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METHODS AND CALCULATIONS FOR REGIONAL, CONTINENTAL, AND GLOBAL DOSE ASSESSMENTS FROM A HYPOTHETICAL FUEL REPROCESSING FACILITY

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ABSTRACT

The Savannah River Laboratory (SRL) is coordinating an interlaboratory effort to provide, test, and use state-of-the-art methods for calculating the environmental impact to an offsite population from the normal releases of radionuclides during the routine operation of a fuel-reprocessing plant. Results of this effort, described in this paper, are the estimated doses to regional, continental, and global populations. Estimates are based upon operation of a hypothetical reprocessing plant at a site in the southeastern United States.

The hypothetical plant will reprocess fuel used at a burn rate of 30 megawatts/metric ton and a burnup of 33,000 megawatt days/metric ton. All fuel will have been cooled for at least 365 days. The plant will have a 10 metric ton/day capacity and an assumed 3000 metric ton/year (82 percent online plant operation) output. Lifetime of the plant is assumed to be 40 years.

* The information contained in this article was developed during the course of work under Contract No. AT(07-2)-1 with the U.S. Department of Energy.

INTRODUCTION

The Savannah River Laboratory (SRL) is actively participating in a current interlaboratory effort to improve calculations of dose-to-man from nuclear facilities. Data bases are being established, models are being tested, and calculations are being made for a hypothetical fuel-reprocessing facility located at several locations.

To calculate the dose to regional and continental populations, you must know the air concentration both with and without deposition. The air concentration with and without deposition accounts for the variety of isotopes and the amount of material deposited on the ground and watersheds around the hypothetical fuel-reprocessing facility. The expertise of several laboratories supported by the U.S. Department of Energy is being used to calculate these concentrations. Lawrence Livermore Laboratory (LLL) is performing regional-scale calculations (out to distances of about 200 km from a model fuel-reprocessing plant). The Air Resources Laboratory (ARL), National Oceanic and Atmospheric Administration, and Battelle Pacific Northwest Laboratories (PNL) are performing the U.S.-scale calculations. ARL is also doing global-scale calculations. Savannah River Laboratory is coordinating the interlaboratory efforts and compiling the results.

The laboratories have now assessed the dose-to-man from a hypothetical facility located on the Savannah River Plant (SRP). The descriptions and results that follow apply to the assessment of dose-to-man from the SRP facility. 1975 meteorological data from a winter period and a summer period were chosen. Using these data, the models were compared to determine which model should be used for annual calculations and to determine if a 200 km square area centered on the site is large enough for dose calculations via the water and food pathways. LLL and the Hanford Engineering Development Laboratory used the deposition data from the models to calculate doses via the water and food pathways. Thus models have been compared, the aerial extent of significant deposition has been determined, and calculated data were compared with observed data. The results follow.

1. The 200 km square area for a site at SRP is considered too small for water and agricultural pathway calculations, because this area contained less than 50% of the deposited material.
2. South Carolina and Georgia will constitute the southeast regional area for water and agricultural pathway calculations.
3. The PNL model will be used for both regional and continental calculations to determine population exposure.

MODEL DESCRIPTION

PNL[1] and ARL[2] models are sequential puff models designed for continental calculations. They determine the air concentration (both with and without deposition) near the surface and also the accumulation of wet and dry deposition on the surface of the ground. Input to these models comes from routine upper-air observations made by the National Weather Service. The LLL model is a regional-scale model (particle-in-cell model, PATRIC). It is a fast running, simplified version of the ADPIC[3] code. Input data are derived from both surface-air and upper-air synoptic data tapes. The SRL[4] model is a regional-scale model and currently provides surface concentrations without depositions. One year's data were used for 50-year dose estimates. All the models used the same meteorological input for two data-rich monthly periods. One of these periods was a winter month (January 26, 1975, through February 25, 1975) and the other was a summer month (June 4, 1975, through July 3, 1975). The release location was a point within the boundaries of the Savannah River Plant. A unit source term (1 Ci/sec) was used in all cases and in the deposition cases, a deposition velocity of 1 cm/sec was used. No comparisons of models were made with the wet deposition cases.

