on assuming the total contemporary ^{137}Cs deposition was due to the Bravo test, an assumption shown to be incorrect in Beck et al. (2010).

While one purpose of this paper is to correct the misconception that urine from children was included in the LASL pooled urine samples measured for ^{131}I, the primary purpose of this publication is broader and includes: 1) to clarify, as well as possible, the historical record on the various sampling efforts, 2) to publish, for the first time, the available bioassay measurement data from the urine sampling conducted by LASL and, to a lesser degree, the other institutions that collected urine samples, 3) to provide a summary of the 1954 interpretation of the assay data (in terms of ^{131}I intakes), and 4) to provide a comparison of various intake estimates that have been reported (Harris**; Lessard et al. 1985; Goetz et al. 1987; Simon et al. 2010b).

METHODS AND REVIEW OF LITERATURE

Urine sampling

One of us (P.S.H.), on behalf of the H Division of LASL, conducted the first collection of urine samples from the Marshallese adults exposed on Rongelap and Ailinginae ([§,**,***]; Langham 1954) who had subsequently been evacuated to Kwajalein Atoll for decontamination and medical care (Bond et al. 1955). The LASL sampling strategy called for collection of complete 24-h urine samples as that was the standard method used at Los Alamos at that time for assessing body burdens of plutonium among workers.[§] However, during the planning stages, it was not known whether individual or pooled samples would be most amenable to the measurement technology available and to the level of activity in the urine.

The first LASL urine collections (16, 17, and 19 March 1954, see Table 1) were carried out with assistance from the medical practitioner of the Rongelap community.[†††] Details about the collection efforts on other days and by agencies other than LASL have not been found.

Four composited urine samples, each a mixture of urine from several persons, were assayed to produce the ^{131}I excretion data upon which all retrospective thyroid dose estimates from Bravo fallout have been based. The urine samples from individuals were collected by LASL from adults and mixed to form the four pooled samples.** One sample was from 35 Marshallese adults on Rongelap Island and was obtained on 16 March 1954 while a second sample from 31 Rongelap adults was obtained on 17 March 1954. A third sample from 9 Rongelap adults exposed on Sifo Island (Ailinginae Atoll) was obtained on 19 March 1954, and a fourth from 9 U.S. military weather observers temporarily resident on Rongerik was collected on 18 March 1954.[‡‡‡,§§§] In addition, nine individual urine samples for plutonium analysis were collected from the American weather observers on Rongerik and a pooled urine sample was obtained on 28 March and 14 April from two sailors of the Japanese fishing vessel, *Lucky Dragon No. 5*,[**,‡‡‡,§§§] that was in the vicinity of Bikini Atoll at the time of the Bravo detonation. The collection of these samples was briefly noted by Harris in 1954.** Table 1 summarizes the various urine collections which totaled at least 19 in 1954. The summary was developed from all literature that could be presently located.

*** Shipman TL. LASL Health Division Progress Report, Nov. 20–Dec. 20, 1954. Los Alamos: Los Alamos Scientific Laboratory (declassified in 1978). Available at: http://www.fas.org/sgp/othergov/doe/lanl/doe_marshall_isl/0671_a.pdf. Accessed 2 June 2010.

††† The medical practitioner was a man named *Jabwe* (P.S. Harris personal notes, also see Sharp and Chapman 1957). At that time, Marshallese typically went by a single name.

‡‡‡ Harris PS. Laboratory notebook, 6465. Verified for declassification, February 1980. Los Alamos, NM: Los Alamos Scientific Laboratory; 1954.

§§§ Harris PS. Laboratory notebook, 6742. Verified for declassification, February 1980. Los Alamos, NM: Los Alamos Scientific Laboratory; 1954.

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Table 1. Summary of collection dates of urine samples in 1954, sample size, and related information.

Institution responsible for sampling	Group sampled	Group code[a]	Date of collection (1954)	Days post detonation	No. of subjects (age category)	Description of samples	References
LASL[b]	Rongelap	LA316R	16 March	15	35 (adults of 48 evacuated)	24-h	**.‡‡‡.§§§ and personal notes of P.S.H.
LASL	Rongelap	LA317R	17 March	16	31 (adults from same group of 48 evacuees)	24-h	**.‡‡‡.§§§ and personal notes of P.S.H.
LASL	American military weather observers on Rongerik	LA318A	18 March	17	9 (adults)	24-h	**.‡‡‡.§§§ and personal notes of P.S.H.
LASL	Rongelap group on Sifo, Ailinginae	LA319S	19 March	18	15 (adults)	24-h	**.‡‡‡.§§§ and personal notes of P.S.H.
Unknown	Japanese fishermen of *Lucky Dragon*	LA328J	28 March	27	2 (adults)	Unknown volume, but assumed as 1 L d^{-1}	**.‡‡‡.§§§ and personal notes of P.S.H.
Unknown	Japanese fishermen of *Lucky Dragon*	LA414J	14 April	44	2 (adults)	Unknown volume, but assumed as 1 L d^{-1}	**.‡‡‡.§§§ and personal notes of P.S.H.
AEC-NYOO (HASL)[c]	Rongelap	NY325R	25 March	24	29 adults[d]	Unknown	**.‡‡‡.§§§ and personal notes of P.S.H., also see footnote d
AEC-NYOO (HASL)	Rongelap/PBM group[e]	NY325RP	25 March	24	13 adults[d] of 16 evacuees	Unknown	**.‡‡‡.§§§ and personal notes of P.S.H., also see footnote f and ‡‡‡
AEC-NYOO (HASL)	Rongelap group on Sifo, Ailinginae	NY325S	25 March	24	15 adults[d] of 18	Unknown	**.‡‡‡.§§§ and personal notes of P.S.H., also see footnote f and ‡‡‡
AEC-NYOO (HASL)	Rongelap	NY401R	1 April	30	31 adults	Unknown	**.‡‡‡.§§§ and personal notes of P.S.H.
AEC-NYOO (HASL)	Rongelap	NY501R	~15 May	~75	Unknown	Unknown	Cronkite et al. (1956)
AEC-NYOO (HASL)	Rongelap	NY601R	~1 June	~90	Unknown	Unknown	Cronkite et al. (1956)
AEC-NYOO (HASL)	Rongelap	NY901R	~1 September	~180	Unknown	Unknown	Cronkite et al. (1956)
NRDL	Rongelap and American military weather observers	NY403RA	3 April–8 April	33–38	26 total: 6 Americans (adults), 20 Marshallese (ages unknown)	24-h	Cronkite et al. (1956)
NRDL	Rongelap	NR413R	13 April	43	27 total: Marshallese: 8 < 5 y[g], 13 of 5–16 y[h], 41 >16 y[i], 25 Americans (adults)	24-h	Cronkite et al. (1956)
NRDL	Rongelap	NR416R	16 April	46	Unknown[j]	24-h	Cronkite et al. (1956)
NRDL	Rongelap	NR515R	~15 May	~75	10 > 16 y	24-h	Cronkite et al. (1956)
NRDL	Rongelap	NR601R	~1 June	~90	10 > 16 y	24-h	Cronkite et al. (1956)
NRDL	Rongelap	NR901R	~1 September	~180	68 total: Marshallese: 8 < 5 y[k], 15 of 5–16 y[l], 45 > 16 y[m]	24-h	Cronkite et al. (1956)

