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INTRODUCTION

The United States conducted a nuclear testing program at Bikini and Enewetak Atolls in the Marshall Islands from 1946 through 1958. Several atolls, including Bikini, Enewetak, Rongelap, and Utirik, were contaminated as a result of the nuclear detonations. Rongelap and Utirik Atolls were inhabited, and the people on these atolls were exposed to fallout on March 1, 1954 from the BRAVO test.

Since 1974 we have conducted an extensive research and monitoring program to determine the radiological conditions at the atolls, identify the critical radionuclides and pathways, estimate the radiological dose to current or resettling populations, and develop remedial measures to reduce the dose to atoll populations.

EXPOSURE PATHWAYS AND RADIONUCLIDES

People residing on the atolls, are exposed to radiation from both external and internal sources that results from background radiation and nuclear test-related radiation.

The external background radiation in the Northern Marshall Island Atolls is 8.5×10^{-10} C kg^{-1} (3.3 μR h^{-1}) or 0.22 mSv y^{-1} (Gudiksen et al. 1976) due to cosmic radiation; the external background dose due to terrestrial radiation is very low in the Marshall Islands because of the composition of the soil. The internal effective dose is about 1.4 mSv y^{-1} for natural occurring radionuclides such as ^{40}K, ^{210}Po, and ^{210}Pb that result from consumption of local and imported foods. The background dose is not included in the doses presented in this paper unless specifically stated.

The radionuclides present today at the atolls that contribute in any significant way to the dose to people are ^{137}Cs, ^{90}Sr, $^{239+240}Pu$ and ^{241}Am.

COMPOSITION OF ATOLL SOILS

The soils at atolls differ from most continental soil in that the mineral matrix consists of sand and more coarse particles of calcium carbonate, containing small amounts of substituted magnesium and strontium. The pH of soil-water slurries ranges from about 7.8 to 8.9. Organic matter content of the surface layers ranges from trace amounts to 14% or more, and is the sole source of cation exchange capacity. The organic content diminishes rapidly with depth through a narrow transition zone.

Total and exchangeable K in atoll soils is very low; the average for several atolls reported by Fosberg and Carroll (1965) is only 300 mg kg^{-1}. Exchangeable or extractable K ranges from 40 to 80 mg kg^{-1} in the higher organic containing surface layers and diminishes rapidly with depth (Robison and Stone 1992).

RADIONUCLIDE TRANSPORT AND DOSE ESTIMATES

As a result of the unique soil system, the transfer of ^{137}Cs and ^{90}Sr from soil to plants is reversed from that observed in temperate, silica-based soils (IAEA, Robison and Conrado). Consequently, the relative contribution of radionuclides and pathways that contribute to the dose differ from those estimates from other soil systems for similar deposition densities.

Using Bikini Atoll as an example, the estimated maximum annual effective dose is 4.0 mSv. The estimated 30-, 50-, and 70-y integral effective dose from the major exposure pathways and radionuclides is listed in Table I. About 90% of the dose is via the terrestrial food chain, the major portion of which is delivered by ^{137}Cs. External gamma exposure is the next most significant pathway, which again is attributable to ^{137}Cs. The other pathways and radionuclides contribute less than 2% of the estimated dose over 70 y.

Consequently, our efforts to develop remedial measures to reduce the dose to island residents have been directed at the ^{137}Cs transport into the terrestrial foods that accounts for about 90% of the total dose.

Table I. The 30-, 50- and 70-y integral effective dose for Bikini Island residents for current island conditions when imported foods are available and when only local foods are consumed.

	Integral effective dose, mSv		
	30 y	50 y	70 y
External	9.1	13	15
Internal			
Ingestion			
^{137}Cs	81	110	130
^{90}Sr	0.85	1.2	1.5
$^{239+240}Pu$	0.011	0.028	0.051
^{241}Am	0.018	0.043	0.075
Inhalation			
$^{239+240}Pu$	0.069	0.16	0.23
^{241}Am	0.050	0.11	0.15
Total[a]	91	130	150

[a] The total dose may vary in the second decimal place due to rounding.

REMEDIAL MEASURES

Several general methods have been evaluated in efforts to find an effective method for reducing the dose to residents at the atolls. They are: Excavation—starting over; KCl—blocking uptake by plants while maintaining useful productivity; Irrigation—(leaching) – removal; Cropping—sustained removal; Clay (mica) and Zeolites (clinoptilolite)—immobilization.