MODEL COMPARISON FOR CALCULATION OF RELATIVE DOSE

Typical dose calculations were performed using output from both PNL and ARL models. Calculations were performed for an actual population distribution and for a unit population assigned to each grid area. With these two types of calculations, it was possible to make some distinction between the effects of a heterogeneous population distribution and the effects of atmospheric dispersion. The average concentration (31 days for the winter case and 30 days for the summer case) times the time interval (the number of seconds in 31 or 30 days) provided the integrated concentration for the period. The integrated concentration was then multiplied by the population within the grid area (0.5° latitude by 0.5° longitude) centered on a grid point. The resulting product was summed over all grids to yield the population-weighted, area-integrated air concentration. The calculated air concentration for the PNL model was then set to 1 for the winter case and the other case values were scaled to the PNL winter case. Table 1 compares the air concentration of the population-weighted data and the unit population data. PNL data in both cases are higher than the ARL data for the same area for summer and winter.

Table 2 is the same type comparison except air concentrations were continuously depleted by deposition. There was no significant difference between PNL and ARL cases when using population-weighted data; when using unit population data, the PNL model calculated higher air concentration than the ARL model. One explanation for this is that population distributions cancel differences in the concentration of the gridded areas at the various grid points.

DEPOSITION COMPARISONS

Deposition comparisons were made for the PNL and ARL grids in a manner similar to the air concentration cases. First we had to make an estimate of how much material was deposited by each model. Table 3 shows what percent of the total emitted was deposited during the entire period. The PNL model yielded higher deposition than the ARL model.

Similar deposition calculations were performed for the LLL regional-area (a 200 × 200 km square area centered on the plant site, a region 2.2° longitude by 1.9° latitude) and a comparable area (2.0° longitude by 2.0° latitude) obtained from the PNL and ARL models. These comparisons are shown in Table 4. The amount of material deposited in the 200 km square was small, so a larger region is being investigated to ensure more complete input to the dose models of agricultural and water pathways. Table 4 also shows these values for the gridded areas within South Carolina and Georgia only.

Dose calculations were needed to account for gamma radiation ("ground shine") from the material accumulated on the ground during the operation of the fuel-recycle plant. Table 5 shows relative surface depositions for the PNL and ARL models as was done for the airborne cases.

ARL AND PNL COMPARISONS

As stated above, ARL model output was consistently lower than similar output from the PNL model. The ARL model was designed to interpret long-range trajectories and to calculate close-in grid values from the individual plumes (calculated, but set to some average value). This "under-calculated" the values at the close-in grid points. The close-in values were, however, the largest on the entire grid; when they were used for dose calculations, differences there dominated the results for the entire field. Figures 1a and 1b are isopleths in powers of ten for the average concentration of the winter case for PNL and ARL models.

COMPARISON WITH KRYPTON-85 DATA

All aspects of the various models were compared to validate them and to determine which model was the best for dose calculation. Our best long-range concentration data were the krypton-85 concentration data obtained by cryogenic samplers located around SRP. The total krypton-85 released during the winter or summer period was assumed to be emitted uniformly over that period. With this assumption and knowing the total amount of krypton-85 released from the plant during the two periods, the model value (air concentration without deposition) of each of the cryogenic stations was compared with the observed value minus the local background (14 pCi/m^3) of krypton-85. The results are shown in Figure 2 (LLL, ARL, SRL, and PNL, respectively). The PNL and ARL values are very close and neither model is superior except that PNL gives slightly higher values for these air concentration cases. There may be a slight preference for ARL since that model is slightly more conservative.

There are at least two sources of errors in this comparison other than inherent model error. One is that krypton-85 is not emitted uniformly, but as a series of puffs throughout the month. A second possible source of error is that occasionally some of the sampling volumes in the cryogenic samplers were unexpectedly decreased. The effect of this error is not known but is under investigation.

