High-level radioactive waste from light-water reactors*

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The production of radioactive nuclei during the operation of a light-water reactor is traced, and their decay history is followed. The potential environmental impacts of this waste are calculated and shown to be comparable to those of other materials we produce. Assuming deep burial, it is shown that there are important time delays which prevent the waste from reaching the biosphere in the first few hundred years while its toxicity is decreasing by several orders of magnitude. In the long term, the most important pathway to man was found to be through groundwater into food and water supplies, with consequences calculated to be 0.4 fatalities in 10^6 years from each year of all-nuclear power in U.S. Other pathways considered and found to be less important include meteorites, volcanism, release through ground water to airborne particulate, and human intrusion by drilling and mining for unspecified materials and for salt. For time scales longer than 10^6 years, nuclear power is shown to reduce man's exposure to radiation by consuming uranium. A cost-benefit analysis is developed for surveillance of buried waste. It is shown that buried high-level waste is environmentally much less dangerous than uranium mill tailings.

CONTENTS

12

In a light-water reactor, uranium enriched to about 3.3% in ^{235}U is exposed to neutrons which induce fission and neutron capture reactions. A great many of the products of these reactions are radioactive, and their radioactivity and that of their daughters is a potential health hazard from a nuclear power industry. In this paper, the problem is explained in some detail, and efforts are made to assess the hazards and place them in perspective.

All calculations were carried out with the Oak Ridge National Laboratory computer program ORIGEN, (Bell, 1973) a highly versatile code which simultaneously solves the coupled differential equations for production and decay of a large number of isotopes. The program includes a very extensive library of nuclear data which was widely used.

Calculations were carried out for one ton (1000 kg) of original fuel charged to the reactor, and many results are given on that basis. Frequent allusion is also made to the total waste generated by one year of all-nuclear power in the U.S. at some future time. This is taken to be 400×10^6 kW (400 GW) average power production, about 80% above total present electric generation (of which 8–9% is now nuclear).

I. PHYSICS ASPECTS: REACTOR PHASE

A typical (Bell, 1973) reactor would produce an average of 30 MW of thermal power per (metric) ton of fuel. The average neutron flux would be 2.9×10^{13} neutrons/cm^2 sec^{-1}. Any given fuel rod would be replaced after about 1100 days in the reactor, which means that one ton of fuel produces 33 000 MW days of thermal energy. The original fuel would contain an average of 3.3% ^{235}U and at discharge this would be reduced to 0.8%, a consumption of 24.9 kg of ^{235}U per ton of fuel. In the process, 2.4% of the ^{238}U, 23.8 kg per ton of fuel, is also consumed, 22.2 kg going to $^{239}\mathrm{U}\rightarrow{}^{239}\mathrm{Np}\rightarrow{}^{239}\mathrm{Pu}$, and 1.6 kg undergoing fission from fast neutrons. The fission reaction releases about 191 MeV of energy per atom (exclusive of neutrino kinetic energy)—168 MeV as fission kinetic energy, 5 MeV as neutron kinetic energy, 5 MeV as prompt gamma rays, 7 MeV as fission product beta particles, and 6 MeV as gamma rays following beta decay. This total converts to 0.91 MW days per gram (910 000 MW days/ton) undergoing fission, so the 33 000 MWdays/ton yield implies that 3.6% of the original fuel undergoes fission.

Depletion and growth of various nuclides is shown as a function of time in Fig. 1, and an understanding of it

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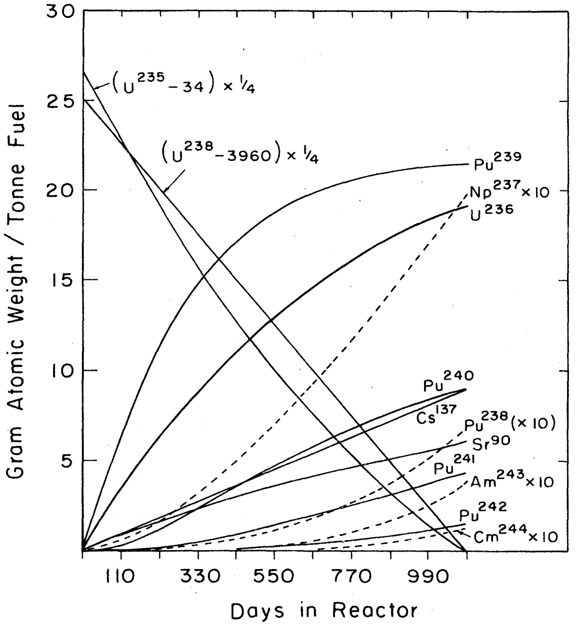

FIG. 1. Quantities of various nuclei in reactor fuel as a function of their time in the reactor. Dashed curves are multiplied by 10.

is aided by the list of cross sections given in Table I. The ^{235}U is burned exponentially, at a rate approximately proportional to the quantity remaining, 80% by fission and 20% by capture to ^{236}U. Because of its small cross section, only 10% of the ^{236}U becomes ^{237}Np which, with its large cross section, is more than 25% converted to ^{238}Pu. (We ignore short half-life beta decays needed to convert ^{237}U to ^{237}Np, ^{238}Np to ^{238}Pu, etc.)

Of the ^{238}U (burned essentially linearly with time because of the small percentage consumption), 7% is consumed by fast neutron fission and 93% by neutron capture to produce ^{239}Pu. The latter, because of its large cross section is largely destroyed, 71% by fission and 29% by capture; about 85 g-at. are produced, but only 23 g-at. survive, so all in all, 0.71 (85 − 23) = 44 g-at./ton of fuel undergo fission as compared with 84 g-at. of ^{235}U (from Fig. 1, 106 g-at. of ^{235}U are consumed, and from Table I, 409/514 of this undergoes fission). The ^{239}Pu that undergoes capture becomes ^{240}Pu, which has a very large cross section and is thus rapidly converted to ^{241}Pu. The latter has very large cross sections both for fission and for capture leading to ^{242}Pu, which may be followed by further neutron captures to produce ^{243}Am and ^{244}Cm. At the end of fuel life, the fissile materials in g-at./ton of fuel are ^{235}U–34, ^{239}Pu–23, and ^{241}Pu–44, so that, after weighting by fission cross sections, we see that the thermal fission reactions at end of fuel life are occurring 32% in ^{235}U, 57% in ^{239}Pu, and 11% in ^{241}Pu.

The buildup of two important fission products, ^{90}Sr and ^{137}Cs, is also shown in Fig. 1. Both of these have long half-lives and modest cross sections and hence build up linearly in time. All in all, for each 1000 kg of uranium charged to the reactor, 44 kg of uranium are converted to 35 g of fission products plus 9 kg of transuranics. In addition, this fuel contains 134 kg of oxygen (the material is UO_2) which is virtually unaffected, and is clad with 272 kg of a Zr alloy containing 249 kg of Zr, 9.6 kg of Ni, 3.7 kg each of Fe, Cr, and Sn, and a few hundred grams of other materials of which the most significant is 54 g of cobalt as 10% of it is converted into ^{60}Co, a potent gamma-ray emitter with a 5.3 yr half-life.

TABLE I. Capture and fission cross sections for various nuclei (in barns). Values are averaged over the neutron spectrum in a light-water reactor (Bell, 1973).

Nucleus	$\sigma(n,\gamma)$	$\sigma(n,f)$	Nucleus	$\sigma(n,\gamma)$	$\sigma(n,f)$
^{238}U	8.4	0.60	^{235}U	105	409
^{239}Pu	443	1060	^{236}U	74	3.2
^{240}Pu	897	2.5	^{237}Np	359	2.5
^{241}Pu	394	1110	^{238}Pu	366	19
^{242}Pu	438	0.2	^{234}U	281	3.9
^{243}Am	566	0.8	^{90}Sr	1.23	⋯
^{244}Cm	223	4.9	^{137}Cs	0.17	⋯

The first step in permanent disposal of these wastes is to send this material to a fuel reprocessing plant where it is to be cut into pieces, dissolved in acid, and chemically processed to extract 99.5% of the uranium and plutonium to be used for fabrication of new fuel. In addition, the gases, H, Kr, and Xe come off at this stage. The remains are what we refer to as "high-level radioactive waste." In practice, cladding hulls do not dissolve and are handled separately, and 99.9% of the Br and I are removed for separate storage, but we treat these materials as though they are part of the waste. In our quantitative discussions we will assume that the fuel is reprocessed 5 months after removal from the reactor. In general, our results are rather insensitive to this time interval, but if it is extended to several years, the decay of ^{241}Pu into ^{241}Am can increase the amount of the latter by an order of magnitude.

II. PHYSICS ASPECTS: WASTE PHASE

The material in the waste continues to undergo radioactive decay and, while the decay chains are simple and

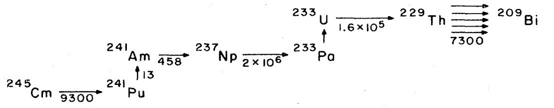

FIG. 2. Principal decay modes of heavy nuclei with half-lives greater than one year. Multiple arrows indicate preceding shorter half-life decays. Horizontal arrows are alpha decays and vertical arrows are beta decays; figures attached to arrows are half-lives in years.

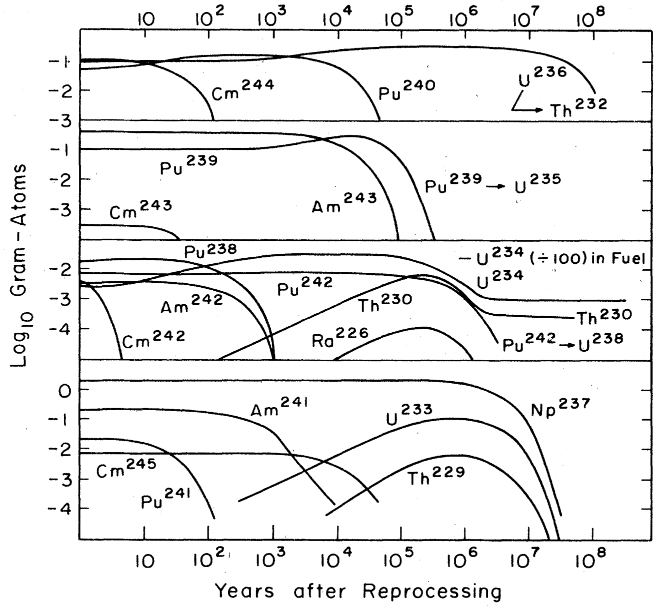

FIG. 3. Quantities of various nuclei in waste as a function of time after reprocessing. The four parts of the figure are for the four radioactive decay series, (from top) $A = 4n$, $(4n-1)$, $(4n-2)$, and $(4n-3)$, where A is the atomic weight, and n is an integer. Note that 99.5% of U and Pu have been extracted.

straightforward for the fission products, the same is not true for the transuranics. The latter may be understood with the help of Figs. 2 and 3 for each of the four decay chains, $A = 4n$, $4n-1$, $4n-2$, and $4n-3$ (where n is integer).

For $A = 4n$, there are initially about equal quantities of ^{244}Cm, ^{240}Pu (recall that 99.5% of the Pu is extracted in reprocessing), and ^{236}U. The ^{244}Cm decays rapidly into ^{240}Pu, which eventually decays into ^{236}U, which in turn very slowly decays into ^{232}Th. (We skip over the decay chains of the naturally radioactive isotopes ^{232}Th, ^{235}U, and ^{238}U because their long half-lives make the hazards from them relatively unimportant. They are, however, included in all calculations.)

For $A = 4n-1$, there is initially about four times as much ^{243}Am as ^{239}Pu, and the former decays into the latter, as does the small amount of ^{243}Cm present. The ^{239}Pu eventually decays into ^{235}U.

For $A = 4n-2$, after one year where Fig. 2 begins, there is a considerable amount of ^{242}Pu, ^{242}Cm, ^{242}Am, ^{238}Pu, and ^{234}U. The ^{234}U is carried over from the original fuel since it is a naturally occurring isotope of uranium, and most of the ^{238}Pu is derived from ^{242}Cm decay in the one year since reprocessing, as 99.5% of the original ^{238}Pu was removed in reprocessing. The ^{242}Pu decays into ^{238}U, but the ^{242}Cm and ^{242}Am bypass ^{238}U by decaying into ^{238}Pu, which then decays directly to ^{234}U, which decays via the chain shown in Fig. 2.

