

Reports

Radioactive Wastes: A Comparison of U.S. Military and Civilian Inventories

Abstract. *Contrary to widespread belief, the accumulated inventory of fission products generated by the still small U.S. civilian nuclear power industry may already be comparable to that generated in the past by U.S. military nuclear programs. Although the volumes of the military wastes are very large, they are on the average almost 100 times more dilute than projected commercial high-level wastes.*

There is a widespread belief within the U.S. nuclear community that the U.S. radioactive waste problem will be dominated until approximately the year 2000 by the wastes that have already been generated in military-related activities. Recently this belief and the policy implications that have often been drawn from it have been summarized as follows (1, p. 52):

Nuclear engineers have generally regarded waste management as a less urgent technical issue than many other practical engineering problems [because] they point out that accumulated commercial wastes will not become comparable in amount to already existing wastes from weapons programs until near the end of the present century and that in the meantime the technologies available for commercial waste management are already far superior to those used in the weapons program. This gives us plenty of lead time to solve the problem, they say.

It is difficult, however, to reconcile such statements with the fact that the projected size of the U.S. commercial nuclear power program, measured in terms of integrated thermal power released, is much larger than "reasonable estimates" of the scale of the U.S. military plutonium production and nuclear submarine propulsion programs. Recently the Energy Research and Development Administration (ERDA) has made information available on the U.S. military radioactive waste inventory which allows us to clarify this situation. It appears that the military radioactive waste inventory is large in comparison

with the current and projected high-level commercial radioactive waste inventories only when measured by volume.

When account is taken of the fact that existing military high-level radioactive wastes are almost 100 times more dilute than projected high-level radioactive wastes from the reprocessing of fuel used in nuclear power plants, one finds that the present activities of the fission products that have been generated by the still relatively small U.S. commercial nuclear energy program may already be approximately equal to those that have been generated in the military programs.

Before we discuss the magnitudes of the military and commercial radioactive inventories, we must make clear the basis on which the comparison will be made. The potential hazards of the radioisotopes in different types of high-level radioactive wastes are conventionally compared in terms of the total amount of water it would take to dilute each of them to the concentration limits specified by the Nuclear Regulatory Commission in the *Code of Federal Regulations* (2). Between a few years and a few hundred years this hazard potential is dominated by ^{90}Sr (half-life, 28 years) (3). We shall therefore take this isotope as our yardstick for comparison between military and high-level wastes in the discussion that follows.

The isotope ^{90}Sr is produced in approximately 5.7 percent of all fissions of ^{235}U (4). The fission yield of ^{90}Sr in highly irradiated commercial reactor fuel is somewhat less, about 4 percent, because a significant fraction of the fissions are of plutonium isotopes created by neutron capture in ^{238}U (5). For our purposes it will be adequate to assume that approximately 5 percent of all fissions yield an

atom of ^{90}Sr . The number of ^{90}Sr atoms produced therefore is roughly proportional to the number of fissions, or correspondingly to the total amount of fission energy released in a particular activity. This energy is conventionally measured in units of megawatt-days of thermal energy (1 Mw-day is approximately equivalent to the fission of 1 g of ^{235}U or other fissionable isotopes).

Since ^{90}Sr is not "burned up" at a significant rate in nuclear reactor fuel (the thermal neutron absorption cross section is almost one thousand times smaller than that of ^{235}U), the quantity of ^{90}Sr produced in a particular program is roughly proportional to the total number of megawatt-days of fission energy released in that program—approximately 3 curies of ^{90}Sr per megawatt-day. The volume of the associated high-level wastes is not simply proportional to the amount of fission power released, however. Instead, it tends to reflect the total amount of uranium in the fuel, because the volume of high-level wastes is roughly proportional to the volume of HNO_3 required to dissolve the uranium fuel in the first step of the reprocessing operation. The concentration of fission products in the high-level wastes thus tends to reflect the concentration of fission products in the spent uranium fuel. Indeed, the number of gallons of high-level acid waste associated with the processing of 1 ton of fuel from the military plutonium-producing "N-reactor" at Hanford is of the same order [250 gallons per ton (1040 liters per metric ton) of uranium in the fuel] [(6), vol. 2, figure II.1-C-3] as that which is projected from the reprocessing of reactor fuel used in commercial power production (7). This is true despite the fact that the N-reactor fuel contains only about 7 percent as many fission products per ton as the commercial fuel (8).