POPULATION EXPOSURE (INHALATION AND IMMERSION)

SRL estimated exposure by examining distributions from immersion in contaminated air and by examining external beta and gamma doses from exposure to contaminated ground. Population exposure from immersion in contaminated air, traditionally, has been given more attention. One objective was to develop a technique to adequately estimate population dose resulting from ground depositions and to establish the relative importance of the contributing factors.

The reference facility had a capacity of 10 metric tons/day and a burnup of 33,000 megawatt days/metric ton. The burnup rate was 30 megawatts/metric ton. The stack effluent was 10^9 ft^3/day and the cooling time was 365 days. The hypothetical reference facility at SRP had an assumed lifetime of 40 years. During this operating period, the dose rate from contaminated air was assumed constant, if we neglect global recirculation; meanwhile, the dose rate from ground depositions was continuously increasing due to a buildup of material on the ground. It was assumed that deposited material remained indefinitely and was depleted only by radioactive decay; shielding due to terrain effects and weathering during the accumulation period were not considered. The dose estimates resulting from facility operation were 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 study it was assumed that the reference facility was located on the Savannah River area. Subsequent studies will also be made for other sites.

A 50-year dose-accumulation period was used in estimating population doses to conform with the general practice of estimating dose commitment for a 50-year period. All results pertained to a 200 km square centered on the SRP.

Figures 3 and 4 are estimates of cumulative exposure from depositions 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 was assumed. 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 hypothetical startup dates, 1980 and 2000, to show the effect of delayed startup. Figure 3 also has a curve which shows the dose from a constant 1970 population to compare with curves based on a population growth factor as a function of time.

Figures 3 and 4 show that the effect of accumulating depositions over time becomes very significant for 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 2.4% of the one-year gamma dose (4.4 man-rem) from immersion in contaminated air. This represents a relatively minor contribution from deposition and 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, and the cumulative air dose reaches 212 man-rem. The deposition dose is now 25% of the air dose value and needs to be considered. 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 years.

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 13.4% 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% of the air immersion value and 56% of the total beta-dose. These estimates allow for the assumption that the air immersion dose is accumulated only over the first 40 years of facility operation.

Total external gamma and beta dose-estimates are given in Figures 5 and 6. The first 50-year total gamma-dose is 264 man-rem for the 1980 startup and 310 for the year 2000 startup. The comparable total beta-dose for the first 50 years is 1540 man-rem and 1795 man-rem.

While the above estimates are intended as conservative estimates 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% of the natural exposure.

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TABLE 1

Relative Air Concentrations (No Deposition)

Data Period	*Lab*	*Population Weighted*	*Unit Population*
Winter	PNL	1.00	1.00
	ARL	0.51	0.43
Summer	PNL	0.87	0.88
	ARL	0.65	0.66

TABLE 2

Relative Air Concentrations (With Deposition)

Data Period	*Lab*	*Population Weighted*	*Unit Population*
Winter	PNL	1.00	1.00
	ARL	0.95	0.64
Summer	PNL	0.97	0.96
	ARL	1.01	0.75

TABLE 3

Percent of Releases Deposited

Data Period	*Lab*	*Deposition, %*
Winter	PNL	85
	ARL	53
Summer	PNL	82
	ARL	62

TABLE 4

Percent of Releases Deposited (SRP 200 km Grid)

Data Period	*Lab*	*Grid Size, Deg*	*SRP Region (200 km) Deposition, %*	*GA-SC Region Deposition, %*
Winter	LLL	2.2 × 1.88	35	-
	PNL	2.0 × 2.0	26	38
	ARL	2.0 × 2.0	13	24
Summer	LLL	2.2 × 1.88	18	-
	PNL	2.0 × 2.0	36	60
	ARL	2.0 × 2.0	13	37