For $A = 4n-3$, the decay is a straightforward chain as $^{245}\text{Cm} \rightarrow {}^{241}\text{Pu} \rightarrow {}^{241}\text{Am} \rightarrow {}^{237}\text{Np} \rightarrow {}^{233}\text{U} \rightarrow {}^{229}\text{Th} \rightarrow {}^{207}\text{Pb}$. The first three are not important contributors to the remainder of the chain, as ^{237}Np is initially present in such large quantities, but they have important potential environmental impacts during their lifetimes.

In summary, all four decay chains contribute important transuranics, and in two of them there are important contributions from lower atomic number species as ^{238}U gets bypassed in the $(4n-2)$ series and there are no half-lives longer than 2.1 million yr in the $(4n-3)$ series.

TABLE II. Long half-life fission products. The last column is the percentage of all fission product atoms that are the designated isotope.

Nucleus	Half-life (yr)	%-g-at.
	A. Half-lives 1–10 yr	
Ru-106 (Rh-106)	1.0	0.51
Sb-125	2.7	0.022
Cs-134	2.1	0.47
Pm-147	2.6	0.25
Eu-154	8.6	0.10
Eu-155	4.8	0.013
	B. Half-lives 10–100 yr	
Kr-85	10.8	0.11
Sr-90	29	2.0
Cd-113*m*	14	0.0001
Sn-121*m*	25	10^{-7}
Cs-137	30	3.0
Sm-151	90	0.10
Eu-152	12	0.0001
Eu-154	16	0.10
	C. Half-lives 100–4×10^{10} yr	
Se-79	7×10^{4}	0.01
Zr-93	9×10^{5}	2.7
Tc-99	2×10^{5}	2.9
Pd-107	7×10^{6}	0.74
Sn-126	1×10^{5}	0.05
I-129	1.6×10^{7}	0.61
Cs-135	2×10^{6}	0.81
Ho-166*m*	1.2×10^{3}	10^{-6}

Fission products with half-lives longer than one year are listed in Table II. We see that, with the unimportant exceptions of ^{151}Sm, ^{166m}Ho, and ^{79}Se, there are no half-lives between 30 yr and 10^5 yr, a truly remarkable gap. Among the shorter-lived group, ^{137}Cs and ^{90}Sr are of predominant importance, especially since ^{85}Kr is by definition not included in the waste. From the sum of the figures in the last column we see that 86% of the fission product atoms have half-lives shorter than one year or longer than 4×10^{10} yr and hence (as we shall see) are not of concern for long-term waste storage.

While there has been no U.S. regulatory decision on long-term handling of nuclear waste, other than a ruling[1] that it be converted into a dry and stable solid within 5 yr and shipped to a Federal Repository within 10 yr, the most commonly discussed plan is to incorporate it into a massive glass rod encased in steel canisters about 10 ft long by 1 ft in diameter and bury it deep underground in some carefully chosen geological formation with horizontal spacings of about 10 m between canisters. These dimensions are dictated largely by heat transfer problems. The thermal power in the waste due to various isotopes is shown in Fig. 4. It may be noted that it decreases by nearly an order of magnitude be-

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[1] Code of Federal Regulations, Title 10, Part 50, Appendix F.

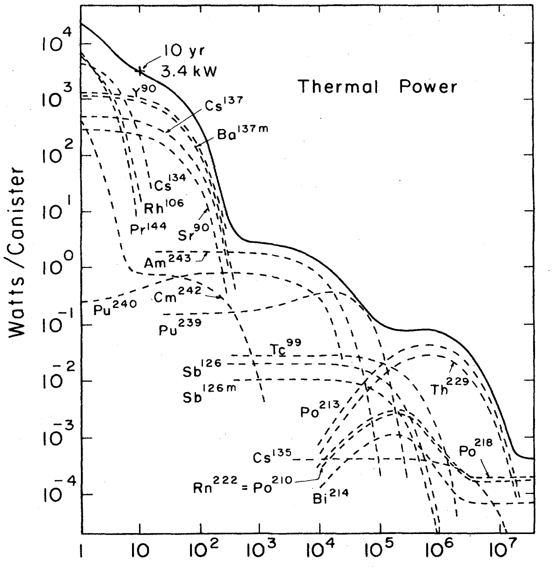

FIG. 4. Thermal power released by the radioactivity in one waste canister. Such a canister contains the waste from 100 MW yr, so a typical large power plant would produce 10 canisters of waste per year. Dashed curves are contributions from individual isotopes, and the solid curve is the sum of these contributions.

tween one and ten years. As shown in Fig. 5, calculations (Schneider and Platt, 1974) of the diffusion of heat into the rock in which it is buried indicate that this translates into nearly an order of magnitude reduction in maximum wall temperature if burial is delayed for 10 yr. For the first 400 yr after burial, the heat is due largely to ^{90}Sr, ^{137}Cs, and their daughters; it decreases by three orders of magnitude during this period. After 400 yr, the dominant contributors are plutonium and americium isotopes for the next 100 000 yr, and their decay daughters thereafter.

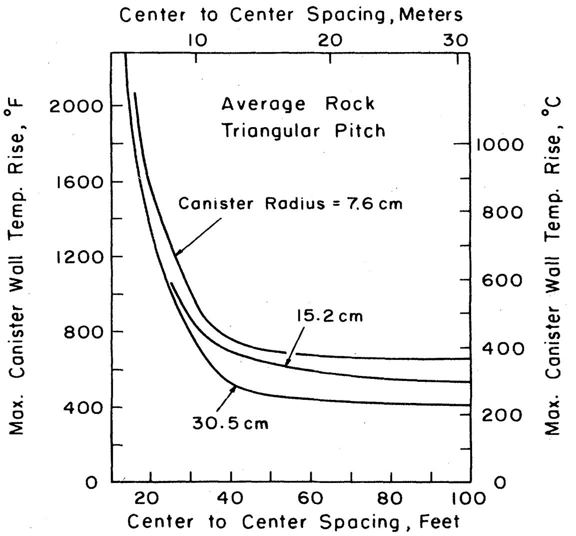

FIG. 6. Effect of spacing and canister radius on maximum canister wall temperature (Schneider and Platt, 1974). It is assumed that each (10 yr aged) canister produces 3.4 KW of heat initially, decaying with a 30 yr half-life.

The reason for the choice of canister diameter and spacing is evident in Fig. 6 (Schneider and Platt, 1974). Larger diameters would be inconvenient for drilling and handling, and larger spacings would be more costly while contributing little to the heat dissipation problem. The height of the canister is chosen to minimize handling problems, with due consideration for the total thermal power of the planar array. For a power of 250 KW/hectare, corresponding to a square array with 11.8 m between rows of canisters with burial at 10 yr, the temperature rise is shown in Fig. 7 (Schneider and Platt, 1974) for various distances above and below the burial plane (assumed infinite in extent) and at various times after burial. It is seen that the maximum temperature does not occur until 40 yr after burial. Also indicated in Fig. 7 is one of the several advantages of burial in rock salt. Its thermal conductivity is about three times higher than that of average rock (1.24×10^{-2} vs 4.1×10^{-3} cal/sec cm^2 °C/cm) which results in a reduction of maximum canister wall temperature rise from 145° to 85 °C.

One interesting and perhaps important feature of burial

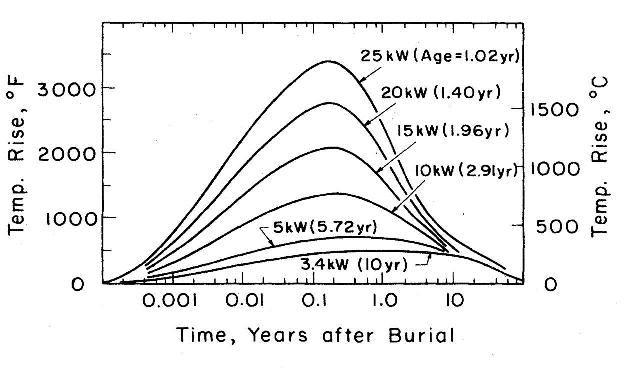

FIG. 5. Effect of waste age (and initial decay heat rate) at burial (labels attached to curves) on the wall temperature of a single buried waste canister (Schneider and Platt, 1974). It is assumed that the surrounding rock has a thermal conductivity of 4.1×10^{-3} cal/cm sec-deg C, which is typical of crystalline rock.

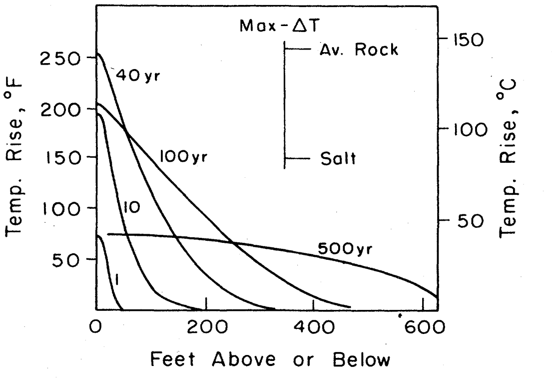

FIG. 7. Rock temperature rise as a function of distance above or below the waste burial plane for various times after burial (Schneider and Platt, 1974). Thermal conductivity is as in Fig. 4. The inset shows this temperature rise at the time when it is a maximum for burial in typical crystalline rock (assumed for curves) and in salt.

in salt is that small brine inclusions (0.1–1 mm) migrate up the temperature gradient due to the increase with temperature of the solubility of salt in water. It is estimated (McClain, 1976) that this will result in the ultimate collection of 20–30 liters of water at each canister; the initial collection rate is 2–3 liters per year, and the collection is complete after 20–25 years, ceasing because of the reduction in temperature gradients with time. Assuming that the repository is kept open for this time period the collected water would flash into steam and be carried away by the ventilation system.

The wastes contained in a single waste canister are those from about 3 ton of original fuel, and represent about $\frac{1}{10}$ of the total waste produced annually by a 1000 MW reactor. A canister has a total volume of about 0.2 m^3 and weighs about 600 kg, including 86 kg of fission products, 14.4 kg of uranium, 1.44 kg of Np, and 0.66 kg of Pu, Am, and Cm combined. The steel casing is designed to contribute to safety in handling and transporting waste, but it is usually assumed to corrode away not long after burial.

If all U.S. electrical power were nuclear (400 GW), about 4000 waste canisters per year would be generated. If they were buried in a square matrix with 10 m spacing between rows, they would occupy 0.4 square kilometers. A thousand years of waste so buried would therefore occupy 400 square kilometers, or an area of 12 miles square if all were colocated.

III. POTENTIAL ENVIRONMENTAL IMPACTS OF HIGH-LEVEL WASTE

The potential environmental impacts of radioactivity that we consider here are those due to external radiation of the human body with gamma rays or with neutrons, and internal radiation if the material is taken into the body by inhalation with air or by ingestion with food or water. All of these effects are governed by the radiation emission rate which, apart from a generally well known branching ratio, is equal to the disintegration rate, often expressed as "curies.". The conversion from "gram-atoms," used in Figs. 1 and 3, to curies depends only on the half-life through the relationships

$$1\ \text{curie} = 3.7\times10^{10}\ \text{disintegrations/second}$$

$$\text{disintegrations/second} = -dN/dt = \lambda N = \frac{0.693N}{\text{half-life}}\ ,$$

where N is the number of atoms (= g-at. × Avogadro number) at any time t. The conversion from curies to thermal watts, used in constructing Fig. 4, simply involves multiplying by the energies of the radiations emitted and converting from MeV to joules.

The biological effects of gamma radiation are roughly proportional to the gamma-ray energy, so the pertinent quantity is gamma-ray energy release per second, or just gamma-ray power expressed in watts. The results for each important isotope as a function of time after reprocessing are shown in Fig. 8. The solid line in Fig. 8 is the sum of all contributions. We see that ^{137}Cs is completely predominant from 10–400 yr, after which ^{239}Np, ^{243}Am, and ^{126}Sb (a daughter of long-lived ^{126}Sn) predominate until Bi daughters of transuranics grow in and become important after 200 000 years.