At any one time the high-level radioactive waste inventory produced by both the civilian and military programs will be found in a number of forms. At present, most of the radioactive wastes from the civilian nuclear power program are still in the spent reactor fuel where they were produced, and the ultimate form of their disposition is still undecided. In contrast, most of the radioactive wastes associated with the military programs have been separated from the original reactor fuel and are stored predominantly in chemically neutralized form. At present, ERDA is engaged in a vigorous solidification program with the objectives of reducing the volume and mobility of this military high-level waste. Because the waste will be in various forms at various

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Table 1. Estimated volumes and ^{90}Sr inventories of military high-level radioactive wastes.

Site	1976		Early 1980's	
	Volume (10^6 gallons)	^{90}Sr inventory (10^6 curies)	Volume (10^6 gallons)	^{90}Sr inventory (10^6 curies)
Hanford	51	114	48	145
SRP	21	150	20	163
INEL	2.3	6	1	8
Total (rounded)	75	270	70	315

times, we will concentrate below on the curies of ^{90}Sr contained in these wastes, a quantity that is affected only by radioactive decay.

Table 1 shows our estimates of current volumes and ^{90}Sr inventories at the principal high-level radioactive waste repositories associated with ERDA's military nuclear activities and projections of these quantities for the early 1980's. These wastes come primarily from plutonium production and the reprocessing of nuclear submarine fuel. The information on which these estimates are based comes primarily from recent ERDA publications on the radioactive waste management operations at ERDA's Hanford site (9), the Savannah River plant (SRP) (10), and Idaho National Engineering Laboratory (INEL) (11). These reports give ^{90}Sr inventories and production rates at various times in the past and projections into the future. In each case we have corrected for the effects of the radioactive decay of the ^{90}Sr in translating quantities from one year to another. According to our estimates, these wastes currently contain an average of only about 3.5 curies of ^{90}Sr per gallon, which is almost 100 times more dilute than the concentration of about 300 curies of ^{90}Sr per gallon projected for the fresh liquid wastes from commercial fuel-reprocessing plants. The total military ^{90}Sr inventory as of 1976 was approximately 270×10^6 curies. There is only a relatively small increase in this inventory projected for the early 1980's. (Table 1 and Fig. 1). Only fairly low rates of plutonium production for military purposes are expected, and radioactive decay of the ^{90}Sr in the currently existing wastes will offset about one half of the projected additions.

As of the end of 1975, the generating capacity of U.S. nuclear-electric power plants was about 40,000 Mw and had produced a cumulative total of about 60,000 Mw-years of electrical energy (12). For a thermal-electric conversion efficiency of approximately one third for nuclear-electric power plants, this amount of electrical energy corresponds roughly to 200,000 thermal Mw-years or an accumulated ^{90}Sr inventory (allowing for decay) of about 200×10^6 curies. On

the basis of current plans of the utilities, each of the next two doublings in U.S. nuclear-electric-generating capacity will take approximately 5 years (13). If these plants are operated, on the average, at 60 percent capacity, the corresponding inventory of ^{90}Sr will increase by approximately an order of magnitude by 1985, at which time the production rate of ^{90}Sr will be about 300×10^6 curies per year.

According to the best estimates we can make from the published data, the civilian and military inventories of ^{90}Sr are currently approximately equal; by 1985 the total civilian inventory will be increasing annually by an amount of the same order as the total military inventory (Fig. 1). The comparison of these numbers in itself therefore offers no basis for complacency concerning the magnitude of the civilian radioactive waste problem. Fortunately, however, the physical and chemical form of civilian radioactive waste (still virtually all in spent fuel) may make its ultimate disposal easier than that of the neutralized radioactive waste concentrates in the tank farms at Hanford and SRP.