[a] Similar to the codes originally used by Harris: institution abbreviation (LA for LASL, NY for AEC-NYOO, NR for NRDL), month, day of month, group identifier: R (Rongelap), J (Japanese), A (Americans), S (Sifo, Ailinginae group from Rongelap).
[b] Los Alamos Scientific Laboratory (H Division).
[c] Atomic Energy Commission-New York Operations Office (AEC-NYOO, renamed HASL, i.e., the Health and Safety Laboratory, 1953).
[d] According to Woodward et al. (1959), 141 samples were collected by NYOO from 24 March through 24 September and analyzed as 19 pooled samples (aggregated by age) though the reference does not provide a breakdown of number of samples collected on each specific day.
[e] Marshallese group evacuated by seaplane, i.e., patrol bomber "Mariner" or PBM.
[f] Eugene Cronkite to Merril Eisenbud, 4 April 1954, Declassified Memorandum. Same as footnote ‡‡. Available from: https://www.osti.gov/opennet/servlets/purl/16366538-uKs8tj/16366538.pdf. Accessed 2 June 2010.
[g] Seven from Rongelap, 1 from Ailinginae.
[h] Eleven from Rongelap, 2 from Ailinginae.
[i] Thirty-one from Rongelap, 10 from Ailinginae.
[j] The number of persons sampled on 13 April and 16 April cannot be discerned due to inadequate explanations in Cronkite et al. (1956).
[k] Eight from Rongelap.
[l] Twelve from Rongelap, 3 from Ailinginae.
[m] Thirty-three from Rongelap, 12 from Ailinginae.

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The most important urine samples from the point-of-view of dose reconstruction were those assayed at the LASL at the end of March 1954 for ^{131}I (see Table 1). No other samples were obtained soon enough after exposure to successfully measure ^{131}I. The value of the samples to dose reconstruction requires careful consideration in regard to their completeness and representativeness, primarily because seemingly small volumes of urine were obtained. The Marshallese health assistant, who directed the collection, assured the on-site LASL investigator (P.S.H.) that complete 24-h urine samples could be collected, and afterwards, were collected. The original interpretation of the urine data by one of us (P.S.H.**) was that urine samples collected were complete 24-h collections. Herein, as well as in Simon et al. (2010b), we support the interpretation that, on average, those are legitimate conclusions.

Radioactivity measurements

Several types of measurements and assays for different radionuclides were performed in 1954 though some of the radiometric analyses were rudimentary by today's standards and, in some cases, the measurement techniques were non-specific or had insufficient detection sensitivity (Table 2 and Appendix). Table A1 summarizes the various assays conducted on urine samples collected in 1954. Here again, this summary was developed from the collection of literature available to us. Because of the importance of the LASL ^{131}I measurements to dose reconstruction, we emphasize those measurements, though in Table A1, we also briefly review the methods and findings of the other various radionuclides measurements based on descriptions that can be found today in archival documents.

LASL samples and measurements. Iodine-131 in adult urine samples following the deposition of Bravo fallout was measured at LASL.**,††,***,‡‡‡,§§§ There were primarily two types of measurements: measurements of gamma-ray emissions made on raw samples and on the volatile fraction.**,‡‡‡,§§§ Only the earliest samples collected by LASL and measured for ^{131}I within 30 d of exposure have offered quantitative measurement data useful for reconstructing doses to the thyroid gland from radioiodines.

Gamma activity measurements used a photomultiplier-based scintillation detector (Fig. 1) developed at LASL coupled to a 100-channel spectral analyzer. The detector system was referred to as the "scintillation arm counter" (SAC), because the operator's arm could be inserted into the chamber that held up to two 500-mL bottles. While no records have been found of the dimensions, wall material, and scintillator thickness, we believe the scintillation fluid to have been diphenyl oxazol (PPO) plus triphenyl dioxazol (POPOP) in toluene as used in the construction of a whole-body counter at Los Alamos (Anderson et al. 1956) with a similar but larger design.

The detector system was used for measurements of ^{131}I in liquid form. Measurements of both the raw urine sample and the volatile fraction were made.**,‡‡‡,§§§ It was found near the end of March 1954 that 77% of the gamma emissions were in the volatile component and it was identified to be ^{131}I through determination of the half-life.**,‡‡‡,§§§ However, the measurements of activity of ^{131}I in urine used for all subsequent intake calculations** (Lessard et al. 1985; Simon et al. 2010b) were

Table 2. Bioassay measurements reported by investigation group (Y indicates that assay was conducted).

Radionuclide	LASL (**,***, Langham 1954)	USNRDL (Cronkite et al. 1956)	Walter Reed (Woodward et al. 1959)
^{137}Cs	Y	—	Y
^{89}Sr	Y	Y	—
^{90}Sr	—	—	Y
^{103}Ru	Y	—	—
^{131}I	Y	—	—
^{140}Ba	Y	Y	—
^{45}Ca	Y		—
^{239,240}Pu	Y	Y	—
Gross beta-activity	Y	Y	—
Pu	Y	—	—

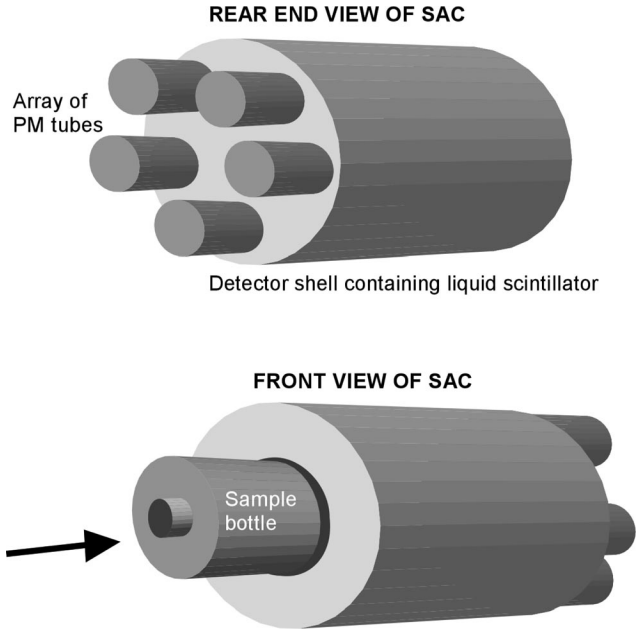

Fig. 1. Drawing of front and rear views of "scintillation arm counter" (SCA) developed at LASL for gamma activity measurements and used to assess ^{131}I activity of pooled urine sample in 1954.

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made on the raw (i.e., unseparated) sample. The detector efficiency for measuring gamma emissions from ^{131}I was estimated to be 39% at the time of the first reporting of results**,‡‡‡,§§§ and later modified by Harris to be 35%.††

Analyses of other radionuclides were also conducted at LASL as described by internal LASL reports from 1954.**,**** These included beta ray measurements of dried urine samples and analyses of plutonium in urine. Analyses for plutonium were based on an unidentified means of chemical isolation which was the procedure in use at LASL at that time, followed by alpha counting for 80 min.****

Assumptions for estimation of radioiodine intake

Intake and internal radiation dose calculations can be made with varying degrees of complexity and realism, and though few parameters are needed to make estimates of intake, calculations generally require assumptions, some that can be made with good assurances and others that can be difficult to verify. One assumption inherent in the interpretations of the ^{131}I assay data by Harris (1954**), was that the excretion followed a single intake of radioiodine from Bravo fallout. This appears as a good assumption because the last test depositing fallout at Rongelap prior to Bravo was the King test in 1952 (Beck et al. 2010), and there were no further tests depositing fallout before the urine samples were collected in mid-March of 1954.

