

Quantities of Fissile Materials  
in  
US and Soviet Nuclear Weapons Arsenals

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PU/CEES Report No. 168

July 1986

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I. INTRODUCTION

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## I. INTRODUCTION

### History of the Proposal to Cut Off the Production of Fissile Material for Nuclear Weapons

Although the original nuclear weapons control proposals such as the 1946 Baruch Plan<sup>1-1</sup> focused on the control of nuclear weapons materials, recent arms control negotiations have focused principally on the control of long-range nuclear weapons delivery systems. This has been in part due to the shift in the focus of the nuclear arms race from the quantitative to the qualitative. In part also it resulted from the Soviet Union's reluctance to allow intrusive verification arrangements. It is relatively easy with "national technical means," such as cameras mounted on satellites, to locate, identify and count large exposed objects such as intercontinental bombers, submarines under construction and missile silos.

Recently, however, there has been renewed interest in the direct control of nuclear warheads. In part, this has been due to the demonstrated widespread public support for the idea of a "freeze on all further testing, production and deployment of nuclear weapons and of missiles and new aircraft designed primarily to deliver nuclear weapons [emphasis added]."<sup>1-2</sup> In part also, it has been due to a trend toward smaller, mobile, and more ambiguous (multiple-purpose) long-range missiles. Such developments, if they are not quickly blocked by new arms control initiatives, will tend to put into doubt the verification capabilities of national technical systems.

### The Verifiability of a Fissile Cutoff

For all these and other reasons, we have been investigating the verifiability of a possible agreement between the superpowers to cut off the production of plutonium and highly-enriched uranium for nuclear weapons.<sup>1-3</sup> All nuclear weapons contain at least a few kilograms of such chain-reacting "fissile" material. A cutoff in the production of such materials for weapons would therefore limit the number of nuclear warheads which could be produced. Perhaps more importantly, it would lay a basis for verifiable reductions in the superpower fissile material stockpiles.

The US repeatedly proposed a fissile cutoff to the Soviet Union as a separate arms control agreement during the period 1956-1969.<sup>1-4</sup> The Soviet Union rejected the idea - apparently in large part because the US stockpile was much larger at the time.<sup>1-5</sup> More recently, however, in 1982, Soviet Foreign Minister, Andrei Gromyko, announced that the Soviet Union now believed that the "cessation of production of fissionable materials for manufacturing of nuclear weapons" could be usefully made one of the "initial stages" of a "nuclear disarmament program."<sup>1-6</sup>

The Importance of Knowing the Amounts of Fissile Materials in the Nuclear  
Arsenals

The verifiability of a fissile cutoff agreement would depend on the size of the violation that would have to be detectable. One obvious way to judge the significance of an hypothetical clandestine program for the production of fissile materials would be to compare the amounts of material being produced over a given period -- say ten years -- with the amounts already in the superpower weapons stockpiles. This has led us to attempt to estimate these stockpiles sizes from publicly available information. We present our analysis and conclusions in this report.

Enough public information about the US nuclear weapons material production complex is available to allow reasonably accurate estimates of the the amounts of fissile materials in the US weapons stockpile. Similar estimates, to ours have been made independently by Cochran, Arkin and Hoenig.<sup>1-7</sup> Insufficient public information is available to make as good estimates for the Soviet Union. We show below, however, that it is possible on the basis of public information to make a rough upper-bound estimate of the amount of plutonium in the Soviet stockpile. We have been unable to make a similar of estimate the amount of highly-enriched uranium in the Soviet nuclear weapons arsenal. Our estimates for the US stockpiles of weapons fissile materials and for the Soviet plutonium stockpile are summarized in Table 1-1.



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Table 1-1: Amounts of Fissile Material in the US Weapons Stockpile and An Upper-Bound Estimate of the Amount of Separated Plutonium in the USSR

<u>Stockpile</u>	<u>Amount</u> (tonnes)
US Weapon-Grade Uranium	550 +/- 60
US Weapon-Grade Plutonium	90 +/- 7
Soviet Plutonium	<140 +/- 25

## II. THE US STOCKPILE OF WEAPON-GRADE URANIUM

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## II. THE US STOCKPILE OF WEAPON-GRADE URANIUM

### The History of US Uranium Enrichment

The uranium in US nuclear weapons is in the form of "weapon-grade" uranium (WGU) -- i.e. uranium enriched up to a level of 93.5 percent U-235.<sup>2-1</sup> This uranium was produced at the government's complex of three huge uranium isotope enrichment plants at Oak Ridge, Tennessee; Paducah, Kentucky and Portsmouth, Ohio. The annual amounts of "separative work" done by this complex and the percentage of U-235 remaining in the depleted uranium produced by these plants -- their "tails assay" -- are matters of public record. These numbers have been translated into equivalent amounts of WGU in Table 2-1.

Of course, the separative work of the US enrichment complex has gone to the production of more than WGU for nuclear weapons. Indeed, since the mid-1970's, most of this work has gone to the production of low-enriched uranium fuel for nuclear power plants. However, the government has made public the fact that all US weapons uranium was enriched during the period 1944-'64,<sup>2-2</sup> and we will conclude below that during that period the demand for enrichment work for all purposes other than the production of weapons uranium was relatively small. This makes possible a rough estimate of the amount of WGU in the US nuclear weapons stockpile. If all uranium enrichment work prior to the end of Fiscal Year 1964 (July 1, 1964) had gone to the production of WGU and all the feed had been natural uranium, about 775 tonnes of WGU would have been produced. If production had ended six months earlier, the corresponding number would be about 740 tonnes. We therefore assume an uncertainty range of 758 +/- 18 tonnes.

Note that, in Table 2-1, the cumulative US requirements for natural uranium for enrichment alone -- even if all the separative work went to producing WGU (thereby maximizing the ratio of enrichment work to uranium feed) -- was approaching the Atomic Energy Commission's cumulative purchases of uranium in 1956 and incremental demand would exceed incremental supply again in 1956. Cochran et al (2-3) therefore suggest that this would have forced the operators to turn to "stripping" U-235 from stockpiled depleted uranium. The low enrichment tails during these two years also suggests that this is what was taking place. Assuming that all the feed to the enrichment plants was depleted uranium during this two-year period, Cochran et al estimate that the production of HEU would have been reduced by 26 tonnes relative to the production which would have occurred with natural uranium feed. This corresponds to the assumption that, by the end of 1956, the depleted tails associated with all enrichment work done prior to 1956 had been "mined" for its U-235 content down to that year's all-time low operating level of 0.163 percent. We therefore reduce by 13 +/- 13 tonnes the range of uncertainty of our estimate of the WGU-equivalent of the enrichment work done before the end of US production of WGU. Combining errors by taking the square root of the sum of the squares gives an estimate of the WGU-equivalent of the separative work done prior to the end of US production of WGU as 745 +/- 22 tonnes (see Table 2-2).

Some of the enrichment work during the period 1944-'64 did go to other purposes, however. Specifically, the US Atomic Energy Commission supplied enriched uranium fuel for many different types of reactors (plutonium-production, naval-propulsion, electric-power, and research) - both in the US and abroad. In addition, some of the weapons uranium was consumed in nuclear weapons tests. We have therefore made first-order corrections to the estimate above of the amount of weapon-grade uranium in the US stockpile by making rough estimates of the amount of separative work required for each of these purposes (see Table 2-2.) The basis for these estimates is given below.

#### Separative Work Used to Produce Fuel for Naval Reactors

US naval ship reactors are fueled with highly-enriched-uranium (HEU) enriched to 97.3 percent U-235.<sup>2-3</sup> In order to supply this highly-enriched uranium, the US DOD purchased during fiscal years 1983-'85 from the Department of Energy an average of 1.27 million separative work units (SWUs) per year.<sup>2-3</sup> At the currently standard enrichment plant tails assay of 0.2 percent U-235, this amount of separative work would suffice to produce highly-enriched uranium for naval fuel containing about 4.7 tonnes of U-235 per year. Part of this HEU would have been used to provide replacement cores for the refueling of already operating reactors and part would have been to provide initial cores for new reactors.

Cochran et al<sup>2-3</sup> have found statistics on US Navy reactor cores given in Congressional testimony by the former chief of the naval reactor program, Admiral Rickover, that indicates that the Navy buys nuclear reactor cores many years before they are actually loaded. On three dates, Rickover gave the cumulative total numbers of cores that the Navy had procured, the total number of naval reactors, and the cumulative number of refuelings that had been accomplished:<sup>2-4</sup>

- o 5 May 1969: 103 operating naval reactors (including land-based prototypes), 297 cores procured, 66 refuelings accomplished;
- o 25 Feb. 1974: 126 naval reactors, 409 cores, 124 refuelings, 1150 reactor-years of operation;
- o 24 April 1979: 153 reactors, 508 cores, 166 refuelings, 1800 reactor-years of operation.

According to these statistics, the Navy had at these dates the equivalent of 1.24, 1.26, and 1.24 extra cores for each operating reactor. A possible approximate explanation of this pattern is that the Navy orders the first core for a new reactor when the reactor is first ordered and the second core when the first core is loaded and so on. In Table 2-3 we show that the number of refuelings cited by Rickover can be approximately reproduced if we assume this pattern of orders and that cores lasted 5 years prior to 1970, then for 6 years through 1974, and then 8 years subsequently. This means that, in 1983 & 1984, for example, the Navy would have been ordering (see Table 2-3):

- 1) First cores for the new ships being ordered in those years (333,000 shaft-horsepower [shp]),
- 2) Second cores for the nuclear reactors starting up in those years (460,000 shp), and
- 3) Replacement cores for all the ships which were being refueled that year. Assuming an average life of 8 years for reactor cores discharged in 1983 & 1984, the reactors being fueled in those years would have been those started in 1975 or 1976 (170,000 shp) or those being refueled in those years (we assume that approximately one eighth of the average fleet shaft-horsepower that was on line in 1967 & 1968 would have been refueled in 1975 & 1976 (360,000 shp).

On average, therefore, in 1983 & 1984 reactor cores associated with 660,000 shp would have been ordered each year. Assuming a ten-year average core life for modern cores, U-235 for an average of 6.6 million shp-years were ordered in each year. This was the equivalent of about 50 percent more than the shp-years accumulated by the fleet in each of those years because the "inventory" of future shaft-horsepower years carried in the fleet cores was growing in those years due to: i) the growing size of the total installed fleet shp, and ii) the growing lifetime of the cores.

Dividing the average of 4.7 tonnes of U-235 ordered for new naval reactors in these years by 6.6 million shp-years gives 0.7 grams of U-235 required per shp-year. (We neglect the delay between the time that the enrichment work is ordered and the reactor core is provided to the Navy.)

That this is not an unreasonable estimate can be seen if we take into account the following information:

- o One gram of U-235, fissioned completely, would yield about 0.96 Megawatt-days or about 3.5 horsepower-years of energy.
- o The actual fraction\* of the U-235 in the fuel that is fissioned is very roughly 35 percent.

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\* High power research reactors, which use high-enriched uranium in metal fuels similar to that used in naval reactors, have fission fractions of 20-50 percent.

A rough estimate of the average U-235 burnup in naval and research reactor fuel is obtainable from the facts that: i) as of Sept. 1976, high-level wastes originally containing approximately 25 million Ci of Cs-137 plus Sr<sup>90</sup> had been calcined at the Idaho Chemical Processing Plant (ICPP); ii) a volume of liquid wastes approximately equal to that already calcined was in storage; and iii) the annual production of new liquid wastes was approximately 5 percent of that which had been produced. By the end of FY 1980, therefore, there would have been accumulated waste originally containing about 60 million Ci of Cs-137 plus

- o Of the fission heat released, perhaps 20 percent would be converted to mechanical shaft power on average (the peak conversion efficiency of a commercial pressurized-water nuclear power plant is 0.33).

This means that the actual annual average mechanical energy output of the naval reactors would be the equivalent of about  $0.7 \times 3.5 \times 0.35 \times 0.2 = 0.17$  shp per rated shaft horsepower. This low value should not be surprising in view of the facts that naval ships are at sea at only a fraction of the time and, while at sea, travel only infrequently at full speed. (The amount of power required to propel an object through a nonviscous fluid increases roughly as the cube of its speed. Therefore, at half speed, a submarine would only require one eighth as much propulsive power. Due to internal energy requirements and losses, however, the heat output of the reactor would not decrease so rapidly with speed.)

One of the few published sets of numbers which bears on the U-235 requirements of early naval reactors, appears in some 1959 hearings of the US Joint Committee on Atomic Energy.<sup>2-10</sup> There, the Atomic Energy Commission submitted for the record a short list of specifications for two alternative submarine cores which were then under consideration to be offered to France as models for French nuclear submarine power plants. One of these cores contained only slightly-enriched uranium, the other contained highly-enriched uranium, however, and was probably quite similar to contemporary US submarine cores.

The core containing highly-enriched uranium was described as containing 40 kg. of U-235 and, at full power, would "burnup" (fission plus convert to U-236 by nonfission neutron capture) 8 kilograms of U-235 in 6 months. Taking into account the fact<sup>2-5</sup> that only about 80 percent of the "burned up" U-235 would be fissioned, the thermal power output of this reactor at full power would therefore have been 34 Mw or 45,000 horsepower. The core was apparently the same as that provided to Britain, whose submarines have all been reported to be rated at 15,000 shaft horsepower (probably the Westinghouse-designed S5W reactor which powered the first US Polaris submarines.<sup>2-11</sup> The peak thermal-to-mechanical energy conversion efficiency of the system would therefore be 33 percent - somewhat higher than the average conversion efficiency of 20 percent assumed above.

(footnote cont.) Sr-90 -- corresponding to approximately 10 tonnes of U-235 fission (see Table 3-1 for the conversion factors). According to Table 2-4a, by that same date, approximately 18 tonnes of U-235 had been recovered at the ICPP. Comparison of these two numbers (allowing for 20 percent non-fission neutron capture in U-235) results in an estimated average fractional U-235 fission of approximately one third.

There is some evidence that the expected burnups of "various future fuels to be processed at the ICPP [Idaho Chemical Processing Plant]" are higher. The ratio of U-236 to U-235 in the projected high-level waste associated with this fuel is 5.5 when measured in Curies<sup>2-9</sup> or 0.18 when measured in atoms. This would suggest an average burnup of about 50 percent. However, it is noted in the quoted report that "the fuel processed during the first 15 years [pre-1980] contains fewer fission products than [is assumed for future wastes]."



The first US ballistic missile submarine with such a core operated for 4.5 years before refueling.<sup>2-11</sup> If we divided this core life into the estimated 6 month core life of the French prototype reactor at full power, we would obtain an average power output of the US ballistic submarine reactor of 11 percent -- which is to be compared with the 17 percent value estimated above for naval reactors. If we divide the 40 kg. of U-235 in the French reactor's core by 15,000\*4.5 shaft horsepower-years, we get 0.6 grams of U-235 per shaft horsepower-year -- quite close to the 0.7 grams estimated above for modern naval propulsion reactors.

Given an average core life of 5 years prior to 1970, and 0.6 grams of U-235 per rated shaft-horsepower (shp) year, each new reactor core would contain about 3 grams of U-235 per rated shp. (This would give 45 kg. for a 15,000 shp reactor in good agreement with the specifications of the French prototype.)

By 1965, all reactors started in 1955 or earlier (0.015 million shp, according to Table 2-3) would have been fueled three times, all reactors started from 1956 through 1960 (0.485 million shp) would have been fueled twice, and all reactors started from 1961 through 1965 (0.740 million shp) would have been fueled only once. If we, in addition, assume the equivalent of 1.25 extra cores available for every reactor in operation in 1965 (1.24 million shp), the total amount of U-235 that would have been required to be provided for the naval reactors would be:

$$3*(0.015*3 + 0.485*2 + 0.740*1 + 1.24*1.25) = 10 \text{ tonnes.}$$

This would be the equivalent, in terms of separative work requirements, of about 11 tonnes of weapon-grade uranium (WGU). (The equivalence ratio is insensitive to the tails assay.) In Table 2-2, we show a 50 percent uncertainty range on this estimate.

#### Separative Work Used in the Production of Fuel for Research and Civilian Power Reactors

The Atomic Energy Commission and its successors, the Energy Research and Development Administration and the Department of Energy, have supplied enriched uranium fuel for many different types of civilian research and power reactors in the United States and abroad. We divide our estimates of the corresponding reductions of the US stockpile of WGU into two parts:

- i) The reductions due to the demands for enriched uranium through 1964. We assume that the corresponding enrichment work would have been done prior to the end of US production of WGU in FY 1964 and therefore would have reduced correspondingly the enrichment work available for the production of WGU; and
- ii) Non-Weapons Demands for high-enriched uranium later than 1964 which might have been supplied from the stockpile of WGU enriched prior to the end of 1964.

## Demands Prior to the End of Fiscal Year 1964

Table 2-5 summarizes our estimates of the WGU-equivalent of the highly-enriched uranium that was delivered to research, testing and power reactors prior to approximately the end of FY 1964 when the AEC stopped building up its WGU stockpile for weapons. We divide our discussion of these estimates into two parts relating to: i) work required for the production of highly-enriched uranium (HEU, more than 20 percent U-235), and ii) work required for the production of low-enriched-uranium (LEU, less than 20 percent enriched).

Highly-Enriched Uranium. The basis for estimates of the WGU-equivalent of the HEU demands of three classes of US reactors are laid out in the following three tables: research and test reactors with powers greater than 1 MWt (2-5a), civilian power reactors (2-5b), and experimental power reactors (2-5c). There were approximately 70 research reactors with powers less than 1 Megawatt thermal power (MWt) which went into operation prior to 1965.<sup>2-12</sup> Such reactors generally have lifetime cores containing about 5 kg. of HEU. They would therefore account for approximately an additional 0.35 tonnes of HEU.

Weapon-grade uranium was also produced for Project Rover, a joint NASA-AEC program during the '50's and '60's whose purpose was to develop a nuclear propulsion reactor for space travel.<sup>2-12</sup> Most of the rocket experiments were conducted prior to 1965.<sup>2-12</sup> By 1985, the DOE had completed the reprocessing of this only slightly-irradiated fuel and recovered 2.8 tonnes of U-235.<sup>2-13</sup>

Not included in the above Tables are the HEU requirements for research and test, experimental power and remote power reactors built for the US military.<sup>2-12</sup> We estimate that less than one tonne of HEU would have been enriched for this purpose prior to the end of FY 1964.

Finally, the US exported through 1964, a total of 1.9 tonnes of HEU containing 1.6 tonnes of U-235 (see Table 2-5f). This is the equivalent (in terms of separative work) of 1.7 tonnes of weapon-grade uranium.

Low-Enriched Uranium. The demand for low-enriched uranium came principally from commercial power reactors. In the US, virtually all these reactors are either pressurized or boiling light water reactors (LWR's). The US has, until recently, also supplied enrichment services for most foreign reactors of this type outside the Soviet Union and Eastern Europe. Most other power reactors outside the Soviet bloc are graphite- or heavy water-moderated and are therefore ordinarily fueled by natural uranium.

Our estimate of the enrichment work used prior to FY 1965 to provide fuel for US power reactors fueled with low-enriched uranium is given in Table 2-5d. The amount of weapon-grade uranium that could have been produced with the separative work used for this purpose would have been about 9 tonnes (assuming 0.32 percent U-235 in the depleted uranium from

the enrichment plants -- see Table 2-1). Through 1964, the US also exported 185.5 tonnes of low-enriched uranium containing an average of 2.7 percent U-235<sup>2-14</sup>. This low-enriched uranium was the equivalent of 2.6 tonnes of weapon-grade uranium. In total, therefore, during the period prior to fiscal year 1965, separative work sufficient to produce approximately 12 tonnes of weapon-grade uranium was used to produce low-enriched uranium for US and foreign power reactors. We associate with this estimate an uncertainty range of +/- 3 tonnes.

For comparison with the above estimates, we have the AEC Annual Report to Congress for 1964 stating (p. 303) that enriched uranium containing 10.2 and 5.8 tonnes of U-235 had, as of the end of that calendar year, been distributed respectively to "licensed [non-government] users within the United States and to nations having agreements for cooperation with the United States." The 10.2 tonnes for nongovernment users within the US is somewhat less than the estimated 3.0 tonnes of U-235 in highly-enriched uranium plus about 14 tonnes of U-235 in low-enriched uranium we estimate to have been supplied to such users in Tables 2-5. The 5.8 tonnes for foreign users is close to the 1.6 tonnes of U-235 in highly-enriched uranium plus 5 tonnes in low-enriched uranium indicated as exported in ref. 2-14.

Highly-Enriched Uranium Used From the Pre-FY 1965 Stockpile After FY 1964. After the end of the buildup of its stockpile of weapon-grade uranium in 1964, the AEC and its successor agencies withdrew highly-enriched uranium from this stockpile to supply much of the demand for highly-enriched fuel of its own research reactors, some university and other private reactors, and some foreign reactors during the subsequent years. Table 2-5 also shows our estimates of the amount of weapon-grade uranium that were taken from the stockpile for these purposes as of the end of 1984.