Removal of Contaminated Surface Soil

The environmental consequences of soil excavation are great and long lasting. First, all vegetation must be removed. Scraping off the surface 40 cm indeed removes the radionuclides, but with them the organic layer developed over centuries. Not only does the organic layers provide nutrient exchange capacity and greatly increase rainfall retention, but it also stabilizes the surface against wind and water erosion, and renders the soil friable for root development. Restoring and maintaining productivity, however, requires a decades-long commitment of effort and expertise that is far from assured. Some of the organic-rich soils contain upwards of 5,000 kg ha^{-1} N and even 10,000 kg ha^{-1} P. The cost of these elements in a fertilizer bag would be several thousand dollars per hectare.

Because of the severe environmental impacts of the excavation option, we have examined other remedial possibilities that might reduce ^{137}Cs in the terrestrial food chain.

Leaching ^{137}Cs from the Soil

A large-scale field irrigation experiment was designed to determine whether a significant fraction of the ^{137}Cs inventory in the soil could be removed by leaching. Atolls simply do not have the large stores of fresh water needed for a single, let alone multiple leachings. The supply of seawater, however, is boundless, and the contained cations might be expected to dislodge any cesium held by simple exchange forces.

We sprayed approximately 80" of seawater on three occasions at two month intervals on a one hectare area while monitoring the downward movement of both seawater and ^{137}Cs. The initial pulse of ^{137}Cs into the ground water diminished in magnitude with each application, and represented only the small fraction that was soluble or readily exchangeable. A final total application of 20 meters (depth) of seawater removed only 3–5% of the total ^{137}Cs inventory. Laboratory experiments with large soil columns gave similar results.

The large effort required for such a small reduction of ^{137}Cs inventory obviously eliminates the treatment as a remedial measure.

Immobilization of ^{137}Cs in Soil

Mica: We applied mica at the rate of 6,000 kg ha^{-1} uniformly over the soil surface of a productive coconut grove. The aim was to immobilize any available Cs already at the surface and thus gradually interrupt cycling of ^{137}Cs from plant to soil to plant again. Sampling of coconuts indicates a reduction in ^{137}Cs after 8 years or so. The reduction in shallow rooted herbaceous vegetation without the large internal inventory of ^{137}Cs is more impressive. For the grass Eustachys *Petraea*, the ^{137}Cs concentration was 1.3 Bq g^{-1} in the mica treated plot and 9.1 Bq g^{-1} in the control plot.

Clinoptilolite: The zeolite study entailed addition of clinoptilolite at the rate of 20, 40, and 80 mt ha^{-1} to soil in small plots. Since the clinoptilolite itself contains appreciable K with some availability to plants, the study included both a conventional control and a control with added K.

Using the control plus a moderate application of K as a standard, the conventional control (without added K) yielded only 61% as much biomass, but it contained 170% more ^{137}Cs. Biomass yields at the lowest clinoptilolite were about equal, but contained only 71% as much ^{137}Cs, or only 41% as much as the conventional control. The highest rate of clinoptilolite produced about twice the biomass as the control, but contained only 12% as much ^{137}Cs (or 6% of the control concentration). However, the studies involved large quantities of materials plus either intensive mixing with bare soil as rather long reaction times. Moreover, immobilization counteracts natural leaching, and largely eliminates removal indicated by an environmental half-life.

Removal by Continuous Cropping

A frequently proposed concept for reducing ^{137}Cs inventory in soil is to repeatedly grow and dispose of successive crops of some plants that have a high uptake of the element. Plant growth factors on atolls demonstrates the limited application of the concept.

The source of fresh water is rainfall that occurs mostly from June through November, which limits natural vegetation growth to that six month interval. The average annual mass of vegetation that can be produced is about 1 kg m^{-2} or perhaps even less with continued cropping. The maximum uptake of ^{137}Cs in vegetation expressed as a concentration ratio (Bq g^{-1} in plants (wet weight)/Bq g^{-1} in soil (dry weight) is about 3. A square meter of soil, 40 cm deep, weights about 440 kg. Thus, the loss of ^{137}Cs is equal to 0.0067 y^{-1}.

The ^{137}Cs concentration in litter fall (fronds and coconuts) has been quantified to determine the annual loss of ^{137}Cs in the coconut grove if all the litter were collected and disposed of annually. This loss rate is $\lambda = 0.0063\ y^{-1}$.