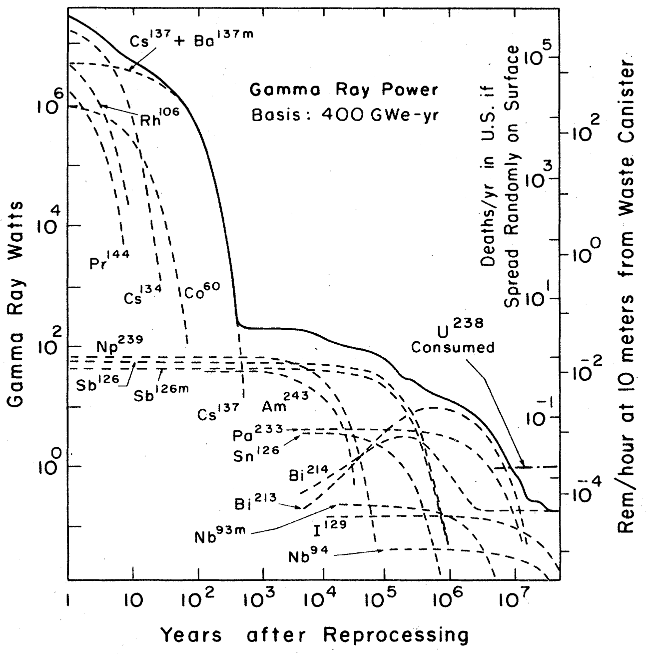

FIG. 8. Gamma-ray power emitted by the waste from 400 GW yr of nuclear electricity. Dashed curves are the contributions from various individual isotopes, and the solid curve is the total emitted power, which is the sum of these individual contributions. The dot-dash curves are what would be emitted by the uranium consumed by the operation. The scale inside the right margin shows the deaths per year expected from gamma-ray induced cancer if all of the waste were spread randomly (or uniformly) over the surface of the U. S., as a function of time when this is done. The scale on the outside of the right margin gives rem per hour exposure at a distance of 10 m from an unshielded waste canister (neglecting self-shielding by the canister material).

Inhalation and ingestion hazards are much more complicated to calculate. In calculating the risk of cancer in body organ j per curie ingested of radioactive isotope i, one must first calculate the dose in rem to organ j per curie of intake, and then multiply it by the cancer risk per rem. The latter, the cancer risk per rem r_j, to various organs j, is obtained from the BEIR Report (NAS 1972), and is listed in Table III. It may be noted that in giving a constant risk per rem, the BEIR Report assumes a linear, no threshold dose-effect relationship. We now turn to the much more complex first part of the calculation, deducing the dose in rem per curie of intake.

Dose to the bone in rem per curie of a given isotope ingested is the energy deposited by radiation in the bone (in units of 100 ergs) divided by the weight of the bone, with the quotient multiplied by the relative biological efficiency (RBE) used to convert rad to rem (ICRP, 1959). The energy deposited per curie is the product of the fractional transfer from the intestines into the bloodstream, the fractional transfer from the bloodstream to the bone, the average residence time in the bone ("biological half-life"), the decay rate (3.7×10^{10} per sec per ci), the energy of radiation emitted per decay, and the fraction of this radiation energy ab-

TABLE III. Lifetime risk of cancer death per rem to various organs. Values from the BEIR Report (NAS, 1972), p. 171, assuming the absolute risk model with a 30 yr plateau period (25 yr for leukemia) for a young adult.

Type of cancer	Risk/rem($\times 10^6$)
Leukemia (bone marrow)	25
Breast	45
Lung	39
G. I. (inc. stomach)	30
Bone	6
Thyroid	6 [a]
All other	24
(Any one other)	≤ 6 [b]
Total	~180

[a] Estimated from NAS, 1972, p. 124, giving credit for a 90% cure rate.

[b] Estimated from fact that all others combined are less than 24.

TABLE IV. Maximum permissible concentrations (MPC) for various isotopes in air (insoluble) and water (soluble). For air, limiting organ is lung; for water, it is bone unless otherwise noted. Units are in Ci/m^3; numbers in parentheses are power of ten multiplication factors.

Isotope	$(MPC)_A$	$(MPC)_W$
^{90}Sr	2(−9)	4(−6)
^{99}Tc	2(−8)	3(−3) [a]
^{129}I	2(−8)	4(−6) [b]
^{137}Cs	5(−9)	2(−4) [c]
^{226}Ra	•••	1(−7)
^{227}Ac	9(−12)	2(−5)
^{229}Th	3(−12)	2(−5)
^{237}Np	4(−11)	3(−5)
^{239}Pu	1(−11)	5(−5)
^{241}Am	4(−11)	4(−5)
^{244}Cm	3(−11)	7(−5)
^{245}Cm	4(−11)	4(−5)

[a] Critical organ is GI tract.

[b] Critical organ is thyroid.

[c] Critical organ is whole body.

sorbed in the bone. The cancer risk is then obtained by multiplying the dose to the bone in rem by the risk per rem (6×10^{-6} from Table III).

This is the risk of bone cancer per curie ingested of a given radioactive isotope. In principle the risk of each other type of cancer should be calculated similarly, and the total cancer risk is the sum of these. However, we have used the ICRP (International Commission on Radiological Protection) procedure of considering only the organ giving the highest cancer risk ("critical organ"). Ignoring the others underestimates the risks (by less than a factor of 2), and we also ignore *in utero* exposures where radiation is an order of magnitude more damaging (Stewart and Kneale, 1968) since only about 1% of exposures are *in utero*. These underestimates are compensated by the fact that we assume that all victims are of the most susceptible age (i.e., they have at least 45 yr of life expectancy after exposure—the 15 yr latent period plus the 30 yr plateau period—and are not children less than age 10 for whom the risks are five times lower for all but leukemia and thyroid cancer), and by the general conservatism in ICRP procedures. In nearly all important cases, the critical organ was the bone; the exceptions are ^{90}Sr (leukemia from bone marrow exposure about equal to bone cancer), ^{129}I (thyroid), and ^{99}Tc (gastrointestinal tract).

All the information needed for the calculation of dose to various body organs per curie ingested of various radioactive isotopes is given in ICRP Publication Numbers 2 and 6 (ICRP, 1959; ICRP, 1962), but the calculations can be short circuited by recognizing the fact that this same information has already been used in just this way in calculating the MPC (maximum permissible concentration for occupational exposure), and the latter is given directly in Tables in those publications. The MPC is defined as that concentration of an isotope in water (in Ci per m^3) which, if water is ingested at a rate of 2.2 li per day (0.8 m^3 w/yr, where w refers to water), would give a dose commitment d_j to body organ j, where d_j is 30 rem for bone and thyroid, 15 rem for lungs, kidneys, gastrointestinal tract, etc. (ICRP, 1959). Some MPC values are listed in Table IV. The number of cancer doses to organ j in the high-level waste produced by one year of all-nuclear power (1.2×10^4 ton of spent fuel) due to radionuclide i, D_{ij}, may then be calculated as

$$D_{ij}\left(\frac{\text{doses}}{\text{year}}\right)=\frac{Q_i(\text{Ci/ton})}{MPC_{ij}(\text{Ci/m}^3\text{w})}\times\frac{dj\,(\text{rem})}{0.8(\text{m}^3\,\text{w})}\times r_j\left(\frac{\text{doses}}{\text{rem}}\right)\times 1.2\times 10^4\left(\frac{\text{ton}}{\text{year}}\right), \tag{1}$$

where Q_i is the curies of isotope i in one ton of spent fuel. For example, ten years after reprocessing, $Q_i=6.0\times 10^4$ for ^{90}Sr, MPC to bone from Table IV is 4×10^{-6}; d_j for bone was given above as 30; and r_j for bone cancer is 6×10^{-6} from Table III, so from (1) we calculate $D_{ij}=4.0\times 10^{10}$. That is, the ^{90}Sr in one year's waste, if fed to people in digestible form, would cause 4.0×10^{10} bone cancers assuming that there are enough people involved and no one person ingests an amount approaching $1/(4\times 10^{10})$ of the total (if these latter provisos are not satisfied, fewer bone cancers would result). Beyond this condition it does not matter how the total ingested is distributed among people; this is a consequence of the linear, no threshold dose-effect relationship used in estimating effects of radiation.

One limitation in this procedure is that ICRP Publication No. 2 does not separately list dose to bone marrow, which is important in causing leukemia. This was obtained from the ratio of bone marrow to outer bone doses listed in document WASH-1400 (NRC, 1975). For ^{90}Sr, this is 0.26; since r_j for leukemia is $\frac{25}{6}$ times that for bone (from Table III), the ratio of leukemias to bone cancers is $0.26\times\frac{25}{6}=1.08$. Thus the ^{90}Sr in one year's waste after 10 years is enough to cause 4.0×10^{10} bone cancers plus 4.3×10^{10} leukemias, or a total of 8.3×10^{10} fatalities.

Calculations of this type were carried out for all important radioisotopes in the waste as a function of time, and the results are shown in Fig. 9. The solid curve in

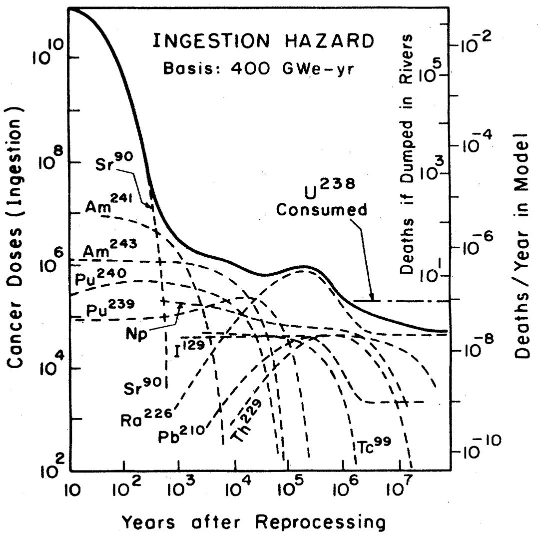

FIG. 9. Number of cancer-causing doses in the waste from 400 GW-yr of nuclear electricity if all of the material were ingested one time by humans in soluble, digestible form. See caption for Fig. 8. The scale inside the right margin shows the number of cancers expected due to ingestion of this material if it were dumped randomly into U. S. rivers. The scale outside the right margin is developed and discussed in Sec. VII.

Fig. 9 is the sum of the curves from the various radioactive nuclides. We see there that the danger for the first few hundred years is from ^{90}Sr; after that it is from Am and Pu isotopes until, after 20 000 yr ^{226}Ra growing in from the decay of ^{234}U becomes predominant.

For inhalation, the calculation is similar. It was assumed that the material inhaled is in insoluble form, and in all cases this makes the lung the critical organ. The MPC in air is based on inhalation at a rate of 7300 m^3 of air per yr to give a dose, d_j, of 15 rem to the lung. Thus, for inhalation,

$$D_{ij}\left(\frac{\text{doses}}{\text{year}}\right)=\frac{Q_i\,(\text{Ci/ton})}{\text{MPC}_{ij}(\text{Ci/m}^3\,\text{A})}\times\frac{dj(\text{rem})}{7300(\text{m}^3\,\text{A})}\times r_j\left(\frac{\text{doses}}{\text{rem}}\right)\times 1.2\times10^4\left(\frac{\text{ton}}{\text{year}}\right)\,, \qquad (2)$$

where m^3 A means cubic meters of air. For ^{90}Sr, $\text{MPC}_{ij}=2\times10^{-9}$ from Table IV, $Q_i=6.0\times10^4$ as before, $d_j=15$, $r_j=39\times10^{-6}$ for lungs from Table III, so the number of deaths expected if this were all inhaled by people is 3.0×10^{10}. Results of this and similar calculations for other isotopes are shown as a function of time in Fig. 10.

In general, the MPC in air is lower for soluble than for insoluble actinides by about an order of magnitude, so it may seem unconservative to consider only insoluble forms. A factor of 3.3 in this difference is made up by the fact that the critical organ for insolubles is the lung, for which the cancer risk/MPC $=r_j\times d_j=39\times10^{-6}/\text{rem}\times15\ \text{rem}=5.9\times10^{-4}$, whereas the critical organ for solubles is the bone, for which the $r_j\times d_j=6\times10^{-6}/\text{rem}\times30\ \text{rem}=1.8\times10^{-4}$. In addition, it is assumed in calculating MPCs that all soluble material deposited in the lung reaches the bone (ICRP, 1959), whereas more recent work shows this to be grossly in error; for most soluble materials, only 10% of that deposited in the lung reaches the bone (ICRP, 1973). Thus the inhalation risk for insoluble material is generally greater than for soluble.