TABLE 5

Relative Surface Depositions

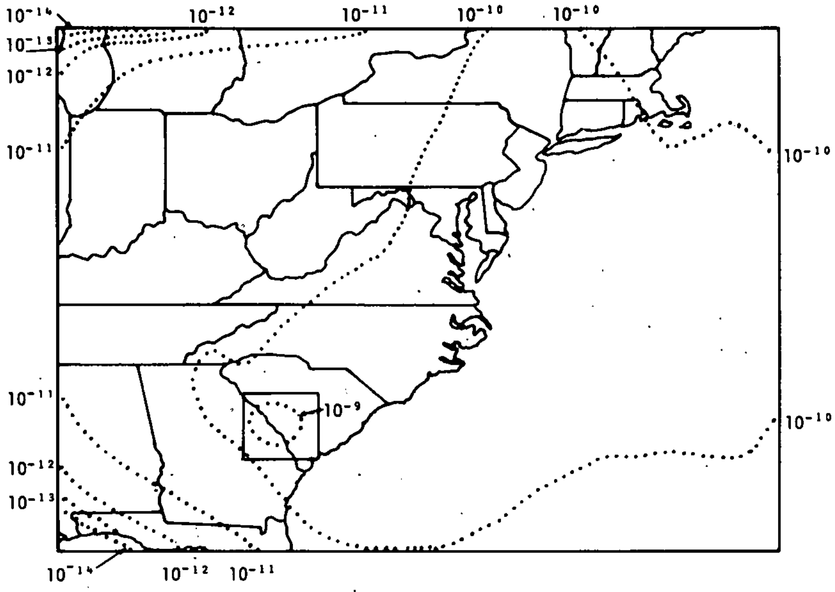
Data Period	*Lab*	*Population Weighted*	*Unit Population*
Winter	PNL	1.00	1.00
	ARL	0.92	0.62
Summer	PNL	0.97	0.96
	ARL	0.98	0.72

TABLE 6

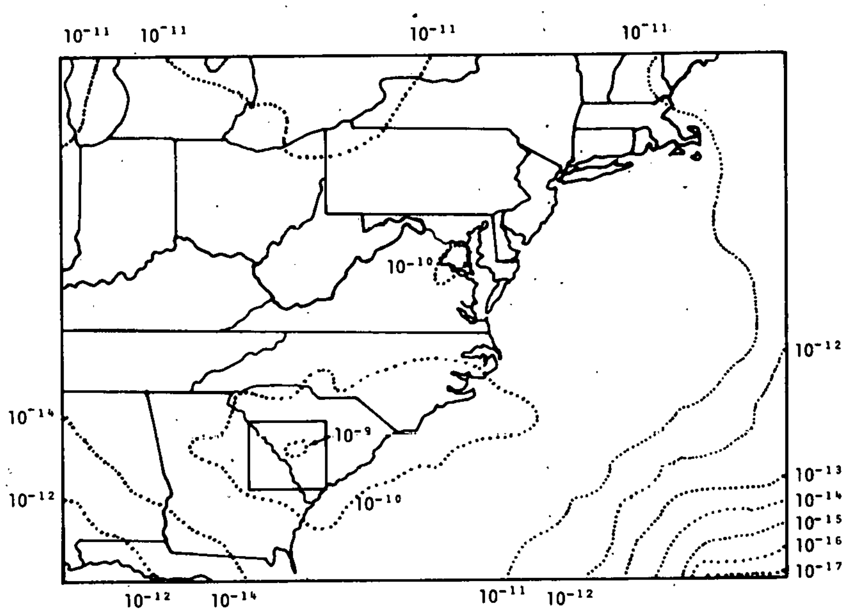
Atmospheric Radioactive Releases

Nuclide	*Curie/year released*
^{3}H	2.42×10^{4}
^{14}C	1.33×10^{-1}
^{85}Kr	3.94×10^{4}
^{129}I	1.39×10^{-3}
^{131}I	7.23×10^{-12}
^{103}Ru	2.26×10^{-3}
^{106}Ru	2.99×10^{-1}
^{89}Sr	2.00×10^{-3}
^{90}Sr	2.83×10^{-2}
^{95}Zr	1.02×10^{-2}
^{95m}Nb	2.16×10^{-4}
^{95}Nb	2.17×10^{-2}
^{134}Cs	6.39×10^{-2}
^{137}Cs	3.84×10^{-2}
^{137m}Ba	3.59×10^{-2}
^{141}Ce	2.10×10^{-4}
^{144}Ce	1.67×10^{-1}
^{144}Pr	1.67×10^{-1}
^{147}Pm	3.1×10^{-2}