From Table 2-5a, it may be seen that from 1965-84 US domestic research and test reactors required about 9 more tonnes of weapon-grade uranium in their fuel that could have been from the pre-1965 stockpile. We assign an uncertainty range of 9 +/- 3 to this number. US exports of weapon-grade uranium to foreign research reactors during this period amounted to approximately 10 tonnes. (This estimate was obtained by subtracting the pre-1965 exports discussed above from the total of 11.5 tonnes of HEU with enrichment above 90 percent [average enrichment of 93 percent] exported through April 1985.<sup>2-14a</sup>)

A significant fraction of this approximately 19 tonnes of WGU equivalent apparently came from the government's stockpile. For example, approximately one half or 700 kg of the highly-enriched uranium exported during the period 1980-83 came from the Y-12 plant at Oak Ridge, Tennessee, -- apparently the location of most of the US stockpile of WGU that is not in weapons (see Table 2-5g). It is likely also, that much of the WGU used in the fuel of domestic research reactors also comes from this stockpile. However, we are unable to make a good estimate of this share. We therefore assume a range of 0-100 percent (see Table 2-5).

### Exports to Britain and France for Military Use

In 1959, the US entered into a barter agreement with Britain to exchange US U-235 in weapon-grade uranium for British plutonium.<sup>2-15</sup> Apparently, exchanges were made under this agreement during the periods 1960-'69 and 1975-1979.<sup>2-16</sup> The total amount of weapon-grade uranium shipped to Britain under this arrangement has been estimated at about 5-6 tonnes.<sup>2-17</sup> A reasonable range of uncertainty would be 6 +/- 3 tonnes. In addition, the US committed itself in 1959 to supply France with up to 0.44 tonnes of weapon-grade uranium " for use in the development and operation of a land based prototype submarine nuclear propulsion plant."<sup>2-15</sup>

### Enrichment Work Used to Produce Fuel for US Plutonium and Tritium Production Reactors

Prior to 1965, some of the separative work of the US uranium enrichment plant went to the production of low-enriched uranium for the production reactors and subsequently some of the weapon-grade uranium produced prior to 1965 was used to fuel the Savannah River production reactors.

Original Eight Hanford Production Reactors: The original Hanford production reactors were started up in the period 1944-55. They were initially fueled with natural uranium being passed once through the reactor. The plutonium was recovered from the irradiated fuel but the uranium, containing perhaps 85 percent of the original U-235 was discarded.<sup>2-18</sup> In the early 1950's, therefore, a new fuel cycle was developed in which the uranium was recovered and recycled several times. In order to maintain the reactivity of the plants, it was necessary to slightly enrich the fresh fuel.<sup>2-18</sup> A plausible assumption would be that the uranium had an original enrichment of 0.9 percent and was recycled four times. Since each pass through the production reactors would reduce this enrichment by about 0.1 percent, if plutonium containing 6 percent Pu-240 were being produced, the average enrichment of the fuel would be about the same as that of natural uranium -- 0.7 percent. During the five pass-throughs, 44 percent of the original uranium would be fissioned.

It is estimated in Table 3-2b below that the original eight Hanford reactors had by the end of calendar year 1964 produced about 43 TWt-days of fission heat. Approximately 90 percent of this fission heat would have come from the fission of 40 tonnes U-235 (most of the remainder from the fission of Pu-239). If 44 percent of the U-235 delivered to the reactors were fissioned, then approximately 10,000 tonnes of 0.9 percent enriched uranium would have been required by the original eight Hanford reactors by the end of 1964. Approximately 56 percent of the U-235 in this uranium would have been fissioned or converted into U-236. [We assume that the remainder is stored somewhere as depleted (0.4 percent U-235) uranium.] At the average plant enrichment tails of about 0.35 percent during that period, the enrichment work required would have reduced the production of weapon-grade uranium during the same period by about 6 tonnes.

Hanford N-Reactor: The ninth and last reactor built at Hanford, the N-reactor, began operation at the beginning of 1964 and was from the beginning fueled by slightly enriched uranium. It too would therefore have required enrichment work to provide its fresh fuel while the US was still producing weapon-grade uranium.

The fresh fuel of the Hanford N-reactor has, until recently at least, had two levels of enrichment: 1.25 and 0.95%.<sup>2-6</sup> Subsequent to its startup period and prior to its recent conversion to the production of weapon-grade plutonium, the fuel of this reactor appears to have operated most of the time at a fuel burnup of 2850 MwD(th)/tonne, at which burnup the irradiated fuel contained<sup>2-20</sup> about 2 kg. of plutonium per tonne of uranium (or about 0.7 grams/MwtD). Assuming that this plutonium averaged 12% Pu-240 and that 2 Pu-239 atoms are fissioned for every one converted into Pu-240, approximately 17 percent of the heat would have come from the fission of Pu-239. Assuming that that 1.25 atoms of U-235 are destroyed for every U-235 atom fissioned, the reduction in the U-235 enrichment in one pass-through of the N-reactor fuel would have been about 0.3%.

According to Table 5-7b, during the years 1964-65, the thermal output of the N-reactor was approximately 0.9 Twt-days. To produce this much heat approximately 300 tonnes of fuel would have been required. Assuming equal amounts of 0.95 and 1.25% enriched uranium and the approximately 0.3 percent enrichment tails of 1963-64, separative work equal to that which would produce approximately 1 tonne of weapon-grade uranium would have been used to produce the fuel for the initial two years operation of the N-reactor.

Savannah River Reactors: According to Cochran et al, the Savannah River production reactors were fueled during their plutonium production runs with either natural or low-enriched uranium until 1968.<sup>2-3</sup> Then the U-235, whose fission generated most of the neutrons and the U-238 which absorbed the neutrons were largely segregated -- with the U-235 in highly-enriched "driver" assemblies and the U-238 in depleted uranium "target" assemblies. According to Table 3-2a, the Savannah River production reactors had through FY 65 produced 27 Twt-days of heat. If we assume, for lack of any better basis, that just as much separative work was associated with a Twt-Day at Savannah River during this period as at Hanford, then an amount of separative work equivalent to that required to produce about 4 tonnes of weapon-grade uranium would have been required. In Table 2-2, we assign a 100 percent uncertainty to this estimate.

The Savannah River reactors were apparently fueled by<sup>2-3</sup> weapon-grade uranium during dedicated tritium-production "campaigns." Cochran et al estimate that<sup>2-3</sup> up to 96 kg of tritium could have been produced in this way through 1964. This would be equivalent to approximately 7 tonnes of weapon-grade plutonium (see Section 3). About 11 tonnes of U-235 would have been fissioned or converted into U-236 in the process. Cochran et al state that the U-235 inventory of one of the Savannah River reactors is 1.6 tonnes and that its residence time in the reactor is 8 months. They also believe that one reactor might have been dedicated to tritium production in 1964. The annual heat output of the Savannah River reactors has averaged

about 0.5 TwtD, corresponding to the fission of about 0.35 tonnes of U-235 in eight months or about 22 percent of the charge. This is not an unreasonable burnup. If we assume two years in the fuel cycle outside of the reactor for every year in the reactor, an additional 4 tonnes of U-235 would have been required to fill the fuel cycle of the reactor that may have been devoted to tritium production in 1964. In this way, we obtain the estimate that up to 17 tonnes of U-235 (19 tonnes of WGU) might have been used to fuel the Savannah River production reactors prior to the end of FY 1964. Allowing for uncertainties in the above estimates, we assume 10 +/- 10 tonnes of WGU.

Cochran et al do not believe that any of the production reactors at the Savannah River Plant were dedicated to tritium production during the period 1965-68. Since 1968, all of the production reactors at the Savannah River production reactors have apparently been fueled entirely with highly-enriched uranium -- independently of whether they were producing tritium or plutonium. This HEU has been obtained from two sources: fresh weapon-grade uranium from the stockpile that was produced prior to 1964 and recycled highly-enriched uranium recovered from irradiated production reactor, naval reactor, and research reactor fuel.<sup>2-21, -22</sup>

According to Table 3-2a, the Savannah River production reactors produced in the period FY 1968-84 approximately 28 Twt-days of heat. About 90 percent of this heat will have been produced by the fission of U-235 at 0.96 TwtD per tonne of U-235 fission. This implies the consumption (80 percent by fission, 20 percent conversion to U-236 -- see Table 3-1 ) of a total of over 33 tonnes of U-235 in these reactors over this period. In addition, if we assume 1.6 tonnes of U-235 in each of the additional three reactors which have been converted to HEU fuel since 1964 and three times as much in a 2-year out-of reactor fuel cycle, an additional 19 tonnes of U-235 would be tied up in the fuel cycle. This gives a total of approximately 52 tonnes of U-235 in HEU required by the Savannah River reactors during the period FY1968-84. We assume an uncertainty of +/- 10 tonnes.

According to Table 2-4a, about 23 tonnes of U-235 were recovered from research and naval reactor fuel at ICPP through FY 1984. In addition, we estimate that an additional 5 +/- 1 tonnes of U-235 have been recovered from research reactor fuel at the Savannah River reprocessing plant and from power-reactor fuel reprocessed by the Nuclear Fuel Services plant (see Table 2-4b). Subtracting the 28 +/- 1 tonnes of recovered U-235 available for recycle into the Savannah River reactors from the 52 +/- 10 tonnes of U-235 that these reactors required during the period 1968-84 means that approximately 24 +/- 10 tonnes of U-235 or the equivalent of 26 +/- 11 tonnes of weapon-grade would have been required from the stockpile to fuel the Savannah River reactors for the period 1968-1984.

### Future Commitments for Research and Production Reactors

The current plan of the Department of Energy is to resume production of highly-enriched uranium metal for weapons and reactors sometime in the period FY1988-90. How much additional HEU will be required to fuel research reactors and the Savannah River reactors during the 5-year period 1985-89? Current requirements for US and foreign research reactors are about 1 +/- 0.5 tonnes per year, of which an unknown portion will be required from the stockpile: we assume 3 +/- 2 tonnes (see Table 2-5g). Assuming that the four Savannah River production reactors operate at a combined level of 2 TwTD per year, they will consume about 2.5 tonnes of U-235 per year. Assuming that 1.5 +/- 0.5 of U-235 in HEU will be recovered annually from spent fuel reprocessed at the Idaho and Savannah River reprocessing plants (the recovery rate at ICPP is projected to increase to almost 1.5 tonnes/yr<sup>2-3</sup>), the net requirements for Savannah River will be 1 +/- 0.5 tonnes/yr for a five year requirement of 5 +/- 3 tonnes.

### Consumption in Nuclear Weapons Tests

The US conducted over 700 nuclear tests in the period 1945-83.<sup>2-25</sup> This corresponds to 3 percent of the number of warheads in the current stockpile.<sup>2-26</sup> We therefore assume that 3 +/- 3 percent of the stockpile estimated in Table 2-2 after subtracting the amounts estimated as being required for reactor fuel. This corresponds to 20 +/- 7 tonnes or 28 +/- 9 kilograms per test.

### Final Estimate

Table 2-2 summarizes the derivation of our estimate of the US stockpile of weapon-grade uranium. As will be seen there, our estimate up to this point is 619 +/- 31 tonnes. This estimate does not allow for process losses or any stockpiles or uses of HEU for nonweapons purposes not accounted for above. If we assume 2 percent losses and an unidentified stockpile of nonweapons HEU of 50 +/- 50 tonnes, then our final stockpile estimate is approximately 550 +/- 60 tonnes. (See Table 2-2.) Cochran et al have gone through a similar exercise and obtained essentially the same estimate. However, they favor<sup>2-3</sup> the value at the lower end of our uncertainty range: 500 tonnes.

## SECTION II. REFERENCES AND NOTES

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Table 2-1. US Uranium Enrichment Operations and Purchases of Natural Uranium as a Function of Time

<u>Year</u> <u>FY<sup>f</sup></u>	<u>Enrichment</u> <u>Work<sup>a</sup></u> (10 <sup>6</sup> kg. SWU)	<u>Tails Assay<sup>b</sup></u>	<u>Weapon-Grade</u> <u>U<sub>3</sub>Equiv.<sup>c</sup></u> (10 <sup>3</sup> kg.)	<u>Equip.</u> <u>Feed<sup>c</sup></u> (10 <sup>6</sup> kg.)	<u>Uranium</u> <u>Purchases<sup>d</sup></u> (10 <sup>6</sup> kg.)
1943			0.08 <sup>e</sup>		
1944			0.1 <sup>e</sup>		3.75
1945	0.05	.005297	.32(0.31) <sup>e</sup>	.17	.39
1946	0.2	.00529	1.27(1.56) <sup>e</sup>	.65	3.16
1947	0.25	.00488	1.53(2.03) <sup>e</sup>	.64	1.36
1948	0.35	.00515	2.20(2.41) <sup>e</sup>	1.05	1.55
1949	0.40	.00506	2.49(2.77) <sup>e</sup>	1.14	1.73
1950	0.50	.00495	3.09	1.34	2.36
1951	0.60	.00506	3.74	1.71	2.84
1952	1.50	.00447	8.83	3.13	2.82
1953	2.00	.00438	11.67	4.00	2.21
1954	4.50	.00366	24.24	6.58	3.57
1955	8.00	.00278	38.34	8.30	4.58
SUBTOTAL				(28.71)	(30.32)
1956	13.70	.00163	53.21	9.11	8.02
1957	14.50	.00199	60.77	11.13	12.37
SUBTOTAL				(48.95)	(50.69)
1958	15.00	.00297	73.90	16.73	20.18
1959	15.50	.00339	80.77	20.34	25.55
1960	16.20	.00337	84.20	21.09	26.50
1961	16.80	.00343	87.99	22.39	24.76
1962	16.00	.00341	83.59	21.16	22.58
1963	15.50	.00313	78.07	18.38	20.77
1964	15.50	.00285	75.06	16.51	14.38
SUB. (156.55)		(.00301)	(775.46)	(185.55)	(205.08)
1965	12.50	.00197	52.19	9.52	11.73
1966	12.50	.00200	52.49	9.64	9.63
1967	10.00	.00200	42.00	7.71	7.54
1968	9.00	.00200	37.80	6.94	6.11
1969	7.00	.00200	29.40	5.40	5.49
1970	6.20	.00200	26.04	4.78	3.09
1971	6.64	.00200			1.00
SUBTOT(220.39)					(249.67)
1972	8.35	.00300			
1973	10.36	.00300			
1974	10.41	.00300			
SUBTOT(249.75)					
1975	11.63				
1976	18.01(15 mo.)				
1977	15.09				
1978	12.55				
1979	13.87				
1980	10.82				
1981	9.62				
1982	9.78				

Table 2-1 -- Notes

a. 1945-1971: James H. Hill and Joe W. Parks, Uranium Enrichment in the United States, (Energy Research and Development Administration, Report #CONF 750324-7, 1975), Fig.1; 1971-1980: Uranium Enrichment, 1980 Annual Report (US DOE, Oak Ridge Operations Office); and 1981-1982: ibid (1982).

b. Cochran, Thomas B., Arkin, William M. and Hoenig, Milton M., Nuclear Weapons Databook, Volume II: The Production Complex (Cambridge, Mass: Ballinger, to be published).

c. These values were calculated assuming that weapon-grade uranium is 94 percent U-235. The changes resulting from switching to 93.5 % were not thought sufficient to justify recalculation. The following formulae relating separative work units (D), tails assay ( $x_w$ ), product quantity (P) enrichment ( $x_p$ ) and feed (F) were taken from Gaseous Diffusion Plant Operations, (USAEC, #ORO-684, 1972), Appendix 2:

$$F/P = (x_p - x_w)/(x_F - x_w), \text{ and}$$

$$D/P = [V(x_p) - V(x_w)] - (F/P)*[V(x_F) - V(x_w)]$$

$$\text{where } V(x) = (2*x - 1)*\ln[x/(1-x)].$$

For 94 percent enriched product we have  $V(0.94) = 2.421$  and, for natural feed, we have  $V(0.00711) = 4.869$ . Consequently, we have, for various values of  $x_w$ :

$x_w$	$V(x_w)$	F/P	D/P
0.00529	5.184	513.6	157.6
0.00335	5.657	249.2	192.9
0.00300	5.771	228.0	202.2
0.00280	5.842	217.4	207.9
0.00275	5.861	214.9	209.3
0.00200	6.188	183.6	238.1
0.00163	6.397	171.2	257.7

Other values of  $V(x_p)$  used in this chapter are:  $V(0.0125) = 4.26$ ,  $V(0.0095) = 4.559$ ,  $V(0.009) = 4.617$ , and  $V(0.973) = 3.391$ .

d. Statistical Data of the Uranium Industry, [US AEC, GJO-100(74), 1974], pp.9, 11, except for the years: 1944 (Hewlett, Richard G. and Anderson, Oscar E., A History of the United States Atomic Energy Commission I: The New World (1939-1946), (US AEC, Report # WASH-1214, 1972). pp. 291-292: Congo, 2850; Canada, 300; US, 600); 1948-1952 (Hewlett, Richard G. and Duncan, Francis, A History of the United States Atomic Energy Commission II: Atomic Shield (1947-1952) (US AEC, Report # WASH-1215, 1972) p. 674); and 1945-1947 and 1953-1955 when the import numbers are from an unpublished report of Robert Pitman Division of Uranium Resources and Enrichment, Energy Research and Development Administration (DOE) via Mike Lopez (Oakland ERDA Office) and Kirk Smith.

(Table 2-1, Notes-cont.)

e. Data for 93.15 percent enriched uranium production obtained in 1980 from the US DOE and reprinted by Peter Vogel in "The Last Wave From Port Chicago," The Black Scholar Spring 1982, p. 30.

f. Fiscal years 1971-'76 (and we assume previous ones as well) began on July 1 of the previous calendar year. Fiscal year 1976 ended on September 30, 1976 and hence was 15 months long. Subsequent fiscal years have begun on October of the previous fiscal year.

Table 2-2. US Stockpile of Weapon-Grade Uranium

	<u>tonnes</u>
Weapon-Grade Uranium Equivalent of All US Enrichment Work Prior to Jan. 1 or July 1 1964	745 +/- 22
Enrichment Work Used to Produce Slightly Enriched Fuel for Production Reactors Through 1964*	
--Hanford (all 9 reactors)	-7 +/- 1
--Savannah River	-4 +/- 4
Enrichment Work Used to Produce Low-Enriched Fuel for Civilian Reactors Through 1964*	-12 +/- 3
Weapon-Grade Uranium Actually Produced Prior to Termination of Production for Weapons	722 +/- 23 <sup>+</sup>
<hr/>	
Requirements by the Savannah River Production Reactors	
--pre-65	-10 +/- 10
--1965-84	-26 +/- 11
--projected for 1985-89	-5 +/- 3
Equivalent to U-235 in Fuel Delivered to US Naval Propulsion Reactors (through 1964)	-11 +/- 5
Delivered to Domestic and Foreign Research Reactors	
--prior to the end of FY 1964	-11 +/- 3
--from the pre-1964 stockpile during 1964-1984	-11 +/- 8
--projected for 1985-89	-3 +/- 2
Export to the UK and France for Military Use	- 6 +/- 3
REMAINING AFTER USES FOR REACTOR FUEL	639 +/- 34 <sup>+</sup>
<hr/>	
Consumption in US Nuclear Weapons Tests	-20 +/- 7
Process Losses Equal to 3 Percent of the Original 745 Tonnes and a Non-Weapons Stockpile of 50+/- 50 tonnes	- 65 +/- 50
ESTIMATED IN WEAPONS OR AVAILABLE FOR WEAPONS	554 +/- 59 <sup>+</sup>

\* Note that these numbers represent the amount of weapon-grade uranium that could have been produced with the enrichment work used to produce this low-enriched uranium. The tonnage of low-enriched uranium produced with this enrichment work and the tonnage of U-235 that it contained were both much larger.

<sup>+</sup> Uncertainties have been combined by taking the square root of the sum of the squares.