While the simplest assumption concerning time of intake is to assume that intake occurs at the onset of deposition, other assumptions are clearly possible. For example, the total estimated intake could be partitioned into fractional intakes at various time following deposition, e.g., at meal-times. Though differences in assumptions about time of intake are not extremely important for ^{131}I, partitioning the total intake may be modestly important for dosimetry of the shorter-lived iodine isotopes. For example, Lessard et al. (1985) calculated total intakes based on the assumption that one-third of the ^{131}I was ingested 5.5 h post-detonation (H+5.5 h or ~12:15 pm) and two-thirds at H+12 h (~6:45 pm). In the earliest LASL assessment by Harris,** no such assumptions were made. In the work of Simon et al. (2010b), intake was considered to occur at the midpoint of the period of deposition which typically takes place for a period of time somewhat less than the elapsed time between detonation and onset of fallout (Simon et al. 2010b).

Another assumption made in 1954 was that ingestion was the primary mode of intake.** This conclusion follows from various arguments including the relatively large size of particles deposited at Rongelap, which would tend to preclude inhalation and has been subsequently supported by Lessard et al. (1985) and Simon et al. (2010b).

Based on these various assumptions, the intake (Bq) of radioiodine on the day of intake can be simply estimated by the quotient of the ^{131}I activity measured in the total daily urine output (decay corrected from time of measurement back to time of collection) and the fractional excretion on the day of collection per unit of intake (unitless).

The urinary fractional excretion on any single day following intake can vary, however, among individuals due to differences in individual metabolism, differences in ambient temperature, and differences in water losses from the body, primarily through the skin (Mao et al. 1990, 2001; ICRP 2002). There are few reported long-term empirical data (beyond a few days) of ^{131}I excreted in urine from an acute intake. Hence, for the most part, the daily excretion fraction must be predicted from a biokinetic model.

Various radioiodine biokinetic models have been published over the years. For example, the International Commission on Radiological Protection (ICRP 1989) published basic metabolic data for iodine in the body based on the description by Riggs (1952) and used that data to develop a three-compartment model with explicit representations for blood, thyroid, and the rest of the body. These models, however, were not available at the time of the exposures to Bravo fallout.

The estimated excretion fraction from an acute intake of ^{131}I (i.e., fraction of original intake excreted on day t) used by Harris in 1954** was 0.001 on H+16 d and H+17 d based on the advice of biokinetics expert, Joseph Hamilton (see, for example, Hamilton and Soley 1939, 1940; Hamilton 1948). However, because good data on the long-term excretion of radioactive iodine were not available at that time, the value of 0.001 was recognized as only an estimate and radioactive decay between time of intake and time of sample counting was accounted for in the decay factor of eqn (1).

Values of the excretion fraction of ^{131}I used by later investigators have varied within a range of two-fold; however, all these implicitly included radioactive decay between day of intake and day of sampling. For example, Lessard et al. (1985) provided an estimate of 1.4×10^{-4} for the early LASL samples as derived from ICRP (1979) and Johnson (1981). Goetz et al. (1987) estimated that the excretion fraction for the American military men on Rongerik was 3.07×10^{-4} on day 17 as derived from

**** Los Alamos Scientific Laboratory, Memo R3574, "Data on plutonium results from urine samples collected in the Marshallese." Undated. Declassified by Atomic Energy Commission, 3/23/1972. Obtained from archives of U.S. DOE Environmental Measurements Laboratory.

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Dunning et al. (1979, 1981) after making an adjustment for ingrowth of ^{131}I. In the work of Simon et al. (2010b), the best estimates of the excretion fraction for the Marshallese were about 1.70×10^{-4}, 1.65×10^{-4}, and 1.43×10^{-4} for days 16, 17, and 19, respectively (see Table A1, Simon et al. 2010b). The estimates in that work were based on a biokinetic model that simulates a relatively high daily water loss through the skin. Decay-correcting the estimate of the iodine excretion fraction of 0.001 originally used by Harris in 1954** for the elapsed time between intake and sampling gives an excretion fraction of 3.0×10^{-4}, a value very close to those used in the other assessments discussed.

While estimates of thyroid dose were presented by Harris in 1954** and others to follow, here we only focus on the data necessary for estimating intake. The reader is referred to Simon et al. (2010b) for a comparison of the dose estimates made over the 55 years since the acute exposures from Bravo fallout took place.

Estimating radioiodine intake

The earliest thyroid dose estimates** used simple estimates of ^{131}I intake derived directly from gamma spectrometric measurements of the count-rate of ^{131}I in each of the LASL pooled urine samples.**,‡‡‡,§§§ The intake (Bq) of ^{131}I can be estimated as shown in eqn (1) from the gamma-ray counting results and other parameter values provided in Table 3. Using the available counting data, only the average intake, $\bar{Q}$, among adults whose urine was sampled (or others for whom the data are a suitable surrogate) can be estimated:

$$\bar{Q}(^{131}\mathrm{I}) = \frac{CR \times K \times \bar{V}}{EF(t) \times \varepsilon_C} \quad (1)$$

where

$\bar{Q}$ = acute intake of ^{131}I intake (Bq, group average);
CR = background adjusted count rate (c s^{-1}) of ^{131}I per mL of urine;
K = correction factor corresponding to the radioactive decay of ^{131}I between time of sampling and time of counting;
$\bar{V}$ = 24-h urine volume (mL) averaged over the sampled population;
$EF(t)$ = urinary excretion fraction for ^{131}I on day of sampling, t being the time elapsed between intake and sampling; and
ε_C = gamma detector counting efficiency (count per decay).

If the estimated excretion fraction is derived from data from stable isotope experiments or is based on short-term observations, the decay correction may need to account for the total time between intake and counting to properly assess the intake of the radioactive isotope. This was the method used by Harris in 1954.** If the excretion fraction pertains specifically to ^{131}I and is derived from reliable measurements or a validated

Table 3. Bioassay data from Harris (**,‡‡‡,§§§) used in 1954 assessment of ^{131}I intake.

Group ID	Group sampled and date of sampling in 1954	Number of days from intake to sampling	Number of days from sampling to counting[a]	^{131}I counting results (c s^{-1} per 500 mL)	Decay correction[b]	Counting efficiency[c] (%)	Assumed urinary excretion on day of collection (%)[d]	Estimate of average daily urine volume (mL)
LA316R	Rongelap adults, 16 March	15	14	70	13.5	35	0.1	500
LA317R	Rongelap adults, 17 March	16	13	76	13.5	35	0.1	500
LA318A	Rongerik (American military weather observers), 18 March	17	12	20	13.5	35	0.1	500[e]
LA319S	Rongelap adults exposed on Sifo, Ailinginae, 19 March	18	11	33	13.5	35	0.1	500
LA328J and LA419J	Japanese fishermen (*Lucky Dragon*), 28 March and April 19	27 and 49	Unknown	0 (not detected above bkgnd)	~13.5	35	0.1	1 L (assumed)

[a] Counting date was 30 March 1954.
[b] Based on elapsed time from intake to counting of approximately 30 d and a half-life of 8 d.
[c] Original estimate estimate was 39% (**), later corrected to 35%††.
[d] Excretion fraction estimate from J. Hamilton (see text) based on data from short-term observations.
[e] Actual mean 24-h urine volume was 1,072 mL (Table 2).