Nuclide	*Curie/year released*
^{232}U	3.06×10^{-9}
^{233}U	5.99×10^{-12}
^{234}U	7.16×10^{-9}
^{235}U	6.21×10^{-9}
^{236}U	9.64×10^{-8}
^{238}U	1.14×10^{-7}
^{237}Np	1.96×10^{-7}
^{239}Np	6.36×10^{-6}
^{236}Pu	1.59×10^{-7}
^{238}Pu	1.07×10^{-3}
^{239}Pu	1.21×10^{-4}
^{240}Pu	1.80×10^{-4}
^{242}Pu	4.97×10^{-7}
^{241}Pu	3.98×10^{-2}
^{241}Am	1.11×10^{-4}
^{242m}Am	1.46×10^{-6}
^{242}Am	1.46×10^{-6}
^{243}Am	6.36×10^{-6}
^{242}Cm	2.19×10^{-3}
^{243}Cm	1.45×10^{-6}
^{244}Cm	8.88×10^{-4}
Total TRU α	4.57×10^{-3}

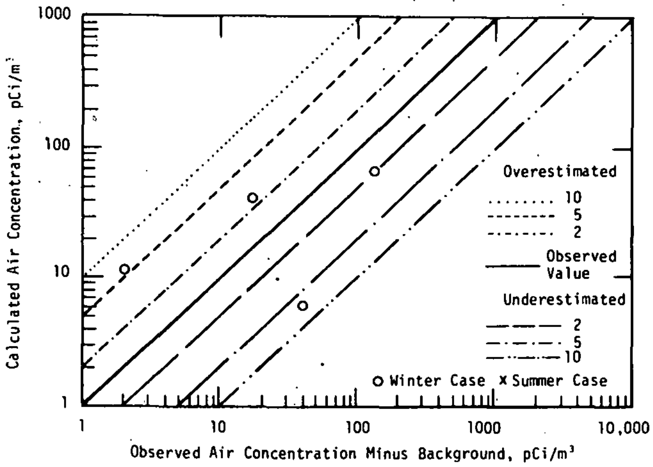


a. Pacific Northwest Laboratory (PNL)

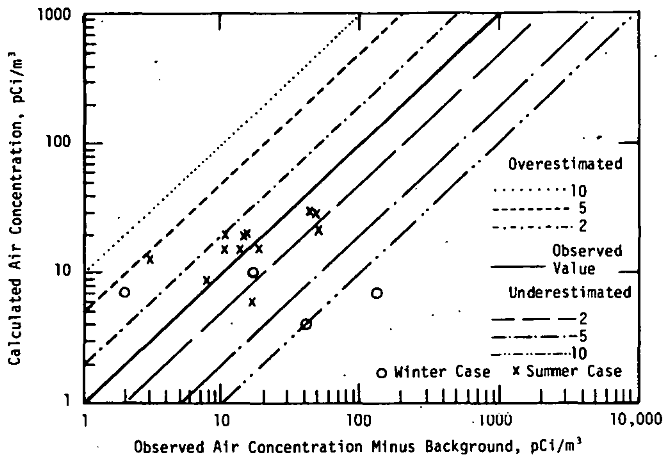


b. Air Resources Laboratory (ARL)