Table 2-3. Power of US and UK Nuclear Ship Reactors

<u>United States</u>						
<u>Year of Startup</u> <sup>b</sup>	<u>Ship</u> <sup>c</sup>	<u>Reactors</u> <sup>d</sup> (#, refuels)	<u>Ship</u> <sup>a</sup>	<u>Shaft hp (x10<sup>3</sup>)</u> Ann. -Incr.	<u>Cum.</u>	<u>Cumulative</u> <u>Shaft-hp-yrs</u>
1954	+Nautalis	(1,0)	15	15	15	15
1955						30
1956						45
1957	Skate	(2,0)	6.6	6.6	21.6	66.6
1958	Swordfish		6.			
	Sargo		6.6			
	Skipjack	(5,0)	15	28.2	49.8	116.4
1959	+Seadragon		6.6			
	+Triton(2 react).		34			
	+Halibut		6.6			
	+G. Washington	(10,1)	15	62.2	112.0	228.4
1960	Enterprise (8 react.)		280			
	+Scorpion		15			
	Snark		15			
	Tullibee		2.5			
	+Patrick Henry		15			
	+Theodore Roosevelt		15			
	+Robert E. Lee		15			
	+Abraham Lincoln		15			
	Seawolf	(27,1)	15	387.5	499.5	727.9
1961	Long Beach (2 react.)		80			
	Sculpin		15			
	Snook		15			
	+Thresher		15			
	+Ethan Allen		15			
	Sam Houston		15			
	+Thomas A. Edison		15			
	Scamp	(36,1)	15	185	684.5	1412.4
1962	Bainbridge (2 react.)		60			
	Plunger		15			
	John Marshall		15			
	+Thomas Jefferson		15			
	Permit	(42,2)	15	120	804.5	2216.9
1963	-Thresher		-15			
	Barb		15			
	Pollack		15			
	Tinosa		15			
	Dace		15			
	Lafayette		15			
	Alexander Hamilton		15			
	Andrew Jackson		15			
	James Monroe		15			
	Nathan Hale		15			
	Woodrow Wilson		15			
	Henry Clay		15			
	Daniel Boone	(54,5)	15	165	969.5	3186.4

(Table 2-3, p. 2)

<u>Year of Startup</u> <sup>b</sup>	<u>Ship</u> <sup>c</sup>	<u>Reactors</u> <sup>d</sup> (#, refuels)	<u>Ship</u> <sup>a</sup>	<u>Shaft hp (x10<sup>3</sup>)</u> <u>Ann.-Incr.</u>	<u>Cum.</u>	<u>Cumulative</u> <u>Shaft-hp-yrs</u>
1964	Haddo		15			
	John Adams		15			
	Daniel Webster		15			
	James Madison		15			
	Tecumseh		15			
	John C. Calhoun		15			
	Ulysses S. Grant		15			
	Von Steuben		15			
	Casimir Pulaski		15			
	Stonewall Jackson		15			
	Sam Rayburn		15			
1965	N. Greene (66,11)		15	180	1149.5	4335.9
	Jack		15			
	Benjamin Franklin		15			
	Simon Bolivar		15			
	Kamehameha		15			
1966	George Bancroft		15			
	Lewis and Clark(72,27)		15	90	1239.5	5575.4
	Guardfish		15			
	Flasher		15			
	Sturgeon		15			
	James K. Polk		15			
	Queenfish		15			
	George C. Marshall		15			
	Henry L. Stimpson		15			
	George Washington		15			
	Carver		15			
1967	Francis Scott Key		15			
	M.G. Vallejo (82,36)		15	150	1389.5	6964.9
	Truxton (2 react.)		60			
	Gato		15			
	Haddock		15			
	Pargo		15			
	Ray		15			
	Will Rogers		15			
	Lapon		15			
	Hammerhead		15			
	Greenling (93,43)		15	180	1569.5	8534.4
1968	-Triton		34			
	-Scorpion		15			
	Whale		15			
	Tautog		15			
	Aspro		15			
	Sunfish		15			
	Gunard		15			
	Sea Devil (96,58)		15	41	1610.5	10144.9



(Table 2-3, p. 3)

<u>Year of Startup</u> <sup>b</sup>	<u>Ship</u> <sup>c</sup>	<u>Reactors</u> <sup>d</sup> (#, refuels)	<u>Ship</u> <sup>a</sup>	<u>Shaft hp (x10<sup>3</sup>)</u> <u>Ann.-Incr.</u>	<u>Cum.</u>	<u>Cumulative</u> <u>Shaft-hp-yrs</u>
1969	Grayling		15			
	Puffer		15			
	Bergall		15			
	Spadefish		15			
	Sea Horse		15			
	Finback		15			
	Narwhal		17			
	Flying Fish (104,75)		15	122	1732.5	11877.4
1970 (6 yr. cores)	Pogy		15			
	Hawkbill		15			
	Pintado		15			
	Trepang		15			
	Bluefish		15			
	Billfish (110,75)		15	90	1822.5	13699.9
1971	Sand Lance		15			
	Drum		15			
	Archerfish		15			
	Silversides (114,81)		15	60	1882.5	15582.4
1972	Guitarro		15			
	William H. Bates		15			
	Batfish		15			
	Cavalla (118,107)		15	60	1942.5	17524.9
1973	California (2 react.)		60			
	Tunny (120,125)		15	75	2017.5	19542.4
1974	Nimitz (2 react.)		260			
	Glennard P. Lipscomb		15?			
	L. Mendell Rivers		15			
	Richard B. Russell		15			
	South Carolina (2. r.)		60			
	Parche (128,140)		15	380	2397.5	21939.9
1975 (8 yr. cores)	(128,140)				2397.5	24337.4
1976	-Halibut		6.6			
	Virginia (2 react.)		100			
	Los Angeles		35			
	Philadelphia		35			
	Los Angeles (132,140)		35	198.4	2595.9	26933.3
1977	D.D. Eisenhower (2 r)		260			
	Memphis		35			
	Omaha		35			
	Texas (2 react.)		100			
	Baton Rouge (138,148)		35	465	3060.9	29994.2
1978	Mississippi (2 r.)		100			
	Groton		35			
	Birmingham		35			
	New York City		35			
	Cincinnati (144,160)		35	240	3300.9	33295.1
1979	(144,176)				3300.9	36596.

(Table 2-3, p. 4)

<u>Year of Startup</u> <sup>b</sup>	<u>Ship</u> <sup>c</sup>	<u>Reactors</u> <sup>d</sup> (#,refuels)	<u>Ship</u> <sup>a</sup>	<u>Shaft hp (x10<sup>3</sup>)</u> <u>Ann.-Incr.</u>	<u>Cum.</u>	<u>Cumulative</u> <u>Shaft-hp-yrs</u>
1980	Arkansas (2 react.)	100				
	Indianapolis	35				
	-Nautalis	(146,192	-15	120	3420.9	40016.9
1981	Carl Vinson (2 r.)	260				
	-Abraham Lincoln	-15				
	Bremerton	35				
	Jacksonville	35				
	Dallas	35				
	La Jolla	35				
	Phoenix	35				
	Boston	35				
	San Francisco	35				
	Atlanta	35				
	Ohio	60				
	-T. Roosevelt(154,202)	-15	570	3990.9	44007.8	
1982	Baltimore	35				
	Houston	35				
	Michigan (157,223)	60				
	Corpus Christi	35				
	Albuquerque	35				
	Florida	60				
	Ethan Allen	-15	245	4235.9	48243.7	
<u>Being Built as of Dec. 31, 1982</u>						
1983	Portsmouth	35				
	Minn-St. Paul (SSN708)	35				
	Norfolk (SSN714)	35				
	Buffalo (SSN715)	35				
	Salt Lake City (SSN716)	35				
	Georgia (SSBN729)	60				
	-Seadragon	-6.6				
	-Patrick Henry	-15				
	-Robert E. Lee	-15				
	-Thomas A. Edison	-15	183.4	4419.3	52663	
1984	Hyman Rickover(SSN709)	35				
	Augusta (SSN710)	35				
	Olympia (SSN717)	35				
	H. Jackson(SSBN730)	60				
	Alabama (SSBN731)	60				
	-George Washington	-15				
	-Thomas Jefferson	-15	195	4614.3	57277.3	

(Table 2-3, p.5)

<u>Year of Startup</u> <sup>b</sup>	<u>Ship</u> <sup>c</sup>	<u>Reactors</u> <sup>d</sup> (#, refuels)	<u>Ship</u> <sup>a</sup>	<u>Shaft hp (x10<sup>3</sup>)</u> <u>Ann. -Incr.</u>	<u>Cumulative</u> <u>Shaft-hp-yrs</u>
1985	Honolulu				
(thru.	Providence				
March 31)	Pittsburgh (SSN720)				
	Chicago (SSN721)				
(not	SSN722				
completed	SSN723				
as of	SSN724				
March	SSN725				
31, 1985)	Newport News (SSN750)				
	SSN751				
	SSN752				
	SSBN732				
	SSBN733				
	SSBN734				
	SSBN735				
	T. Roosevelt (CVN71)				
	CVN72				
	CVN73			(1405)	

Additional Naval Reactors Being Built as of March (31?) 1985

	SSN751				
	SSN752				
	SSN753				
	SSN754				
	SSN755				
	SSN756				
	SSN757				
	SSN758				
	SSN759				
	SSBN736				
Total Added				(375)	

(Table 2-3, p.6)

		<u>United Kingdom</u>				
<u>Year</u>	<u>Com-</u> <u>missioned<sup>e</sup></u>	<u>Ship</u>	<u>Ship</u>	<u>Shaft</u> <u>Ann. Incr.</u>	<u>hp (x10<sup>3</sup>)</u> <u>Cumulative</u>	<u>Cumulative</u> <u>Shaft-hp-yrs</u>
1966	1	Valiant	1x15	15	15	15
1967	1	Resolution	1x15			
		Valiant	1x15	30	45	60
1968	2	Resolution	2x15	30	75	135
1969	1	Resolution	1x15	15	90	225
1970	1	Valiant	1x15	15	105	330
1971	2	Valiant	2x15	30	135	465
1972					135	600
1973	1	Swiftsure	1x15	15	150	750
1974	1	Swiftsure	1x15	15	165	915
1975					165	1080
1976	1	Swiftsure	1x15	15	180	1260
1977					180	1440
1978	1	Swiftsure	1x15	15	195	1635
1979	1	Swiftsure	1x15	15	210	1845
1980					210	2055
1981	1	Swiftsure	1x15	15	225	2280

Table 2-3. Notes and References

- a. Jane's Fighting Ships, 1982-83, (London, Jane's Yearbook) pp. 598-627.
- b. Nuclear Reactors Built, Being Built, or Planned (Washington DC: US Department of Energy, Report # DOE/TIC-8200-R47, 1983).
- c. + Means now retired.
- d. The number of cores replaced are estimated by assuming that cores were replaced after 5 years until 1970, then 6 years until 1975 and 8 years subsequently -- except that double counting is avoided for the transition periods.
- e. Jane's Fighting Ships, 1983-84, pp. 581-583.

Table 2-4a. U-235 Recovered at Idaho Chemical Processing Plant<sup>a</sup>

<u>Fiscal Year</u>	<u>Amount (kg)</u>	
	<u>Total Uranium</u>	<u>Contained U-235</u>
1953	161	146
1954	165	149
1955	930	819
1956	914	816
1957	746	656
1958	1593	1390
1959	2931	2534
1960	941	828
1961	45	35
1962	690	614
1963	191	166
1964	677	589
SUBTOTAL	(9984)	(8742)
1965	685	588
1966	625	527
1967	102	82
1968	737	623
1969	0	0
SUBTOTAL	(12133)	(10562)
1970	1275	997
1971	800	600
1972	354	284
1973	1543.6	802.2
1974	414.9	315.3
SUBTOTAL	(16520.5)	(13560.5)
1975	2724	1812.8
1976+transition quarter	1825.9	1008
1977	1680.7	1318.7
1978	461.4	358.7
1979	34.6	24.8
SUBTOTAL	(23247.1)	(18083.5)
1980	123.7	91.2
1981	1375.9	1000.5
1982	693.3	499.4
1983	882.7	793.2
1984	2464.1	2294.4
TOTAL	(28786.8)	(22762.2)

Table 2-4a. References and Notes

a. Cochran et al, Nuclear Weapons Databook, II (Cambridge Mass: Ballinger, to be published).

Table 2-4b. Uranium-235 Recovered in the US from the Spent Fuel of Domestic and Foreign Civilian Reactors (through February 1985)

<u>Reprocessing Facility</u>	<u>Amount of U-235 Recovered</u> <u>(kg)</u>
<u>Savannah River Plant (SRP)</u>	
Domestic Fuel	3,200 <sup>a</sup>
<u>Foreign Fuel</u>	<u>1,250<sup>b</sup></u>
SUBTOTAL	4,200
<u>Idaho Chemical Processing Plant (ICPP)</u>	
Research, Test and Power Reactor Fuel Originally Enriched to Over 90 percent U-235	7,530 <sup>c, d</sup>
EBR-2 Fuel (originally enriched to 60 % U-235)	3,400 <sup>e</sup>
<u>Project Rover Space Propulsion Reactor Fuel</u>	<u>2,820<sup>c</sup></u>
SUBTOTAL	13,750
<u>Nuclear Fuel Services (NFS)</u>	<u>800<sup>f</sup></u>
TOTAL	19,000

Table 2-4b, Notes

a. Based on the data in Tables 2-5a, -b and -c. Where more than one reprocessor is listed, it is assumed that half of the fuel went to each. We assume that, on average, 70 percent of the U-235 in the original fresh fuel was recovered. Over half of the U-235 recovered at the SRP was recovered from the fuel of the High Flux Isotope Reactor (HFIR) located at Oak Ridge.

b. In total, 1.5 tonnes of U-235 were returned to the US through 1983 according to Table 2-5f. In 1982 and 1983, INEL received 130 kilograms of U-235 in HEU spent fuel (average enrichment was 72 percent) while, from the beginning of 1978 until 1982 it received little foreign spent fuel (DOE, Nuclear Materials Management and Safeguards System, Transaction Journal-21 [TJ-21], Mod 3, year-end report for these years). The large amount received in 1982 and 1983 was due to increased requirements for Kr-85, which ICPP can recover during reprocessing. From 1964, when foreign fuel was first returned, through 1967, 50 kilograms of U-235 in HEU (average enrichment of 65 percent) were received at INEL (Annual Report to Congress of the AEC, 1964, 1965 and 1967, and Major Activities of the Atomic Energy Commission, 1966). Ignoring 1982 and 1983, the average rate of return of spent fuel to INEL during the remaining eight years for which we have data was about 6 kg/yr. We assume this average rate for the years for which we have no data: 1968-77. On this basis, we estimate that INEL received, in total, about 250 kg of U-235 in foreign civilian HEU reactor fuel. This leaves about 1.25 tonnes for the SRP.

c. Letter from John L. Meinhardt, Director, Office of Nuclear Materials Production, Department of Energy, to DA, May 6, 1985. Includes an estimated 250 kg U-235 recovered from foreign fuel (see note b).

d. An independent estimate, using the information in Tables 2-5a; -b and -c, as described for the SRP in note a, yields an estimate of 6.9-7.6 tonnes.

e. Meinhardt (ref. c), as corrected in a telephone conversation with DA, 21 May 1985.

f. Derived from the information in Table 2-5b using the same approach as for the SRP case described in note a.

Table 2-5. Weapon-Grade Uranium Equivalent of Pre-FY-1965 US Enrichment Work Used to Produce Fuel for US and Foreign Civilian Reactors  
(tonnes)

Reactors Fueled With HEU

US Reactors

Research reactors (Table 2-5a) <sup>+</sup>	3
Power reactors	3
--Utility-Operated (Table 2-5b) <sup>+</sup>	2.6
--Experimental (Table 2-5c) <sup>+</sup>	0.7
Rocket Propulsion reactors (Table 2-5b) <sup>+</sup>	2.5 +/- 0.5
Military reactors other than naval and production reactors	<1
Foreign Research Reactors (Table 2-5f) <sup>+</sup>	1.7
SUBTOTAL	11 +/- 3

Reactors Fueled with LEU

US Power Reactors (Table 2-5d)	9 +/- 2
Foreign Power Reactors (see text)	2.6 +/- 1
SUBTOTAL	12 +/- 3

Weapon-Grade Uranium from the Stockpile Shipped After 1965.0

Domestic Research Reactors (Table 2-5a) <sup>+</sup>	6 +/- 6
Foreign Research Reactors (see text)	5 +/- 5
SUBTOTAL	11 +/- 8
TOTAL	34 +/- 9

+ Note that the tables referred to show tonnes of U-235 which are converted to the equivalent number of tonnes of 93.5 percent HEU in the present table.



Table 2-5a. Uranium Demand of US Civilian Research Reactors With Powers Greater Than 1 Mwt and With Metal Fuel Which Were Fueled from the HEU Stockpile Enriched Through 1964

Reactor	Power <sup>a</sup> (Mwt)	Startup <sup>a</sup> Date	Shutdown <sup>a</sup> Date	-----U-235 Demand(kg)----- Annual <sup>b</sup> Per Mwt-yr	Reproc./ Store	
					1950- -1965.0	1950- -1985.0

Reactors With Powers of 10 Mwt or Greater

Operating

			Department of Energy (DOE)				
ATR	250-->125 <sup>d</sup>	1967 <sup>d</sup>	---	175 <sup>e</sup>	1.4 <sup>e</sup>	0	3900 INEL
HFIR	100	1965	---	140 <sup>f</sup>	1.4	0	(2800)* SRP
HFBR	40	1965	1982	(40 <sup>g</sup> )	[1.00]	0	720 SRP& <sup>h</sup>
"	60	1982	---	59		0	120 INEL
ORR	30	1958	---	18	0.60	126	490 IN&SR
			Other Government				
NBSR	10	1967	---	13	1.3	--	230 SRP
			Private				
MURR	5	1966	1974	(9, scaled)	1.8	--	80 ?
	10	1974	--	19	1.9	--	170 SRP
SUBTOTALS				(424)		(126)	(5730)

Shutdown

ETR	175	1957	1972 <sup>i</sup>	DOE 180 <sup>i</sup>	1.03	1440	2700 INEL
MTR	40	1952	1970	(40 <sup>g</sup> )	[1.00]	520	720 INEL
			Other Government				
NASA-TR	60	1963	1974	30 <sup>j</sup>	0.5	60	330 IN&SR <sup>h</sup>
			Private				
GETR	30	1958	1966	(30 <sup>g</sup> )	[1.0]	210	270 INEL
"	50	1966	1977	(50 <sup>g</sup> )	[1.0]	0	550 INEL
WTR	60	1959	1962			80 <sup>k</sup>	80 INEL
NONGOVERNMENT SUBTOTAL						(290)	(900)*
SUBTOTAL, THIS PAGE						(2436)	10380*

\* The HFIR is fueled with oxide fuel. According to its staff<sup>f</sup> the HEU is enriched and converted from uranium hexafluoride to U<sub>3</sub>O<sub>8</sub> at the Oak Ridge Y12 plant. Its HEU therefore does not come from the pre-1965 stockpile of HEU metal and consequently is not included in the running sums. However, the HEU recovered from the HFIR fuel is recycled to fuel the Savannah River production reactors.

(Table 2-5a, cont.)

Steady-State Reactors Fueled With HEU Metal Fuel and Powers of 1-9.9 Mwt

<u>Reactor</u>	<u>Power<sup>a</sup></u> (Mwt)	<u>Startup<sup>a</sup></u> <u>Date</u>	<u>Shutdown<sup>a</sup></u> <u>Date</u>	<u>U-235 Demand(kg)</u>		<u>Store</u>		<u>Loc.<sup>c</sup></u>
				<u>Annual<sup>b</sup></u>	<u>Per Mwt-yr</u>	<u>1950-</u> <u>-1965.0</u>	<u>1950-</u> <u>-1985.0</u>	

Operating

				<u>DOE</u>				
OWR	8	1956	--	5.4	0.68	49	160	INEL
BMRR	5	1959	--	0.2	0.04	1	5	?
BSR	2	1950	--	0.5	0.25	8	17	SR/IN
TSR-2	1	1960	--	0.2	0.2	1	5	"

NRC(formerly AEC)-Licensed

MITR	5	1958	--	5.4	1.1	38	146	SRP <sup>h</sup>
UCNR	5	1961	--	5.4	1.1	22	130	SRP
GTRR	5	1965	--	1.9	0.4	0	38 <sub>1</sub>	?
FNR	2	1957	--	3.3	1.7	26	80 <sup>1</sup>	SRP
RINSC	2	1964	--	2.5	1.3	3	53	SRP
UVAR	2	1960	--	1.3	0.7	7	33	SRP
ULR	1	1974	--	0.2	0.2	-	2	?

SUBTOTAL, THIS PAGE SO FAR

(26.3)

(155)(669)

Shutdown

				<u>DOE</u>				
ALRR	5	1965	1977	5 <sup>g</sup>	[1.0]	5	60	SRP
CP-5	5	1954	1979	5 <sup>g</sup>	[1.0]	55	125	SRP
SER	5	1961	1970	5 <sup>g</sup>	[1.0]	20	45	INEL
LPTR	3	1957	1980	3 <sup>g</sup>	[1.0]	27	69	INEL
LITR	3	1950	1968	3 <sup>g</sup>	[1.0]	45	57	?

NRC(formerly AEC)-Licensed

BAWTR	6	1964	1971	6 <sup>g</sup>	[1.0]	6	42	?
ILR	5	1958	1975	5 <sup>g</sup>	[1.0]	35	85	INEL
LPR	1	1958	1981	1 <sup>g</sup>	[1.0]	7	23	

SUBTOTAL, THIS PAGE

(352)(1172)

TOTAL, TABLE 2-5a

450(in '85)

(2788)11552)

+2800 from  
HFIR)

Table 2-5a, Notes

a. US DOE, Nuclear Reactors Built, Being Built, or Planned (US DOE, Technical Information Center, Report #DOE/TIC 8200-R47, 1983). The full name of each reactor is given in this report. We assume that the reactors started up at the beginning of the year.

b. Unless otherwise indicated, the uranium requirements are from Matos, J.E. (Argonne National Laboratory), September 1982: RERTR Program Summary, attached to letter to K.L. Mattern (US DOE). In all cases the enrichment is 93 percent, except in the cases of the ILR and LITR where the enrichment is given as 90 percent, and the BMRR where it is 90-93 percent.

c. Unless otherwise noted, the spent fuel is reprocessed at either the Savannah River Plant (SRP) or the Idaho National Engineering Laboratory (INEL). Information on the locations where the fuel from specific reactors is reprocessed may be found in several sources: Annual Report to the Congress of the AEC (years, 1964, -65, 67); Major Activities of the Atomic Energy Programs, Jan.-Dec.1966 (US Atomic Energy Commission); R.R. Burn, Research, Training, Test, and Production Reactor Directory (American Nuclear Society, 1983); "Department of Energy News: Factsheet on Foreign/Domestic Research Reactor Spent Fuel Shipments," (DOE, Savannah River Operations Office, years 1982-84); letter to Ted Harris, Public Research Foundation, Columbia, S.C. from Robert C. Webb, Deputy Director, Office of External Affairs, DOE, Savannah River Operations Office, 22 July 1982; and private communications with the operators of certain major research reactors. It has been assumed that the fuel from reactors located at INEL is reprocessed there. The location of the reprocessing site for a few of the small reactors is an educated guess.

d. The ATR ran at 250 Mwt in 1967, at about 220 Mwt in 1967 and after that the power was reduced gradually to 120-130 Mwt by 1975 (INEL, private communication to DA, May 1985).

e. 150-175 new fuel elements, each containing 1.075 kg of U-235, are loaded into the ATR each year (INEL, private communication to DA, May 1984). At 125 Mwt, this demand corresponds to 1.3-1.5 kg/Mwt-year.

f. Personal communication from HFIR staff to DA, May 1984. An independent estimate may also be obtained from the fact that, from 1978 through 1984, the average annual amount of HEU in HFIR spent fuel shipped to SRP for reprocessing was 120 kg (DOE News, op. cit., footnote c). The average burnup of HFIR fuel is 30 percent [R.R. Burn, Research, Training, Test, and Production Reactor Directory, (American Nuclear Society, 1983)].

g. 1 kg/Mwt-year assumed, based on average value for reactors in ref. b.

h. HFBR: No fuel shipped off-site between 1976-Jan. 85. This fuel is being sent to INEL (Nuclear Fuel, 18 June 1984 and 28 January 1985). Prior and subsequent years probably to SRP. MITR: No fuel shipped off-site since 1975. Shipments prior to 1975 went to SRP (personal communication to DA from MITR staff, Feb. 1986).

