POPULATION DOSE ESTIMATES RESULTING FROM
RADIOACTIVE MATERIALS DISCHARGING FROM A
NUCLEAR REPROCESSING FACILITY

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INTRODUCTION AND SUMMARY

The Savannah River Laboratory (SRL) is currently involved as the focal point of an inter-laboratory effort to estimate the environmental effects of operating a nuclear reprocessing facility.¹ As a part of this effort SRL has assumed the responsibility of providing estimates of population exposure resulting from radioactive discharges to the atmosphere. This responsibility does not include contributions from drinking water and food ingestion which are being estimated by Hanford Engineering Development Laboratory (HEDL) and Lawrence Livermore Laboratory (LLL), two of the participating laboratories for the overall environmental effects study. The SRL effort in exposure estimation is confined to contributions resulting from immersion in contaminated air and external beta and gamma dose from exposure to contaminated ground. Population exposure resulting from immersion in contaminated air traditionally have been given more attention. A primary objective of this report is to develop a technique to provide an adequate estimation of population dose resulting from ground depositions and to establish the relative importance of the contributing factors.

The reference facility is assumed to have an operating lifetime of 40 years with annual discharges as indicated in Table I. During this operating period the dose rate from contaminated air is assumed constant, neglecting global recirculation, while the dose rate from ground depositions is continuously increasing due to a buildup of material on the ground. It is assumed that material deposited remains indefinitely and is depleted only by radioactive decay. No credit is taken for shielding due to terrain effects nor for weathering during the accumulation period. The dose estimates resulting from facility operation will be strongly dependent on the population as a function of time during and after the operating period. Since startup time is indefinite a time scaling parameter and a time dependent representation of population is included in the model. In this report it is assumed that the reference facility is located on the Savannah River Plant (SRP) area. Subsequent studies will also be made for other sites.

A 50-year dose accumulation period is being used in estimating population doses in conformance with the general practice of estimating dose commitment for a 50-year period. Results of computations are given in Figures 4 through 9 for two different start-up times, 1980 and 2000. All results pertain to a 200 km square area centered on the Savannah River Plant.

Calculations show that the first year gamma dose from deposition is about 0.11 man-rem which is about 2.4 percent of the 4.4 man-rem resulting from immersion in air. Due to the cumulative effect of depositions, the cumulative gamma dose from deposited material over a 50-year period is about 52 man-rem, and about 25 percent of the cumulative 212 man-rem from air immersion.

The first year beta (skin) dose estimate from deposition is 1.7 man-rem or 13.4 percent of the air immersion beta dose. Cumulative beta dose over a 50-year period is estimated to be 850 man-rem from deposition. This is 125 percent of the cumulative beta dose from air immersion. These calculations tend to show the ground depositions resulting from nuclear reprocessing plants to be a significant factor to the total population exposure from operations.

DISCUSSION

STATEMENT OF THE PROBLEM

During the operating lifetime of a nuclear fuel reprocessing facility there will be small, more or less continuous, releases of radioactive contaminants to the atmosphere. Most of this material (excluding noble gases) will be deposited onto surfaces at or near ground level and will therefore represent a long term exposure potential to residents where the material is deposited. The deposited material will generally be various isotopes having long decay times and, unless there is a significant removal mechanism, deposition will build up during the operating lifetime of the facility. As a result of buildup the exposure rate at any given location increases as a function of time. After operations are terminated the exposure rate becomes a function of radioactive decay. Total population exposure over a given time interval is the product of population and exposure rate (rems) integrated over that time interval. Population is expected to vary significantly over the operating period of the facility and beyond. This variation should therefore be incorporated in the exposure estimation procedure. The objective of this report is to develop an exposure estimation procedure capable of accounting for a dynamic population and allowing flexibility in startup time and length of periods over which exposures are accumulated. The deposition data that were utilized in the development of this report were obtained from the Lawrence Livermore Laboratory (LLL) as part of the AFCT¹ program to define the environmental effects of a fuel reprocessing program. These data pertain to a 200 km square region centered on the Savannah River Plant (SRP).

PROCEDURE

Table 1 lists the assumed effluents from the generic fuel reprocessing facility by individual nuclides. Annual surface deposition rates are then determined over a region of interest by some suitable method. Cumulative population exposure $E(t)$ is then determined by

$$E(t) = \sum_{i=1}^I \sum_{j=1}^J P_j V_i(t) W_{i,j} \quad (1)$$

where

P_j = population of subregion j , people

$W_{i,j}$ = surface deposition of nuclide i in subregion j , Ci/yr

$V_i(t)$ = dose conversion factor for nuclide i , cumulative rem per Ci deposited per year

The dose conversion factors were obtained from Reference 2 for beta and gamma dose.