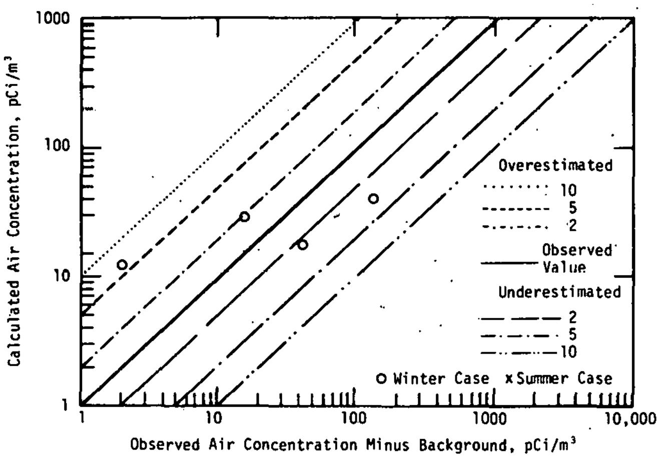
FIGURE 1. Average Air Concentration in Ci/m^3 for Winter Case. (Source Term of 1 Ci/sec.)



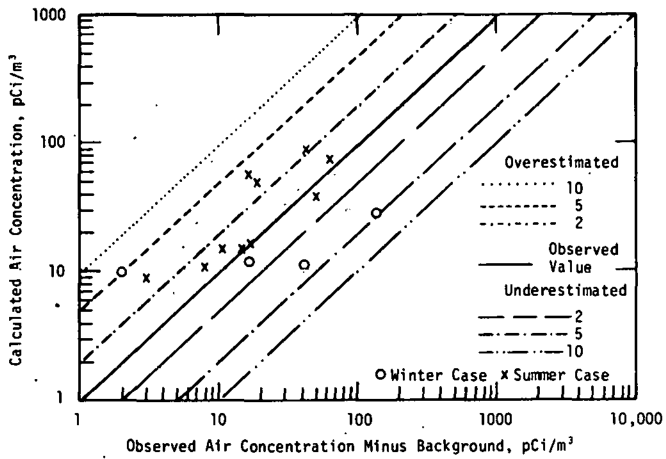
a. Comparison of the Lawrence Livermore Laboratory regional model



b. Comparison of the Air Resources Laboratory model



c. Comparison of the Savannah River Laboratory model



d. Comparison of the Pacific Northwest Laboratory model

FIGURE 2. Comparison of Model Output *versus* Observed Excess Above Background (background was 14 pCi/m^3). The diagonal line shown at 45° from the origin is the case in which the model output exactly equals the observed excess. The three lines above this curve show the model overestimating by a factor of two, five or ten. The three lines below the line show the model underestimating by a factor of two, five and ten.