Over 90 y, radioactive decay ($\lambda = 0.023\ y^{-1}$) reduces the inventory of ^{137}Cs to 12% of its initial value. The effective decay constant is $\lambda_{eff} = 0.023 + 0.0068 \cong 0.030\ y^{-1}$ which over 90 y reduces the ^{137}Cs inventory to 6.7% of its initial value. Thus, cropping the entire island for 90 y and disposing of the vegetation in some manner—an enormous task—would achieve only an additional 6% loss of ^{137}Cs. Practically, this would never happen. Moreover, our field experiments demonstrate that repeated cropping depletes the pool of available potassium so that growth greatly diminishes. This is a self-defeating cycle. Adding K to increase productivity reduces the ^{137}Cs concentration in plants (see previous and next sections), and consequently, the total removal of ^{137}Cs by cropping becomes miniscule.

Potassium (K) Treatment

We have conducted many large-scale field experiments with coconut and other plants evaluating the effect of K additions, their times of application, and the longevity of the effect. Figure 1 illustrates some of the results obtained when 1260 and 2520 kg ha^{-1} of K were added to a portion of the coconut grove. In a separate experiment the addition of 660 kg ha^{-1} reduced the ^{137}Cs in coconuts to about 20% of pretreatment levels (Fig. 2). A second application of 660 kg ha^{-1} shows a further reduction that is consistent with the 1260 kg ha^{-1} application. Near maximum effectiveness is achieved with the addition of the 1260 and 2520 kg ha^{-1}, which reduces the ^{137}Cs in coconuts to bout 5% of pretreatment levels. Such reduction endures because K content increases and ^{137}Cs content decreases in the large stem of the fruit trees.

The large and prompt reduction of ^{137}Cs in coconuts and all other food crops tested, the ease of application, the duration of the effect, and the relatively low cost makes K addition the preferred treatment option although this was unknown 12 years ago. Added K also increases growth and productivity of plants on the island.

REDUCTION IN DOSE FROM A COMBINED OPTION

As an alternative to removing the top 40 cm of soil from the entire island, we have recommended a “combined option” which consists of treatment of the coconut grove and agricultural areas with KCl, and the limited removal of soil only in the housing and village areas. The KCl treatment reduces the dose from ^{137}Cs in locally grown foods, that contribute most of the dose. The limited soil removal, however, decreases the external exposure from ^{137}Cs where people spend most of their time. It also helps reduce the internal exposure from $^{239+240}$Pu and ^{241}Am.

The results of the combined option on the estimated doses at the atolls is shown in Figure 3 for Bikini, Enewetak, and Rongelap Atolls. The reduction is nearly a factor of ten in the case of Bikini Island and Enjebi Island at Enewetak, and about a factor of 5 for Rongelap Island.

The population average annual effective total dose at the atolls, which consists of the background dose plus the weapons-related dose, is shown in Figure 4, relative to the annual population average effective background dose in the U.S. In all cases, the annual total dose at the atolls using the combined option is less than the annual average background dose around the rest of the world.