(Table 2-5a Notes, cont.)

i. INEL, personal communication to DA, May 1985. The ETR was shut down in 1972 to install a sodium loop for fast breeder research. Subsequently, until the ETR shut down in 1981, it used very little HEU. The ETR might be restarted and would require about 180 kg of U-235 per year.

j. Estimate based on the average annual amounts of U-235 reported recovered from NASA-TR at SRP & INEL and the enrichment of the recovered HEU (Annual Report to the Congress of the AEC, 1964, 1965, 1967 and Major Activities in the Atomic Energy Programs, January-December 1966).

k. WTR: Annual Report to Congress of the AEC for 1964, p. 61.

l. Converted to LEU fuels in the early 1980's (LEU Study Group, Assessment of the Implications of Conversion of University and Training Reactors to Low Enriched Fuel, D.R. Harris, chairman. Study sponsored by the Nuclear Regulatory Commission, Nov. 15, 1983).

Table 2-5b. Amounts of HEU Used in US Civilian Power Reactors Through 1964  
(enrichment in all cases 92-93 percent)

<u>Reactor</u>	<u>Power</u> <sup>a</sup> (Mwt)	<u>Startup</u> <sup>a</sup> <u>Date</u>	<u>Shutdown</u> <sup>a</sup> <u>Date</u>	-----U-235 Demand(kg)-----		<u>Store</u> <u>Loc.</u> <sup>c</sup>
				<u>Annual</u> <sup>b</sup>	<u>Per Mwt-yr</u>	
Shippingport	240	1957	1982	680 <sup>b</sup>	1020 <sup>b</sup>	INEL <sup>b</sup>
Indian Pt. I	615	1962	1980	1100 <sup>c</sup>	1100 <sup>c</sup>	NFS <sup>d</sup>
Pathfinder	190	1964	1967	50 <sup>e</sup>	100 <sup>e</sup>	INEL <sup>f</sup>
Peach Bottom	115	1966	1974	220 <sup>g</sup>	440 <sup>g</sup>	INEL <sup>h</sup>
<u>Elk River</u>	<u>58</u>	<u>1962</u>	<u>1968</u>	<u>344</u> <sup>i</sup>	<u>344</u> <sup>i</sup>	Italy&
				2400	3000	SRP <sup>j</sup>

Table 2-5b, Notes

a. US DOE, Nuclear Reactors Built, Being Built, or Planned (US DOE, Technical Information Center, Report #DOE/TIC 8200-R47, 1983).

b. The first core contained 345 kg U-235 in HEU and the second contained 672 kg. The HEU fuel for the first and one half of the second were fabricated before the end of FY 1964 [M.T. Simnad, Fuel Element Experience in Nuclear Power Reactors (AEC Monograph, Gordon and Breach, 1971) p. 211; and F. Duncan and J.M. Holl, "Shippingport: The Nation's First Atomic Power Station," (History Division, Department of Energy, undated, reprinted in Naval Nuclear Propulsion Program -- 1983, Hearings before the House Armed Service Committee (print H.A.S.C. 98-25) pp. 204-206. The highly-enriched "seeds" from the first core were most likely reprocessed at INEL. The seeds from the second core are scheduled for reprocessing at INEL after 1985 (INEL [DOE, Idaho Operations Office, undated, p. 14]).

c. Only the first core used HEU, Simnad, op cit, pp. 288-289.

d. NFS - Nuclear Fuel Services. G. Rochlin et al, "West Valley: Remnant of the AEC," Bulletin of the Atomic Scientists, January 1978, Table 2.

e. Only two cores were fabricated [AEC Authorization Legislation for Year 1965 and ibid, 1967]. The U-235 content of the cores is given in Simnad, op cit, pp. 405-406.

f. About 50 kg. of slightly-irradiated fresh fuel were still stored, unprocessed at INEL as of the end of 1983 [US DOE, Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics (DOE/RW-0006, September 1984)].

Notes, Table 2-5b, cont.

- g. Only two cores were fabricated [H.L. Brey and H.G. Olson, "Fort St. Vrain Experience," Nuclear Energy 22, April 1983, p. 120. The amount of U-235 in each core is given in Simnad op. cit., p. 116. The second core was loaded after 1968 ["Operating History of US Nuclear Power Reactors," AEC Authorizing Legislation, FY 1970, Part II, Hearings before the Joint Committee on Atomic Energy, April 24 - 25, Appendix 4, p. 1561.] (Table 2-5b Notes, cont.)
- h. As of the end of 1983, spent fuel containing 220 kg. of U-235 in 330 kg-U was stored at INEL [DOE, Spent Fuel...].
- i. One core had been inserted and another was being fabricated in March 1964 [AEC Authorizing Legislation for FY 65 Hearing before the Joint Committee on Atomic Energy, p. 781]. Each core contained 172 kg. of U-235 [Simnad, op cit, p. 378].
- j. A small amount of this fuel was reprocessed at the ITREC facility in Italy during the 1970's [S. Cao et al, "Italian Experience with Pilot Reprocessing Plants" in IAEA-CN-36/304, May 1977. As of the end of 1983, 190 kg. of 83 percent enriched spent fuel was stored at SRP [DOE, Spent Fuel...].

Table 2-5c. Amounts of U-235 in HEU Used in US Experimental-Power Reactors Through 1964  
(enrichment in all cases 90-93 percent)

Reactor	Power <sup>a</sup> (Mwt)	Startup <sup>a</sup> Date	Shutdown <sup>a</sup> Date	----- Annual <sup>b</sup>	U-235 Demand(kg)		Store Loc. <sup>c</sup>
					Per Mwt-yr	1950- 1965.0	
EBWR	100	1956	1967	--	----	30 <sup>b</sup>	30 SRP <sup>c</sup>
HWCTR	61	1962	1964	--	----	50 <sup>d</sup>	50 SRP <sup>e</sup>
EOCR	40	1962(scheduled--terminated)				65 <sup>f</sup>	65 INEL <sup>g</sup>
VBWR	33	1957	1963	--	----	35 <sup>h</sup>	35 INEL <sup>i</sup>
SRE	20	1957	1964	--	----	150 <sup>j</sup>	150 SRP <sup>j</sup>
OMRE	12	1957	1963	--	----	75 <sup>k</sup>	75 INEL <sup>g</sup>
MSRE	8	1965	1969	8	[1.0] <sup>l</sup>	32 <sup>m</sup>	32 ?
HRE-1	1	1952	1954	1	----	5	5 ?
HRE-2	5	1957	1961	5	[1.0] <sup>l</sup>	25	25 ?
UHTREX	3	1968	1970	--	----	--	11 <sup>n</sup> ?
LAPRE-1	2	1956	1957	--	----	4 <sup>o</sup>	4 ?
LAPRE-2	1	1959	1959	--	----	4 <sup>o</sup>	4 ?
EBR-1	1.4	1951	1962	13.6	9.7	150 <sup>p</sup>	150 INEL <sup>p</sup>
BORAX 1,2,3,5	--	1953	1958	--	--	50 <sup>r</sup>	50 INEL <sup>r</sup>
				TOTALS		675	686

Table 2-5c, Notes and References

a. US DOE, Nuclear Reactors Built, Being Built, or Planned (US DOE, Technical Information Center, Report #DOE/TIC 8200-R47, 1983). The full names of the reactors are given in this reference.

b. Only the second core contained HEU [M.T. Simnad, Fuel Element Experience in Nuclear Power Reactors (AEC Monograph, Gordon and Breach, 1971) pp. 339, 345].

c. About 27 kg of U-235 in EBWR spent fuel, still enriched to 92 percent, was stored at SRP as of the end of 1983 [US DOE, Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics (DOE/RW-0006, September 1984)].

d. 24 kg. in the driver elements of each core [R.R. Burn, Research, Training, Test and Production Reactor Directory (American Nuclear Society, 1983), p. 777]. Two cores were used [AEC Authorizing Legislation for FY 1969, Hearings before the Joint Committee on Atomic Energy (JCAE); Jan. 30, 31 and Feb. 5, 1968, p. 908].

e. Spent HWCTR fuel containing about 32 kg of U-235 was stored at SRP as of the end of 1983 [DOE, Spent Fuel...., 1984].

f. We assume that one core was produced. The amount of U-235 in the core is from R. R. Burn, op cit, p. 669.

(Table 2-5c, Notes, cont.)

- g. Since EOCR and OMRE were located at INEL, we assume that their fuel was reprocessed there.
- h. The first core used HEU. In 1960, the reactor was modified to operate on LEU [M.T. Simnad, op cit, p.349].
- i. About 33 kg of U-235 was recovered from VBWR spent fuel at INEL in the early 1960's [Annual Report to Congress of the Atomic Energy Commission, 1964, p. 61.]
- j. Only the second core of the SRE contained HEU [Simnad, op cit, p. 465. SRE spent fuel containing 143 kg of U-235 at an enrichment of 92 percent was stored at SRP as of the end of 1983 [DOE, Spent Fuel..., 1984]. We assume that this was the entire core.
- k. Each core of the OMRE contained about 25 kg. of U-235 [Simnad, op cit, p. 437]. Three cores were fabricated [AEC Authorizing Legislation for Fiscal Year 1965, p. 782].
- l. Assumed.
- m. We assume only one core and that it was enriched prior to 1965.
- n. The core of the UHTREX contained 11 kg. of U-235 [Simnad, op cit, p. 149].
- o. Accelerating Civilian Reactor Program, Hearings before the JCAE, May 23, 24, 25, 28 and 29, 1956, pp. 54-57.
- p. The EBR-1 used 3 cores -- each containing about 50 kg. of HEU [Simnad, op cit, p. 516]. Since the EBR-1 was located at INEL, we assume that its fuel was reprocessed there.
- r. These were safety experiments involving testing to core destruction. The BORAX-1 and 2 are each estimated to contain about 15 kg. of U-235 in their cores and BORAX-3 contained about 15 kg (Accelerating Civilian Reactor Program, op cit. p. 54. The BORAX-5 core contained about 20 kg of U-235, most of which was still in storage at INEL as of the end of 1983 [DOE, Spent Fuel..., op. cit., 1983]. The BORAX-4 core contained 7 percent enriched uranium (Simnad, op. cit. p. 384).



Table 2-5d. Enrichment Work Expended in Producing Low-Enriched Fuel for US Power Reactors Through 1964

<u>Reactor</u> <sup>a</sup>	<u>Core</u> <sup>b</sup> (MtU)	<u>#</u> <sup>c</sup>	<u>U-235</u> (kg)	<u>Enrich</u> <sup>b</sup> (%)	<u>F/P</u> <sup>d</sup>	<u>D/P</u> <sup>d</sup> (SWU/kg)	<u>D</u> (Mt-SWU)	<u>F</u> (MtU)
Indian Pt. 1	20	1	680	3.4	7.88	4.00	80	158
Yankee-Rowe	20.8	2	1414	3.4	7.88	4.00	166	328
	20.8	3 <sup>b</sup>	2496	4.0	9.41	5.09	317	588
Big Rock Pt.	11.7	3 <sup>b</sup>	1123	3.2	7.37	3.64	128	259
Dresden 1	51.5	2	2060	2.0	4.30	1.62	162	442
Humboldt Bay	13.8	1	359	2.6	5.83	2.60	36	78
	13.8	0.5 <sup>e</sup>	152	2.2	4.81	1.94	13	34
La Crosse	8.6	1	312	3.63	8.47	4.41	38	73
Bonus								
-Boiler	2.81	1	67	2.4	5.32	2.27	6	15
-Superheater	1.79	1	58	3.25	7.49	3.73	7	13
Hallam	29.2	1	1051	3.6	8.39	4.36	127	245
	29.2	1	1431	4.9	11.71	6.76	197	342
Piqua	6.6	1	125	1.9	4.04	1.47	10	26
Carolina-Va								
Tube Reactor	3.28	2	115	1.75	3.66	1.24	8	24
Pathfinder	6.56	1	144	2.2	4.81	1.94	13	32
EBR-2	0.35	2	343	49	124.50	98.35	69	88
Fermi-1	2.0	2	1024	25.6	64.66	48.85	195	258
EBWR	5.6 <sup>f</sup>	1	81	1.44 <sup>f</sup>	2.86	0.79	4	16
EVESR	2.2 <sup>f</sup>	2 <sup>g</sup>	238	5.4 <sup>f</sup>	13.03	7.74	34	58
EBOR	0.16 <sup>h</sup>	1	100	62.5 <sup>h</sup>	159.4	127.68	20	26
EGCR	9.86 <sup>i</sup>	1	251	2.55 <sup>i</sup>	5.70	2.52	25	56
Saxton	1.27	1.5 <sup>e</sup>	109	5.7	13.76	8.28	16	26
SRE	3.00	1	84	2.8	6.34	2.95	9	19
	3.00	1	156	5.2	12.48	7.33	22	37
VBWR	0.8	1	24	3.0	6.85	3.29	3	6
TOTALS			13997				1710	3247

This much separative work with 0.32 % tails would produce 8.7 tonnes of 93.5 % HEU.

Table 2-5d, Notes and References

a. US DOE, Nuclear Reactors Built, Being Built, or Planned (US DOE, Technical Information Center, Report #DOE/TIC 8200-R47, 1983). The dates of operation and full names of the reactor are given in this reference. The very low-power reactors were ignored.

b. Unless otherwise noted, the source is M.T. Simnad, Fuel Element Experience in Nuclear Power Reactors (AEC Monograph, Gordon and Breach, 1971).

c. Unless otherwise noted, based on AEC tables listing power reactor fuel elements fabricated through various years, reprinted in hearings by the Joint Committee on Atomic Energy (JCAE) -- both authorization hearings for Atomic Energy Committee (AEC) appropriations and oversight hearings on the AEC-fostered development of the atomic energy industry.

d. The following formulae relating separative work units (D), tails assay ( $x_w$ ), product quantity (P) enrichment ( $x_p$ ) and feed (F) were taken from Gaseous Diffusion Plant Operations, (USAEC, #ORO-684, 1972), Appendix 2:

$$F/P = (x_p - x_w)/(x_f - x_w), \text{ and}$$

$$D/P = [V(x_p) - V(x_w)] - (F/P)*[V(x_f) - V(x_w)], \text{ where}$$

$$V(x) = (2*x - 1)*\ln[x/(1-x)].$$

We have assumed  $x_w = 0.32$ , which is an average value for 1960-64 -- the period during which the bulk of this fuel was enriched (see Table 2-1). We have assumed natural uranium feed:  $x_f = 0.00711$ .

$$V(0.00711) = 4.869, V(0.0032) = 5.705, \text{ Diff} = -0.836, V(0.935) = 2.32.$$

e. Sum of partial reloads.

f. R.R. Burn, Research, Training, Test, and Production Reactor Directory, (American Nuclear Society, 1983), p. 646.

g. "Operating History of the U.S. Nuclear Power Program," in AEC Authorizing Legislation, Fiscal Year 1970 in "Reactor Development and Technology Program," Hearing before the JCAE, April 24 and 25, 1969, Part 2.

h. R.R. Burn, op cit, p. 667.

i. US AEC, Office of Operations Analysis and Forecasting, Costs of Nuclear Power, (Washington, DC: TID-8531, January 1961).

Table 2-5e. US Exports of Highly-Enriched Uranium for Civilian Purposes by Country  
(1 January 1954 -- 28 February 1983)

<u>Country</u>	<u>Amount</u> <sup>a</sup> (kg U-235)	<u>Average Enrichment</u> <sup>a</sup> (percent)	<u>Returned</u> <sup>b</sup> ----- (kg U-235)-----
Argentina	59.15	63	0.02
Austria	7.32	75	4.57
Australia	9.16	90	0.05
Belgium	159.17	85	385.70 <sup>c</sup>
Brazil	7.70	93	0.0
Canada	1861.49	93	355.12
Columbia	2.82	91	0.0
Denmark	23.55	90	26.70 <sup>c</sup>
Finland	0.77	20	0.0
France	4655.46	74	1738.65
W. Germany	6612.51	66	325.23
Greece	6.15	93	0.0
IAEA	0.25	81	0.0
India	0.08	80	0.0
Indonesia	0.02	??	0.0
Iran	5.16	93	0.0
Israel	17.09	91	0.0
Italy	306.80	80	36.63
Japan	964.81	48	146.41
S. Korea	18.47	62	0.0
Mexico	12.32	42	0.4
Netherlands	56.52	89	115.77 <sup>c</sup>
Pakistan	5.18	90	0.0
Phillipines	3.07	93	0.0
Portugal	7.14	93	0.0
Rumania	36.56	93	0.0
South Africa	30.21	92	25.75
Spain	8.30	88	0.0
Sweden	133.34	90	170.42 <sup>c</sup>
Switzerland	7.96	91	4.36
Taiwan	9.21	93	0.0
Thailand	4.77	90	0.0
Turkey	4.78	90	0.0
United Kingdom	2141.35	93	82.79
Venezuela	0.01	??	0.0
Vietnam	0.08	21	0.0
Yugoslavia	5.91	35	0.05
Zaire	0.28	21	0.0
<u>TOTALS</u>	<u>17184.92</u>		<u>3418.62</u>

Table 2-5e. References and Notes

a. US Department of Energy, Nuclear Materials Management and Safeguards System, NMMSS Report TJ-25, 28 March 1983, as quoted in Cochran et al, The Nuclear Weapons Databook II: The Production Complex (Cambridge, MA: Ballinger, to be published).

b. US. Department of Energy, Nuclear Materials Management and Safeguards System, NMMSS Report, US Origin Imports, 29 November 1984, enclosure in a letter to Thomas B. Cochran from Robert O'Brien, Jr., 13 December 1984, as quoted in Cochran et al, The Nuclear Weapons Databook II: The Production Complex (Cambridge, MA: Ballinger, to be published).

c. Returns exceed exports. This may reflect retransfers between foreign countries associated with the fabrication and reprocessing of the fuel abroad.

Table 2-5f. U.S. Civilian HEU Exports and Returns by Year<sup>a</sup>

<u>Year</u>	<u>Exports</u> (kg U-235)	<u>Average Enrichment</u> (percent)	<u>Returns</u>	
			<u>In Spent Fuel</u> ----(kg U-235)-----	<u>Total</u>
1954-56	0		0.0	0.0
1957	24.59	93	0.0	0.0
1958	12.33	90	1.32	1.32
1959	16.18	91	0.0	0.0
1960	47.53	90	2.24	3.22
1961	212.32	80	1.81	2.17
1962	296.04	90	1.47	1.74
1963	296.20	82	13.49	18.92
1964	675.18	84	37.96	52.32
SUBTOTAL	(1580.35)	(85)	(58.29)	(79.69)
1965	1346.92	53	15.59	29.51
1966	927.71	62	29.42	74.81
1967	1260.61	47	83.69	168.68
1968	806.72	89	5.09	73.52
1969	1450.28	74	3.34	12.60
1970	962.09	75	26.48	43.64
1971	1358.91	61	38.61	340.22
1972	795.73	90	31.82	216.13
1973	823.03	93	7.90	316.85
1974	1221.62	79	7.65	255.52
1975	618.72	93	19.94	135.57
SUBTOTAL	(13152.01)	(69.5)	(327.82)	(1746.74)
1976	556.85	93	44.14	296.70
1977	1367.99	73	61.80	103.56
1978	346.81	86	278.37	593.06
1979	336.43	93	128.17	167.35
1980	424.24	86	198.39<-?->	151.40
1981	485.51	91	164.70	195.74
1982	349.04	88	207.00 <sup>c</sup>	??
1983	116.00 <sup>b</sup>	93	92.00 <sup>d</sup>	??
TOTALS	(17134.88)	(75)	(1502.39)	(3254.55+??)

Table 2-5f. References and Notes

- a. Except where indicated, from US Department of Energy, Nuclear Materials Management and Safeguards System, NMMSS Report TJ-25, 28 March 1983, as quoted in Cochran et al, The Nuclear Weapons Databook II: The Production Complex (Cambridge, MA: Ballinger, to be published).
- b. US Department of Energy, Nuclear Materials Management and Safeguards System, 29 November 1984, as quoted in Cochran et al, ibid.
- c. Estimate by Cochran et al, ibid.
- d. US DOE, "News Fact Sheet on Foreign/Domestic Research Fuel Shipments," (Savannah River Operations Office, June 1983 and February 1984).