Figures 1 and 2 show cumulative exposure estimates by evaluating Equation 1 over the above described region using a constant 1970 census population over an assumed operating period of 100 years. In this study it is assumed that the lifetime of the facility is 40 years. Thus, these Figures show a solid line for the first 40 years and a dotted line continuation to show the trend if operations continued. The solid line after the first 40 years represents a continuing dose accumulation from the material deposited during facility operation. These and subsequently presented data assume that after the material is initially deposited it remains in place indefinitely. The data of Figures 1 and 2 will be modified to reflect a changing population base and variable facility operating period. It is immediately obvious from these figures that the cumulative effect of long term operation yields a significant exposure estimate relative to the annual estimate for the first year of operation, about 0.1 man-rem for gamma exposure and about 1.5 man-rem for beta exposure. After only 20 years of operation these values have increased about a hundred-fold to 10.7 and 157 man-rem.

To include the changing population effect and to facilitate the determination of estimates for arbitrary starting times and operating periods it is required to develop a mathematical model of cumulative exposure versus time. The data representing the operating periods of Figures 1 and 2 may be modeled adequately by the hyperbolic expression

$$D_k(x) = \left(a_k x^2 - b_k x + c_k \right)^{1/2} - d_k \quad (2)$$

$k = 1$ for gamma exposure

2 for beta exposure

where

$D(x)$ = cumulative man-rem as a function of time

a_k, b_k, c_k, d_k = fitting coefficients

x = time, yr

The fitting coefficients were obtained by utilizing the data of Table II from which Figures 1 and 2 also were obtained. The circled points of the Figures are calculated from Equation 2 indicating a good fit to the data of Table II. Coefficients for this particular case are given in Table III. Since these data were obtained by using a constant 1970 population base it is necessary to modify Equation 2 to account for population growth as shown in Figure 3. These are projected population changes for 50 years following 1950 as obtained from Reference 3. From these data the projected population change can be expressed as a linear function of time relative to the 1970 population used in the above cumulative exposure data,

$$G(t) = gt - h \quad (3)$$

where $G(t)$ is a growth factor to be applied at time t , and g and h are the linear fitting coefficients. Equation 3 was developed to represent a growth factor rather than the actual population since it must be used to modify the data of Figures 1 and 2 which already reflect a 1970 population. $G(t) = 1.0$ for $t = 1970$. Since it is impossible to have the fuel reprocessing facility operating where the population would equal the 1970 population $G(t)$ will always be greater than unity in the subsequent analyses.

If the cumulative man-rem over any time interval is given as the integral,

$$M = \int_{t_1}^{t_2} M'(t) dt \quad (4)$$

where

M = cumulative man-rem and

M' = accumulation rate for man-rem

This leads to three types of time periods over which the integral of Equation 4 must be performed to properly estimate exposure. In the first case t_2 of Equation 4 is less than the time at which the facility is shut down to terminate operations. Then,

$$M'(t) = G(t) D'(t,s) \quad (5)$$

where $G(t)$ accounts for the variable population and $D'(t,s)$ is obtained as follows to adjust the accumulation period. From Equation 2 let

$$x = t - s \quad (6)$$

where s is the time difference in years between 1970 (the population base year) and the startup time for facility operations. Equation 2 then becomes

$$D(t,s) = \left[a_k t^2 - (2a_k s + b_k) t + (a_k s^2 + b_k s + c_k) \right]^{1/2} - d_k \quad (7)$$

and $D'(t,s)$, the time derivative, is

$$D'(t,s) = \frac{2a_k t - [2a_k s + b_k]}{2 \left[a_k t^2 - (2a_k s + b_k) t + (a_k s^2 + b_k s + c_k) \right]^{1/2}} \quad (8)$$