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model, the decay correction need only to account for the elapsed time between sampling and counting. This was the method used by Lessard et al. (1985), Goetz et al. (1987), and Simon et al. (2010b).

RESULTS AND DISCUSSION

Urine volumes

The distributions of individual urine volumes in the three groups of samples collected by LASL from Rongelapese on March 16 ($n = 35$), March 17 ($n = 31$), and April 15 ($n = 21$) in 1954 are presented in Table 4 and Fig. 2.[‡‡‡,§§§] The mean urine volumes from the LASL collections in mid-March 1954 were similar; the average values were 427 mL (March 16, Rongelap), 448 mL (March 17, Rongelap), and 385 mL (March 19, Sifo). The distributions and mean urine volumes of the HASL samples (Fig. 3) from March were similar in magnitude to the LASL samples, but slightly higher, 596 mL (March 24, Rongelap), 523 mL (March 25, Rongelap), 756 mL (March 24, Sifo), 603 mL (March 25, Sifo), and 573 mL (April 15, Rongelap) (Cronkite et al. 1956).[‡‡]

Individual urine volumes from two groups of the American military weather observers resident on Rongerik at the time of Bravo who provided samples in 1954 on March 19 ($n = 9$ for beta activity measurements and $n = 10$ for Pu activity measurements) are shown in Fig. 4 and are also summarized in Table 4.[‡‡‡,§§§]

The urine volumes collected from the Marshallese were, on average, small compared to the usual range of 800 to 2,000 mL d^{-1} reported for populations with a typical fluid intake of about 2 L d^{-1} (MedlinePlus 2002). Not all Marshallese sampled, however, excreted these extremely small samples. For example, of those sampled on March 16, the volume for one urine sample was greater than 800 mL, of those sampled on March 17, three (12%) were greater than 800 mL, and of those sampled on June 15, six (30%) were greater than 800 mL.

Lower than average urine volumes are, in general, a result of either reduced fluid intake and subsequent dehydration or high water losses through feces or, more commonly, through the skin. Hence, one possible explanation for low urine volumes among the Marshallese was a well documented drought that had been underway in the northern Marshall Islands for a number of months prior to the Bravo test. Sharp and Chapman (1957) reported that "for many weeks prior to 1 March, the natives had been rationed to one pint cup per individual per day." The shortage of fresh water would have also affected those on Ailinginae and Utrik. The average urinary excretion for the American weathermen on Rongerik was significantly greater compared to the Marshallese (Table 4) and averaged about 1,100 to 1,200 mL per day.[‡‡‡,§§§] However, Rongerik, where the American military weather observers were located, had a water distillation unit and drinking water was available in 5-gallon cans at the time of evacuation (Sharp and Chapman 1957).

Another plausible explanation for the small average values of urine is the reduction of daily urine volume in

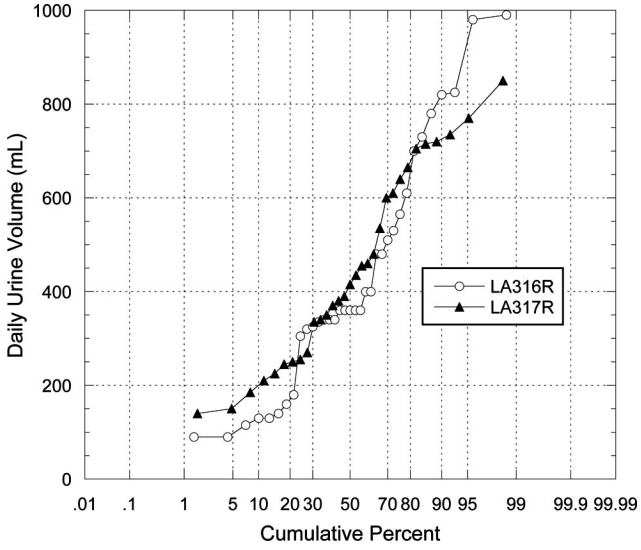

Fig. 2. Empirical cumulative probability distributions of urine volumes obtained from two Los Alamos Scientific Laboratory (LASL) samplings of Rongelapese greater than 16 y old at time of exposure (see Table 4 for a summary of the data).

Table 4. Summary statistics of sampled urine volumes (see Table 1 for references). All values are nominally mL per 24 h (na = not available).

	Group ID									
	LA316R	LA317R	LA319A	LA319AP[a]	LA319S	NY324R	NY325R	NY324S	NY325S	NY416R
No. of samples	35	31	9	10	15	40	43	12	15	21
Minimum	90	140	730	760	na	70	95	320	90	47
Maximum	990	850	1,345	1,525	na	980	980	965	985	980
Mean	427	448	1,072	1,197	385	596	523	756	603	573
Median	360	415	1,130	1,250	na	653	480	805	750	540
Std Error	42	37	71	86	na	47	44	57	76	59

[a] LASL sample on March 19 from American (A) military for plutonium (P) analysis.

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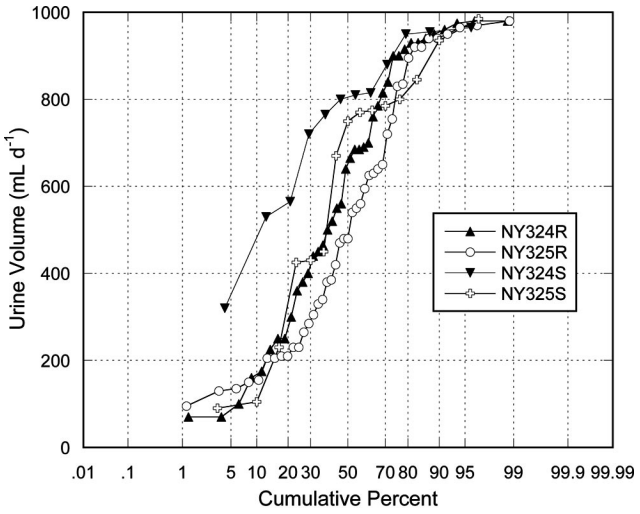

Fig. 3. Empirical cumulative probability distributions of urine volumes obtained from Rongelap and Ailinginae groups by the Health and Safety Laboratory (HASL AEC-NYOO) (see Table 4 for a summary of the data).

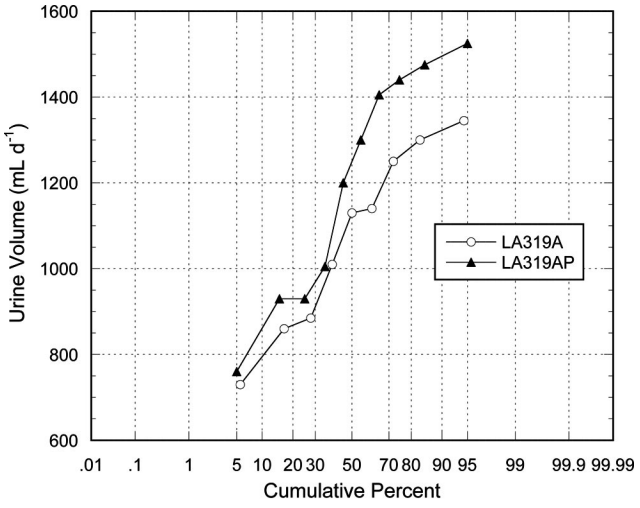

Fig. 4. Empirical cumulative probability distributions of urine volumes obtained from two Los Alamos Scientific Laboratory (LASL) samplings of American military weather observers on Rongerik (see Table 4 for a summary of the data).

the tropics that occurs as a consequence of perspiration and insensible water losses through the skin due to high humidity and temperatures. It is known that urine volumes in humid, tropical climates are typically much smaller than more temperate climates (Elebute 1973; Latham 1997); see the Appendix of Simon et al. (2010b) for a detailed discussion of that phenomenon. For these reasons, we believe that the relatively small volumes obtained from the Marshallese were, on average, reasonable.