Work performed under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

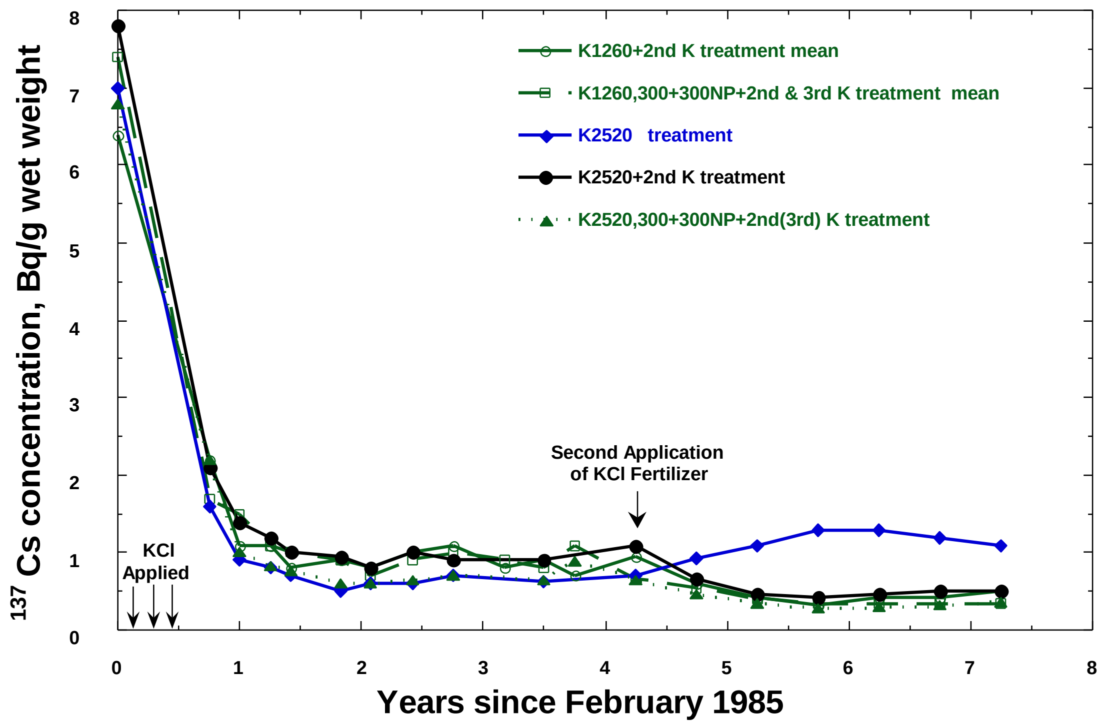

Figure 1. The reduction in the ^{137}Cs concentration in drinking coconut meat as a result of two applications of potassium (as KCl) at 1260 and 2250 kg ha^{-1}.

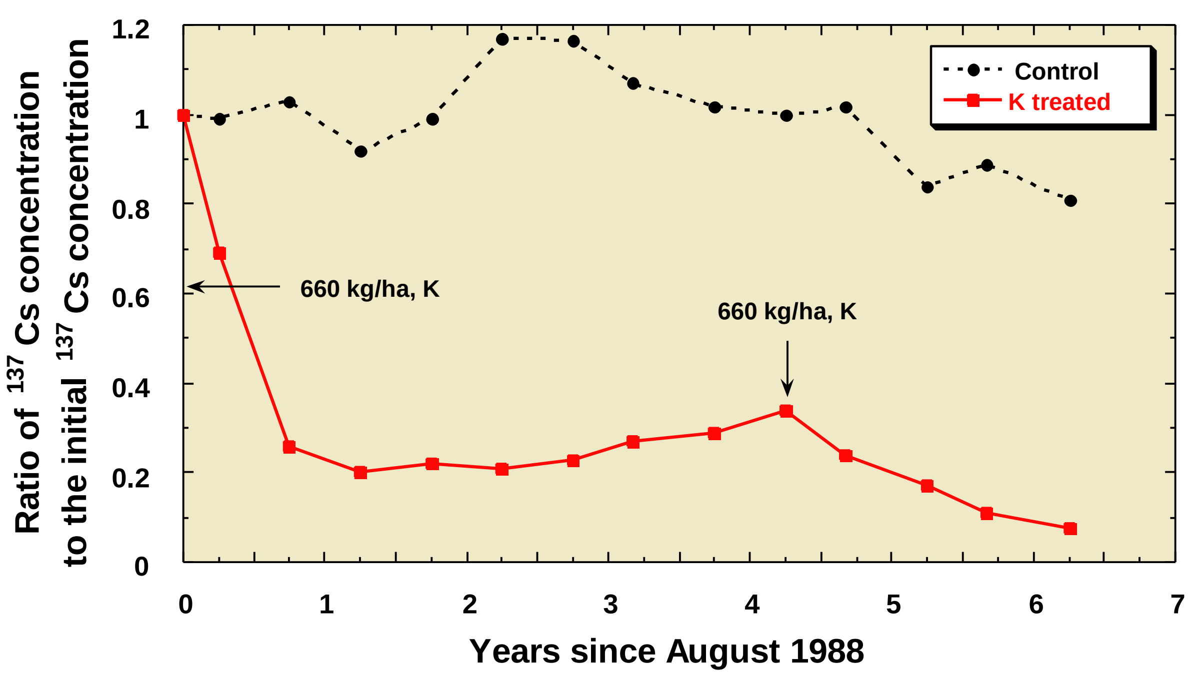

Figure 2. The effect of two applications of lower amounts of potassium (as KCl) on the ^{137}Cs concentration in drinking coconut meat.