Note added in proof: Recently figures have been made public for the quantities of ^{90}Sr plus ^{137}Cs in military radioactive wastes at the principal ERDA sites as of

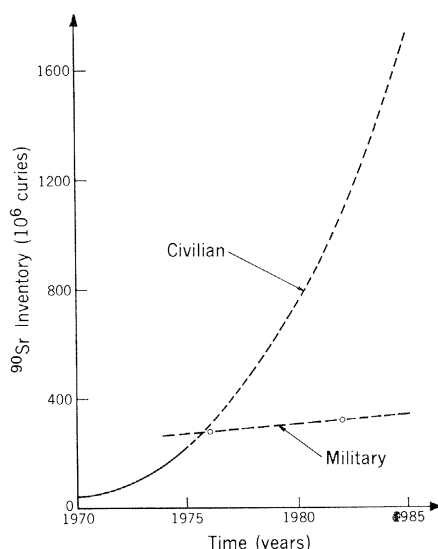


Fig. 1. Estimated civilian and military high-level waste inventories measured in terms of their ^{90}Sr content as a function of time.

the end of 1976: Hanford, 360; SRP, 210; INEL, 20; and total, 590 (in 10^6 curies). Dividing by 2 to get the approximate inventories of ^{90}Sr gives numbers in reasonable agreement with our estimates in the second column of Table 1 (14).

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References and Notes

1. H. Brooks, in *Proceeding of the International Symposium on the Management of Wastes from the Light Water Reactor Fuel Cycle* (CONF-76-0701, Energy Research and Development Administration, Washington, D.C., 1976).
2. Code of Federal Regulations, Title 10, Part 20: Standards for Radiation Protection, Appendix B, table II.
3. *Environmental Survey of the Reprocessing and Waste Management Portions of the Light Water Reactor Fuel Cycle* (NUREG-0116, Nuclear Regulatory Commission, Washington, D.C., 1976), figures 4.9 and 4.11.
4. K. A. Varteressian and L. Burris, *Fission Product Spectra from Fast and Thermal Fission of ^{235}U and ^{239}Pu* (ANL 7678, Argonne National Laboratory, Argonne, Ill., 1970). This is the "chain yield," that is, it includes ^{90}Sr atoms which result from the β -decay of the following short-lived fission products: ^{90}Br (half-life, 1.6 seconds), ^{90}Kr (33 seconds), and ^{90}Rb (2.9 minutes).
5. *Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors* (NUREG-0002, Nuclear Regulatory Commission, Washington, D.C., 1976), chap. IV, table IV H-17.
6. *Final Environmental Statement, Waste Management Operations, Hanford Reservation* (ERDA-1538, Energy Research and Development Administration, Washington, D.C., 1975).
7. Between 170 and 300 gallons per metric ton of uranium in the spent fuel [Alternatives for Managing Wastes from Reactors and Post Fission Operations in the Light Water Reactor Fuel Cycle (ERDA-76-43, Energy Research and Development Administration, Washington, D.C., 1976), vol. 2, p. 6.1].
8. About 1380 Mw-days of fission per ton of uranium in the N-reactor fuel [see (6), vol. 2, p. III-D-3] versus a typical "burnup" of about 20,000 Mw-days per ton of uranium in commercial power reactor fuel. The burnup of the fuel in the plutonium production reactor is kept so low in order to minimize the contamination of ^{239}Pu by the higher-mass plutonium isotopes which result from neutron capture by ^{239}Pu .
9. Numbers for the waste volume and radioactive inventory are given by D. D. Wodrich [Hanford Long Term High-Level Waste Management Program Phase I: Technology Development (ARH-LD-142, Atlantic Richfield Hanford Company, Richland, 1976), p. 2] for the early 1980's. Because of a separation program for ^{137}Cs and ^{90}Sr , about 90 percent of this ^{90}Sr will actually not be in the large-volume, high-level wastes but will be stored in water-cooled capsules elsewhere on the site. The number of curies quoted in the above report, 265×10^6 curies, is the sum of the activities of the separated ^{90}Sr and ^{137}Cs isotopes. We have obtained the number of curies of ^{90}Sr by taking into account the fission yield of the ^{90}Sr and ^{137}Cs isotopes from thermal fission of ^{235}U [5.7 and 5.9 percent, respectively (4)], their specific activities, and the separation factor of 90 percent for the process used to separate cesium and strontium from the wastes as quoted in (6), vol. 1, p. II.1-31. While the 1976 volume is provided in the above report, the ^{90}Sr activity for 1976 has been obtained by calculating back from the inventory given for the early 1980's on the assumption that 8.43×10^6 curies of ^{90}Sr will be generated per year between 1976 and 1982 [(6), vol. 1, p. iii, and vol. 2, Appendix D] and by allowing for radioactive decay.
10. *Draft Environmental Statement: Waste Management Operations, Savannah River Plant, Aiken, South Carolina* (ERDA-1537, Energy Research and Development Administration, Washington, D.C., 1976). For high-level waste volumes, see p. I-7. The ^{90}Sr activity in fresh high-level waste is typically 3 curies per gallon (table C-2). Past and projected high-level waste gener-