Table 2-5g. Origins of US HEU Exports, 1980-84<sup>a</sup>  
 [kg-U (percent U-235)]

<u>Source:</u>	<u>Stockpile<sup>b</sup></u>	<u>Toll-Enriched<sup>c</sup></u>
<u>Destination (Year)</u>		
Canada(1980)	93(WG <sup>d</sup> )	4(70 <sup>e</sup> )
(1981)	183(WG)	
(1982)	95(WG)	
(1983)	65(WG)	
France(1980)	10(WG)+ 48(45%)	
(1982)	5(WG)	
(1983)	20(WG)	
Japan (1980)	30(WG)	
(1981)	40(WG)	
(1982)	63(WG)	
(1983)	40(WG)	
W. Germany (1980)		287(WG)
--		19(47)
--		19(WG) <sup>e</sup>
(1981)		280(WG)
--		22(45)
--		7(WG) <sup>e</sup>
(1982)		150(WG)
--		43(47)
	<hr/> 644(WG) + 48(45)	<hr/> 743(WG) + 88(48)

Table 2-5g. References and Notes

- a. Source: US Department of Energy, Nuclear Materials Management and Safeguards System, Transaction Journal-21 (TJ-21), Mod 3, year-end report for these years.
- b. The origin is given as the "Y-12" plant at Oak Ridge. This plant is the original US plant for the production of highly-enriched uranium and the location where highly-enriched uranium hexifloride gas was reduced to metal prior to the end of US production of weapon-grade uranium in fiscal year 1964.
- c. Except where indicated, the source is the enrichment plant at Portsmouth, Ohio, where the US currently produces all of its highly-enriched uranium (for nonweapons purposes).
- d. WGU is weapons-grade uranium. The source gives the U-235 content in kilograms. When this is divided into the total amount of uranium the result is within rounding uncertainties of 93.5 percent.
- e. In this case, the source is given as United Nuclear Corporation, which fabricates naval fuel and evidently recovers HEU from scrap.



III. THE US STOCKPILE OF WEAPON-GRADE PLUTONIUM

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## Estimate Based on Fission Power Output

Our estimates of the amount of weapon-grade plutonium produced by the US start with our estimates of the amounts of U-235 fissioned in US plutonium production reactors. We base these estimates on recently-released information concerning the historical thermal power outputs of US production reactors at the Department of Energy's Hanford site on the Columbia River near Richland, Washington and the Savannah River site near Aiken, South Carolina.

Before this data became available, we based our estimates on public information about the quantities of 30-year half-life Strontium-90 and Cesium-137 present in the high-level radioactive waste at these sites. These estimates are quite close to those obtained from the thermal power data. We therefore preserve them in Appendix A for reasons of methodological interest.

The approximations used in making these estimates are summarized in Table 3-1.

A plutonium production reactor produces approximately one gram of "weapon-grade" plutonium (plutonium with 6 percent Pu-240<sup>3-3</sup>) per gram of U-235 fissioned. For graphite-moderated, water-cooled reactors of the<sub>3</sub> type used to produce plutonium at Hanford, the ratio is approximately 0.92. We will assume an uncertainty of 5 percent in this coefficient and the same value for the heavy-water-moderated production reactors at Savannah River. (Cochran *et al* assume a value of 0.96.<sup>3-2</sup> If these reactors were as neutron-efficient as Canadian heavy water "CANDU" reactors, the ratio would be slightly over one.<sup>3-1</sup> However, the available information about their design and operation suggests that the Savannah River reactors are probably less efficient plutonium producers.<sup>3-2</sup>

Weapon-grade plutonium is defined as plutonium with about 94 percent Pu-239.<sup>3-3</sup> Pu-239 is the first fissile isotope that is formed as a result of neutron capture on the abundant but non-fissile uranium isotope, U-238. The other six percent of weapon-grade plutonium is almost entirely Pu-240, which is formed as a result of the capture of a second neutron on Pu-239.

For every neutron capture that forms a Pu-240, approximately two cause fissions of Pu-239.<sup>3-1</sup> We will therefore assume that, for each gram of weapon-grade plutonium produced in a production reactor, 1.09 grams of U-235 and 0.12 grams of Pu-239 fission. These fissions are the principal source of the fission products in the radioactive waste at Hanford and Savannah River. They are also the source of the fission heat. If we assume that the fission of a gram of U-235 (Pu-239) releases 0.96 (0.98) Mwt-days of heat,<sup>3-4</sup> then the production of a gram of weapon-grade plutonium is accompanied by the release of approximately 1.16 Mwt-days of heat. Alternatively, for every terrawatt-day (1 TwtD = 10<sup>12</sup> Watt-days) of thermal power generated, US production reactors other than the N-reactor produced the equivalent of 0.86 +/- 0.04 tonnes of weapon-grade plutonium.

The information that has recently been made public by the Department of Energy gives the fission heat output of its operating Savannah River reactors for the fiscal years 1955-'83 and of the eight Hanford graphite-moderated reactors for the years 1950-1971 (see Tables 3-2). The cumulative estimated thermal outputs of the Savannah River (Hanford graphite reactors) shown in Table 3-2 is 57.5 (57.2) Twt-days -- corresponding (at 0.86 +/- 0.04) tonnes per Twt-day) to 49.4 (49.2) or a total of 99 +/- 5 tonnes of weapon-grade plutonium.

#### Correction for Tritium Production

We have assumed that the production reactors at Hanford and Savannah River were devoted entirely to plutonium production. This is, however, not the case. Perhaps most importantly, the production reactors at Savannah River have been used to produce tritium as well as plutonium. Tritium is used in the tiny electrostatic accelerator neutron generators that initiate the fission chain reaction in nuclear weapons and it supplies neutrons via the deuterium-tritium fusion reaction to "boost" the fraction of the fissile material in a weapon that is actually fissioned. In the past few years, tritium production rates have been increased to supply tritium for enhanced-radiation or "neutron" weapons. According to Cochran *et al*<sup>3-2</sup> tritium is produced in the Savannah River reactors as a biproduct of plutonium production in lithium control rods and blankets. They estimate a production rate of 0.002 grams (the neutron equivalent of 0.144 grams of weapon-grade plutonium) per thermal megawatt-day. This corresponds to an assumption that about 0.15 neutrons per fission -- almost one half of all\* neutrons not captured in U-235, U-238 or Pu-239 -- are captured in Li-6.

In addition to the production of tritium as a byproduct of plutonium production, Cochran *et al* have found references to the use of Savannah River reactors to produce tritium as a primary product during 1972 and

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\* We assume that 0.86 grams of weapon-grade plutonium are produced per MWt-day of fission, of which 6 percent (0.052 grams) are Pu-240. Since, for the production of one atom of Pu-240, two atoms of Pu-239 are fissioned, 0.104 grams of Pu-239 are fissioned per MWt-day. Given 0.98 (0.96) MWt-days of fission-heat per gram of Pu-239 (U-235) fissioned, this means that: 10 percent of the fission heat is due to the fission of plutonium; 9.8 percent of the fissions are fissions of Pu-239; and that 1.04 grams of fission occur per MwtD of fission heat and that 0.79 net atoms of Pu-239 and 0.05 atoms of Pu-240 are produced per atom fissioned. A fission of U-235 (Pu-239) at thermal (0.025 eV) energies releases 2.44 (2.90) neutrons. A weighted average fission therefore releases 2.49 neutrons. For each fission in U-235, there are 0.175 neutrons absorbed without fission -- yielding U-236. (U-235 and Pu-239 neutronics from ref. 3-4, Table 3-2. The fraction of nonfission captures shown in our Table 3-1 are somewhat different because in real reactors some captures happen before the neutrons are slowed to thermal energies.) This leaves a net of 2.31 neutrons -- of which: 1 is absorbed sustaining the chain reaction, 0.79 in producing Pu-239 and 0.10 in producing Pu-240 (two per atom). This leaves

to the use, since calendar year 1982, of one of the Savannah River production reactors to exclusively produce tritium. Cochran et al have also interpreted the increased rates of routine tritium releases per Mwt-day during the period 1956-'64 and during the year 1972 (see Table 3-2a) as indications of the dedicated tritium production required to create the initial US tritium stockpile. In the absence of dedicated production, the annual byproduct production, estimated by Cochran et al at about 2 kilograms per year during the period FY1977-1980 would have been sufficient to offset the losses by decay of a stockpile of about 40 kilograms. In any case, Cochran et al estimate that up to approximately 115 kg. of tritium might have been produced in dedicated production "campaigns" as of the end of 1983. This would reduce our estimate of the plutonium production from Savannah River by up to 8 tonnes.

#### Other Corrections

Other significant corrections to the US plutonium inventory as of 1984.0 come from the facts that:

- 1) The US may have imported on the order of one tonne of weapon-grade plutonium from Britain prior to 1964.<sup>3-7</sup>
- 2) We assume losses of 2 tonnes of plutonium. Losses in the Nuclear Fuel Services commercial fuel reprocessing plant which operated at West Valley New York during the period 1966-71 were 3 percent.<sup>3-9</sup> The Energy Research and Development Administration (the predecessor agency to the Department of Energy) reported in 1977 that, since the establishment of the Atomic Energy Commission in 1946 through Sept. 30 1976, there had been an accumulated Inventory Difference at its facilities (not including the Rocky Flats Plant near Golden, Colo.) of 1.49 tonnes of plutonium.<sup>3-8</sup>
- 3) Cochran et al estimate that other isotopes equivalent to up to 2 tonnes of plutonium were produced at Savannah River.<sup>3-2</sup>

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(footnote continued from previous page)

0.42 neutrons (17 percent of all neutrons) after absorption. For comparison, in the CANDU heavy water reactor, all but 10 percent of the neutrons are absorbed by the U-235, U-238, plutonium and fission products in the fuel. Of that ten percent: 0.85 percent are absorbed in the fuel cladding, 1.8 percent are absorbed in the heavy water moderator and coolant, 4.6 percent are absorbed in the structural material of the reactor and 2.5 percent leak out of the core.<sup>3-5</sup>

\* It takes one neutron to make either an atom of tritium or Pu-239. Pu-239 is 80 times heavier. However, the equivalence ratio is not 80 for weapon-grade plutonium because an extra neutron is required to produce an atom of Pu-240. Furthermore, the difference between the neutronics of U-235 and Pu-239 affects the neutron economy of the reactor. The resulting official equivalence factor is 72 grams of weapon-grade plutonium per gram of tritium.<sup>3-6</sup>)

- 4) Some plutonium (3 +/- 1 percent of the inventory if we correct by the ratio of the number of tests to the number of weapons currently in inventory [see Section II]) would have been consumed and dissipated in the testing of nuclear weapons.
- 5) As of March 31, 1983, the Department of Energy had about 10 tonnes of separated and 6 tonnes of unseparated (mostly in unprocessed N-reactor fuel) fuel-grade plutonium.<sup>3-10</sup> The DOE plans to upgrade some of this plutonium to weapon-grade by diluting it with "super-grade" plutonium (3 percent Pu-240) or subjecting it to laser isotope separation.<sup>3-11</sup> (However, 4 tonnes of the US stockpile of fuel-grade plutonium are from British civil reactors -- obtained during the 1960's under a U.S./U.K barter agreement for highly enriched uranium and tritium to be used in British military nuclear programs.<sup>3-12, 3-2</sup> In 1964, the United States gave assurances to Britain that this civilian plutonium would not be used for weapons purposes.<sup>3-13</sup>) As of 1984.0 the US inventory of weapon-grade plutonium had been increased by approximately 1 tonne,<sup>3-2</sup> as a result of blending of fuel-grade with super-grade plutonium.
- 6) Miscellaneous uses such as the exports, irradiation of weapon-grade plutonium to make high-burnup plutonium, and use for domestic R&D may have reduced the total by the order of another tonne.
- 7) The US also reprocessed some Canadian research reactor fuel in the late 1950's and early 1960's and recovered a fraction of a tonne<sup>3-14</sup> of weapon-grade plutonium that was added to the US stockpile.

#### Final Estimate

In Table 3-3, we take into account the largest of the above corrections, imbed the rest in an assumed uncertainty range of about 10 percent and conclude that the US stockpile of weapon-grade plutonium as of the beginning of 1984 was 90 +/- 7 tonnes. Including the up to 15 (8 +/- 8) tonnes of fuel-grade plutonium that is already produced and that the Department of Energy plans to convert to weapons-grade would raise the total to 98 +/- 11 or approximately 100 tonnes.

REFERENCES -- SECTION III

- 3-1. S.E. Turner et al, Southern Science Applications, Inc, Criticality Studies of Graphite-Moderated Production Reactors, (Report # SSA-125, prepared for US Arms Control and Disarmament Agency, Contract No. AC9NX707, January 1980).
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Table 3-1: Approximations Used in Deriving Estimates of Production of  
Weapon-Grade Plutonium  
(See text for references.)

- o Weapon-grade Pu is 94% Pu-239 and 6% Pu-240;
- o 0.92 +/- 0.05 grams of weapon-grade Pu were produced per gram of U-235 fissioned in US plutonium-production reactors (except for those reactors that were dedicated to tritium-production campaigns and with the exception of the Hanford N-reactor).
- o Heat released by fission:
  - 0.96 Megawatt-days per gram of U-235, and
  - 0.98           "           "           "           " Pu-239.
- o Ratio of total to fission captures of neutrons:
  - 1.25 on U-235, and
  - 1.50 on Pu-239.

Assumptions Below Used Only in Appendix A Approach

- o Based on the above, and Table 3A-1, 7.5 Curies of (Sr-90 plus Cs-137) would have been produced per gram of plutonium produced.
- o Inventories of Sr-90 plus Cs-137 were:
  - 250 MCi at Hanford in 1974.0 (when reprocessing stopped), and
  - 260 MCi at Savannah River in 1985.0.

Table 3-2a. Power History of Savannah River Production Reactors

<u>Fiscal Year</u> <sup>a</sup>	<u>Routine Releases</u> <u>of Tritium from</u> <u>Separations Areas</u> <sup>b</sup> (MCI) [MCI/TwtD] <sup>d</sup>	<u>Total Energy</u> <u>Output</u> <sup>c</sup> (TwtD)	<u>Subtotals</u> (incl. Hanford) [TwtD (end of CY)]
			4.66(CY54.5)
1955	0.02 [0.04]	0.51	
1956	0.42 [0.34]	1.23	
1957	1.12 [0.61]	1.84	20.67(CY57.5)
1958	2.25 [1.08]	2.08	
1959	0.82 [0.28]	2.92	
1960	0.645 [0.20]	3.15	
1961	0.654 [0.20]	3.25	
1962	0.736 [0.23]	3.15	
1963	0.736 [0.24]	3.12	
1964	0.936 [0.29]	3.25	62.04(CY64.0)
1965	0.311 [0.15]	2.14	
1966	0.301 [0.14]	2.21	
1967	0.308 [0.12]	2.63	
1968	0.411 [0.16]	2.51	
(number of production reactors reduced to three)			
1969	0.272 [0.16]	1.75	
1970	0.246 [0.17]	1.49	
1971	0.379 [0.27]	1.42	95.81(CY71.5)
1972	0.530 [0.30]	1.75	
1973	0.312 [0.17]	1.87	
1974	0.189 [0.10]	1.90	
1975	0.143 [0.10]	1.42	
1976 (5 quarters)	0.125 [0.08]	1.64	
1977	0.192 [0.15]	1.30	
1978	0.192 [0.16]	1.23	
1979	0.180 [0.15]	1.19	
1980	0.200 [0.14]	1.46	
1981	0.231 [0.17]	1.38	
1982	0.257 [0.14]	1.87	
1983	0.407 [0.22]	1.87	113.76(CY83.0)
1984		1.90 <sup>e</sup>	
TOTAL		59.42	
PER REACTOR-YEAR (through 1984 115 RY)		0.517	
PER REACTOR-YEAR (1969-'84)		0.53	
AVERAGE (FY1969-84)		1.59	

Notes for Table 3-2a

- a. In 1976, the end of the fiscal year was changed from June 30 to September 30.
- b. Values for 1955-'78 from Ashley, C; and Ziegler, C.C.: Releases of Radioactivity at the Savannah River Plant, 1954 Through 1978 (DPSPU 75-25-1, 1980) p. 102, as quoted in ref. 3-2. Later values estimated in ref. 3-2.
- c. Department of Energy National Security and Military Applications of Nuclear Energy Authorization Act of 1985, Hearings before the Procurement and Military Nuclear Systems Subcommittee of the House Committee on Armed Services, Feb.-Mar. 1984, p. 333.
- d. One MCi of tritium is approximately equal to 100 grams.
- e. From House Armed Services Committee, Fiscal Year 1986 Authorization Hearings, p. 240 (cited in ref. 3-2).

Table 3-2b. Power History of the Original Eight Hanford Reactors

<u>Calendar</u> <u>Year</u>	<u>Output</u> <sup>a</sup> (Twt-days)
1944	0.021
1945	0.120
1946	0.141
1947	0.128
1948	0.160
1949	0.213
1950	0.307
1951	0.50
1952	1.00
1953	1.25
1954	1.50
1955	2.25
1956	2.50
1957	3.40
1958	2.90
1959	4.25
1960	4.75
1961	4.30
1962	4.50
1963	4.90
SUBCY64.0	39.16
1964	4.02
1965	3.14
1966	3.55
1967	2.40
1968	2.20
1969	1.95
1970	0.75
1971	0.00
TOTAL	57.10

a. Values from Cochran et al, Nuclear Weapons Databook II: The Production Complex (Cambridge Mass: Ballinger, to be published). Values for 1951-1971 from letter from John L. Meinhardt, Director, Office of Nuclear Materials Production (Department of Energy) to Cochran. Earlier values are estimates by Cochran, based on the assumption that the original reactors were rated at 250 Mwt each and operated at 70 percent capacity.

Table 3-3. Steps in Making the Final Estimate of US Weapon-Grade Plutonium

(as of 1 January 1984)

(tonnes)

Equivalent of Fission Heat Produced by Savannah River and Original Eight Hanford Production Reactors (0.86 gms/Mwtd)	99 +/- 5
Imports from Britain	1
Increase Due to "Blending" of Fuel-Grade Plutonium	1
Reductions for Estimated Tritium Production at Savannah R.	-4 +/- 4
Reductions for the Production of Other Isotopes	-1 +/- 1
Process losses	-2
Consumption in Nuclear Weapons Tests	-3 +/- 1
Miscellaneous	-1
FINAL	90 +/- 7*

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\* The cumulative uncertainty is calculated as the square root of the sum of the squares of the contributing uncertainties.

Appendix A. Estimate Based on the Sr-90 and Cs-137 Inventories at the Department of Energy's Production Sites

Strontium-90 is produced in 5.8 percent of all fissions of U-235 (2.1 percent of all fissions of Pu-239 by thermal neutrons) and has a half-life of 29 years. Cs-137 is produced by 6.2 percent of all fissions of U-235 (5.8 percent of all fissions of Pu-239 by thermal neutrons) and has a half-life of 30.1 years (see Table 3-A1.) Therefore, approximately 3.6 Curies of Sr-90 and 3.9 Curies of Cs-137 or 7.5 Curies of the combined isotopes would have been produced by the plutonium production reactors per gram of weapon-grade plutonium produced.

As of the end of 1974.0, after the reprocessing of production reactor fuel at Hanford had stopped, the high level radioactive waste storage tanks there contained approximately 250 million Curies of Sr-90 and Cs-137.<sup>3A-1</sup> As of 1985 (we assume 1985.0), it was expected that the high level radioactive waste tanks at Savannah River would contain approximately 260 million Curies of Sr-90 and Cs-137.<sup>3A-2</sup>

These are not, of course, the original quantities of Sr-90 and Cs-137 produced because there has been some considerable amount of radioactive decay since the fission products were produced. Table 3-A2 gives the dates of the Hanford and Savannah River reactors. The fourth column shows the fraction that remained out of the original amount of Sr-90 and Cs-137 that would have been produced by each reactor as of the end of 1973 at Hanford and as of the end of 1984 at Savannah River if each reactor had operated at constant power throughout its life.

In fact, the average output of the individual production reactors increased considerably with time -- especially at Hanford. To show the sensitivity of the decay factors to such an effect, we also give in Table 3-A2 decay factors for each reactor for a case in which the reactor power would have increased linearly with time and reached a plateau only after ten years. It will be seen that the decay factors increase by only about 5 percent.

There is also the fact that the later production reactors built at Hanford had much larger capacities than the first three. One cannot therefore simply take an average of the individual Hanford reactor decay factors weighted by the associated equivalent full-power reactor-years and obtain an overall decay factor for the Hanford radioactive waste. However, the weighted average of the decay factors is relatively insensitive to the individual weights assigned. For example, giving the first three Hanford reactors a relative weight of zero would only change the range of the weighted average decay factor for the Hanford reactors from 0.71-0.74 to 0.74-0.78. To allow for this uncertainty, we take a range of uncertainty for the weighted-average Hanford decay factors to be 0.71-0.78.

Using the weighted average decay factors in Table 3-A2, we translate the inventories of Sr-90 plus Cs-137 at Hanford and Savannah River into estimates of the equivalent of 91-98 tonnes of weapon-grade plutonium produced and separated at the two sites: 43-47 at Hanford and 48-51 at

Savannah River. This corresponds to about 0.3 and 0.44 tonnes per average production reactor-year respectively. The corresponding average power outputs are 940 and 1430 Mw(thermal) per reactor at Hanford and Savannah River respectively.