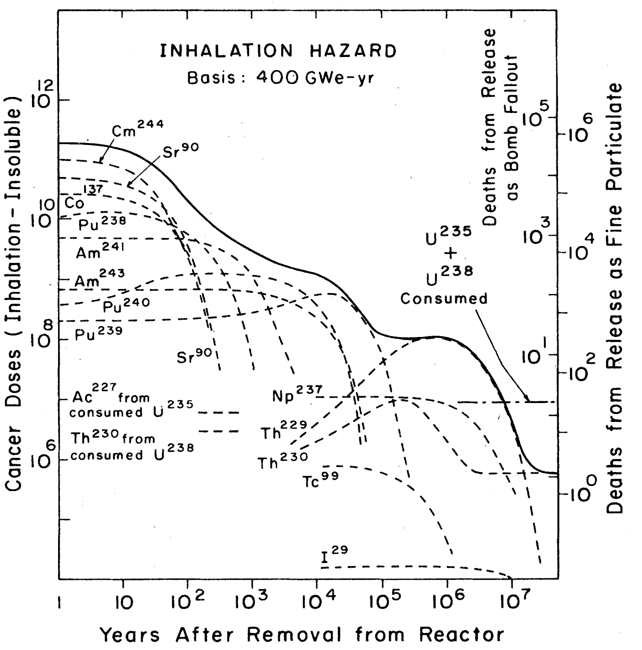

FIG. 10. Number of cancer-causing doses in the waste from 400 400 GW-yr of nuclear electricity if all of the material were inhaled one time by humans as a fine, insoluble particulate. See caption for Fig. 8. The scales on the right side give the effects of releasing the material as bomb fallout, and as a fine particulate dispersed from a point source at ground level from a location in Illinois. The ^{227}Ac from ^{235}U and the ^{230}Th from ^{238}U are the principal components of the dot-dash line for ^{235}U plus ^{238}U burnup.

In Figs. 9 and 10, the ingestion and inhalation hazards are based on cancer induction. In addition, radiation causes genetic effects. In order to calculate these, the transfer of the various radionuclides from the bloodstream to the gonads, the elimination rate from the gonads, etc. would be required. These generally are not well known but estimates have been made for various purposes. It is generally found (NRC, 1975) that the total number of eventual genetic defects is comparable with, but somewhat less than, the number of cancers.

One other property of the waste that has some potential for environmental impact is the neutron emission This arises from two sources, spontaneous fission and (α, n) reactions induced by alpha particles emitted from heavy nuclei. The calculation for spontaneous fission is straightforward as a product of curies, fractional decay by spontaneous fission, and neutrons per fission. The calculation for (α, n) uses an expression developed for alpha emitters in a UO$_2$ matrix (Bell, 1973) inducing reactions in oxygen,

$$\frac{\text{neutrons}}{\text{alpha disintegration}}=1.0\times10^{-10}E_\alpha(\text{MeV})^{3.65}\,. \qquad (3)$$

While this is a crude approximation for use with waste, it should be valid at least as an order of magnitude esti-

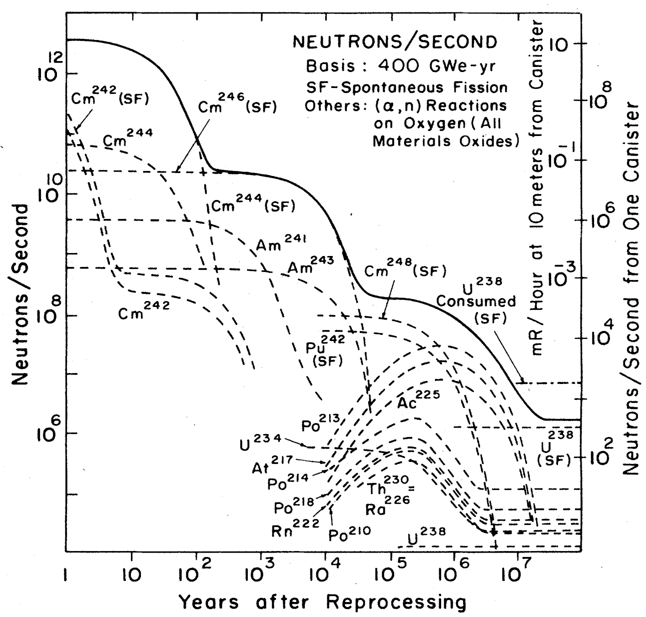

FIG. 11. Neutrons per second emitted by the waste from 400 GW-yr of nuclear electricity. See caption for Fig. 8. SF indicates spontaneous fission sources, and those not so marked are from (α, n) reactions induced by the alpha particles emitted by these nuclei striking oxygen nuclei. The scales on the right give the neutrons per second from a single waste canister and the mR per hour at 10 m from an unshielded waste canister.

mate. The results are shown in Fig. 11. We see that spontaneous fission of curium isotopes is the dominant source for the first few hundred thousand years, after which (α, n) from ^{237}Np daughters assumes importance until 5 million years, when spontaneous fission of ^{238}U takes over.

Before closing this section, it should be recalled that 99.5% of the uranium and plutonium are assumed to have been removed in the reprocessing for burning in other reactors, so the long-term equilibrium situation would be one in which plutonium is used as the fuel in a breeder (LMFBR) reactor. After 500 yr of decay, wastes from an LMFBR would have about an order of magnitude higher potential hazards than those discussed here. For example, the ratio of amounts of a few important isotopes in an LMFBR to those in present day reactors are (Bell and Dillon, 1971): ^{239}Pu–10.8, ^{240}Pu–9.0, ^{242}Pu–9.4, ^{241}Am–10.6, ^{243}Am–2.7; but ^{237}Np–0.27. On the other hand, we will find that ^{90}Sr is the most important contributor to waste hazards for the first 500 yr and only 60% as much of it is produced in an LMFBR. After some tens of thousands of years, we will find ^{226}Ra to be predominant, and it is about equally abundant in LWR and LMFBR wastes. We limit our discussion here to wastes from present day reactors, but almost all aspects of the problem are readily translatable to LMFBR wastes. Plutonium recycle in light-water reactors would give hazards intermediate between the two.

IV. PERSPECTIVE ON POTENTIAL HAZARDS

Toxicologists are quick to point out that potential hazards such as those plotted in Figs. 8–11 are meaningless without a treatment of the possible pathways to man. However, in discussions of environmental impacts of nuclear power, just such potential hazards have often been emphasized. We therefore offer a short discussion of other potential hazards, where "potential" carries essentially the same meaning as in Figs. 8–11. It should be clearly understood, however, that we are comparing only *potential* hazards, which are not necessarily related to actual hazards.

TABLE V. Lethal doses produced or consumed per year in the U. S.

Inhalation (Simmons *et al.*, 1974)

Chlorine—4 $\times 10^{14}$
Phosgene—1.8 $\times 10^{13}$
Ammonia—6 $\times 10^{12}$
Hydrogen cyanide—6 $\times 10^{12}$
Nuclear waste—10 yr = 1.6 $\times 10^{11}$
—500 yr = 5 $\times 10^{9}$

Ingestion (Christensen, 1974)

Barium—9 $\times 10^{10}$
Arsenic—1 $\times 10^{10}$
Nuclear waste—10 yr = 8 $\times 10^{10}$
500 yr = 10^{7}

The most obvious comparisons are with chemical poisons, for which there is a great deal of information, mostly from animal studies but including some human experience. Data on industrially produced chemicals are given in Table V. The largest potential hazards occur if materials are administered in single large doses by inhalation; the chlorine gas produced annually in the U.S. (10^7 tons) is enough to administer an acute lethal dose (1000 ppm min in air inhaled) to 4×10^{14} people, and other gases like phosgene, ammonia, and HCN are not very far behind (Simmons *et al.*, 1974).

Poisoning by ingestion is somewhat less hazardous. To construct the entries in Table V, we have used the values shown in Table VI for the quantities of various elements or compounds which have a 50% chance of being lethal (LD-50) if ingested. The lethality in man is assumed to be derivable from that in animals by scaling in proportion to body weight. We see that the toxicity of the waste is not much greater than that of other common materials even after 10 yr, and after 500 yr it is much less. The values in Table VI are used in conjunction with annual U.S. consumption to derive the ingestion values in Table V. For example, the barium compounds produced annually (1.6×10^6 tons) are enough to provide orally ingested lethal doses (18 g) to 9×10^{10} people.

Poisonous gases like chlorine would have only a limited lifetime in our environment, but that is not true of barium and arsenic. It might be claimed that the barium compounds were present in the ground and all we do is change their form, but about half of our barium and arsenic are imported, so we are indeed introducing them artificially into our environment, and in any case making them more readily available.

A potential hazard in our environment more like radioactive waste is the natural radioactivity in the

TABLE VI. Lethal quantities (LD-50) for various elements if ingested orally (Christensen, 1974). Extrapolation to man assumes 70 kg body weight.

Element	Compound	LD-50(Av) mg/kg	Animal	LD-50 Man (g) (extrapolated)
Selenium	Na_2SeO_3	5	Rabbit, mouse Rat, guinea pig	0.35
(Cyanide)	KCN	10	Rat	0.7
Mercury	$HgCl_2$	23	Rat, mouse	1.6
Arsenic	As_2O_3	45	Mouse, rat	3
Barium	$BaCl_2$, $Ba(NO_3)_2$	250	Rat	18
Copper	CuO, $CuCl_2$	300	Rat	21
Nickel	$Ni(NO_3)_2$	1620	Rat	110
Aluminum	$AlCl_3$ $Al_2(SO_4)_3$	4000	Rat, mouse	280
Nuclear waste				
10 yr				0.03
500 yr				170

ground. If the waste is buried at a depth of 600 m (a typical plan), it might be reasonable to assume that its accessibility to the biosphere is no greater than that of the uranium in the rock and soil in our country down to a depth of 600 m. There are 3.5×10^{16} g of ^{238}U in U.S. soil down to this depth, and the 1.2×10^{10} g of ^{226}Ra in equilibrium with them is 3×10^{13} cancer doses by ingestion (Appendix A), 400 times more than the ingestion hazard in one year's waste at time of burial. If we used nuclear power for hundreds of years, the combined waste would contain only 40 times more radioactivity than one year's waste at burial, because the hazard in this period is dominated by the 28 yr half-life of ^{90}Sr.

If one were to extend this argument to include all the poisons in the top 600 m of the earth's crust, one would obtain far higher potential hazards. For example, the chromium and nickel in the U.S. down to a depth of 600 m is more than 10^{16} lethal doses for each, and that of the aluminum (estimated as chloride or sulfate) is an order of magnitude higher. The combined toxicity of the biological agents in the ground is much larger still.

It should be re-emphasized that the purpose of this section is not to compare *actual* hazards of the waste and that of other things, but rather to show that the *potential* hazards of the waste are not of unprecedented, or even of unusual, magnitude.

V. ROUGH SCOPING OF HAZARDS

The ordinates used on the left sides of Figs. 8–11 are those derived from physical calculations, but they have no direct relevance to environmental impacts. This is obvious for the gamma-ray power and the neutron emission rate, but it is also true for the ingestion and inhalation hazards, as there is no conceivable way in which all of the waste could be ingested or inhaled by people.

In order to give some scope to the potential environmental hazards, we consider here what the consequences would be if the waste were handled in the most careless conceivable manner. Scales corresponding to this are included on the right sides of the figures, and their origin will now be explained. For the gamma-ray hazard we take this most careless handling to be uniform dispersal on the ground throughout the U.S., or what is completely equivalent in view of the linearity hypothesis used in dose-effect calculations, a random dispersal, or arbitrary dispersal with uniform population density. We make the computation for ^{137}Cs, the most important single nuclide, but its 0.66 MeV energy is also typical of the other important nuclides. For ^{137}Cs, one watt of gamma-ray power spread evenly over the U.S. corresponds to 0.032 mCi/km^2 (Appendix B). Using a dose rate of 0.033 mrem/yr to unshielded people per mCi/km^2 of ^{137}Cs on the ground, and a factor for shielding by buildings of 0.4 (UNSCEAR, 1972) then gives an average whole-body dose of 0.44×10^{-6} rem per yr. Multiplying by the 2×10^8 population and 180×10^{-6} cancers per man-rem of whole-body radiation (from Table III) then gives 0.016 deaths per yr in the U.S. for each watt of gamma-ray power lying on the ground. This was used to generate the scale on the right of Fig. 8.