^{131}I count rates and estimates of intake

Gamma-ray count rates per 500 mL from each of the four pooled urine samples (Table 3) and the related estimates of intakes in 1954** are presented here (Table 5). Urine samples were counted on numerous days to check for consistency of the decay rate.[‡‡‡,§§§] The count rate data reported here were specifically from 30 March 1954. Count rates from ^{131}I in the pooled urine samples obtained from adult Marshallese on March 16 and 17 on Rongelap were nearly identical: about 70 counts per second (c s^{-1}) per 500 mL obtained on 30 March 1954. The count rate from the pooled sample of Rongelapese on Sifo Island, Ailinginae Atoll, at the time of the Bravo test was 33 c s^{-1} per 500 mL, about one-half of the value for the adults exposed on Rongelap.[‡‡‡,§§§]

The count rate in a pooled urine sample from the American military weather observers on Rongerik, made on an equal volume of urine as that for the Marshallese samples, was less than one-third of that of the Rongelap samples, about 20 counts per second per 500 mL,**[‡‡‡,§§§] though when adjusted to the approximate 1 L per day urinary excretion of the Americans, the count rate per 24-h excretion was 40 c s^{-1}, higher than on Ailinginae. This is explained by the two-fold higher deposition of ^{137}Cs on Rongerik compared to Ailinginae (see Table 7, Simon et al. 2010b).

The intakes of ^{131}I originally estimated in 1954 by Harris** were 2.1 MBq (56 μCi) for the Rongelapese and 0.65 MBq (17.5 μCi) for the Rongerik group. Revised estimates by Harris[††] that included minor revisions for detector efficiency were 2.8 MBq for the Rongelap group, 1.3 MBq for the Ailinginae group, and 0.78 MBq for the Rongerik group (Table 5).

Comparison of estimates of intake of radioiodines

A comparison of adult male and female average intakes of ^{131}I for Marshallese exposed on Rongelap and Ailinginae to Bravo fallout is made in Table 6 from estimates derived by Harris in 1954** and later modified,[††] Lessard et al. (1985), and Simon et al. (2010b). Similarly, a comparison of estimates of intake for American military weather observers exposed to Bravo fallout on Rongerik is made in Table 6 from estimates presented by Harris in 1954,** Goetz et al. (1987), and Simon et al. (2010b).

Average estimates of intakes of ^{131}I among adults, depending on the assessment, ranged from 2.8 to 3.5

Table 5. Original and revised estimates of intakes (MBq) of ^{131}I by Harris.[a]

Sampled group	Original estimates (**)	Revised estimates (††)
LA316R and LA317R	2.1	2.8
LA318A	0.65	0.78
LA319S	nr	1.3

[a] "nr" is not reported.

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Table 6. Comparison of estimates of average acute intake (MBq) of ^{131}I among exposed Marshallese and American groups following deposition of Bravo fallout. All values are rounded to two significant digits.

Group and source of estimates	Gender (adults)	^{131}I (MBq)
Rongelap Island group		
Harris (**,††)	Male-female average	2.8
Lessard et al. (1985)	Male-female average	3.4
Simon et al. (2010b)[a]	Male-female average	3.5
Sifo, Ailinginae group		
Harris (**,††)	Male-female average	1.3
Lessard et al. (1985)	Male-female average	0.69[b]
Simon et al. (2010b)[c]	Male-female average	1.2
American military on Rongerik		
Harris (**,††)	Male	0.78 (1.7)[d]
Goetz et al. (1987)	Male	0.56 (1.2)[e]
Simon et al. (2010b)	Male	1.7

[a] Intake estimates here use the Simon et al. (2010b) model assumptions and the Table 3 bioassay data (Harris 1954, footnote §) from 3/16/54 and 3/17/54 for Rongelap, while intake estimates for Rongelap in Simon et al. (2010b) used a weighted value of intake per unit deposition from Rongelap and Ailinginae.
[b] The intake estimate by Lessard et. al. (1985) at Sifo is not solely based on urinary ^{131}I measurement but is weighted by fallout particle sizes, external exposure rate measurements, and considers meal times, and time-of-arrival.
[c] Intake estimates here use the Simon et al. (2010b) model assumptions and the Table 3 bioassay data for Ailinginae while intake estimates for Ailinginae in Simon et al. (2010b) used a weighted value of intake per unit deposition from Rongelap and Ailinginae.
[d] 500 mL urine volume (same as for Marshallese) was used in this calculation; use of 1,100 mL urine volume for LA319A (see Table 2) would have given 1.7 MBq.
[e] 500 mL urine volume (same as for Marshallese) was used; use of 1,100 mL urine volume for LA319A (see Table 2) would have given 1.2 MBq.

MBq for the adults exposed on Rongelap, from 0.7 to 1.3 MBq for the Rongelap adults exposed on Ailinginae, and from 0.6 to 1.7 MBq for the American military weather observers exposed on Rongerik (Table 6). In this context, "average" refers to the mean value for the group of adults sampled and that contributed to the pooled urine sample. Those data can be reasonably assumed to be applicable to other adults on the atoll who were not sampled on that day.

An average of male and female intake estimates is presented in Table 6 for comparison with estimates from other investigators. The estimated intake of ^{131}I among adults on Rongelap was 2.8 MBq by Harris, 3.4 MBq by Lessard et al. (1985), and 3.5 MBq by Simon et al. (2010b). The primary differences can be attributed to the assumed volume of urine excreted in 24 h and the fractional excretion on the day of sampling (eqn 1). A comparison of the estimated intakes of ^{131}I among the military weather observers on Rongerik by Harris, Goetz et al. (1987), and Simon et al. (2010b) showed significant differences but can be primarily attributed to the volume of urine assumed to have been excreted in 24 h. The actual average urine volume for American military weather observers was 1,072 mL (Table 4). Harris in 1954** and Goetz et al. (1987) both used 500 mL, while Simon et al. (2010b) used the actual mean value.

Quality of assays of radionuclides other than ^{131}I

Table A1 of the Appendix summarizes some information about the assay methods for other radionuclides as implemented by LASL, Walter Reed, and the NRDL. Little detail on these methods could be found. We have attempted to evaluate the reliability of those measurements from sparse documentation with the following conclusions (adapted from Ibrahim 2007††††):

1. Walter Reed data on ^{137}Cs and the Harris data on ^{131}I are good due to the specific measurements employed. Both data sets are likely to have modest-sized measurement uncertainties;
2. ^{90}Sr data, analyzed only at Walter Reed, are also good due to the specific radiochemical separation used;
3. ^{89}Sr measured by NRDL and LASL is satisfactory within a modest range of uncertainty;
4. ^{140}Ba measured by LASL was not satisfactory due to interference from ^{140}La ingrowth;
5. ^{103}Ru measured by LASL during the initial phase of the work was not satisfactory, but improved somewhat afterwards. Even at best, the associated uncertainty with ^{103}Ru measurements was high; and
6. Plutonium measurements conducted in 1954 were not reliable and were recognized as such in 1954.§

Various counting results from these assays can be found in some of the historical documents previously noted. Tables A2 through A6 present these data, though we do not attempt use or interpret these data since many of those data were either already described or, in some cases, found to be unreliable. This documentation is primarily for historical purposes.