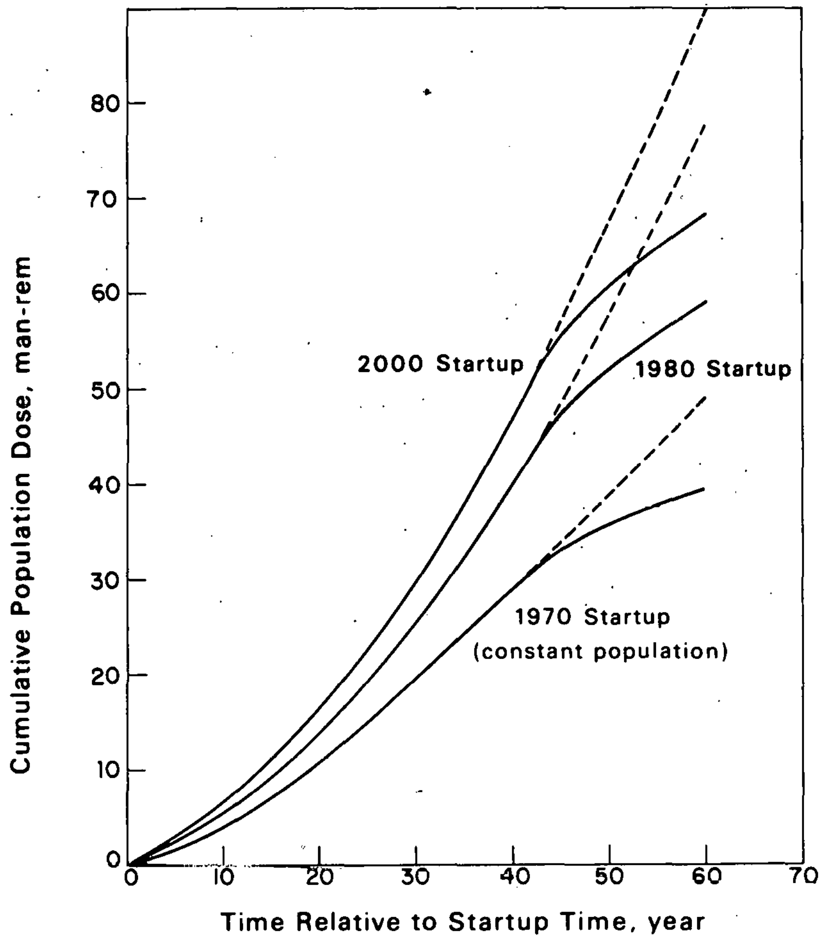


FIGURE 3. Gamma Dose to Population Resulting from Ground Deposition

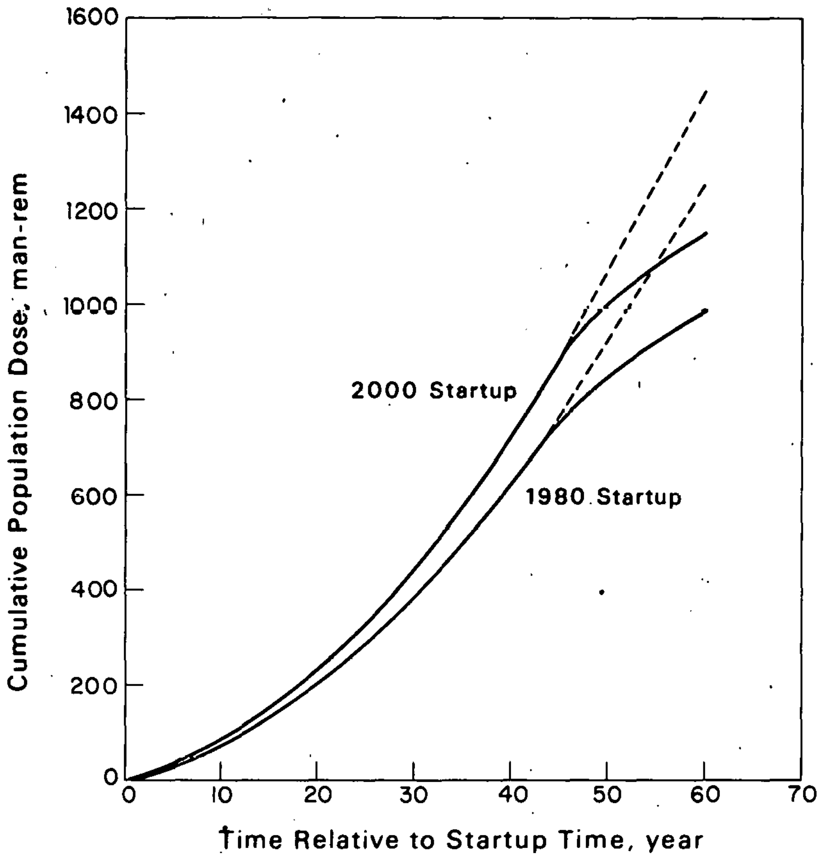


FIGURE 4. Beta Dose to Population Resulting from Ground Deposition