Equation 5 now becomes

$$M'_1(t) = \frac{2a_k g t^2 - [2a_k g s + b_k g + 2a_k h] t + h [2a_k s + b_k]}{2 \left[a_k t^2 - (2a_k s + b_k) t + (a_k s^2 + b_k s + c_k) \right]^{1/2}} \quad (9)$$

which incorporates both the variable population and the time shifting parameter s which allows a variable starting time. Man-rem for any period during facility operation is given as

$$M_1(t_1, t_2) = \int_{t_1}^{t_2} M'_1(t) dt$$

$$M_1(t_1, t_2) = \left\{ a_k g \left[\left(\frac{t}{2A} - \frac{3B}{4A^2} \right) \sqrt{X} + \left(\frac{3B^2 - 4AC}{8A^{5/2}} \right) \lg Z \right] - \left(\frac{2a_k g s + b_k g + 2a_k h}{2} \right) \left[\frac{\sqrt{X}}{A} - \frac{B}{2A^{3/2}} \lg Z \right] + \frac{h [2a_k s + b_k] \lg Z}{2\sqrt{A}} \right\} \bigg|_{t=t_1}^{t=t_2} \quad (10)$$

where

$$X = At^2 + Bt + C \quad (10a)$$

$$Z = 2\sqrt{AX} + 2At + B \quad (10b)$$

$$A = a_k \quad (10c)$$

$$B = -\{2a_k s + b_k\} \quad (10d)$$

$$C = a_k s^2 + sb_k + c \quad (10e)$$

In the second case it is assumed that the exposure period begins after the facility has terminated operations, $t_1 \geq t_s$. The cumulative man-rem distributions of Figures 1 and 2 no longer follow the hyperbolic curves representing the period when material was being deposited continuously. The cumulative distributions now tend to level off after deviating tangentially from the operating curve. These distributions are also shown on Figures 1 and 2 as solid lines beyond the first 40 years.

The shape of the shutdown curves, as well as the magnitude of the cumulative distribution at shutdown, depends on the length of the operating period. In this analysis it is assumed that the operating period is to be 40 years, the stipulated lifetime of the facility for study purposes. Under this condition it was determined that the shutdown distributions could be adequately represented as a parabolic function.

$$D(t) = \frac{q_k + \sqrt{[q_k^2 - 4p_k r_k - 4p_k s] + 4p_k t}}{2p_k} \quad (11)$$

with time derivative,

$$D'(t) = \left[[q_k^2 - 4p_k r_k - 4p_k s] + 4p_k t \right]^{-1/2} \quad (12)$$

where the p_k , q_k , r_k are the parabolic fitting coefficients.

Applying the population growth factor,

$$M_2'(t) = (gt-h) \left[[q_k^2 - 4p_k r_k - 4p_k s] + 4p_k t \right]^{-1/2} \quad (13)$$

$$M_2(t_1, t_2) = \left[g \frac{(2Rt - 4Q)}{3R^2} - \frac{2h}{R} \right] \sqrt{Q + Rt} \Big|_{t_1}^{t_2} \quad (14)$$

$$Q = q_k^2 - 4p_k r_k - 4p_k s \quad (14a)$$

$$R = 4p_k \quad (14b)$$

It is to be noted that since Equations 10 and 14 represent definite integrals there is no need for integration constants.

The third case is a composite of the first and second cases just described. In this case it is assumed that the accumulation period of interest begins before the termination of operations but carries over into the shutdown period. The exposure over this period is then given as

$$M_3(t) = M_1(t_1, t_s) + M_2(t_s, t_2) \quad (15)$$

where t_s is the time at shutdown.

Population dose resulting from immersion in contaminated air is given as

$$M_a(t_1, t_2) = D_a(1970) \int_{t_1}^{t_2} (gt - h) dt \quad (16)$$

where the subscript a denotes air immersion and $D_a(1970)$ is the annual population dose estimate for the population base year. Upon integration this becomes

$$M_a(t_1, t_2) = D_a(1970) \left[\frac{gt^2}{2} - ht \right]_{t_1}^{t_2} \quad (17)$$

COMPUTATIONAL RESULTS

Figures 4 and 5 are solutions to Equations 10 and 14 as a function of time so that the total exposure over a given period may be evaluated easily. In these and subsequent Figures the 40-year plant lifetime is assumed and a continuous cumulative population dose is computed. To obtain an exposure estimate for any period over the range of these curves it is only necessary to take the difference between the ending and beginning times for the period of interest. In each Figure, curves are shown for two hypothesized startup dates, 1980 and 2000, to show the effect of delayed startup. Figure 4 also contains a reproduction of Figure 3 to show the difference

between assuming a constant population and applying a population growth factor as a function of time. Figures 6 and 7 show cumulative dose from immersion in contaminated air for the same periods as Figures 4 and 5.