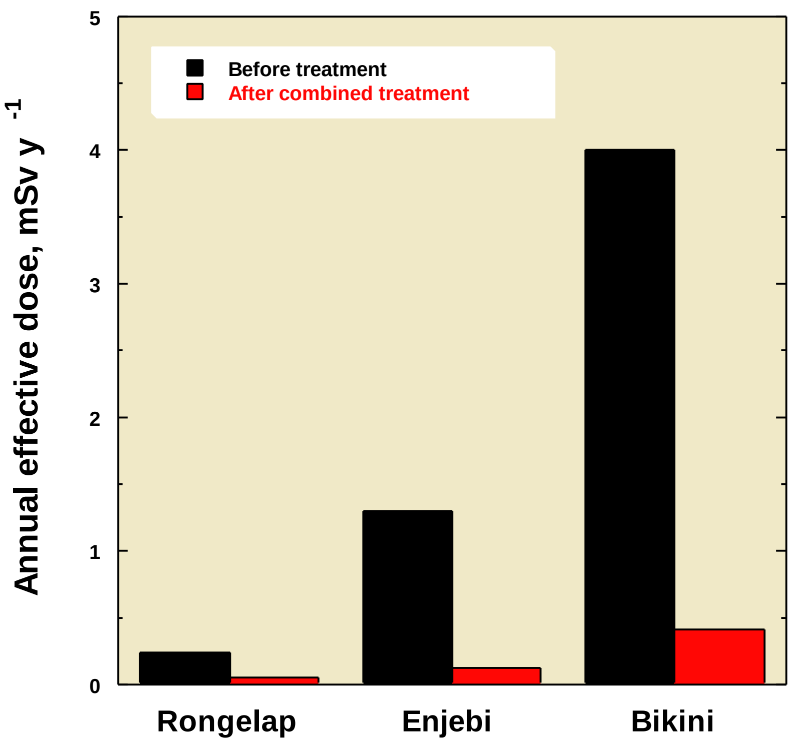

Figure 3. The reduction in the annual effective dose using the combined option.

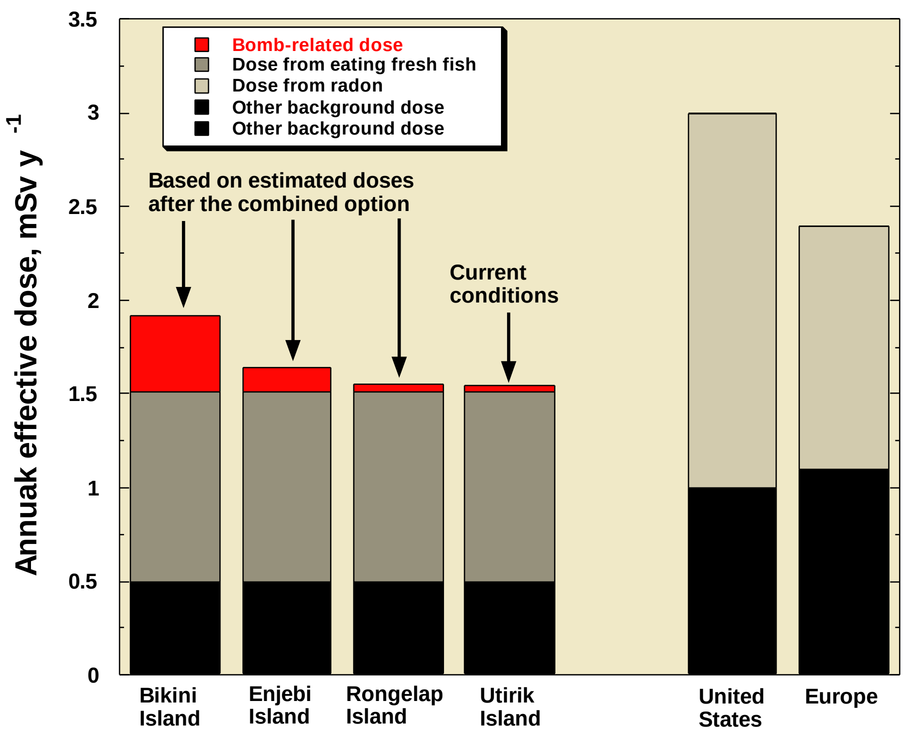

Figure 4. The total annual effective dose (background plus weapons-related) at the atolls compared to the United States and Europe annual background effective dose.