Another scale on the right side of Fig. 8 is useful in assessing the problems in handling waste canisters. The radiation level at 10 m from a source emitting 1W of gamma rays is about 1 rem/h ignoring shielding (the shielding by the waste itself reduces this by about a factor of 3). Exposure anywhere near a fresh canister would result in a lethal dose in an hour or so without shielding.

A scale similar to the above is also included in Fig. 11 for the hazard from neutron emission. It is based on the equivalence of 7.2 neutrons per cm^2 sec to 1 mrem/h (Lamarsh, 1975, page 400). A comparison of these scales for Figs. 8 and 11 indicates that the neutrons are never as dangerous as the gamma rays, and in the early years when handling would be most common, the gamma rays are orders of magnitude more dangerous. We will therefore ignore the neutron hazard.

With regard to ingestion hazard, it is assumed that the most careless credible handling would be to dump the material in soluble form randomly into rivers. The total annual runoff in the U.S. is 1.5×10^{15} l per yr, and the total water ingested is 1.6×10^{11} l per yr (2.2 l/day $\times$ 365 days $\times 2 \times 10^{8}$ population), so the probability for human ingestion of a random sample of river water is about 10^{-4}. However, some fraction of the material is removed by flocculation and filtering processes (UNSCEAR, 1972). For example, rivers contain 0.2 pCi per l of ^{226}Ra (Gera, 1975),[2] whereas tap water in U.S. cities contains typically 0.03 pCi per l (Eisenbud, 1973), so only 1.5×10^{-5} of the Ra in rivers is ingested by people. Since ^{226}Ra is one of the two most important radionuclides in the waste and the other (^{90}Sr) is chemically similar to it, we multiply the scale on the left by 1.5×10^{-5} to obtain our rough scoping scale shown inside the right margin of Fig. 9.

We see from Fig. 9 that if the wastes generated by one year of all-nuclear power were randomly dumped into rivers shortly after reprocessing, almost a million deaths would result from the ingestion of radioactivity with drinking water, but if the dumping were delayed for 500 yr the death toll would be reduced to 150. These consequences of dumping wastes into rivers are five orders of magnitude larger than the consequences of dumping into oceans (Burnett *et al.*, 1976).

From the standpoint of inhalation hazard, perhaps the worst credible handling would be to release all the material as a fine particulate from ground level. Calculations on this sort of release from a point in northern Illinois (upwind from the populous northeastern U.S.) indicate (AEC, 1974a) that about 4×10^{-6} of the material released is inhaled by people. One scale on the right side of Fig. 10 is calculated on that basis. Since the material would never exist as a fine powder, this estimate is somewhat unrealistic. A perhaps more realistic basis would be to assume that the material might be dispersed by a very energetic explosion and thus be released in a manner similar to nuclear bomb fallout. An evaluation can then be made by use of plutonium data from fallout (UNSCEAR, 1972). A typical integrated level of ^{239}Pu in surface air in the temperate zone of the Northern Hemisphere is 4×10^{-15} Ci yr/m^3, so the 2×10^{9} people breathing 7300 m^3/yr of this air have inhaled the product of these, or a total of 6×10^{-2} Ci. The total ^{239}Pu released in bomb tests is 3×10^{5} Ci (AEC, 1974a), so the fraction of the release that has been inhaled is 2×10^{-7}. This factor is used for the other scale on the right side of Fig. 10.

VI. BURIED WASTE: GENERAL CONSIDERATIONS AND TIME DELAYS

From the results of the last section it is clear that the radioactive waste must be isolated from man's environment for at least a few hundred years. At first thought this conclusion appears to be a fearful one, as few things in our experience have longevity of this magnitude. Typical time constants for change in structures and institutions in our society are of the order of a few tens of years, and even most natural formations undergo important changes with similar frequency. However, such time constants are typical only of the environment on the earth's surface; the situation is very different in a deep underground environment where time constants for change are of the order of tens of millions of years. The basic rational for deep underground burial is to benefit from this difference in time constants. But beyond such generalities, any analysis must delve into a study of detailed pathways by which the wastes, once buried deep underground, might be released.

The most widely considered mechanism is that the wastes might be contacted by ground water, be leached into solution, travel through aquifers, and eventually reach surface waters from which they might get into food or drinking water supplies. In order to avoid this possibility, care should be exercised in choosing a burial site to attempt to minimize the chance for ground water to reach the waste. Salt beds offer a measure of security in this regard, as their very existence over time spans like 250 million years is proof that water has not entered them, and in many situations all geological evidence indicates that this situation should continue far into the future.[3] There are several other types of rock that are largely free of water and impervious to it (Schneider and Platt, 1974), including most intrusive igneous rocks like granite, some volcanic rock like tuff, some selected shales, and several types of low silica content metamorphic rocks, especially in the Precambrian shield (large deep mines in this shield rock operate without pumps). While generalities such as these are useful, the particular geology, lithology, and hydrology of an area would be thoroughly explored before it is chosen for use as a burial site.

On the other hand, if ground water does enter the geological formation in which the rock is buried, disaster is not imminent, as there are several important time delays before the waste can reach the surface. The nonporous rock formation in which the waste is encased must be leached or dissolved away, and then the waste-containing glass must be leached before the waste gets into the ground water. Under most circumstances this water moves rather slowly and must travel long distances before coming into contact with surface waters, and the radioactive materials are held up by ion exchange processes so that they move much more slowly than the water. We now consider these time delay processes in some detail.

The first factor is most easily understood for burial in salt; the average time delay before the water reaches the waste is simply the time required to dissolve away half of the salt formation in which it is enclosed. In the site under consideration in New Mexico, if all the ground water now flowing through aquifers above the salt were diverted to flow through it, this time would be

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[2] Gera (1975) gives the average Ra content of rivers as 2×10^{-13} g/l, referencing Tokarev and Sheherbakov (1956) and Heinreich (1958).

[3] Evidence on this for the New Mexico site is reviewed by Claiborne and Gera (1974); the general rationale is discussed in NAS, 1957; NAS, 1961; NAS, 1966; NAS, 1970.

about 30 000 yr for the salt enclosing the waste from one year of all-nuclear power (Appendix C). Even this would saturate the water with salt, making it incapable of further dissolution in downstream areas containing buried waste.

If the burial is in a rock that is not self-sealing like salt, the repository would probably be backfilled with concrete or some other impermeable and nonporous material, and the rock itself would also have these properties. Great care should be taken in the burial to minimize the probabilities of scenarios in which water could enter and have direct access to a large fraction of the waste without first having to leach away a great deal of this surrounding material.

The leach rate for the waste glass is not well understood and there is some controversy among experts on the subject. The choice of glass is based on the ease and cheapness of the conversion, and glass is somewhat less than ideal (Roy, 1976). It is not the most thermodynamically stable form of the material, so given sufficient activation energy, it can devitrify to a crystalline form, and because of the high sodium concentration (from $NaNO_3$ introduced in the chemical processing), the latter may be much more leachable. Moreover, the activation energy may be available from the radioactive heating, and even the glass is much more leachable at the higher temperatures caused by this heating for the first century or so (cf. Figs. 5 and 6).

On the other hand, blocks of waste glass buried in the water table (at ambient temperatures) are leaching at a rate of only about 10^{-8} per yr (Merritt, 1976), and most rock within the water table lasts for millions of years. The glasses in current use do not seem to devitrify below 700 °C, and with current handling plans there should be little difficulty in keeping surface temperatures, at least, well below this (Ross, 1976). In laboratory tests, these glasses are apparently less leachable than nearly all rock (McElroy, 1975), and when they have been allowed to devitrify, their leach rates have increased only by a factor of 4 to 10 (Ross, 1976). Leachability is apparently not affected by radiation damage (Mendel *et al.*, 1976). Leach rates are proportional to surface area, and glasses with waste-containing composition have been found not to suffer greatly increased surface area due to fracturing in the production process (McElroy, 1975). If it should be decided that the glass form is inadequate, there are other forms under development which have improved stability and will not be much more costly to implement (Roy, 1976).

Once the material is leached, the water transport problem is encountered, and it introduces further important delays. A typical speed for water travelling through a deep aquifer is not more than 0.3 m per day (0.1 km per yr)—more than 1 m per day is quite exceptional, and velocities as low as 0.03 m/day are not unusual (Galley, 1972). Water from this depth would have to travel typically 100 km before reaching the surface (Galley, 1972 gives "tens or hundreds of kilometers"). In typical situations, it would therefore take the water into which the waste is leached many hundreds, or even thousands, of years to reach the surface (EB, 1970).

Denham *et al.* (1973) list typical values for ion exchange hold-up factors—the ratio of velocity of ions to velocity of the water—as 10^{-2} for Sr, 10^{-3} for Cs, 10^{-4} for Pu and Am, and 2×10^{-3} for Ra. Both the slow movement of water and the ion exchange hold-up can be bypassed by cracks in the rock. However cracks at these depths are largely sealed by the pressure and by cementing with ground-up rock. If the water is saturated with salt, as would be the case if the waste were buried in salt, ion exchange hold-up would be substantially reduced (Jansen and Burkholder, 1975) and should perhaps not be considered as a time delay factor.

There are at least two situations in which ^{90}Sr has been released into aquifers. In a Canadian experiment (Merritt, 1976), the ^{90}Sr "front" moved 33 m in 11 yr, or 3 m/yr. Our typical rate was 0.3 m/day (water speed)$\times$0.01 (ion exchange hold-up for ^{90}Sr)$\times$365 day/yr$=$1m/yr. This is not necessarily inconsistent, as the "front" presumably moves faster than the bulk of the material, and with shallow burial cracks would be more important. The other situation is in the Idaho Chemical Processing Plant where ^{90}Sr was injected into the Snake River aquifer, an extraordinarily fast flowing one (3.5 m/day) about 150 m below ground. About 1% of the ^{90}Sr migrated 1 km in 10 yr (Robertson and Barraclough, 1973; Barraclough and Jensen, 1976), an average of 100 m/yr, as compared with our estimate for typical ion exchange hold-up as $3.5\times0.01\times365=13$ m/yr. Again this is not inconsistent, as it pertains to only 1% of the material (the bulk of the material has not moved more than a few percent of this distance) and the burial depth is only $\frac{1}{4}$ of the 600 m we have been considering, so transmission through cracks would be more important.

It should be noted that two radionuclides of some importance are not held up appreciably by ion exchange, namely ^{99}Tc and ^{129}I. Neither of these contributes as much as 1% to the hazard during the first 500 yr when time delays are most important. Iodine is physically separated from the other waste, so its handling is really a special problem,[4] and ^{99}Tc never contributes more than 10% to the hazard.

In summary, we have three completely independent factors—burial in a formation expected to be free of water, the leaching time for the surrounding material and for the waste itself, and the travel time for the waste once it is dissolved in ground water—any one of which would ordinarily be sufficient to prevent any appreciable fraction of the waste from reaching surface waters in the first few hundred years when the potential hazards are so large. In fact, our discussions would lead one to believe that even if the waste were contacted by ground water and leached into solution, most of it would not reach man's environment for a million years or so. If this statement seems incredible, one might consider the petroleum which we are now exploiting. It is a liquid with a flow rate considerably faster than that of the waste when ion exchange is considered, but much of it has remained underground for tens of millions of years.

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[4] It will presumably also be buried, but volumes are small and there are no heat or radiation problems in handling, so more security can be provided.

One final guarantee against disaster is the high detectability of radioactivity. There is routine radioactivity monitoring in many rivers now, and it would be extremely easy and cheap to maintain such monitoring in the region of the waste repository.

If the dangers over the first 500 yr can be discounted, it is interesting to put the residual dangers into perspective. From Fig. 9 we see that the waste from one year of all-nuclear power in the U.S. after 500 yr contains 10^7 cancer doses. From Sec. II we calculate the mass of this waste to be 600 kg/canister ×10 canisters/GW×400 GW $=2.4\times10^6$ kg, so the mass per lethal dose is 0.24 kg. The LD-50 for this material would then be about 170 g. From Table VI we see that this makes it two orders of magnitude less toxic than materials commonly found in homes. To have material of this toxicity buried deep underground in a low leachability form does not seem intuitively to be highly dangerous, but we now proceed to quantify its hazard.