For the following comparisons and results a 50-year dose accumulation period was used to estimate population exposures in conformance with the prevailing practice of estimating dose commitment for 50-year periods which corresponds to the average adult lifetime. While it is true that an individual is not irrevocably committed to a 50-year exposure, there is definitely a long term commitment to the surrounding population. This seems to be an equitable means of accounting for total exposure to a population even in a continual state of flux, and provides a more meaningful comparison between air immersion exposure and exposure to deposited material.

Figures 4 and 5 show the effect of accumulating depositions over time becomes very significant with respect to exposure estimates. Assuming startup in 1980, the gamma dose to the population for the first year is only 0.106 man-rem from deposition. This is about 2.4 percent of the one year gamma population dose (4.4 man-rem) from immersion in contaminated air. This represents a relatively minor contribution from deposition and conceivably could be ignored with little loss in accuracy. However, for a 50-year dose, the cumulative dose from deposition for the first 50 years reaches 52 man-rem while the cumulative air dose goes to 212 man-rem. The deposition dose is now about 25 percent of the air dose value and most definitely needs to be taken into account. The maximum deposition dose over any 50-year period would be slightly higher since the contribution during the first few years is less than the accumulation over a comparable number of years after the first 50.

The significance of deposition dose becomes even more apparent for beta exposure estimates. The first year beta dose from deposition is 1.67 man-rem. This is about 13.4 percent of that due to immersion in air (12.5 man-rem). The 50-year beta dose is 850 man-rem from deposition and 680 man-rem from air immersion. Thus, the contribution from deposition is 125 percent of the air immersion value and about 56 percent of the total beta dose. These estimates take into account the assumption that the air immersion dose is accumulated only over the first 40 years while the facility is in operation.

Total external gamma and beta dose estimates are given in Figures 8 and 9. The first 50-year total gamma dose is about 264 man-rem for the 1980 startup and 310 for the year 2000 startup. The total beta dose for the first 50 years is 1540 man-rem and 1795 man-rem for the same startup times. In each of the estimates for total dose the first 50-year period yields a higher dose than any other possible 50-year period since the accumulation rate near startup is greater than that after shutdown.

While the above estimates are intended as a conservative estimate of the environmental effects of facility operations it is interesting to compare these estimates to exposure from natural radiation. Taking a value of 120 millirem as an average annual whole body exposure rate to an individual, the population exposure for the 50-year period beginning in 1980 is estimated to be 8.78×10^6 man-rem. Thus the total body gamma dose from operations (264 man-rem) over the same period is about 0.003 percent of the natural exposure.

REFERENCES

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2. G. G. Killough and L. R. McKay, "A Methodology for Calculating Radiation Doses from Radioactivity Released to the Environment," Oak Ridge National Laboratory, ORNL-4992, March 1976.
3. W. Lerner, "Statistical Abstract of the United States 1973," U.S. Department of Commerce, Social and Economic Statistics Administration.

TABLE I
SOURCE TERM FOR FUEL REPROCESSING PLANT

Capacity: 10 metric ton/day (3000 metric ton/year)
 Burnup: 33,000 megawatt days/metric ton
 Burn Rate: 30 megawatts/metric ton
 Stack Effluent: 10^9 ft³/day
 Cooling Time: 365 days