ation can be approximated by 3.3×10^6 gallons per year between 1957 and 1976, and by 2×10^6 gallons per year from 1976 to 1982 (figure I-1). Calculated ^{90}Sr activities for 1976 and 1982 take into account radioactive decay.

11. *Draft Environmental Statement: Waste Management Operations, Idaho National Engineering Laboratory, Idaho* (ERDA-1536, Energy Research and Development Administration, Washington, D.C., 1976). Waste volumes are derived from information on pp. I-3 and I-4. Inventories of ^{90}Sr have been calculated as follows: the 300,000 gallons of liquid waste calcined in 1974 had a fission product concentration of 6.3 curies per gallon before calcination (figure II-18). If we make the conservative (high) assumption that this fission product activity is exclusively due to the isotope pairs $^{90}\text{Sr}/^{90}\text{Y}$ and $^{137}\text{Cs}/^{137}\text{Ba}$, then ^{90}Sr would be responsible for one quarter of this activity, that is, about 1.6 curies per gallon. Further assumptions are a high-level waste generation of 0.24×10^6 gallons per year between 1956

and 1976, and 0.34×10^6 gallons per year from 1976 to 1982 (pp. I-3 and I-4). Radioactive decay is also accounted for.

12. *Statistical Abstract of the United States, 1975* (Department of Commerce, Washington, D.C., 1975), table 886; Department of Interior, Bureau of Mines, News Release, "Annual U.S. energy use drops again" (5 April 1976), table 2.
13. *Electr. World* **186** (No. 6), 43 (15 September 1976).
14. J. A. Lieberman, W. A. Rodger, F. P. Baranowski, "High-Level Waste Management," prepared testimony presented to the California Energy Resources Conservation and Development Commission, 21 March 1977. We would like to thank T. Lash for bringing this testimony to our attention.
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Plant-Animal Mutualism: Coevolution with Dodo Leads to Near Extinction of Plant

Abstract. *An endemic sapotaceous tree Calvaria major found on the island of Mauritius is nearly extinct because its seeds apparently required passage through the digestive tract of the now-extinct dodo Raphus cucullatus to overcome persistent seed coat dormancy caused by a specially thickened endocarp.*

Coevolution can lead to mutualism in plant-animal systems, and occasionally one or both of two mutualistic species develops a considerable degree of dependence on the other member of the pair (1). Sometimes the elimination of a particular plant species can result in a marked numerical response in an associated animal population (2). However, there seem to be no documented examples of the reverse situation, in which a plant declines in abundance as a result of the extirpation of an associated animal, even though such an occurrence is obviously possible (3). In this report, I present evidence that the near extinction of a plant species may have been the direct result of the extinction of an animal with which that plant had coevolved. The two species in this example of obligatory mutualism are a now nearly extinct tree *Calvaria major* and the extinct dodo *Raphus cucullatus*, each of which is or was endemic to the island of Mauritius in the western Indian Ocean.