CONCLUSION

For the first time in the refereed literature, the bioassay measurement data on ^{131}I and volumes of urine samples collected are presented for samples collected in 1954 following exposure of Marshallese and American military weather observers to radioactive fallout from the Bravo nuclear test. The data reported here include individual and group-average urine volumes, count rates

†††† Ibrahim SA. Summary of urine bioassay conducted at the Marshall Islands in the 1950's and evaluation of reliability and quality of measurements techniques. Report to the National Cancer Institute. 10 October 2007.

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from ^{131}I gamma spectrometry measurements, and estimates of concentration of various radionuclides, in particular, ^{131}I in pooled urine samples. These data, though long overdue in easily accessible literature, are vitally important to assessing the doses received by Marshallese and Americans from the Bravo nuclear test of 1954.

In addition, herein, we have corrected the erroneous assumptions first presented by James (1964) that the LASL urine samples were partially composed of urine from children. Limited data were acquired on children in other samplings leading to estimates of excreted ^{137}Cs and ^{90}Sr. For completeness, we also present most of the available bioassay counting data from the HASL and NRDL previously available only in laboratory reports, though we do not interpret all of those data as there were many methodological- and instrumentation-related limitations to the data.

Urine volumes, on an individual or population-average basis, were smaller than expected based on bioassay experience in locations with temperate climates, but are, nonetheless, reasonable when considering perspiration and insensible water losses through the skin which typically occur at much higher rates in a tropical climate. Moreover, there was considerable consistency of the distributions of urine volumes over many different sampling dates and by four different institutions, particularly when the limitations imposed by relatively small numbers of samples are considered.

Estimates of intake of radioiodines and iodine-precursor radionuclides for the Marshallese groups do not vary greatly among several assessments conducted over the period of 55 y and all appear within the range of the likely uncertainty of estimation. These radioiodine assay data are a particularly cogent example of the importance of bioassay data following events involving environmental contamination and exposure of the public and clearly indicate that the uses of such data may continue well beyond the immediate events that caused the exposure.

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REFERENCES

Anderson EC, Schuch RB, Perrings JD, Langham WH. The Los Alamos human counter. Nucleonics 14:26–29; 1956.

Beck HL, Bouville A, Moroz BE, Simon SL. Fallout deposition in the Marshall Islands from Bikini and Enewetak nuclear weapons tests. Health Phys 99:124–142; 2010.

Bond VP, Conard RA, Robertson JS, Weden EA. Operation Castle-Addendum Report Project 4.1A: Medical examination of Rongelap people six months after exposure to fallout. Bethesda, MD: Naval Medical Research Institute and U.S. Naval Radiological Defense Laboratory; WT-937; 1955.

Bouville A, Beck HL. The HASL gummed-film network and its use in the reconstruction of doses resulting from nuclear weapons tests. Technol 7:355–379; 2000.

Bouville A, Beck HL, Simon SL. Doses from external irradiation to Marshall Islanders from Bikini and Enewetak nuclear weapons tests. Health Phys 99:143–156; 2010.

Breslin AJ, Cassidy ME. Radioactive debris from Operation Castle islands of the mid-Pacific. New York: United States Atomic Energy Commission, New York Operations Office, Health and Safety Division; NYO-4623; 1955. Available at: http://www.hss.energy.gov/HealthSafety/IHS/marshall/collection/.

Conard RA. A twenty-year review of medical findings in a Marshallese population accidentally exposed to radioactive fallout. Upton, NY: Brookhaven National Laboratory; BNL-50424; 1975.

Conard RA, Cannon B, Huggins CE, Richards JB, Lowery A. Medical survey of Marshallese two years after exposure to radiation. Upton, NY: Brookhaven National Laboratory and Naval Medical Research Institute; BNL 412; 1956.

Cronkite EP, Bond VP, Dunham CL. Some effects of ionizing radiation on human beings: a report on the Marshallese and Americans accidentally exposed to radiation from fallout and a discussion of radiation injury in the human being. Washington, DC: U.S. Atomic Energy Commission; TID-538; 1956. Available at: http://www.hss.energy.gov/HealthSafety/IHS/marshall/collection/.

Defense Nuclear Agency. Compilations of local fallout data from test detonations 1945–1962 extracted from DASA 1251, Volume II—Oceanic U.S. tests. General Electric Company-TEMPO; 1979.

Dunning Jr DE, Bernard SR, Walsh PJ, Killough GG, Pleasant JC. Estimates of internal dose equivalent to 22 target organs for radionuclides occurring in routine releases from nuclear fuel-cycle facilities, Vol. II. Oak Ridge, TN: Oak Ridge National Laboratory; NUREG/CR-0150, Vol. 2, ORNL/NUREG/TM-190/V2; 1979.

Dunning Jr DE, Killough GG, Bernard SR, Pleasant JC, Walsh PJ. Estimates of internal dose equivalent to 22 target organs for radionuclides occurring in routine releases from nuclear fuel-cycle facilities, Vol. III. Oak Ridge, TN: Oak Ridge National Laboratory; NUREG/CR-0150, Vol. 3, ORNL/NUREG/TM-190/V3; 1981.

Elebute EA. Determinants of evaporated fluid loss in a tropical environment. Bulletin de la Societé Internationale de Chirurge (Surgery) 2:2–6–213; 1973.

Goetz J, Klemm J, Phillips J, Thomas C. Analysis of radiation exposure—service personnel on Rongerik Atoll, Operation Castle–Shot Bravo. McLean, VA: Science Applications International Corp.; SAIC-86/1608; 1987.

Hamilton JG. The metabolic properties of the fission products and actinide elements. Reviews Modern Phys 20:718–728; 1948.

Hamilton JG, Soley MH. Studies in iodine metabolism by the use of a new radioactive isotope of iodine. Am J Physiol 127:557–572; 1939.

Hamilton JG, Soley MH. A comparison of the metabolism of iodine and of element 85 (Eka-Iodine). Proceedings of the National Academy of Sciences (PNAS) 26:483–489; 1940.

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International Commission on Radiological Protection. Report of Committee IX on Limits for Intakes of Radionuclides by Workers. Oxford: Pergamon Press; ICRP Publication 30; 1979.

International Commission on Radiological Protection. Age-dependent doses to members of the public from intake of radionuclides: Part 1. Oxford: Pergamon Press; ICRP Publication 56; 1989.

International Commission on Radiological Protection. Basic anatomical and physiological data for use in radiological protection: reference values. New York: Elsevier; ICRP Publication 89; 2002.

James RA. Estimate of radiation dose to thyroids of the Rongelap children following the Bravo event. Livermore, CA: Lawrence Radiation Laboratory; UCRL-12273; 1964. Available at: www.hss.energy.gov/healthsafety/ihs/marshall/collection/.

Johnson JR. Radioiodine dosimetry. J Radioanal Chem 65:223–238; 1981.

Langham WH. Annual report, Biomedical Research Group, Health Division, 1954. Los Alamos, NM: Los Alamos Scientific Laboratory; LA-1889; 1954.