The above estimates should be subjected to two relatively small corrections stemming from the following facts:

- 1) Some research reactor and early power reactor fuel has been reprocessed at Savannah River and Hanford and has contributed the fission product equivalent of one to two tonne of U-235 fission to the radioactive waste products stored there. We neglect this correction (see Section II).
- 2) The Hanford N-reactor has, until recently, been producing "fuel-grade" (9-12 percent Pu-240) almost exclusively rather than weapon-grade plutonium. Assuming a one-year delay between fission and reprocessing, an amount of N-reactor fuel with a total burnup of about 5.2 TwtDs would have been reprocessed during the period prior to 1972 while the Hanford Purex plant was operating. Of this an amount with a total burnup of 0.8 TwtDs was reprocessed at the short-lived commercial reprocessing plant that operated at West Valley New York. The N-reactor produced about 0.7 grams of plutonium per MwtD of fission energy released. Therefore, approximately 8 percent of the Cs-137 plus Sr-90 in the Hanford wastes in 1974 would have been from reprocessed N-reactor fuel and our estimate of the amount of weapon-grade plutonium produced at Hanford should be reduced by approximately 3.5 tonnes.

Taking these corrections into account, results in an estimate of US production of weapon-grade plutonium through 1983 of 87-95 tonnes. This estimate should be compared with the estimate of 99 +/- 5 tonnes estimated on the basis of the thermal outputs of the Savannah River and original eight Hanford reactors before corrections (see Table 3-3).

#### APPENDIX A. REFERENCES

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- 3A-2. US Department of Energy, 1978, Long-Term Management of Defense High-Level Radioactive Waste, Savannah River Plant, Draft Environmental Impact Statement, (DOE/EIS-0023-D), Table IV-6.

Table 3-A1. Fission Yields of Cs-137, Sr-90 and Kr-85

<u>Fissioning Isotope</u>	<u>Neutron Energy</u>	<u>Fission Yield</u> [Percent(Curies/gram)]			
		<u>Cs-137</u>	<u>Sr-90</u>	<u>Kr-85</u>	<u>%Kr/%Sr</u>
U-235	thermal	6.19(3.14)	5.77(3.03)	0.285(0.405)	0.05
U-235	fission	6.22(3.15)	5.47(2.88)	0.273(0.388)	0.05
U-235	fusion	4.92(2.49)	4.58(2.41)	0.347(0.493)	0.08
Pu-239	thermal	6.71(3.34)	2.10(1.09)	0.127(0.177)	0.06
Pu-239	fission	6.51(3.24)	2.02(1.04)	0.125(0.174)	0.06
U-238	fusion	5.00(2.50)	3.16(1.59)	0.200(0.281)	0.06

Source: Rider, D.F., 1981: Compilation of Fission Product Yields, (Nuclear Fuel and Services Engineering Department, General Electric Co., Vallecitos Nuclear Center, Pleasanton, CA 94566, NEDO-12154-3(C).



Table 3-A2. Dates of Operation of US Plutonium Production Reactors

<u>Reactor</u>	<u>Startup</u> <sup>a</sup>	<u>Shutdown</u> <sup>a</sup>	<u>Decay Factors</u> <sup>b</sup>		<u>Equivalent Full-power reactor-years</u>	
			<u>Step</u>	<u>Ramp</u>	<u>Total</u>	<u>Before 1966</u>
<u>Hanford</u>			<u>As of 1974.0</u>		<u>Step(Ramp)</u>	<u>Step(Ramp)</u>
B	1944.7	1968.1	0.67	0.70	23.4(18.4)	21.3(15.3)
D	1944.9	1967.5	0.67	0.70	22.6(17.6)	21.1(16.1)
F	1945.1	1965.5	0.65	0.68	20.4(15.4)	20.4(15.4)
H	1949.8	1965.3	0.68	0.72	15.5(10.5)	15.5(10.5)
DR	1950.8	1965.0	0.69	0.72	14.2 (9.2)	14.2 (9.2)
C	1952.9	1969.3	0.74	0.78	16.4(11.4)	13.1 (8.1)
KW	1955.0	1970.1	0.77	0.81	15.1(10.1)	11.0 (6.0)
KE	1955.3	1971.0	<u>0.78</u>	<u>0.82</u>	<u>15.7(10.7)</u>	<u>10.7 (5.7)</u>
TOTAL (WEIGHTED AVERAGE) (weighted by full-power reactor-years)			0.70	0.73	143.(103.3)	127.3(86.3)
N	1964.0	1971.7 <sup>c</sup>	<u>0.87</u>	----	<u>7.7</u>	<u>2.0</u>
<u>Savannah River</u>			<u>As of 1985.0</u>		<u>Total Before 1984.0</u> <sup>d</sup>	
R	1954.0	1964.5	0.55	0.57	10.5 (5.5)	
P	1954.1	----	0.70	0.74	29.9(24.9)	
L	1954.5	1968.1	0.58	0.60	13.6 (8.6)	
K	1955.0	----	0.71	0.75	29.0(24.0)	
C	<u>1955.2</u>	<u>----</u>	<u>0.71</u>	<u>0.75</u>	<u>28.8(23.8)</u>	
WEIGHTED AVERAGE			0.68	0.72	111.8(86.8)	

NOTES: TABLE 3-A2

- a. See e.g. Cochran, Thomas B., Arkin, William M. and Hoenig, Milton M., Nuclear Weapons Databook, Volume II: The Production Complex (Cambridge, Mass: Ballinger, to be published).
- b. Fraction of original Cs-137 plus Sr-90 remaining as of end of 1973 for Hanford and as of the end of 1984 for Savannah River. Given a power curve which rises linearly from zero at time  $T_0$  to a plateau at time  $T_1$  and then stays constant to a time  $T_3$ , the fraction of the original Sr-90 plus Cs-137 which will remain undecayed at time  $t$  after shutdown is

$$F_t = [2/(2c_{30} - c_{10})] * (\exp(-c_{t3}) + [\exp(-c_{t0}) - \exp(-c_{t1})] / c_{10})$$

$$\begin{array}{l} \text{---> } (\exp(-c_{t3}) - \exp(-c_{t0}) / c_{30} \quad \text{where} \\ T_1 \text{--->} T_0 \end{array}$$

$$c_{ij} = (T_i - T_j) / 42.6$$

and 42.6 years is the average of the exponential decay lives of Sr-90 and Cs-137. For the "step" case, we have assumed  $T_1 - T_0 = 0$ . For the "ramp" case, we have assumed that  $T_1 - T_0 = 10^1$  years.

- c. End of reprocessing at Hanford (see ref. a) minus one year for fuel cooling.
- d. We assume a year's delay between fission and addition of the fission products to the high-level waste inventory.

IV. DISPOSITION OF THE US GOVERNMENT'S STOCKPILE OF NATURAL URANIUM

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- 4-1. Disposition of the US Government's Stockpile of U-235
- 4-2. Commercial Nuclear Power Plants with Low-Enriched Fuel Outside the Soviet-bloc
- 4-3. Excess Uranium Feed Requirements of US Enrichment Plants Planned in 1975 for the Period 1975-'85
- 4-4. Enrichment Requirements of the Light Water Reactors Outside of the Centrally-Planned Economies

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## IV. DISPOSITION OF THE US GOVERNMENT'S STOCKPILE OF NATURAL URANIUM

### Introduction

This section is provided as an overall consistency check on our various estimates of US production of highly-enriched uranium and plutonium for weapons and uranium of various levels of enrichment for reactor fuel. The check is to see whether these various estimated uses and the associated waste streams can account approximately for the disposition of the government's stockpile of natural uranium.

### US U-235 Purchases

From 1944 through 1971, the US government bought approximately 250,000 tonnes of natural uranium containing 0.711 percent U-235 for a total of approximately 1780 tonnes of U-235 (see Table 2-1). The government has not bought any more uranium since.

### Dispositions

Stockpiles of Natural and Depleted Uranium. Some of the government's U-235 seems to be stockpiled in the form of natural or low enriched uranium. As of 30 September 1980, the amount of uranium in this form was equivalent to 67,702 tonnes of natural uranium.<sup>4-1</sup> This quantity of natural uranium would contain 481 tonnes of U-235.

### Weapon-Grade Uranium -- Pre-1965

From the numbers in Table 2-2 we can estimate that 675 +/- 22 tonnes of U-235 ended up either in weapon-grade (93.5 percent U-235) or in 97.3 percent enriched uranium for naval reactors produced prior to 1965. Assuming that the ultimate enrichment of the associated depleted uranium were 0.2 percent, the associated depleted uranium would contain 216 +/- 7 tonnes of U-235 (see Table 2-1, note c).

### Naval Reactor Fuel -- 1965-1980

In Section II, we estimate that the US Navy orders 0.7 +/- 0.35 grams of U-235 per shaft-horsepower year and that the fuel in 1980 was being ordered about 10-15 years ahead of the average date of fission. (The first core is ordered when the reactor is ordered. Subsequent cores are ordered when the previous core is loaded. Average core life is currently about 10 years.) In Table 2-3, we estimate that the US nuclear fleet had accumulated 40 million shaft-horsepower years as of the end of 1980 and accumulated an additional 4.0 million during 1984. If we assume that the fleet averages 5 +/- 1 million shaft-horsepower per year over the 10-15

years after 1981.0, then the US Navy should have bought enough highly-enriched uranium by 1981.0 to fuel the nuclear navy for 100 +/- 16 million shaft-horsepower years. The associated amount of U-235 would be 70 +/- 35 tonnes of which we have attributed 11 +/- 5 tonnes to the period prior to 1965 (see Table 2-2). This leaves 60 +/- 30 tonnes for the period 1965-80, with which we associate 23 +/- 11 tonnes of U-235 in 0.002 percent depleted uranium.

#### Original Eight Hanford Reactors

In Table 3-2b, we estimate that the total energy output of the original Hanford reactors was 57.1 Twt-days, corresponding to the fission of about 53.5 tonnes of U-235 (see Table 3-1 for assumptions). In Section II, we assume that this U-235 was in uranium enriched to 0.9 percent and that 44 percent of the U-235 originally in the fuel was fissioned. Assuming that one atom of U-235 was converted to U-236 for every four atoms of U-235 fissioned, then 69 tonnes of U-235 would have been consumed in the reactors, 55 tonnes would remain in the "burned out" uranium fuel and 9 tonnes would have ended up in the associated 0.002 percent depleted uranium.

#### Low-Enriched Uranium for Savannah River Reactors

According to Cochran et al,<sup>4-2</sup> with the exception of tritium-production campaigns, the Savannah River reactors were fueled with either natural or low-enriched uranium until 1968, when they were converted to highly-enriched uranium. As of the end of fiscal year 1967, the Savannah River reactors had generated 31.5 Twt-days of heat (see Table 3-2a). According to section III, the tritium equivalent of up to 7 tonnes of weapon-grade plutonium may have been produced in this period with (according to Table 3-1) an associated 7.6 Twt-days of fission heat released in highly-enriched uranium. We therefore subtract 4 +/- 4 Twt-days for this purpose giving 28 +/- 4 Twt-days of fission heat that would have been released in the natural or low-enriched fuel. If we assume that the Savannah River reactors had the same types of fuel cycles as the original eight Hanford reactors when not operating on high-enriched fuel, then associated with the 28 +/- 4 Twt-days of fission heat would have been 36 +/- 5 tonnes of U-235 fissioned or converted to U-236, 29 +/- 4 tonnes of U-235 in burned out uranium fuel, and 5 +/- 1 tonnes in the associated 0.002 percent depleted uranium.

#### Demand by the N-Reactor

The<sup>4-3</sup> fresh fuel of the N-reactor has two levels of enrichment: 1.25 and 0.95%. Subsequent to its startup period and prior to its recent conversion to the production of weapon-grade plutonium, the fuel of this reactor seems to have operated most of the time at a fuel burnup of 2850 MwD(th)/tonne, at which burnup<sup>4-4</sup> the irradiated fuel contained about 2 kg. of plutonium per tonne of uranium. Assuming that this plutonium averaged

12% Pu-240 and that 1.25 atoms of U-235 are destroyed for every U-235 atom fissioned, the corresponding reduction in the U-235 enrichment in one pass-through of this fuel through the N-reactor would be about 0.3%.

The obvious interpretation, therefore, is that the 0.95% enriched fuel is once-recycled uranium. The twice-recycled fuel would then have an enrichment of 0.65% which, prior to 1971, might have been used as a substitute for natural uranium in one of the eight original production reactors. We will assume that this was so. (The last of the eight original Hanford reactors was shut down at the beginning of 1971.)

Due to the shut-down of the Hanford Purex reprocessing plant in 1972, an amount of irradiated N-reactor fuel containing approximately 2440 tonnes of uranium was in spent fuel storage at Hanford as of the end of 1980.<sup>4-2</sup> Given the rated power of 3850 Mw(thermal) and the average capacity factor for electricity generation of .425 for the period 1966-1981,<sup>4-2</sup> and assuming the above burnup, this amount of spent fuel would correspond to 11.6 years' production at this rate. We assume this means that the reprocessing of N-reactor fuel ended in 1969.4. In that case, at the same rate, the N-reactor would have consumed approximately 3.4 tonnes of U-235 in the period 1964-1969.4 in addition to the 7.3 tonnes fissioned or converted to U-236 in the unprocessed fuel stored at Hanford at the end of 1980. In addition, there would be approximately 6 tonnes of U-235 in the (0.2 percent) enriched tails associated with the production of this fuel and 20 tonnes of U-235 remaining in the unprocessed fuel (see Table 4-1).

#### Power Reactor Demand Prior to 1969

The US first began offering toll-enrichment services -- i.e.,<sup>4-5</sup> the enrichment of uranium not owned by the US government -- in 1969. Prior to that date we assume that all the uranium enriched for power reactors by the US came from the government's own stockpile. Prior to 1969, the US government was essentially the only source of low-enriched uranium for power reactors in the US, Western Europe and Japan.<sup>4-6</sup> We therefore assume that, through 1970, essentially all the capacity shown in Table 4-2 was supplied by uranium from the US government stockpiles enriched prior to the beginning of toll enrichment in 1969.

According to Table 4-2, 23 thermal Gw of power reactor capacity fueled with low-enriched uranium was operating outside of the Soviet bloc at the end of 1970 and had operated for 77.12 GWt capacity-years. According to ref. 4-7, a large pressurized or boiling water reactor requires 100-120 tonnes of natural uranium to provide its startup core and, if operated at an average capacity factor of 70 percent will require an additional 40 tonnes/year. On this basis, about 5600 tonnes of natural uranium from the US government's stockpiles would have been used by the end of 1969 to supply fuel for the nuclear power plants shown in Table 4-2. Assuming an average enrichment of 3 percent, 77 percent or 31 tonnes of the U-235 would have ended up in the fuel and 9 tonnes in the 0.002 percent depleted tails. These estimates are quite uncertain for a number of reasons: the average

"burnup" of the U-235 in early reactor fuel was typically 12 Gwt-days/metric-tonne-uranium (see Table 5-8) versus the approximately 30 Gwt-days/MTU assumed in ref. 4-7 and the average capacity factor was typically closer to 50 percent than 70 percent.<sup>4-8</sup> We therefore double the above estimate and assume a 50 percent uncertainty in Table 4-1.

#### Power Reactor Demand After 1969

Partly because of concern that the demand for the US government's enrichment services might soon exceed the supply, after 1969, the US government operated its plants at a higher tails assay, requiring less enrichment work but more uranium feed per unit of product. The government supplied private and foreign utility customers with some of the additional U-235 required in lieu of some enrichment work. The government charged these customers for enrichment work and requested them to supply natural uranium calculated according to "transaction tails" for the enrichment plants that were lower than the "operating tails" that were actually in effect.

We assume that the government did carry out its enrichment program through 1979 as projected in 1975 (see Table 4-3) and subsequently, as the prospect of excess enrichment demand faded, kept its enrichment tails transaction assay in line with its operating assay. Then the actual reduction of the stockpile would have been only by the amount of government uranium in low-enriched uranium actually shipped off site due to difference between the operating and transaction tails during the period 1972-1979. (Actually, the programmed difference between operating and transaction tails was to be very small in 1977 and zero in 1978 and 1979. Virtually all of the shipments would therefore have occurred during the period 1972-'76.)

If we assume that the low-enriched fuel for power reactors had an average enrichment of 3 percent during this period (the result would differ by less than 10 percent if we had assumed 2 percent), then by the end of fiscal 1977, 7,200 tonnes of the government's natural uranium would have been used to produce low-enriched fuel for US and foreign power reactors (see Table 4-3). The resulting low-enriched uranium would have contained 33 tonnes of U-235 and there would be 18 tonnes of U-235 in the associated enrichment plant tails (approximating the enrichment of the depleted uranium as 0.003 percent during this period). We assume an uncertainty of 30 percent on this estimate.

#### Comparison of Purchases With Estimated Dispositions

It will be seen from Table 4-1 that the consistency between our estimated dispositions of the government's U-235 in Table 3 and the amount of U-235 in the natural uranium originally purchased is remarkably good, given the uncertainties in the estimates and our neglect of process losses. This provides a good check of our understanding of the government's nuclear enterprise and its relationships with commercial nuclear power.



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- 4-5. Brewer, Shelby T., Assistant Secretary for Nuclear Energy, US Department of Energy, Answers Submitted for the Record, in Energy and Water Development Appropriations fo 1985, Hearings before a Subcommittee of the House Committee on Appropriations, 19 March 1984, p. 939.
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- 4-8. Howles, L.R., "Nuclear Station Achievement, Annual Review, 1978" Nuclear Engineering International, March 1979, pp. 66-67.
- 4-9. Hill, James H. and Joe W. Parks, Uranium Enrichment in the United States, (Washington DC: Energy Research and Development Administration, Report #CONF 750324-7, 1975).

Table 4-1. Disposition of the US Government's Stockpile of U-235  
(as of the end of 1980)

	<u>tonnes</u>
<u>Amount of U-235 in Natural Uranium Purchased (1944-71)</u>	<u>1780</u>
<u>Disposition:</u>	
Associated With Government Stockpiles of Natural and Low-enriched Uranium	481
In highly-enriched uranium produced prior to 1965	675 +/- 22
-- In the associated depleted uranium	216 +/- 7
In naval reactor fuel produced during the period 1965-80	60 +/- 30
-- In the associated depleted uranium	23 +/- 11
Consumed in the original 8 Hanford production reactors	69
-- In the associated "burned out" uranium	55
-- In the associated depleted uranium	9
Consumed in the Savannah R. reactors	
-- Consumed in natural or low-enriched fuel	36 +/- 5
-- In the associated burned out uranium	29 +/- 4
-- In the associated depleted uranium	5 +/- 1
Consumed by the N-reactor	11
-- In its unreprocessed spent fuel (as of the end of 1980)	23
-- In the associated enrichment plant tails	6
In low-enriched uranium shipped to power reactors prior to 1969	62 +/- 31
-- In the associated enrichment plant tails	18 +/- 9
In low-enriched uranium shipped to power reactors because of "split" diffusion plant "tails" 1972-'77	33 +/- 10
-- In the associated enrichment plant tails	<u>18 +/- 6</u>
<b>TOTAL DISPOSITIONS</b>	<b>1823 +/- 53</b>

Table 4-2. Capacity of Nuclear Power Plants Using Low-Enriched Fuel in NonCommunist Nations

<u>Year</u>	<u>Capacity (Gwth)<sup>a</sup></u>					<u>Total</u>	<u>Cumulative (Gwt-yr)</u>
	<u>US</u>	<u>FGR</u>	<u>Japan</u>	<u>Sweden</u>	<u>Other</u>		
1957	0.24					0.24	0.24
1958	"					"	0.48
1959	"					"	0.76
1960	0.94					0.94	1.70
1961	1.54					1.54	3.24
1962	2.16	0.06				2.22	5.46
1963	2.61	"	0.09			2.76	8.22
1964	"	"	"		0.51	3.27	11.47
1965	"	"	"		1.38	4.14	15.61
SUB	(13.19)	(0.24)	(0.27)		(1.89)	(15.61)	(15.61)
1966	"	0.86	"		"	4.94	20.53
1967	"	"	"		2.42	5.98	26.51
1968	5.79	2.43	"		"	10.73	37.24
1969	9.73	"	"		4.88	17.13	54.37
1970	13.26	"	2.18		"	22.75	77.12
1971	22.2	"	3.56		6.26	34.45	111.57
1972	34.8	6.23	5.02	1.375	8.39	55.82	167.39
1973	55.9	"	"	"	9.76	78.29	245.68
1974	87.6	"	11.22	"	"	116.19	361.87
1975	108.5	9.75	15.31	7.22	13.61	154.39	516.26
1976	120.2	12.25	25.02	9.49	13.61	180.57	696.83
1977	139.8	18.28	26.67	11.19	16.27	212.21	909.04
1978	146.0	20.86	37.16	11.19	25.22	240.43	1149.47

Table 4-2. References

a. "Power Reactors, 1982", Nuclear Engineering International, August 1982, Supplement. See also Thomas L. Neff and Henry D. Jacoby, The International Uranium Market, (MIT Energy Lab Report # MIT-EL 80-014, 1980), Table A-9.

Table 4-3. Excess Uranium Feed Requirements of US Enrichment Plants  
 (Actual through 1974, projected thereafter)  
 (10<sup>3</sup> metric tonnes)

<u>Fiscal</u> <u>Year</u>	<u>Tails</u> <sup>a</sup>		<u>Due to Diff.</u> <u>in Op. &amp; Trans.</u> <u>Tails</u> <sup>b</sup>	<u>Contract</u> <u>SWUs</u> <sup>c</sup>
	<u>Operating</u>	<u>Transaction</u>		
1972	0.003	0.002	0.72	2.835
1973	0.003	0.002	1.14	4.505
1974	0.003	0.002	1.24	4.911
1975	0.003	0.002	1.82	7.2(5.8)
1976	0.003	0.002	2.07	8.2(6.4)
1977	0.0028	0.00275	0.21	12.31
1978	0.00275	0.00275	0.00	0.00
1979	0.00275	0.00275	0.00	0.00
TOTAL			7.20	

Table 4-3, References

a. Projections of enrichment supply and demand, and operating and transaction tails from: James H. Hill and Joe W. Parks, Uranium Enrichment in the United States, (Energy Research and Development Administration, CONF-750324-7, 1975).