VII. RELEASE THROUGH GROUND WATER: PROBABILITY ESTIMATES

Once the waste is buried underground, it seems reasonable to compare it with natural radioactivity in the ground, and as has been shown in Sec. IV, such a comparison makes the hazards from the waste seem mild. A widespread reaction to such a comparison is to point out that the radioactivity in the waste is far more concentrated. However, this concentration is of no importance in estimating the average expected effects so long as the linear, no threshold dose-effect relationship is maintained. The concentration does not increase the probability for any given radioactive atom to find its way into a person, and with the linearity hypothesis only the total number of radioactive atoms in people is relevant. Of course the linear, no threshold theory may well be incorrect, but calculations based on it at least give a valid upper limit on effects to be expected.

In order to quantify the comparison of the waste with natural radioactivity in the soil we introduce calculational models. Burial of the waste will probably be at a depth between 300 and 3000 m (Schneider and Platt, 1974). In the New Mexico salt bed now being considered for a waste repository, the depth would be 600 m so we use that number in our examples. We assume that the waste is buried at random locations throughout the U.S. (contiguous 48 states), but always at a depth of 600 m. We then assume that an atom of waste is no more likely to be released than an average atom of radium or uranium in the rock or soil above it.[5] The reason for the assumption of random burial is that it allows us to use the whole United States as our laboratory. We can estimate the total amount of uranium and radium in U.S. soil down to a depth of 600 m[31]; as noted in Sec. IV, there are 3.5×10^{16} g of uranium and 1.2×10^{10} g of radium. The annual runoff water in the U.S. is 1.5×10^{15} l, and the radium content in rivers is typically 2×10^{-13} g/l (Gera, 1975), so about 300 g of radium is leached per yr. The average probability of leaching is then $300/1.2\times10^{10}=2.5\times10^{-8}$ per yr; the average life of rock in the top 600 m is the inverse of that, or 40 million yr. The Ra/U ratio in rivers is also the equilibrium one, so the leaching probability for uranium per year is also 2.5×10^{-8}. We therefore take this to be the leaching probability for the buried waste. The scale on Fig. 9 for deaths per year if wastes were dumped in rivers is therefore multiplied by 2.5×10^{-8} to obtain an estimate of the deaths per year expected in our random burial model. This corresponds to multiplying the original scale of potential cancer doses $2.5\times10^{-8}\times1.5\times10^{-5}$ (transfer from rivers to people from Sec. V) $=4\times10^{-13}$. This calculation could have been simplified by just noting that the ratio of the total Ra ingested in the U.S. annually with water (0.03×10^{-12} g/l $\times2.2$ l/day $\times365$ day/yr$\times2\times10^8$ population $=4.8\times10^{-3}$ g) is 4×10^{-13} of the Ra in the top 600 m of the U.S. (1.2×10^{10} g).

There is some degree of question in this procedure. In the first place, it ignores the pathway through food, and in the second place it does not take into account the digestibility of the radium which may vary from that assumed in estimating the number of cancer doses in Fig. 9.[6] These difficulties may be bypassed if we go directly from the amount of radium in the soil to the amount of radium in human bone as determined from measurements on corpses. The average bone dosage from ^{226}Ra is 10 mrem per yr (EPA, 1972),[7] which, combined with the U.S. population and the bone cancer risk from Table IV, gives an estimate of 12 bone cancers per year from this source. We have seen in Sec. IV that the number of cancer doses from ^{226}Ra in the top 600 m of U.S. soil is 3×10^{13}, so the ratio between the actual hazard per year and the total potential hazard is $12/3\times10^{13}=4\times10^{-13}$ per yr, in agreement with our previous result. It may be viewed as the fraction of the radium in the top 600 m that is "effectively" ingested annually.

The *total* number of eventual deaths that may be expected via the pathway under discussion from one year of all nuclear power in the model can be obtained by integrating over time the curve in Fig. 9 using the scale on the (outside) right. This integration is, of course, sensitive to its upper and lower limits. Figure 12 gives

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[5] The choice of 600 m as the depth to which we average for the source of radium in the environment is the most unfavorable possible here. If less than 600 m is used, it is clearly unfair to assume that the waste would escape as easily, and if more than 600 m is used, the probability for release of the waste would be reduced.

[6] It was assumed that 3% of ingested Ra ends up in the bone. This is in accordance with ICRP Publications 6 (1962) and 10 (1968), which give values for the most digestible form. Actually, about 1.5×10^{-12} Ci/day $=3.5\times10^{-8}$ Ci/lifetime is ingested, but only 7×10^{-11} Ci ends up in the bone (UNSCEAR, 1972), so transfer from ingestion to bone is only 0.2%, or 15 times less than assumed in these ICRP publications. For uranium, on the other hand, the ratio of that ingested to that in bone is roughly equal to estimates in the ICRP publications.

[7] This number can also be calculated from the quantity in a human body, 36 pCi from UNSCEAR, 1972. Dosage to other organs from ^{226}Ra is relatively negligible. Other radionuclides giving appreciable internal dosage to the body reach man principally by inhalation (Lauder, 1975).

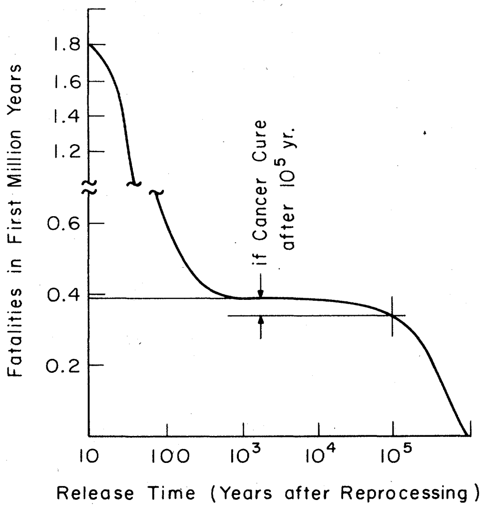

FIG. 12. Integration of Fig. 9 up to 10^6 yr after reprocessing using the scale for our model (outside right margin), as a function of the initial time for the integration. If the discussion in the text of time delays is accepted as precluding release within the first few hundred years, about 0.4 eventual fatalities are expected from 400 GW-yr of nuclear power. If a cure for cancer is found at some time in the future (example shown for 10^5 years), the fatalities obtained using that as the initial time should be subtracted off.

the integration for the first million years as a function of its lower limit, N_1, the number of years before waste first begins to reach man.

In view of our previous discussion of time delays, it would be most unlikely for much of the waste to reach the surface in less than 200 yr, so our result is $\underline{0.4}$ deaths in the first million years. In addition to the lower limit we have been discussing, the integration in Fig. 12 may also have an upper limit, based on a cure being found for cancer. Of course if this cure is found in a number of years less than N_1, there will be no deaths from the wastes. If it is found in 10^5 yr, the number of deaths is reduced by about one order of magnitude.

VIII. CRITIQUE OF PROBABILITY ESTIMATES

A. Use of 4 $\times 10^{-13}$ as annual transfer rate from rock to "effective" ingestion

The transfer per year from average rock to *actual* ingestion for various elements is given as (Cook, 1976)[8]: Sr, 2.7×10^{-11}; I, 2.3×10^{-9}; U, 3.8×10^{-12}; Th, 1.4×10^{-12}; Ra, 1.3×10^{-11}. An alternative estimate gives U, 2×10^{-12}; Ra, 5×10^{-12}.[9] The high values for Sr and I are due to their well known concentration in the food chain so these elements should be given special consideration. Our model should therefore probably not be used in short time periods when ^{90}Sr is very important. Fortunately, ^{129}I is separated from the rest of the waste and handled separately, and this factor should be carefully considered in deciding on its disposal (cf. footnote 4).

However, for the purposes of our integration leading to Fig. 12, we are primarily concerned with Ra and actinides. For Ra and U, the transfer functions are *known* from measurements of quantities in bone to be 4×10^{-13} and 2×10^{-12}, respectively. Since Ra is the predominant contributor to the integration, it is important to use the correct value for it. We might have used a higher value for the other materials, like perhaps the uranium value or something larger. If we had used 2×10^{-12}, it would have increased our result from 0.4 fatalities to 0.8; if we had used the value for Sr, it would have increased it to about 10 fatalities. The reason for not adopting these procedures are the following:

(a) Our value of 4×10^{-13} was shown to be correct for ingestion with water, with all higher values coming from food intake. But food intake is expected to be much less important than water intake for released waste (unlike the situation for natural elements in the soil): plants take their materials from the soil above the water table whereas the released waste would be largely confined to below the water table. Indeed this consideration would substantially reduce the effect of Ra, more than 90% of which is ingested with food, so 4×10^{-13} may even be considerably too high.

(b) The reason why the transfer rate for Ra from rock to *effective* ingestion is smaller than for *actual* ingestion is, as noted previously (footnote 6), that ICRP estimates of how the human body transfers Ra from average food to bone and retains it there are extremely conservative. Such over-conservatism is more a rule than an exception in ICRP estimates, and may well be present for other isotopes; the purpose of ICRP is to set public health limits, and conservatism is clearly justified in that regard.

B. Burial operations introduce release mechanisms that invalidate comparisons with undisturbed rock

This question is really beyond the scope of this paper, and one interpretation of the thrust of this paper may be that this is the problem on which to concentrate. However, we offer what information is available on it.

The problem has been studied mostly with regard to burial in salt; in fact one of the principal advantages of salt is that it flows plastically and seals cracks, so that the waste ends up sealed inside a gigantic crystalline mass. This still leaves the problem of the vertical shaft through which the waste is brought in. This has a miniscule area in comparison with the area of the repository, and it is widely believed that if reasonable care is taken to assure good contact along the sides, this shaft can be satisfactorily plugged with concrete and cement (Gormley, 1976). There is some work in progress on the possibility of using fused rock to better simulate the original geological situation (Gormley, 1976). In any case, since the shaft dead-ends in salt, water leaking in would become saturated, after which

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[8] Estimates are based on annual intakes with food from ICRP Pub. 23 and abundances in rock.

[9] The amounts in the top 600 m of the U. S. are given above as 3.5×10^{16} g of U, and 1.2×10^{10} g of Ra. Average annual ingestion is 10^{-12} g/day for Ra and 1 μg/day for U (UNSCEAR, 1972). The ratios of these numbers must be multiplied by the U. S. population (2×10^8) and the number of days per year. Thus, for Ra, the transfer rate is $10^{-12}\times2\times10^8\times365/1.2\times10^{10}=5\times10^{-12}$.

it would be incapable of further dissolution. Burial in rock other than salt would require considerable research, but it is hoped that backfilling the repository with concrete or some other material would handle the situation.

C. Implied assumption that radioactive waste is no more likely to be leached than average rock

Burial in an area with low probability for water intrusion supports this as does the work in developing a low-leachability form for the waste.

D. Conservatism in the probability estimate

While considering the above criticisms which are aimed at increasing the probability estimate, it seems appropriate to point out some of the conservatism in the estimate that tend to make it high already. Firstly, the model assumes random burial at a depth of 600 m, which includes many areas saturated with water. It would seem that by use of all the experience and knowledge of geologists, lithologists, hydrologists, etc., a far more secure location than random burial may be expected. Secondly, the model assumes that the release probability for an atom of buried waste is equal to that of an average atom of radium in the rock or soil above it. But the latter includes radium near the surface and that is where most erosion takes place. There we have rivers and winds which are both powerful erosive forces, water soaking in, freeze-thaw cycles breaking up the soil, vegetation and animals bringing materials up to the surface, etc. Material buried at 600 m depth is much less likely to be released than average material above it which includes material near the surface.

A third element of conservatism arises from use of the linear, no threshold dose-effect relationship. This is widely conceded to be a conservative procedure, much more likely to overestimate than to underestimate the effects of radiation (NAS, 1972). A committee of leading experts in the field recently recommended a dose reduction factor for low doses (NRC, 1975) which would reduce our results by a factor of 5.

IX. COMPARISON OF HAZARDS FROM WASTE WITH HAZARDS OF URANIUM CONSUMED

In assessing effects of the radioactivity in the waste over very long time periods, it seems reasonable to give credit for the fact that uranium, which also produces long-term effects from its radioactivity, is *consumed* in producing the waste. The magnitude of this credit is shown by the dot-dash lines in Figs. 8–10. From Fig. 9 we see that this saves about 0.1 lives per million years for each year of all-nuclear power, and the rate at which this saves lives exceeds the rate at which the waste takes lives after one million years.