Latham MC. Human nutrition in the developing world. FAO Food and Nutrition Series, No. 29 [online]. Rome: United Nations; 1997. Available at: http://193.43.36.103/docrep/W0073e/w0073e00.htm.

Lessard E, Miltenberger R, Conard R, Musolino S, Naidu J, Moorthy A, Schopfer C. Thyroid absorbed dose for people at Rongelap, Utrik and Sifo on March 1, 1954. Upton, NY: Brookhaven National Laboratory, Safety and Environmental Protection Division; BNL 51882; 1985.

Martin EJ, Rowland RH. CASTLE Series 1954. Washington, DC: Defense Nuclear Agency; DNA 603F; 1982.

Mao IF, Chen ML, Ko YC. Electrolyte loss in sweat and iodine deficiency in a hot environment. Archives Environmental Health 56:271–277; 2001.

Mao IF, Ko YC, Chen ML. The stability of iodine in human sweat. Japanese J Physiol 40:693–700; 1990.

MedlinePlus Health Information, National Library of Medicine. Urine 24-hour volume [online]. Bethesda, MD: National Library of Medicine; 2002. Available at: http://www.nlm.nih.gov/medlineplus/ency/article/003425.htm. Accessed 2008.

Musolino SV, Greenhouse NA, Hull AP. An estimate by two methods of thyroid absorbed doses to Bravo fallout in several northern Marshall Islands. Health Phys 73:651–662; 1997.

Riggs DS. Quantitative aspects of iodine metabolism in man. Pharmacol Rev 18:516–521; 1952.

Sharp R, Chapman WH. Report to the scientific director: Exposure of Marshall Islanders and American military personnel to fallout, Operation Castle-–Project 4.1 Addendum. Bethesda, MD: Naval Medical Research Institute; WT-938; 1957. [Extract version prepared for Defense Nuclear Agency, 1980.] Available at: http://www.hss.energy.gov/healthsafety/ihs/marshall/collection/.

Simon SL, Bouville A, Land CE, Beck HL. Radiation doses and cancer risks in the Marshall Islands associated with exposure to radioactive fallout from Bikini and Enewetak nuclear weapons tests: summary. Health Phys 99:105–123; 2010a.

Simon SL, Bouville A, Melo D, Beck HL, Weinstock RM. Acute and chronic intakes of fallout radionuclides by Marshallese from nuclear weapons testing at Bikini and Enewetak and related internal radiation doses. Health Phys 99:157–200; 2010b.

Takahashi T, Trott KR, Fujimori K, Nakashima N, Ohtomo H, Schoemaker MJ, Simon SL. Thyroid disease in the Marshall Islands, findings from 10 years of study. Sendai, Japan: Tohoku University Press; 2001.

Woodward KT, Schrodt AG, Anderson JE, Claypool HA, Hartgering JB. The determination of internally deposited radioactive isotopes in the Marshallese people by excretion analysis. Washington, DC: Walter Reed Institute of Research Division of Nuclear Medicine; DASA-1180; 1959. Available at: http://www.hss.energy.gov/healthsafety/ihs/marshall/collection/.

APPENDIX

Several types of measurements and assays for different radionuclides were performed in 1954 though some of the radiometric analyses were rudimentary by today's standards and in some cases had chemical interferences or the measurement techniques were non-specific or had insufficient detection sensitivity (see Tables A1–A6).

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Table A1. Summary of analytical methods used for radionuclide assays of urine from Marshallese (adapted from [‡‡‡‡]).

Source of data	Radionuclide	Year of collection	Chemical separation method (where applicable)	Counting method	Comments on measurement methods	Data quality
LASL (**,††,***)	^{131}I	1954	None	Gamma counting with SAC counter (see text).		Satisfactory, with low to moderate uncertainty
LASL (**,††,***)	^{131}I	1954	Urine aliquots evaporated to dryness after adjusting pH to 8–9	Total beta activity counting and decay rate measurements (8-d half-life) to confirm ^{131}I. Counting instruments not specified.	Correction for self absorption/or use of calibration standard were not mentioned. Other beta emitters will interfere.	Satisfactory, with moderate uncertainty
			Extraction/isolation method (not specified). Method used for limited data sets on 100 mL urine aliquots	Total beta activity counting and decay rate measurement to confirm ^{131}I.	Yield for extraction was thought to be about 60% of all isotopes, ^{131}I had the most significant activity in urine.	Only satisfactory within an order of magnitude due to uncertain chemical yield
	^{89}Sr		Dry or wet ashed urine of 100 mL urine aliquots (no chemical separation	Total nonvolatile beta activity and decay rate measurements (55 d) to confirm ^{89}Sr. It was predominant isotope after the decay of ^{131}I.	^{90}Sr/^{89}Sr ratio of 0.1 is assumed. Other beta emitters (^{137}Cs) will interfere.	Satisfactory, with moderate uncertainty
			Extraction/isolation method (not specified)	Total beta activity and decay rate measurement to confirm ^{89}Sr.	Chemical yield uncertain due to high variability.	High degree of uncertainty
	^{45}Ca		Extraction/isolation method (not specified)	Total beta activity counting, instruments not specified.	Data corrected for self absorption and decay. Other beta emitter can interfere.	High degree of uncertainty
	Ru (likely ^{103}Ru)		Extraction/isolation method (not specified)	Total beta activity counting, instruments not specified.	Uncorrected for self absorption. Levels of ^{103}Ru in urine are expected to be low due to poor absorption in humans (likely introducing large error in measurements).	Unreliable
	Plutonium	1954	Alpha counting	None known.	Lack of required sensitivity and non-specific to plutonium.	Unreliable
Walter Reed (Woodward et al. 1959)	^{137}Cs	1954–1957	Precipitation on nickel ferrocyanide from urine made strongly alkaline	Na I well counter with 20 channel gamma spectrometer to count precipitate.	Spectral analysis calibrated with standard source Reported measurement error of ± 5%	Satisfactory, reliable set of data
		1958	No chemical separation	Urine counted directly on 8×4 cm Na I crystal.	Large sample volume (2.5 L) Gamma-ray spectral analysis	Satisfactory, reliable set of data
	^{90}Sr	1954–1958	Precipitation as carbonate, ^{90}Y was separated and identified by its half-life	Precipitation counted using thin-window gas flow counter.	Specific separation for ^{90}Sr Data corrected for decay to time of collection Low to moderate degree of uncertainty	Satisfactory, reliable set of data

(*Continued*)

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Table A1. (*Continued*)

Source of data	Radionuclide	Year of collection	Chemical separation method (where applicable)	Counting method	Comments on measurement methods	Data quality
U.S. Naval Radiological Defense Laboratory (Cronkite et al. 1956)	^{89}Sr		Oxalate precipitation using a small urine aliquot Alternative method: Carbonate precipitation of the entire 24-h urine sample, if collected later than 2.5 mo past detonation	Precipitate counted using thin-window G-M counter Standard source was used for calibration of counter Self-absorption correction was made using Sr standard	The method eliminates the normal ^{40}K The precipitation method is non-specific and ^{90}Sr could interfere if present	Satisfactory if ^{89}Sr activity is much greater than ^{90}Sr (likely to have been the case)
	^{40}Ba		Mentioned, but no information was provided	—	—	Not evaluated
	Gross beta		Mentioned, but no information provided	—	—	Not evaluated

Table A2. Summary of USNRDL and HASL urine analysis (volumes in mL and gross beta activity in d m^{-1}) at four times post-detonation (Cronkite et al. 1956).