Table 4-3 -- Notes (cont.)

b. The following formulae relating separative work units (D) and tails assay ( $x_w$ ) to product quantity (P) enrichment ( $x_p$ ) and feed (F) were taken from Gaseous Diffusion Plant Operations, (USAEC, #ORO-684, 1972), Appendix 2:

$$F/P = (x_p - x_w)/(x_F - x_w), \text{ and}$$

$$D/P = [V(x_p) - V(x_w)] - (F/P)*[V(x_F) - V(x_w)]$$

where

$$V(x) = (2*x - 1)*\ln[x/(1-x)].$$

For 3 percent enriched product we have  $V(0.03) = 3.268$ , for 2 percent, 3.736 and, for natural feed, we have  $V(0.00711) = 4.869$ . Consequently, we have, for various values of  $x_w$ :

<u>Product Enrichment</u> $x_w$	<u><math>V(x_w)</math></u>	<u>3 percent</u>			<u>2 percent</u>		
		<u>F/P</u>	<u>D/P</u>	<u>F/D</u>	<u>F/P</u>	<u>D/P</u>	<u>F/D</u>
0.00335	5.657	7.088	3.198	2.216	4.428	1.568	2.824
0.00300	5.771	6.569	3.424	1.919	4.136	1.696	2.439
0.00280	5.842	6.311	3.569	1.768	3.991	1.777	2.246
0.00275	5.861	6.250	3.607	1.733	3.956	1.799	2.200
0.00200	6.188	5.479	4.306	1.272	3.523	2.195	1.605

For split tails, the excess uranium feed requirements per SWU are:

$$\text{Excess Feed} = [(F/P)_O - (F/P)_T]x(P/D)_T x(\text{SWUs}).$$

where the subscript O denotes operating and T denotes transaction. This ratio is only weakly dependent upon the product enrichment in the range considered here, so we have assumed an enrichment of 3 percent.

c. We use the SWU demand calculated in table 4-4 prior to 1975 and the demand projected in ref. a subsequently. The values in parenthesis for 1975 and 1976 show that the values calculated in ref. a are about 25 percent higher than those calculated in Table 4-4.

Table 4-4. Enrichment Requirements of the Light Water Reactors Outside of the Centrally-Planned Economies

Year	Tails Assay <sup>a</sup>	V(x <sub>w</sub> ) <sup>b</sup>	SWU/Gw(th)		Gw(th)		SWU <sup>c</sup>	
			Annual	Initial	Annual	Initial	Total	
			(10 <sup>3</sup> )					
1955	.00278	5.85	26.7	47.9	--	--	0.011	0.011
1956	.00163	6.397	43.3	78.3	--	0.010	--	0.010
1957	.00199	6.193	36.9	66.7	.24	0.009	--	0.010
1958	.00297	5.782	24.8	44.2	.24	0.006	0.031	0.037
1959	.00339	5.645	20.7	37.1	.24	0.019	0.022	0.042
1960	.00337	5.651	21.0	37.4	.94	0.032	0.025	0.058
1961	.00343	5.633	20.5	36.5	1.54	0.046	0.020	0.065
1962	.00341	5.639	20.6	36.8	2.22	0.057	0.019	0.076
1963	.00313	5.728	23.2	41.4	2.76	0.076	0.036	0.112
1964	.00285	5.824	25.9	46.5	3.27	0.107	0.037	0.144
1965	.00197	6.203	37.3	67.3	4.14	0.184	0.069	0.253
1966	.00200	6.188	36.8	66.4	4.94	0.220	0.315	0.535
1967	.00200	6.188	36.8	66.4	5.98	0.395	0.425	0.820
1968	.00200	6.188	36.8	66.4	10.73	0.630	0.373	1.004
1969	.00200	6.188	36.8	66.4	17.13	0.837	0.777	1.614
1970	.00200	6.188	36.8	66.4	22.75	1.268	1.419	2.687
1971	.00200	6.188	36.8	66.4	34.45	2.054	1.492	3.546
1972	.00300	5.771	24.4	43.7	55.82	1.910	0.925	2.835
1973	.00300	5.771	24.4	43.7	78.29	2.835	1.669	4.504
1974	.00300	5.771	24.4	43.7	116.19	3.767	1.144	4.911
SUBTOTAL								(23.27)
1975	.00300	5.771	24.4	43.7	154.39	4.406	1.383	5.788
1976	.00300	5.771	24.4	43.7	180.57	5.178	1.233	6.411
TOTAL								(35.47)
1977					212.21			
1978					240.43			

Table 4-4. References

- a. See Table 2-1.
- b. See Table 4-2, note b.
- c. We assume that enrichment is provided two years before the fuel is loaded.
- d. We assume an annual requirement of 8.6 tonnes of 3 percent enriched uranium per GWth capacity (see ref. 4-7). This assumption corresponds to a 70 percent capacity factor which is somewhat higher than actually experienced but it also corresponds to an average fuel burnup of 30 GWt-days/tonne-U which is also somewhat higher than the norm during this period.
- e. We assume an initial requirement of 25.9 tonnes of 2.2 percent enriched uranium per GWth capacity (see ref. 4-7).

Table 4-5. Enrichment Conversion Factors

<u>F.Y.</u>	<u>Tails Assay</u> <sup>a</sup>	<u>P/D(0.935)</u> (kg/SWU)	<u>R(0.935/0.973)</u> <sup>b</sup>
1945	.00529?	0.00641	1.05
1946	.00529	0.00641	
1947	.00488	0.00616	
1948	.00515	0.00632	1.05
1949	.00506	0.00627	
1950	.00495	0.00621	
1951	.00506	0.00627	
1952	.00447	0.00597	
1953	.00438	0.00591	
1954	.00366	0.00546	
1955	.00278	0.00485	
1956	.00163	0.00393	1.05
1957	.00199	0.00424	
1958	.00297	0.00499	
1959	.00339	0.00528	
1960	.00337	0.00527	
1961	.00343	0.00531	
1962	.00341	0.00529	
1963	.00313	0.00510	
1964	.00285	0.00490	

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Table 4-5. References

a. See Table 2-1.

b. The ratio of the amount of product that can be produced with a given amount of enrichment work for an enrichment of 93.5 percent (weapon-grade uranium) and for an enrichment of 97.3 percent (the level used in naval reactor fuel).

$$V(0.935) = 2.32, V(0.94) = 2.42, V(0.973) = 3.39, V(0.00711) = 4.87$$





## V. THE SOVIET PLUTONIUM STOCKPILE

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## Measuring Soviet Plutonium Production Through Associated Releases of Kr-85 to the Atmosphere

An upper-bound estimate of Soviet plutonium production can be obtained by estimating the Soviet contribution to the buildup of Krypton-85 (Kr-85) in the earth's atmosphere. The connection comes from the fact that plutonium is produced by the capture of neutrons on U-238 in nuclear reactor fuel. The neutrons are generated principally by the fission of U-235 with some small contribution from the fission of Pu-239. And Kr-85 is one of the fission products produced when U-235 or Pu-239 fissions.

When U-235 is fissioned by a thermal neutron, Kr-85 (or a short-lived atom which decays into Kr-85) is produced with a 0.285 percent probability. This translates into the production of 0.405 Curies of Kr-85 per gram of U-235 thermal fission. Kr-85 is also produced with a probability of approximately 0.127 percent by the fission of Pu-239. (See Table 5-1.) The Kr-85 accumulates in nuclear reactor fuel (on average, much less than one percent leaks out of power reactor fuel<sup>5-1</sup>) and is ordinarily released to the atmosphere only when such fuel is chemically "reprocessed." Its accumulation in the atmosphere, when corrected for radioactive decay, is therefore a rough indication of the amount of fission that has occurred in reprocessed fuel. Lesser amounts of Kr-85 have also been released to the atmosphere by nuclear weapons tests and leakage from unprocessed fuel. Because Kr-85 is chemically inert, it accumulates in the atmosphere. Because it has a relatively long radioactive half-life (10.76 years), it has time to mix relatively uniformly throughout the atmosphere - a fact which facilitates estimates of its global inventory.

### Measurements of the Concentration of Kr-85 in the Atmosphere

Measurements of the concentration of Kr-85 in the atmosphere have been reported since<sub>3</sub>1954. (The unit of concentration which will be used in this paper is pCi/m<sup>3</sup>-air at standard pressure and temperature. Table 5-2 presents the factors required to convert to the other units used in the literature.) Unfortunately, most of the accurate published measurements have been made in Europe within a few hundred kilometers of the major nuclear fuel reprocessing centers of France and the United Kingdom. There is a three-fold problem in extracting a global average concentration from these measurements:

- 1) Kr-85 concentrations can be more than doubled hundreds of kilometers downwind of reprocessing plants.
- 2) Even when the wind is blowing from another direction, the background concentration of Kr-85 in ground level air in mid-Northern latitudes is elevated because releases from the world's reprocessing plants occur there. This air circulates relatively rapidly around the globe (weeks) but takes much longer to mix with the air at higher altitudes and other latitudes.

- 3) The concentration of Kr-85 in Northern troposphere air is elevated relative to stratospheric and Southern Hemispheric air because the mixing across the tropopause and across<sup>5-2</sup> the intertropical convergence near the equator is particularly slow.

These considerations make non-European measurements particularly useful - even when they are somewhat less accurate. Several north-south profiles of the concentration of Kr-85 taken on sea-voyages in the Atlantic Ocean have been reported for: 1964, 1971 and 1972, late 1980, early 1981, winter 1981-'82, winter 1982-'83, spring 1983, and fall 1983. Two of the profiles (for 1964 and 1972) are given in Table 5-3a. The six profiles from the early 1980's are given in Table 5-3b.

As can be deduced from the data in Table 5-3, the sea-level average concentration of Kr-85 increased at an average annual rate of about 0.5 pCi/m<sup>3</sup>-air between 1964 and 1984. Interpolation gives a value of about 12.6 (14.4) pCi/m<sup>3</sup> for the global (North Hemisphere) average in 1973.25, the center of the period for<sup>5-3</sup> which Telegdas and Ferber report Kr-85 measurements for the stratosphere. The North Hemisphere number falls in the middle of the range of 13.2-16.2 pCi/m<sup>3</sup> which Telegdas and Ferber quote for January-June 1973 for "10 stations in the contiguous United States." They estimate a North Hemisphere tropospheric average for 1973.6 which is somewhat lower, 13.8 pCi/m<sup>3</sup>, and a stratospheric average which is lower still, 10 pCi/m<sup>3</sup>.

Since there are no other published measurements of the stratospheric concentration of Kr-85, we have assumed in Table 5-4 that the ratios between the stratospheric concentration and the Northern troposphere ground-level concentration are constant and equal to the levels measured in 1973.

The time development of the ratios between the Northern and Southern Hemisphere Kr-85 concentrations does not show an easily interpretable pattern. To make possible the extrapolation of the Kr-85 concentrations in the Southern hemisphere to the period before 1964, we we have therefore used a simple two-box diffusion model suggested by Weiss et al.<sup>5-2</sup> We have fixed the one free parameter, the mixing rate between Northern and Southern hemisphere air using the average for the concentration ratio in the 1980's. The model is described in Appendix 5-A.

Early reference points for the<sup>3</sup> Northern Hemisphere surface level Kr-85 concentration are 0.5 and 1.9 pCi/m<sup>3</sup> in 1955.5 and 1958.5, respectively, obtained from a curve interpolating earlier measurements (ref. given in Table 5-6, note b). We use these values and the two-box model to estimate the corresponding concentrations in the Southern hemisphere (see Table 5-5). The concentrations in the various compartments of the stratosphere are obtained from the corresponding Northern Hemisphere ground-level averages using ratios measured by Telegdas and Ferber in 1973 (see Table 5-4).

## Reconstruction of Total Releases of Kr-85 into the Atmosphere

The concentrations in the different compartments were weighted by the fractions of the atmosphere's mass in these compartments (see Table 5-4) to calculate in Table 5-6 the global atmospheric Kr-85 inventories for the years 1955.5, 1958.5, 1964.9, 1972.5, 1981.0 and 1984.0. We also give in Table 5-6 our estimates of the corresponding amounts of Kr-85 that decayed in the intervening periods. This makes it possible to make the estimates, also shown there, for the cumulative total releases of Kr-85 into the atmosphere. Estimates by Jacob *et al* for 1981.0 and 1984.0, made using a global circulation model to obtain a self-consistent atmospheric distribution from the same Atlantic north-south concentration profiles, the estimates in this report of non-Soviet Kr-85 sources and derived Soviet Kr-85 sources gives results for the global inventory 1.5 and 1.8 percent higher respectively than ours -- well within our assumed 5 percent overall uncertainty (see Table 5-6).

### Contributions to the Atmosphere's Kr-85 from Sources Other Than Soviet Nuclear Reactors

Reprocessing of US Plutonium Production Reactor Fuel. The annual total releases of Kr-85 from the Savannah River site have been published for the period 1971-1983 (see Table 5-7a) and for the Hanford site for the year 1972 (the last year of reprocessing until 1983 -- see Table 5-7b). The releases from these sites for other years have been estimated from the data on the power output of the associated reactors as a function of time, assuming a one-year delay between fission and release of the Kr-85 to the atmosphere. A check on this methodology in the case of the Savannah River reactors shows that the average releases reported during 1971-83 and the predictions obtained from the reported thermal power output and one-year decay time are in quite good agreement.

Reprocessing at West Valley, New York. During the period 1966-'71, a short-lived commercial reprocessing operation existed at West Valley, New York. Data is available on the quantities and types of fuel reprocessed and their "burnup" measured in megawatt-days of thermal energy released per kilogram of fuel. (See Table 5-8.) On this basis, one may estimate the associated Kr-85 releases. Some of the fuel that was reprocessed was from the Atomic Energy Commission's N-reactor at Hanford, which was being used for both electricity and plutonium production. We assume a delay of two years between the production of Kr-85 by fission in this fuel and the release of the Kr-85 to the atmosphere by reprocessing. On the basis of the initial full-power operating dates of the reactors which provided most of the commercial reactor fuel, we have estimated for their fuel an average 5-year delay between fission and reprocessing.

Reprocessing of US Naval and Research Reactor Fuel. To our knowledge, all US naval reactor fuel is reprocessed at the Idaho Chemical Processing Plant (ICPP). Most US research reactor fuel is reprocessed there as well (see Table 2-5a). Table 5-9 shows the reported data on Kr-85 releases from the ICPP during the period 1953-'83. There is a gap during the period 1957-'63

but the resulting probable uncertainty is not very large. Not all of the Kr-85 in the fuel reprocessed at Idaho is released, as this facility is equipped with a "rare gas recovery facility." However, "the actual quantity of krypton removed from the ICPP off-gas stream is small compared to the total released."<sup>5-4</sup>

Some research reactor fuel has been reprocessed at the Savannah River Plant (see Table 2-5a). We estimate the corresponding fission products as the equivalent to 2 tonnes of U-235 fission (see Table 2-4b). Assuming an average five-year delay between fission and reprocessing, this fuel would have released about 0.6 MCi of Kr-85 to the atmosphere. We assume that half of this Kr-85 was released during the period 1965-72.5 and half during the period 1972.5-1983 (see Table 5-17).

Nuclear Explosions. The 1977 report of the United Nations' Scientific Committee on the Effects Atomic Radiation (UNSCEAR) presents an estimate of the release of Kr-85 to the atmosphere, scaled from measurements of Sr-90 assuming a Kr-85/Sr-90 fission ratio of 0.07. (The ratios in Table 5-1 would suggest a value of 0.06 [assuming that most of the fission was the fission of U-238 by fusion neutrons] but an estimated 28 percent of the fission yield in pre-1963 tests occurred in tests which were surface-burst and for which therefore a significant fraction of the Sr-90 would not have gone into global fallout.) Based upon cumulative Sr-90 injections into the stratosphere of 16 MCi, this report arrives at an estimate of 3 MCi of Kr-85 injected into the atmosphere by nuclear explosions (0.015 MCi/Mt).<sup>5-5</sup> According to the 1964 UNSCEAR report (ref. 5-6, Table IV), the total Sr-90 inventory in global fallout was already 13 MCi in January 1964. Table 5-10a shows the time distribution of the total fission yield of 194 Mt in atmospheric tests reported before the US-Soviet-UK treaty limiting nuclear testing to the underground.

There have subsequently been approximately twice as many tests. Most have these have been underground and the average yield has been less than for the atmospheric tests. It is also not clear how much of the Kr-85 is trapped underground. In Table 5-10b, we estimate that, if the Kr-85 all leaked to the surface within a time short in comparison with its half-life, US and Soviet underground tests would have released less than one MCi to the atmosphere.

Nuclear Fuel Reprocessing in Britain. Britain reprocesses virtually all of its nuclear reactor fuel. We can therefore estimate the Kr-85 releases from the British reprocessing plant at Windscale from the amount of fission heat generated by British power reactors. In Table 5-11, we show our estimates of the cumulative amount of fission heat released in these reactors through 1981. We also show the increase in the estimated fuel "burnups" in British power reactors, and the associated decline in the amount of Kr-85 produced per Mwt-day because of the increasing fraction of the heat coming from the fission of Pu-239 and the increased time delay between the average time of formation of the Kr-85 and its release at the reprocessing plant. The resulting estimated annual releases of Kr-85 from the Sellafield (formerly Windscale) reprocessing plant are shown on Table

5-12. As a check, we compare in Table 5-12 our estimates of the releases from Sellafield with the reported annual Kr-85 releases since 1971, when release figures began to become available. The agreement between the averages is very good.

Nuclear Fuel Reprocessing in France. In Table 5-13, we show the available data on the fuel reprocessed at France's two reprocessing plants. In Table 5-14, we compare the estimated Kr-85 releases from these plants, assuming no delay between fission and reprocessing and the data reported by France to the European Economic Community for the years 1972-1980. It will be seen that, through 1978, even before allowing for decay of the Kr-85, the estimates of the amount of Kr-85 in the reprocessed fuel are lower than the reported data on releases. Obviously, either the data on the reprocessed fuel or the reported Kr-85 releases are incorrect. The average reported releases at Marcoule are about 10-15 percent higher than we would estimate if we assumed a one year delay between fission and reprocessing -- as would be appropriate for the early years when the fuel had low burnups (see the corresponding assumptions on Table 5-11). The reported releases at La Hague are about 50 percent higher than those which we would estimate, if we assume 3 years average decay before reprocessing (a low value considering the burnups of the fuel reprocessed there). There have been no responses to our repeated requests to the French government for clarification of this anomaly. Fortunately, however, since the discrepancy seems to have disappeared in the data for 1979-80, the uncertainty in the French releases introduced by this mystery is less than one MCi.

Together, the United Kingdom and France have released about two thirds as much Kr-85 to the atmosphere as has the US. A much smaller tonnage of fuel has been reprocessed in Europe, however, since the European commercial fuel had on average a much higher burnup and therefore released much more Kr-85 to the atmosphere per tonne than the US production reactor fuel.

Other Reprocessing Plants: Finally, a number of smaller reprocessing plants have operated in Europe and Japan. We show the available information about these operations in Table 5-15 and, on this basis, estimate that they have released a total of about 1.5 MCi of Kr-85 to the atmosphere.

Leakage from Unreprocessed Fuel. In Table 5-16, we give annual amounts of electrical energy generated in the nuclear power plants of the non-Communist countries between 1960 and 1983 and an estimate of the nuclear-electric energy generated in the USSR through 1982. We also give in Table 5-16 an estimate of the amount of Kr-85 that has been produced in this fuel. During 1979, the production of approximately  $11.2 \times 10^3$  GwD(e) of nuclear-electric energy by US light water reactors was associated with the release of 0.019 MCi of Kr-85 -- approximately 0.2 percent of the 11.5 MCi produced. <sup>5-7</sup> If we assume that this has been the average leakage rate for all the fuel in the NonCommunist states, then the cumulative leakage would be about 0.7 MCi. However, the amount of Kr-85 which has accumulated in unreprocessed fuel is now so large that our results for recent years would be significantly affected if the leakage rate were 1 percent or higher.

The amount of Kr-85 that has been produced in Soviet electric power reactors in recent years is about one tenth as great as in the power reactors of nonCommunist states and the Soviet Union accounts for most of the nuclear electric power capacity in Communist countries. Leakage from unprocessed fuel in Communist states is therefore unlikely to have been a major contributor to the atmosphere's inventory of Kr-85 -- unless the percentage leakage from this fuel is an order of magnitude higher than that from nuclear fuel in nonCommunist states. We have no reason to believe that this is so -- but the paucity of data from the USSR means that we also are not certain that it is not so. Some further investigation on this point would be useful.