This is based on the premise that the original uranium ore was as securely buried as the waste. The fact that all uranium is not so buried is evident from the natural radon, a uranium daughter, which pervades our atmosphere. This natural radon administers a dose of 150 mrem per yr to the tracheobronchial tree of the average person (UNSCEAR, 1972). Coupling this with the lung cancer dose from Table III and the U.S. population gives an estimated death toll of $0.15\times2\times10^8\times39\times10^{-6}=1200$ per year.[10] One year of all-nuclear power would consume 2.9×10^8 g of ^{238}U, which is 4×10^{-8} of that in the top 100 m (we assume that the ore being mined is from depths uniformly distributed over the top 100 m), so consuming this ^{238}U saves $4\times10^{-8}\times1200=48$ lives per million years in the U.S.

In this treatment we have ignored the fact that about 4% of the uranium mined is left with the ore-processing mill tailings (EPA, 1973), which are piled about 4 m high on the surface, so the amount of uranium within 4 m of the surface is about the same as before the mining. This would seem to cancel out the lives saved from Ra inhalation by burning up uranium. However, in all probability these tailings piles will be effectively covered to inhibit the much larger radon emissions from the radium in the piles (Sears, *et al.*, 1975). One could postulate that covers will erode away, but it would then only be fair to assume that the top 100 m of soil from which the uranium was originally mined would also erode away over some millions of years, so that all uranium rather than just that in the top few meters becomes responsible for radon inhalation fatalities. In either case, there would be 48 net lives saved per million years from burning up the uranium.

A far more important point here is that we have given no credit for removal from the ground of uranium which has been mined and concentrated but not burned up in the reactor; there are 250 g concentrated and purified for every gram burned. It would be very cheap and easy to bury this material deep underground with the wastes, thereby saving $250\times48=12\,000$ lives per million years in the U.S. A much better use would be to burn it in breeder reactors, which would produce an amount of waste comparable with that from the light-water reactors (without producing mill tailings), so the ratio of 48 lives saved per million years from uranium burn-up to 0.4 lives lost due to waste in the first million years, is preserved.

Thus, on a million year scale, nuclear power is a device for *cleansing* the earth of radioactivity. While this may seem strange at first, the underlying logic is clear. Alpha particle emitters are a hundred times more dangerous than beta-gamma emitters where ingestion or inhalation are the principal pathways to man—they have typically 10 times higher energy and 10 times higher biological efficiency (50 times in bone) for doing damage—and every uranium nucleus is destined to decay with the emission of 8 of these alphas. Instead, nuclear power converts it into two beta-gamma emitters (fission products), 80% of which have decayed away before they leave the reactor.

X. RELEASE AS AIRBORNE PARTICULATE

Since the potential hazards due to inhalation (Fig. 10) are considerably larger and more persistent in time than those due to ingestion (Fig. 9), it is important to consider possible mechanisms for release of the buried

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[10] Many estimates are much larger than this. EPA uses 6000 fatalities/year from Ra (letter from W. H. Ellett, dated July 2, 1975), which would increase the result by a factor of 5.

waste as airborne particulate. The first thing that comes to mind is a nuclear bomb explosion, but for the largest bombs yet exploded (50 megaton), the crater depth would be only 340 m and the fracture zone would reach only to 500 m (Claiborne and Gera, 1974), so the waste buried at 600 m would not be affected. There are no publicly announced delivery systems for bombs larger than this size and no evident military reason to justify developing them. Releasing the radioactive waste would clearly not qualify in this regard, as far greater destruction could be wrought by using even a much smaller bomb on a city.

A natural event with the potential for releasing the waste would be the impact of a giant meteorite; one leaving a crater 2 km in diameter would be required to reach a depth of 600 m, and the probability of this has been estimated to be about $2\times10^{-14}/\text{km}^2\,\text{yr}$ (Claiborne and Gera, 1974; Gera and Jacobs, 1972). Since the effective area of such a meteorite crater is on the order of 1 km^2 and the waste occupies a smaller area, the release probability itself is on the order of $2\times10^{-14}/\text{yr}$. Such an event would have many characteristics of a nuclear bomb explosion, including a fireball, so the release would be similar to nuclear fallout.

The average number of fatalities expected from such a release may be obtained by multiplying the integral over time of the solid curve in Fig. 10 (read with the scale on the inside of the right margin) by the probability per year for such a release.[11] The integral works out to 4×10^7 in the first million years, and 12×10^7 if extended to 10^8 years. Multiplying these by 10^{-14} gives an average fatality total of the order of 10^{-7} for each year of all-nuclear power, millions of times lower than through the ground water–ingestion pathway. If there are worries about release through meteorite impact, it should be noted that the maximum number of fatalities from Fig. 10 is 30 000, whereas a direct impact on a city could easily kill millions, and an impact on water would form a tsunami that could easily kill hundreds of thousands.

Another natural release mechanism would be through volcanic action. Areas where volcanism may be expected are well known, and intrusive dykes give further evidence on the volcanic potential for any particular area. For the central plateau of the United States, where the probability is minimal, it is estimated (Bickerman, 1976) to be of the order of $10^{-14}/\text{km}^2\,\text{yr}$. Most of the magma comes up through "pipes" which break through the rock, but a few meters of rock around the circumference of these pipes may be melted and carried up. This cross sectional area is estimated to be about 10^{-3} km^2, so the risk of particular rock being brought to the surface is of the order of 10^{-17} per yr. This danger is therefore three orders of magnitude smaller even than the risk from meteorite impact.

Since these effects are so small, probably the principal mechanism for release as airborne particulate would be secondary effects from the 2.5×10^{-8}/yr released into surface waters, resulting in some of the material's ending up as a dry, finely divided powder on the surface, which may be stirred up to become airborne particulate. Methods have been developed to calculate the resuspension of airborne particulate that has been deposited on the ground (AEC, 1974a) so we use them here. The resuspension coefficient K, defined as g/m^3 resuspended in air per g/m^2 deposited on the ground, is taken initially to be 10^{-5} m^{-1}, and to decrease with a 50 day half-life until it reaches 10^{-9} m^{-1} after something less than two years, and thereafter to decrease with a 10–100 yr half-life[12] as the dust sinks into, and becomes part of, the soil. In this model, the exposure integrated to infinity is as though it were at the initial rate ($K=10^{-5}$ m^{-1}) and extended for 70 (i.e., 50/ln2) days. For each gram deposited randomly over the U.S. (area $=8\times10^{12}$ m^2), the average surface deposit is 1.2×10^{-13} g/m^2, whence the average concentration of dust initially suspended in air is 1.2×10^{-18} g/m^3. In 70 days, the average person inhales 1400 m^3 of air, which then contains 1.6×10^{-15} g of dust, so the 2×10^8 people in the U.S. inhale a total of 3×10^{-7} of the material initially deposited on the ground. Thus the hazard in spreading material on the ground in readily suspendable form is essentially the same as releasing it as bomb fallout (for which 2×10^{-7} is inhaled by people), and may be read on the scale inside the right margin of Fig. 10.

Returning now to our original problem, let us assume that 4% of the material released to rivers ends up spread randomly over the surface of the U.S. as a readily suspendable dust; this is probably an overestimate, as most of the material would sink into river bottoms or banks, or be carried out to sea, and that which does end up on land surfaces would generally be combined with mud or other sediments which would leave very little of it in particle sizes less than about 5 μm, the maximum size for suspension in air and deposition in the lung on inhalation. With this assumption, the probability for waste to end up as a suspendable dust is 0.04 times the 2.5×10^{-8} per yr probability for release into rivers, or 1×10^{-9} per yr. To obtain the eventual fatalities resulting, this must be multiplied by the integral of the curve in Fig. 10, as read from the scale inside the right margin, which we have seen is 4×10^7 in the first million years. The result is 0.04 eventual fatalities via this pathway from the wastes generated by one year of all-nuclear power, still an order of magnitude less than our estimate for the ingestion pathway.

XI. EXTERNAL RADIATION RESULTING FROM RELEASE OF BURIED WASTE

If we assume as in the last section that 4% of the waste released via ground water ends up spread over the surface of the U.S. and remains there for T yr, the effects of external gamma-ray exposure are 2.3×10^{-8} (fractional annual release) $\times0.04$ (fraction on surface) $\times T$ $\times$the integral of the curve in Fig. 8 using the scale on

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[11] If p is the probability/year for release and $E(t)$ is the effect of such a release, the average effect, $\bar{E}$, is

$$\bar{E}=\int pE(t)dt=p\int E(t)dt$$

since we take p to be constant.

[12] The decrease below 10^{-9} was not used in AEC, 1974a, but it is implied by data given therein.

the right side (inside the margin). This integral is 4×10^5 for the first million years, so the total number of deaths is $4T\times10^{-4}$. Thus if T is less than 1000 yr, external radiation is less important than the ingestion pathway. If consideration is extended beyond a million years, the relative shapes of the curves in Figs. 8 and 9 indicate that ingestion becomes relatively still more important.

Most people live in cities where radioactive materials on paved surfaces are rapidly washed away and where new construction frequently covers or replaces surfaces, so T would be no more than a few tens of years. In rural areas radioactive material may be shielded by being washed deep into the soil or by draining away, either of which would probably have time constants of 1000 yr or less. The initial assumption of 4% being spread over the surface is probably pessimistic, as explained in Sec. X. For all these reasons, external radiation seems to be less important than the ingestion pathway.

XII. RELEASE THROUGH HUMAN INTRUSION

Once the repository is sealed, the wastes would not be an attractive target for sabotage. It would take many days of effort with large machinery to remove them, and those working on the project would run grave risks of injury from the radiation. We therefore consider only release through inadvertent human intrusion. Even this possibility would be excluded without the collapse of civilization and social institutions. As long as these remain, the burial site would be remembered, retained in government ownership, and probably kept under surveillance. In addition there are plans for permanent markers explaining the dangers, which it is hoped would outlast lapses in civilization.

Nevertheless, let us assume for the moment that all memory of the burial site is lost. The two most likely modes of intervention would be drilling or mining. Drilling for water to a depth of 600 m would be most unusual, and one of the criteria for selecting a repository is that it be in an area unattractive for oil or other mineral resources. It therefore seems reasonable to assume that the probability for a drill hole in the repository would not be larger than for an average location in the U.S. if the rate of exploratory drilling were equal to that of current "rank-wildcat" oil drilling. Averaged over the U.S., this is 3×10^{-4} drill holes per yr per km^2 (Searle, 1975). An 18 in diameter drill hole removes an area of 2×10^{-7} km^2, so the probability for an atom of waste to be brought to the surface by drilling is the product of these, or 6×10^{-11} per yr. Its subsequent behavior in the environment would depend on a large number of imponderable factors, but a realistic guess might be that its effects are equivalent to those of release into rivers; this release mode would then be almost three orders of magnitude less probable than the natural release to rivers we have been assuming (2.5×10^{-8} per yr). Another possibility is that it would be spread out over the ground in a form which allows some fraction of it to become suspended as airborne particulate, but it was shown in Sec. VIII that this leads to lesser consequences than release into rivers.[13]

If one considers future mining that might take place in the waste repository for unspecified minerals (presumably ones that are not now recognized to be economically interesting), it is difficult to conceive of operations more than five percent as extensive as our current coal mining operation. The latter annually involves 2×10^{-8} of the rock in the U.S. down to a depth of 1000 m,[14] so the probability of the waste's being mined is not more than 10^{-9} per yr. If the effect is equivalent to release into rivers, it is still 20 times less important than the natural release we have assumed.

An important exception to the above argument would be if the waste were buried in salt, which is, after all, a material we now recover by mining. There is enough salt underground in the U.S. for about 25 million yr at current rates of usage, so the probability for any particular small volume to be mined is 4×10^{-8} per yr. Most of this would probably eventually find its way into rivers or other surface waters, so this source would seem to be about equal to what we have assumed for natural releases into surface waters (2.5×10^{-8} per yr). However, the waste released through salt mining would be in insoluble form, and insoluble radioactive material is orders of magnitude less dangerous if ingested than soluble material such as leached waste in natural releases—for the two most important nuclei in Fig. 9, the ratio (ICRP, 1959) is 20 for ^{90}Sr and 600 for ^{226}Ra—so release through salt mining is at least an order of magnitude less important than natural releases.