Nuclide	Form	Atmospheric Radioactive Releases			
		Mass/day inventory (g)	Curie/day inventory (Ci)	Retention Factor	Curie/year released
³ H	HTO	.69*	6.63×10^3	10^2	2.42×10^4
¹⁴ C	CO ₂	.83*	3.65	10^4	1.33×10^{-1}
⁸⁵ Kr		276	1.08×10^5	10^3	3.94×10^4
¹²⁹ I	I ₂	$2.12 \times 10^{3*}$	.346	10^5	1.39×10^{-3}
	CH ₃ I	210*	.034		
¹³¹ I			1.98×10^{-7}		7.23×10^{-12}
¹⁰² Ru	RuO ₄	.654*	$6.20 \times 10^{2**}$	10^8	2.26×10^{-3}
Ru	RuO	813*	$8.20 \times 10^{**}$		2.99×10^{-1}
⁸⁹ Sr			5.47×10^4	10^{10}	2.00×10^{-3}
⁹⁰ Sr			7.75×10		2.83×10^{-2}
⁹⁵ Zr			2.79×10^5	10^{10}	1.02×10^{-2}
^{95m} Nb			5.92×10^3	10^{10}	2.16×10^{-4}
⁹⁵ Nb			5.93×10^5		2.17×10^{-2}
¹³⁴ Cs			1.75×10^6	10^{10}	6.39×10^{-2}
¹³⁷ Cs			1.05×10^6		3.84×10^{-2}
^{137m} Ba			9.82×10^{-5}	10^{10}	3.59×10^{-2}
¹⁴¹ Ce			5.7×10^3	10^{10}	2.10×10^{-4}
¹⁴⁴ Ce			4.56×10^6		1.67×10^{-1}
¹⁴⁴ Pr			4.56×10^6		1.67×10^{-1}
¹⁴⁷ Pm			8.5×10^5	10^{10}	3.1×10^{-2}
²³² U		3.92×10^{-3}	8.39×10^{-2}	10^{10}	3.06×10^{-9}
²³³ U		1.73×10^{-2}	1.64×10^{-4}		5.99×10^{-12}
²³⁴ U		3.17×10^1	1.96×10^{-1}		7.16×10^{-9}
²³⁵ U		7.95×10^4	1.70×10^{-1}		6.21×10^{-9}
²³⁶ U		4.08×10^4	2.59		9.46×10^{-8}
²³⁸ U		9.4×10^6	3.13		1.14×10^{-7}
²³⁷ Np			5.37	10^{10}	1.96×10^{-7}
²³⁹ Np			1.74×10^2		6.36×10^{-6}

TABLE I (Cont'd)

Atmospheric Radioactive Releases					
Nuclide	Form	Mass/day inventory (g)	Curie/day inventory (Ci)	Retention Factor	Curie/year released
^{236}Pu		8.2×10^{-3}	4.36	10^{10}	1.59×10^{-7}
^{238}Pu		1.68×10^3	2.92×10^4		1.07×10^{-3}
^{239}Pu		5.38×10^4	3.30×10^3		1.21×10^{-4}
^{240}Pu		2.17×10^4	4.92×10^3		1.80×10^{-4}
^{242}Pu		3.49×10^3	1.36×10^1		4.97×10^{-7}
^{241}Pu		9.75×10^3	1.09×10^6		3.98×10^{-2}
^{241}Am			3.01×10^3		1.11×10^{-4}
$^{242\text{m}}\text{Am}$			4.01×10^1		1.46×10^{-6}
^{242}Am			4.01×10^1		1.46×10^{-6}
^{243}Am			1.74×10^2		6.36×10^{-6}
^{242}Cm			$6. \times 10^4$		2.19×10^{-3}
^{243}Cm			3.96×10^1		1.45×10^{-6}
^{244}Cm			2.43×10^4		8.88×10^{-4}
Total TRU α					4.57×10^{-3}

* Mass is grams of element

** 3 percent of Ru in off-gas system

TABLE II
 CUMULATIVE POPULATION^a DOSE ESTIMATES FOR THE SRP
 VICINITY RESULTING FROM GROUND DEPOSITION

<i>Years Discharged</i>	<i>Cumulative Gamma Dose</i>	<i>Man-rem Beta Dose</i>
1	0.95	1.49
5	1.36	20.4
10	3.92	56.2
20	10.7	157.
30	19.0	289.
50	39.0	623.
100	100.	1680.

^a 1970 population (1,034,825) for 200 km square area
 centered on SRP

TABLE III
ANALYTICAL FITTING COEFFICIENTS USED
IN POPULATION EXPOSURE MODEL

<i>Data</i>	<i>Coefficient Designation</i>	<i>Value</i>
Gamma Population Dose	a	2.11860×10^0
	b	8.29943×10^3
	c	8.13407×10^6
	d	7.91366×10^1
Beta Population Dose	a	6.41983×10^2
	b	2.52051×10^6
	c	2.47571×10^9
	d	1.3338×10^3
Population	g	1.18227×10^{-2}
	h	2.22906×10^1

FIGURE 1

CUMULATIVE GAMMA POPULATION* DOSE RESULTING FROM GROUND DEPOSITIONS

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CUMULATIVE BETA POPULATION* DOSE RESULTING FROM GROUND DEPOSITIONS

FIGURE 2

FIGURE 3

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FIGURE 4

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FIGURE 5

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FIGURE 6

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FIGURE 7

BETA DOSE TO POPULATION RESULTING
FROM IMMERSION IN CONTAMINATED AIR

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FIGURE 8

TOTAL EXTERNAL BETA POPULATION DOSE

FIGURE 9