Calvaria major is a large monoecious tree in the family Sapotaceae (4). Historical forestry records indicate that *Calvaria* was formerly common and frequently exploited for lumber on Mauritius (5). However, by 1973, only 13 old, overmature, and dying trees were known to survive in the remnant native forests of the island. The age of each of these trees was estimated to be more than 300 years by experienced Mauritian foresters (6). No younger specimens are known to exist despite the fact that the surviving trees produce well-formed, apparently fertile seeds each year. None of these

seeds now germinate naturally, and if the total absence of young plants is a valid indication, there has been no germination of *Calvaria* seeds for hundreds of years. Even when planted under nursery conditions, the seeds remain dormant (6). Low seed germination rates are not uncommon among tropical island plants (7), but the extraordinarily long period of time during which no *Calvaria* seeds have apparently germinated seems too excessive to be normal.

The fruits of *Calvaria* are large, single-seeded drupes about 50 mm in diameter. Anatomically, the fruit is composed of a thin exocarp; a pulpy, succulent mesocarp; and a hard, woody, thick-walled endocarp. The seed is depresso-globose in shape and is completely enclosed in a stone or pit formed by the walls of the endocarp, which can be as thick as 15 mm. Some other sapotaceous plants also have relatively thick endocarps covering their seeds, but the endocarp surrounding a *Calvaria* seed is extraordinarily thick even for this family. Apparently, *Calvaria* seeds fail to germinate because the thick endocarp mechanically resists the expansion of the embryo within.

At the time of its discovery, Mauritius supported a remarkable endemic avifauna which has subsequently been decimated through the direct or indirect activities of man (8). Perhaps the most unusual of the original endemic birds was the dodo. We know disappointingly little of the biology of this fascinating bird; it became extinct by 1681, less than two centuries after it was discovered. We do know that the dodo was a huge, flightless

bird that attained an estimated body weight of at least 12 kg. It had a large, strong beak, and the reports of early explorers indicate that dodos fed on fruits and seeds, especially seeds of palms and large forest trees. The dodo possessed a well-developed gizzard that contained large stones, which were used to crush tough food items (8).

The temporal coincidence between the extinction of the dodo 300 years ago and the last evidence of natural germination of *Calvaria* seeds led me to hypothesize the following mutualistic relationship between *Calvaria* and the dodo. In response to intense exploitation of its fruits by dodos, *Calvaria* evolved an extremely thick endocarp as a protection for its seeds; seeds surrounded by thin-walled pits would have been destroyed in the dodo's gizzard. These specialized, thick-walled pits could withstand ingestion by dodos, but the seeds within were unable to germinate without first being abraded and scarified in the gizzard of a dodo.

Fossil *Calvaria* pits have been found among skeletal remains of dodos in the mud of the Mare aux Songes marsh, which suggests that dodos ate the fallen fruits of *Calvaria*. Even today the fruits are frequently consumed by endemic frugivorous animals like the Mauritius parakeet *Psittacula echo* and the Mauritius flying fox *Pteropus niger*. Unlike these smaller surviving animals that eat only the fleshy mesocarp and leave the large pit untouched, the dodo was large enough to have swallowed the entire fruit. Many birds will retain in their digestive tract hard objects that the gizzard cannot readily crush; after a period of time, these objects are subsequently either regurgitated or reduced in size and passed through the intestinal tract (9). The hard pits of *Calvaria* may have similarly been regurgitated or excreted by dodos.

Germination rates in some plant species that show seed-coat dormancy can be increased significantly by passage through the digestive tracts of animals (10). In this way, animals can play an important role in overcoming seed-coat dormancy and in dispersing the consequently germinable seeds. Although there seem to be no recorded instances in which passage through an animal's gut is an absolute necessity for seed germination, in many cases germination rates are extremely low without such treatment (10).

If the *Calvaria*-dodo coevolution hypothesis is correct, *Calvaria* pits must have been able to withstand the crushing forces that were presumably generated by a dodo's gizzard. Measurements of