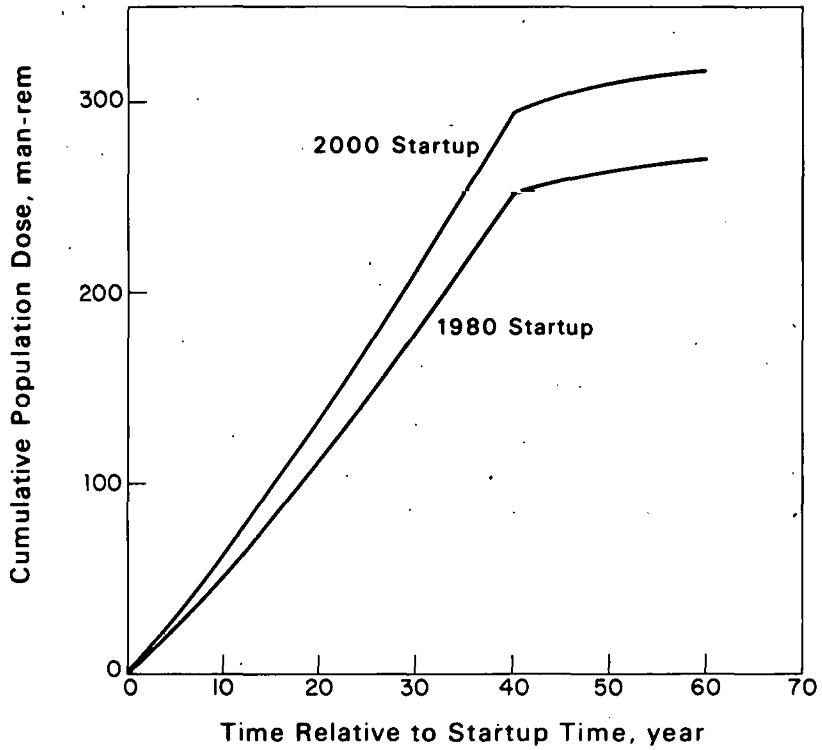


FIGURE 5. Total Gamma Population Dose

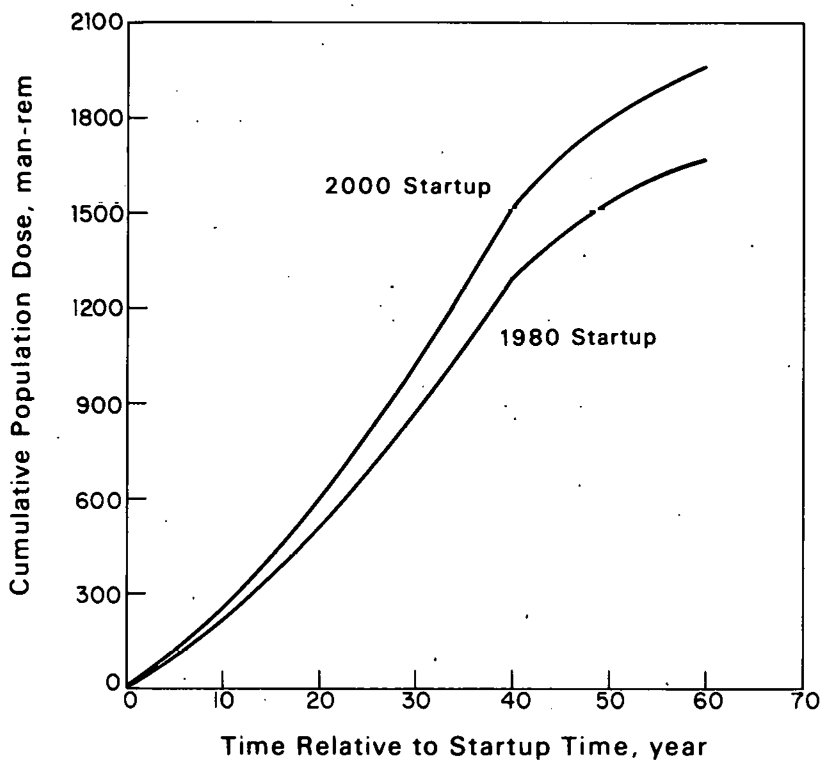


FIGURE 6. Total External Beta Population Dose