	Approximate time post-detonation and bioassay data											
	1 1/2 mo			2 1/2 mo			3 mo			6 mo		
Population and age group	Number of subjects	Avg. vol. (mL) per 24 h	Avg. dis min^{-1}	Number of subjects	Avg. vol. (mL) per 24 h	Avg. dis min^{-1}	Number of subjects	Avg. vol. (mL) per 24 h	Avg. dis min^{-1}	Number of subjects	Avg. vol. (mL) per 24 h	Avg. dis min^{-1}
Rongelap												
<5 y	7	165	404	—	—	—	—		—	8	360	12
5–16 y	11	439	758	—	—	—	—		—	12	510	5
>16 y	13	581	1,208	10	824	705	10	379	339	33	625	0
Ailinginae												
<5 y	1	150	217	—	—	—	—	—	—	—	—	—
5–16 y	2	275	126	—	—	—	—	—	—	3	400	0
>16 y	10	722	553	—	—	—	—	—	—	12	655	0
American military weather observers (adult)	25	1,158	309	—	—	—	—	—	—	—	—	—

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Table A3. Gross beta activity in urine of individuals from Rongelap, sampled at 46 d post-detonation in USNRDL analyses (Cronkite et al. 1956).

Age and case no.	Urine vol. (mL per 24 h)	Beta activity (dis min^{-1})
Age <5 y		
2	120	712
3	150	894
5	155	313
23	40	223
33	260	0
54	80	385
69	455	301
Mean =	**165**	**404**
Age 6–15 y		
20	265	1,900
24	550	0
26	650	1,032
35	255	0
36	190	236
39	280	1,100
47	650	1,705
67	450	674
72	110	507
75	440	0
76	980	1,180
Mean =	**439**	**758**
Age >16 y		
4	455	634
7	810	1,700
9	355	201
10	980	549
11	450	1,583
13	340	1,677
14	780	2,460
18	455	1,670
22	47	77
20	960	438
34	750	570
37	480	792
40	550	1,450
46	330	495
49	425	0
52	780	0
55	320	1,080
56	700	3,220
57	550	1,095
58	750	2,170
60	810	580
62	980	1,985
63	635	2,260
66	855	1,715
68	300	2,010
71	290	1,450
73	230	0
78	965	52
79	465	2,038
80	540	1,353
82	670	2,140
Mean =	**581**	**1,208**

Table A4. Gross beta activity in urine of individuals from Rongelap people on Ailinginae and American military on Rongerik sampled at 46 and 44 d post-detonation, respectively, in USNRDL analyses (Cronkite et al. 1956).

Ailinginae (H+46 d)

Age group and case no.	Urine vol. (mL per 24 h)	Beta activity (dis min^{-1})
Age <5 y		
6	—	—
8	—	—
44	150	217
Mean =	150	217
Age 6–15 y		
48	180	164
53	—	—
81	370	88
Mean =	**275**	**126**
Age >16 y		
1	900	765
16	880	827
28	680	1202
29	780	0
31	260	846
41	920	62
43	610	754
45	850	680
51	410	400
70	440	0
Mean =	**722**	**553**

American military weather observers on Rongerik (H+44 d)

Age group and case no.	Urine vol. (mL per 24 h)	Beta activity (dis min^{-1})
Adults		
1	1,970	0
2	650	0
3	1,224	820
4	440	78
5	735	0
6	900	248
7	1,340	0
8	1,410	1,260
9	—	—
10	—	—
11	1,580	385
12	1,460	0
13	1,810	965
14	720	438
15	1,380	830
16	1,930	0
117	945	—
18	1,520	0
19	1,300	466
20	1,070	0
21	550	353
22	—	—
23	1,180	0
24	1,160	750
25	1,380	187
26	510	323
27	565	—
28	1,220	0
Mean =	**1,158**	**309**

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Table A5. Radionuclide analysis of urine from Rongelap people 45 d post-detonation (H+45) in NRDL analyses (Cronkite et al. 1956).

Sample no.	Beta activity (dis min^{-1})			
	Gross beta	^{89}Sr	^{140}Ba	Rare earth activity
1	1,370	490	120	197
2	1,260	510	130	244
3	1,020	480	120	324
4	1,210	626	150	284
5	1,460	328	110	474
6	1,200	727	170	353
Mean =	1,253	562	134	312
Percent of total beta activity	100	42	10.7	25.5

Table A6. Urine bioassay counting (d m^{-1} L^{-1}) data for plutonium from Rongelap adults.****

Subject No.	Collection dates, disintegration rates, and urine volumes[a]					
	3/24/1954			3/25/1954		
	dis min^{-1} L^{-1}	1 σ	Urine volume (mL)	dis min^{-1} L^{-1}	1 σ	Urine volume (mL)
1	1	2	965	0	2.5	670
4	0	2.5	—	—	na	—
7	0	2.5	—	0	2.5	—
9	1	2	—	0	2.5	—
10	0	2.5	—	1	2	—
11	0	2.5	—	0	2.5	—
12	0	2.5	—	1	2	—
14	4	2.5	—	0	2.5	—
16	0	2.5	955	0	2.5	800
18	—	na	—	5	4.5	—
19	0	2.5	—	—	na	—
22	7	2.5	—	0	2.5	—
25	0	2.5	—	—	na	—
26	1	2	—	0	2.5	—
28	0	2.5	810	0	2.5	785
29	1	2	565	0	2.5	845
30	—	na	—	0	2.5	—
32	—	na	—	3	2.5	—
33	—	na	70	2	4	210
34	0	2.5	685	0	2.5	895
35	0	2.5	250	—	na	155
37	0	2.5	550	0	2.5	385
39	—	na	175	—	—	—
40	0	2.5	700	0	2.5	650
41	1	2	950	12	3	985
43	0	2.5	720	0	2.5	775
44	—	na	—	—	na	90
45	0	2.5	680	0	2.5	935
46	—	na	—	0	2.5	205
47	0	2.5	975	0	2.5	540
49	17	3	980	—	na	130
50	1	2	765	100	8	770
51	8	2.5	320	0	2.5	230
52	—	na		0	2.5	285
53	3.5	2.5	530	—	na	105
55	0	2.5	400	0	2.5	720
56	—	na	—	0	2.5	595
57	—	na	360	0	2.5	830
58	3	2.5	665	0	2.5	640
59	0	2.5	800	0	2.5	430
60	4.5	2.5	900	—	na	150
61	0	2.5	250	0	2.5	480
62	0	2.5	560	0	2.5	755
63	0	2.5	840	2.5	2	420
64	2	2	685	1	2	550
66	0	2.5	785	7	2.5	950
67	0	2.5	500	—	na	—
68	3	2.5	940	1	2	590
69	1.5	2.5	160	—	na	—
70	—	na	—	7.5	2.5	750
71	0	2.5	225	0	2.5	210
72	—	na	—	0	2.5	265
73	1	2	450	—	—	—
74	—	na	380	0	2.5	630
75	0	2.5	300	—	na	—
76	0	2.5	980	0	2.5	470
77	—	na	—	2.5	2	965
78	0	2.5	640	0	2.5	340
79	0	2.5	915	0	2.5	480
80	2.5	2	690	0	2.5	560
81	6	2.5	815	0	2.5	450
82	—	na	—	1	2	205

[a] Dash (—) indicates no data; "na" means not applicable.

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