In reaching this conclusion we have ignored the use of salt in food which provides a direct pathway into man, thereby bypassing the 1.5×10^{-5} probability for materials in rivers to be ingested. However, only one percent of our salt is used for food. Moreover, it is purified by solution techniques allowing ample time for insolubles to settle out (Mulkey, 1975); this should eliminate all but perhaps 10^{-3} of the waste (nearly all of the waste should be in large, glassy, highly insoluble lumps), so it reduces the importance of salt used for foods relative to other salt[15] by a factor of 10^5

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[13] In addition to the effects of wastes brought to the surface, a drill hole would introduce a possible flow path connecting aquifers above and below the burial formation. If the latter is salt, this would cause its dissolution. If all the water normally flowing through a 1 m wide strip were to be diverted to this path, a 100 cm^2 cross section extending vertically through the formation could be dissolved away each year. At this rate, the salt enclosing one year's waste would be dissolved in 5×10^7 yr, but as soon as the cavity opened by the water became much larger than the hole through the impervious formation above the salt, dissolution would stop until plastic flow of the salt closes the hole. The process would thus take much longer than 5×10^7 yr.

[14] About 3×10^8 tons per yr are now obtained from underground coal mining. Assuming that half of the volume removed is coal, there are 1.5 tons of coal per cubic meter mined, so there are 2×10^8 m^3 per yr excavated. The rock in the U. S. to a depth of 1000 m occupies 10^{16} m^3, so 2×10^{-8} of this is mined for coal annually.

[15] In addition to factors discussed here, there are other reasons why mining salt for food is unlikely. The process requires a great deal of water, so mining in arid regions would be unlikely. Salt for food is mined only near population centers to minimize transport costs—the price of salt is so low that transport costs are an important factor.

(0.01×10^{-3}). Hence the two are of comparable importance and still an order of magnitude less important than what we have assumed for natural releases.

As further reinforcement for this conclusion it should be noted that we have ignored sea water as a competing source for salt. In a low technology civilization, sea water would seem to be a much more practical source than mining at 600 m depth, and furthermore such a civilization would not use salt at nearly the current rate. In a high technology society, radioactivity would undoubtedly be familiar, so there is an excellent chance that the problem would be discovered. Moreover, it should be recalled that there would be little difficulty with human intrusion if the burial site is not forgotten.

XIII. REQUIREMENTS FOR SURVEILLANCE

We now consider the question of what type of surveillance would be desirable for a waste repository. While the technology is being developed, of course, close surveillance would be important, but we consider here the situation after the technology is established and the repository is sealed.

It is important to recognize that in the model we used in Sec. VII to obtain our estimate of 0.4 eventual deaths from one year of all-nuclear power, there was no surveillance assumed. No one is watching to see that uranium ore is not getting into streams, and our estimate was based on a comparison with that process. Any surveillance would then be only to decrease the fatality toll below our estimate of 0.4.

If we are willing to place a dollar value on a random human life (average age-55, since cancers occur 15 to 45 yr after exposure), we can use Fig. 9 to estimate the maximum amount it is worth to watch one year's waste. It would be very difficult to justify a figure higher than $10 million per life saved[16]—money spent on medical research, medical care, or public health could easily save one life for each $10 million spent, and there are countless examples in which people submit their own lives to a 10^{-6} risk to save $10, or to a 10^{-5} risk to save $100.[17] If we accept the $10 million figure and use Fig. 9, we find that after 2000 yr, watching might save 10^{-6} lives per yr, so it would not pay to spend more than $10 per yr on the operation. Similarly it would not pay to spend more than $100 per yr after 400 yr, $30 per yr after 600 yr and $1 per yr after a half million yr.

A realistic surveillance program might consist of periodic inspections to maintain warning markers and to prevent deep drilling or mining in the repository area, and periodic water sample collections from nearby streams and wells for radioactivity measurements. A program of this type could maintain surveillance over a thousand years of waste (occupying an area of 500 km^2) with the part-time service of a single employee.

It is often said that watching our wastes will impose a great burden on our progeny. According to our model, the waste will increase their average radiation exposure by one part in 10^{10} for each year of all-nuclear power.[18] It is interesting to compare this with another burden we place on our progeny in consuming most of the world's rich mineral resources within a few generations, including coal, oil, and gas which would be useful feedstock for our progeny in producing substitute materials (plastics, organic chemicals), but which are now being burned up at a rate of millions of tons per day. In comparing the burden of increased radiation with the burden of lack of raw materials it should be kept in mind that the latter can be compensated technologically by cheap and abundant energy, the only source of which we can now guarantee is nuclear power.

XIV. COMPARISON WITH URANIUM MILL TAILINGS

After being mined, the uranium ore is shipped to an ore processing mill where it is concentrated to produce "yellow cake" which is 80% U_3O_8. In this operation, the uranium decay daughters ^{230}Th, ^{226}Ra, ^{222}Rn, etc. are disposed of with the other rock and soil material in large piles referred to as "mill tailings." A typical pile of this sort, formed from producing fuel for 106 reactor-yr of 1000 MW LWR operation, would cover about 250 acres (Sears *et al.*, 1975). It is of considerable interest to make a comparison between the hazards in these mill tailings and those in the waste.

The ingestion and inhalation hazards in the tailings from 400 GW are shown in Fig. 13, where they are compared with the hazards from the waste. We see that for the ingestion hazard, which we have found to be most important for the waste, the mill tailings surpass the waste as a hazard after only 250 yr. Since the waste is much more securely buried than the mill tailings, and is especially secure against release for the first several hundred years, it must be clearly evident that the mill tailings are a far larger potential hazard than the waste.

The rough scoping estimates from Sec. V are included on the right sides of the graphs in Fig. 13. We see that if 0.1% of the tailings become suspended as airborne fine particulate over the next 100 000 yr or so, we may expect about three lung cancers for each year of all-nuclear power, and if that amount is released to rivers, we may expect about one bone cancer from ingestion. It is therefore important that the mill tailings be

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[16] The U. S. Nuclear Regulatory Commission requires expenditures up to $1000 per man-rem of radiation exposure averted (Federal Register 40, 19441, 5/15/75). From Table IV, the cancer risk/man-rem is 180×10^{-6}, so this corresponds to a value of life $= 1000/180 \times 10^{-6} =$ $6 million.

[17] For example, if a life is worth $10 million, riding in an automobile which gives an average risk of 2×10^{-8} per mile, costs 20 cents per passenger-mile. This is twice the cost of air travel, which is considerably safer, so if a family drives somewhere to save airline fare they are effectively saying their lives are not worth $5 million.

Another approach is to recognize that $10 million is equivalent to about 500 man-yr of 40 hr/week labor or 100 man-yr of around-the-clock labor. If this adds 20 yr of life expectancy for the would-be victim, equivalently five people are working around-the-clock to keep this person alive.

[18] In our model the waste causes 0.4 deaths per 10^6 yr. From NAS, 1972, natural radioactivity causes about 3000 fatalities per year, so the waste increases average exposure to our progeny by $0.4 \times 10^{-6}/3000$ or 10^{-10} from one year of nuclear power.

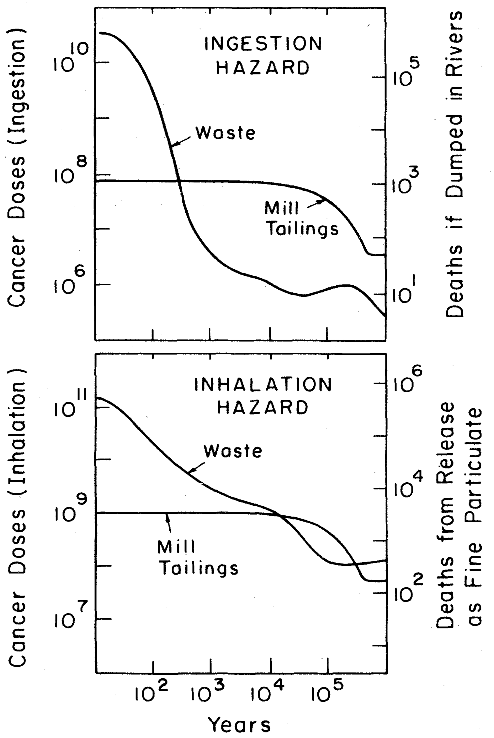

FIG. 13. Comparison of hazards from high-level waste and from uranium ore-processing mill tailings with respect to ingestion and inhalation. The curves for the waste are the solid curves from Figs. 9 and 10.

handled in such a way as to assure against releases greater than this. This may be a matter of some difficulty.

In this discussion, we have been ignoring the effects of the ^{222}Rn escaping from the tailings piles. This problem has been pointed out by others (EPA, 1973; Pohl, 1975) and further emphasizes the fact that uranium mill tailings represent a far greater hazard than the high-level waste. Solutions to the mill tailings problem are discussed in Sears, 1975 (see also Cohen, 1976).

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APPENDIX A: CANCER DOSES FOR INGESTION DUE TO RADIUM IN THE EARTH

The mass of uranium, U, in the U.S. rock and soil to a depth h, is

$$\mathrm{U} = Ahdf ,$$

where

A is the area of the U.S. (excluding Alaska) $= 8\times10^{12}$ m^2

h is the depth considered, in this case 600 m

d is the average density of rock, 2.7×10^{6} g/m^3

f is the fraction of average rock that is uranium, 2.7 ppm (McGraw-Hill, 1971)

Inserting these, $\mathrm{U} = 3.5\times10^{16}$ g.

The equilibrium ratio of radium to uranium, Ra/U, is

$$\frac{\mathrm{Ra}}{\mathrm{U}} = \frac{\mathrm{HL(Ra)}}{\mathrm{HL(U)}} \times \frac{\mathrm{A(Ra)}}{\mathrm{A(U)}}$$

where HL are the half-lives, 1.6×10^{3} yr for Ra and 4.5×10^{9} yr for U; and A are the atomic weights, 226 for Ra, 238 for U. Inserting these gives

$$\frac{\mathrm{Ra}}{\mathrm{U}} = 3.3\times10^{-7} .$$

Multiplying this by the above result for U gives

$$\mathrm{Ra} = 1.2\times10^{10}\ \mathrm{g} .$$

From Table IV, the cancer risk for Ra is 2.5×10^{3}/g (1 Ci = 1 g). Multiplying by the amount of Ra gives 3×10^{13} cancer doses in the top 600 m of the U.S.

APPENDIX B: CONVERSION FROM WATTS OF GAMMA-RAY POWER SPREAD OVER THE U. S. TO mCi/km^2

The power in gamma-ray emission, P, is

$$P = rE_\gamma$$

where

r is the rate of emissions/sec $= \dfrac{3.7\times10^{10}/\mathrm{sec}}{\mathrm{Ci}}$.

Here E_γ is the gamma-ray energy which, for ^{137}Cs, is

$$0.66\ \mathrm{MeV}\times10^{-13}\ \mathrm{joules/MeV} = 1.06\times10^{-13}\ \mathrm{joules}.$$

Inserting these,

$$P = 3.9\times10^{-3}\ \frac{\mathrm{W}}{\mathrm{Ci}} .$$

Thus

$$1\ \mathrm{W} = 2.6\times10^{2}\ \mathrm{Ci} = 2.6\times10^{5}\ \mathrm{mCi} .$$

Averaged over the area of the U.S., $A = 8\times10^{6}$ km^2, one watt is $2.6\times10^{5}/8\times10^{6} = 0.032$ mCi/km^2.

APPENDIX C: TIME TO DISSOLVE AWAY THE SALT ENCLOSING ONE YEAR'S WASTE

The mass of salt m_s is

$$m_s = A_s h_s d_s ,$$

where

A_s is the area covered by one year's waste, 4×10^{5} m^2

h_s is the thickness of the salt formation, 600 m

d_s is the density of salt, 2.2×10^{6} g/m^3

Substituting these gives $m_s = 5.3\times10^{14}$ g.

The volume of water required to dissolve this, V_w, is

$$V_w = m_s/S_s ,$$

where S_s is the solubility of salt, 350 g/l. Inserting these gives $V_w = 1.5 \times 10^{12}$ l.

Ground water flow in the New Mexico region is 3.7×10^4 l/yr per m of width (Claiborne and Gera, 1974). If the area is square, it is 600 m wide, so the total water flow over the area, F is the product of these, or $F = 2.3 \times 10^7$ l/yr.

The time to dissolve away the salt, T_s, is

$$T_s = V_w/F$$

Substituting from above, $T_s = 6 \times 10^4$ yr.

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