#### Estimate of Soviet Plutonium Production

In Table 5-17, we subtract our estimate of the total releases of Kr-85 into the atmosphere from reprocessing plants and leaking nuclear reactor fuel in nonCommunist countries and from all nuclear weapons explosions from the results shown for the cumulative releases of Kr-85 into the atmosphere. As of the end of 1983 we obtain a range of estimates for Kr-85 releases from the Soviet Union of  $60 \pm 10$  MCi. If this release were all due to the reprocessing of fuel from Hanford-type reactors, with a release of 0.43 MCi of Kr-85 per gram of weapon-grade plutonium produced (see Table 5-7a, note a) the corresponding amount of weapon-grade plutonium would be  $140 \pm 25$  tonnes. This assumption is, of course, an overestimate since, as in the US, some of the Soviet Kr-85 releases must come from the reprocessing of naval, research, commercial power, and tritium production reactor fuel. In the case of the US, the same approach would result in an estimate of 110 tonnes of weapon-grade plutonium, whereas our estimate based on other data which takes into account reprocessing not associated with the production of weapon-grade plutonium reduces this estimate to about  $90 \pm 7$  tonnes -- with, however, up to 13 tonnes of US-origin plutonium available from the upgrade of fuel-grade and reactor-grade plutonium (see Section III).

In an effort to obtain information about the rate of Soviet releases of Kr-85 into the atmosphere as a function of time, we have, in Table 5-17, divided the Western contributions to the atmosphere's inventory into the same time periods over which we have estimated the total additions. For the first two periods, because of uncertainties in the atmosphere's inventory of Kr-85, we can only estimate upper bounds on the Soviet contribution. This contribution rises clearly out of the background associated with nonSoviet releases by 1965, however. Between 1972.5 and 1984.0, it appears that both these releases and those from reprocessing in Western Europe exceeded those from the US. A plausible interpretation is that, unlike the US, the Soviet Union had not, as of the end of 1983, cut back the rate of its production of plutonium. For the period 1972.5-1984.0, our estimate of Soviet Kr-85 releases would correspond to an annual production of  $7.4 \pm 2.8$  tonnes of weapon-grade plutonium -- comparable to the peak rate of US production in the early 1960's.



Soviet Reprocessing of Power Reactor Spent Fuel. Based on rather sparse indications, it appears that, with regard to the reprocessing of power reactor fuel, the Soviet situation is more similar to that in the US (i.e. very little civilian reprocessing) than to that in Western Europe (quite a bit). The primary projected use of plutonium recovered from the fuel of current-generation reactors is apparently to fuel plutonium breeder reactors and, due to delays in breeder reactor deployment, plans for the reprocessing of large amounts of spent power reactor fuel have apparently also been delayed.

According to the report of a US Department of Energy contractor, the Soviet Union has not yet built a commercial-size reprocessing plant for power reactor fuel. A 3 kg U/day experimental-scale facility began operation in 1973 that reportedly processes spent fuel from pressurized water reactors and fast breeder reactors.

Nuclear Fuel reported in 1983, citing Soviet documents, that the spent fuel from the RBMK-1000 graphite-moderated (Chernobyl-type) reactors is being stored in at-reactor pools. According to the same report, a firm choice between reprocessing the fuel or storing it in an away-from-reactor facility has not yet been made.

Concern has been expressed that the Soviet Union might use its RBMK power reactors to produce weapon-grade plutonium. The fact that these reactors are refueled while they are in operation means that more frequent refueling could be accomplished without frequent reactor shutdowns. The resulting weapon-grade plutonium could presumably be recovered at military reprocessing facilities. The first set of 100 Mw(e) reactors of this type, of which the first came into operation in 1958, were, in fact, "dual purpose," in that they produced plutonium as well as electricity. However, in 1983 Congressional testimony, a Department of Energy official stated that he was not "personally aware of any good evidence that would say that they [the Soviets] are" using the RBMKs to obtain weapon-grade plutonium.

Plutonium Utilization and Production by Soviet Breeder Reactors. The Soviet Union operates two demonstration-size breeder reactors. The BN-350 began operation in 1973. Originally, it was designed to produce the equivalent of 350 MWe but, due to problems with the steam generators soon after it began operation, it was derated. Since 1980 the reactor has generated 700 MWt of heat which provides for the generation of 120 MWe of electricity and 85,000 tonnes of distilled water per day (equivalent to 280 MWe of electricity). The BN-600 (600 MWe and 1470 MWt) breeder reactor began operation in April 1980 and reached nominal power in late 1981. Through 1981 it had produced 420 MWe-yr of electricity. Through June 1, 1985 it had produced 1,600 MWe-yr of electricity. Both these breeder reactors use medium enriched uranium fuel instead of plutonium fuel.

Each of the two Soviet breeder reactors creates super-grade plutonium in its uranium "blanket" surrounding its core. Using French data, about 360 kilograms of plutonium are produced per GWe-yr on the basis of a 100 percent capacity factor.<sup>5-17</sup> Through 1983, the BN-350 would have produced roughly 600 kilograms of super-grade plutonium. Using the electricity data stated above for the BN-600, through 1983 about 400 kilograms of super-grade plutonium could have been produced in its blanket.

Kr-85 Capture at Soviet Reprocessing Plants. It appears that the Soviet Union, like the US, captures and uses some of the Kr-85 released at its reprocessing plants. According to a 1982 press release issued by the Idaho National Engineering Laboratory,

Krypton-85 is in short supply. It now sells for about \$70 a curie in the U.S. The only other world supplier, the Soviet Union, sells the gas at approximately \$300 a curie.<sup>5-18</sup>

We assume that, as in the case of the US, the amount of Kr-85 captured in the Soviet Union is a negligible fraction of that released from reprocessed fuel.

#### Acknowledgement

We would like to thank Daniel Jacob of Harvard University's Center for Earth and Planetary Physics for pointing out some errors in an earlier version of this section.

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Appendix 5-A. Model for the Ratio of the Kr-85 Inventories of the Northern and Southern Hemispheres

The equation for the total amount of Kr-85 ( $N_+$ ) in the atmosphere is

$$\frac{dN_+}{dt} = -\frac{N_+}{T_d} + I.$$

Here  $T_d$  is the exponential decay lifetime of Kr-85 (15.5 years) and  $I$  is the rate of input. The solution to this equation that is zero at  $t = 0$  is:

$$N_+ = \exp(-t/T_d) \times [\text{Integral from 0 to } t \text{ of } I(t') \times \exp(t'/T_d)].$$

If we approximate the atmosphere by a two boxes with equal volume which exchange their contents with a characteristic time constant  $T_e$ , then the equation for the (north-south) difference in the Kr-85 contents of the two boxes ( $N_-$ ) is

$$\frac{dN_-}{dt} = -N_- \left[ \frac{2}{T_e} + \frac{1}{T_d} \right] + I,$$

where we have made the (good) approximation that all the sources of Kr-85 are in the Northern Hemisphere.

We now note that, to a good approximation, the global average ground-level concentration (which we assume to be proportional to  $N_+$ ) can be approximated by (see Tables 5-3)

$$N_+ = 0.553 \cdot t,$$

where  $t$  is the number of years since 1950. The corresponding

$$I = 0.553 \cdot (1 + t/T_d)$$

and the corresponding solution for

$$r_{-/+} = \frac{N_-}{N_+} = (T'/T_d) \cdot \left( 1 + (1/t) \cdot (T_d - T') \cdot [1 - \exp(-t/T')] \right)$$

where

$$T' = 1 / \left[ \frac{1}{T_d} + \frac{2}{T_e} \right].$$

Since  $T_e$  is of the order of one year, one may solve for it by fitting  $r_{-/+}$  at  $t = 32$  years (see Table 5-3b). This gives  $T_e = 1.8$  years, which we may then use to solve for the S/N concentration ratio as a function of time

$$S/N = (1 - r_{-/+}) / (1 + r_{-/+}).$$

(See Table 5-5.) It will be seen from Table 5-4 that the predictions of this model fall within the range of uncertainty for the most recent measurements. They differ with the measurements in 1965 and 1972 by

approximately the quoted uncertainty of the individual points (about 10 percent). This may reflect either some systematic errors in the earlier measurements or an oversimplification of our two-box model. In either case, an uncertainty in the Southern Hemisphere inventory of 10 percent would translate into an uncertainty of the global inventory of about 5 percent.

Table 5-1 (Table 3-2 is ident.). Fission Yields of Cs-137, Sr-90 and Kr-85<sup>a</sup>

<u>Fissioning Isotope</u>	<u>Neutron Energy</u>	<u>Fission Yield</u> [Percent(Curies/gram)]			
		<u>Cs-137</u>	<u>Sr-90</u>	<u>Kr-85</u>	<u>%Kr/%Sr</u> <sup>b</sup>
U-235	thermal	6.19(3.14)	5.77(3.03)	0.285(0.405)	0.05
U-235	fission	6.22(3.15)	5.47(2.88)	0.273(0.388)	0.05
U-235	fusion	4.92(2.49)	4.58(2.41)	0.347(0.493)	0.08
Pu-239	thermal	6.71(3.34)	2.10(1.09)	0.127(0.177)	0.06
Pu-239	fission	6.51(3.24)	2.02(1.04)	0.125(0.174)	0.06
U-238	fusion	5.00(2.50)	3.16(1.59)	0.200(0.281)	0.06

Other Assumptions:

Conversion from grams fissioned to Mwt-days thermal. We assume 0.96 and 0.98 Mwt-days/gram for U-235 and Pu-239 respectively (see Table 3-1).

Treatment of Other Plutonium Isotopes. We make the approximation that all plutonium fissioned is Pu-239.

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a. Rider, D.F., 1981: Compilation of Fission Product Yields, (Nuclear Fuel and Services Engineering Department, General Electric Co., Vallecitos Nuclear Center, Pleasanton, CA 94566, NEDO-12154-3(C)).

b. The half-lives of Cs-137, Sr-90 and Kr-85 are 30.1, 29 and 10.73 years respectively. In order to obtain relative yields expressed in Curies, it is necessary to multiply the fission yield ratio by the corresponding ratio of the decay rates (the inverse of the half-lives).

Table 5-2. Conversion Factors Between Different Measures of the Krypton-85 Concentration in the Atmosphere<sup>a</sup>

<u>Unit</u>	<u>Factor to Convert to Equivalent in pCi/SCM-Air</u> <sup>b</sup>
dpm/cm <sup>3</sup> -Kr	0.514
dpm/mmol-Kr	0.0229
Bq/SCM	27.03
pCi/kg-Air	1.2929
dpm/kg-Air	0.582

a. Based on a concentration of krypton in the atmosphere of 1.14 ppm (Lawrence Berkeley Laboratory: Environmental Instrumentation Survey, Vol. I: Radiation, Second Edition [Wiley, 1983].)

b. pCi =  $10^{-12}$  Curies = 0.037 disintegrations/sec; SCM = a standard cubic meter of dry air at one atmosphere (760 mm Hg) pressure and 20 °C temperature; dpm = disintegrations per minute; mmol =  $10^{-3}$  moles; Bq = Becquerel = 1 disintegration/sec.

Table 5-3a. Measured North-South Profiles of the Concentration of Kr-85 at Sea-Level in 1964 and 1972 (pCi/SCM-Air)

<u>Latitude</u>	<u>Area Weighting<sup>a</sup> Factor</u>	<u>1964.9<sup>b</sup></u>	<u>1972.5<sup>c</sup></u>
90-N			
55-N	0.103		15.3
50-N	0.046		16.0
47-N	0.156	9.9	
40-N	0.049	10.6	
39-N	0.093		14.9
32.5-N	0.055	8.8	
25-N	0.057	8.0	
23-N	0.074		13.4
20-N	0.045		14.2
18-N	0.062	8.9	
12-N	0.047		13.3
10-N	0.090	8.1	
9-N	0.026		13.3
6-N	0.078		11.1
3-S	0.139	8.0	
9-S	0.104		10.5
18-S	0.062		10.6
22-S	0.111	9.3	
24-S	0.048		10.3
30-S	0.049	8.2	
30-S	0.088		10.1
35-S	0.053	6.2	
45-S	0.081	6.8	
48-S	0.071		9.2
53-S	0.024		9.2
57-S	0.017		8.9
60-S	0.074		9.4
62-S	0.062	7.8	
<u>74-S</u>	<u>0.036</u>	<u>10.1</u>	
90-S			
Area-Weighted Averages		8.6	12.0
Linear Interpolation <sup>d</sup> (to 1964 -1983 data)		(8.3)	(12.5)
Area and Troposphere Height- Weighted Averages <sup>e</sup>		8.6	12.1
Troposphere and Stratosphere Weighted Averages (using the stratospheric weighting factors in Table 5-4)		8.2	11.6
Uncertainty Range (based on approx. +/- 10 % uncertainties of indiv. measurements shown in original papers)		7.3-9.3	10.6-12.6



(Table 5-3a, cont)

<u>Year</u>	<u>1964.9</u>	<u>1972.5</u>
N. Hemisphere Area- Weighted Averages (Weiss Interpolation) <sup>f</sup>	9.2 (9.7)	14.0 (15.4)
Average Annual Increase	---0.64----	
S. Hemisphere Area- Weighted Averages (Weiss Interpolation) <sup>f</sup>	8.0 (6.5)	10.0 (12.0)
S/N Ratio	0.87	0.71

a. Each measurement is weighted by a factor proportional to the area of the globe between a latitude midway between the latitude of measurement ( $L$ ) and the latitudes of the adjacent measurements to the north ( $L_+$ ) and south ( $L_-$ ):  $\{\sin[(L_+ + L)/2] - \sin[(L + L_-)/2]\}/2$ .

b. Pannetier, R.: "Original Use of the Radioactive Tracer Gas Krypton 85 to Study the Meridional Atmospheric Flow," Journal of Geophysical Research 75 (1970), p. 2985. See also, Distribution, Atmospheric Transfer and Balance of Krypton-85 (French Thesis, Faculty of Sciences, Orsay, 1968 and Report# - CEA-R-3591). [Translation, BNWL-TR-34]. (Original numbers reported as dpm/cm<sup>3</sup>-Kr.)

c. Gudkov, A.N., V.I. Ivanov, L.L. Karol', V.M. Kolobashkin, O.I. Leypunskiy, V.I. Nekrasov, V.P. Novichkov, Yu. A. Serbulov and N.P. Ushakova, "Latitudinal Distribution of Krypton 85 in the Atmospheric Surface Layer (from the Results of R/V Akademik Kurchatov Expeditions). Abstracts of International Conference on "Physical Aspects of Air Pollution," Vil'nyus, June 1974. As shown in Karol, I.L.; Babanova, V.V.; and Romanovskaya, L.A.: "Global Dispersion of 'Inert' Gases in the Atmosphere from Ground Sources," Atmospheric and Oceanic Physics 12 (1976), p. 787. The same data is plotted in a more easily read form in Rozanski, K.: "Krypton-85 in the Atmosphere 1950-1977: a Data Review," Environment International 2 (1979), p. 139. (Original numbers reported as dpm/mmol-Kr.)

d. We use the fit  $-27.6 + 0.553xY$  where  $Y$  is the year of the 20th century. This is close to the best linear fit to the North Hemisphere averages in Tables 5.3.

e. We follow Telegdas and Ferber<sup>5-3b</sup> and assume that the troposphere has a height of 0.9 of the atmosphere between 30-S and 30-N latitudes (one half of the earth's surface) and 0.74 of the atmosphere elsewhere. The corresponding weighting factors are therefore 1.10 for 30-S to 30-N and 0.9 elsewhere.

f. W. Weiss, private communication, Freiburg, W. Germany, 29 January 1985.

Table 5-3b. Measured North-South Profiles of the Concentration of Kr-85 at Sea-Level -- Measurements in the Early 1980's  
(Measurement [pCi/SCM-Air]xArea Weighting Factor<sup>a</sup>)

<u>Latitude</u>	<u>1980.8<sup>b</sup></u>	<u>1981.3<sup>b</sup></u>	<u>1982.0<sup>c</sup></u>	<u>1983.1<sup>c</sup></u>	<u>1983.2<sup>c</sup></u>	<u>1983.8<sup>c</sup></u>
53.5-N			20.3x0.114			
50.0-N					20.6x.123	22.0x.131
48.0-N					20.8x.021	
47.5-N		20.1x0.150	19.4x0.034			
46.8-N	18.9x0.154					
45.0-N						20.0x.032
44.0-N				20.4x0.179		
43.0-N					20.9x.032	
41.9-N		20.0x0.032	19.2x0.034			
40.6-N	18.2x0.055					
39.5-N						19.7x.035
38.0-N					20.2x.038	
36.9-N		19.1x0.040	19.3x0.034			
36.0-N				19.2x.049		
34.5-N						19.8x.036
32.0-N			19.2x0.037		20.6x.039	
31.3-N	18.7x0.075					
30.7-N		19.2x0.046				
30.0-N				19.2x0.037		
29.5-N						20.1x.036
27.5-N					20.0x.039	
27.0-N			19.2x0.039			
26.0-N				19.9x0.056		
25.0-N						20.1x.040
24.5-N		19.6x0.055				
22.0-N			19.1x0.040		19.3x.042	
20.8-N	17.8x0.086					
19.5-N						19.8x.043
17.0-N			19.3x0.042		19.7x.042	
16.8-N		18.5x0.060				
16.0-N				19.8x0.069		
14.5-N						18.9x.043
12.0-N			19.0x0.041		19.4x.047	
9.9-N		19.0x0.061				
9.4-N	16.4x0.085			19.3x0.058		17.9x.063
7.5-N			18.6x0.043			
6.0-N					19.2x.032	
4.5-N					18.7x.014	
2.8-N		16.4x0.063			18.5x.017	
2.5-N				19.2x0.041		
2.0-N			17.6x0.046			
0.9-N	16.4x0.094					
0.5-N					18.4x.030	
0.0-N				18.2x0.031		18.1x.065
3.0-S			17.4x0.044			
4.0-S					17.9x.055	

(Table 5-3b, cont. 1)

Year:	<u>1980.8</u> <sup>b</sup>	<u>1981.3</u> <sup>b</sup>	<u>1982.0</u> <sup>c</sup>	<u>1983.1</u> <sup>c</sup>	<u>1983.2</u> <sup>c</sup>	<u>1983.8</u> <sup>c</sup>
4.7-S		16.6x0.054		17.5x0.052		
5.5-S						17.4x.046
8.0-S			16.7x0.043			
9.6-S		16.5x0.050				
10.5-S						17.4x.043
12.1-S	16.5x0.090			17.4x0.059	17.8x.068	
13.0-S			16.7x0.045			
15.2-S		16.5x0.040				
15.5-S						17.5x.042
18.5-S			16.9x0.043	17.5x0.047		
19.9-S		16.4x0.039				
20.0-S	16.0x0.068				17.3x.057	
20.5-S						17.5x.345
23.5-S			16.9x0.044	17.4x0.041		
24.9-S		16.2x0.040				
26.0-S					17.2x.045	
28.8-S	15.9x0.293			17.3x0.035		
29.5-S			17.0x0.047			
30.1-S		16.2x0.038				
31.5-S					17.4x.037	
32.5-S				16.9x0.041		
35.0-S		16.4x0.231				
36.0-S			16.6x0.037		17.3x.027	
39.0					17.2x.033	
40.0-S			16.8x0.056	17.0x0.049		
46.0-S					17.5x.037	
47.0-S				16.9x0.046		
51.0-S					17.2x.026	
53.5-S			16.5x0.060			
55.5-S				17.0x0.041	17.4x.023	
60.5-S					17.0x.019	
62.5-S			16.4x0.028			
63.5-S				17.1x0.030		
64.5-S					17.4x.015	
67.0-S			16.5x0.015			
68.5-S					17.5x.015	
71.0-S			16.7x0.008	16.9x0.018		
72.5-S			16.7x0.008			
74.0-S					16.9x.011	
75.5-S				16.9x0.007		
77.0-S			16.4x0.006			
77.5-S				16.8x0.014	17.2x.015	
78.0-S			16.6x0.012			
Area-Wtd Aye.	17.0	17.8	18.0	18.5	18.7	18.7
Linear Fit <sup>d</sup>	(17.3)	(17.6)	(17.9) (17.7) <sup>e</sup>	(18.6)	(18.6)	(19.0)
Year	1980.8	1981.3	1982.0	1983.1	1983.2	1983.8

Year (Y)	Table 5-3b, Notes					
	'80.8	'81.3	'82.0	'83.1	'83.2	'83.8
N. Hemisphere Area- Weighted Averages (-23.6 +0.52*Y)	18.3 (18.4)	19.2 (18.7)	19.2 (19.0)	19.8 (19.6)	20.0 (19.7)	20.0 (20.0)
(Freiburg Interp.) <sup>f</sup>	(20.4)	(20.7)	(21.1)	(21.4)		
(Nevada Interp.) <sup>g</sup>	(22.2)	(22.6)	(23.2)	(24.1)		
(Ghent Interp.) <sup>h</sup>	(17.7)	(18.2)	(18.8)	(19.9)	(20.0)	(20.6)
S. Hemisphere Area- Weighted Averages (Weiss Interpolation) <sup>f</sup>	15.7 (16.1)	16.4 (16.4)	16.8 (16.8)	17.2 (17.5)	17.4	17.5
S/N Ratio	0.82	0.85	0.88	0.87	0.87	0.88

a. Each measurement is weighted by a factor proportional to the area of the globe between a latitude midway between the latitude of measurement (L) and the latitudes of the adjacent measurements to the north ( $L_+$ ) and south ( $L_-$ ):  $(\sin[(L_+ + L)/2] - \sin[(L + L_-)/2])/2$ . At the northernmost (southernmost) measurement points,  $L_+$  ( $L_-$ ) is taken to be 90-N (90-S).

b. Weiss, W.; Sittkus, A; Stockburger, H; and Sartorius, H., "Large-Scale Atmospheric Mixing Derived From Meridional Profiles of Krypton-85", Journal of Geophysical Research, Vol. 88, No. C13, pp. 8574-8578 (1983).

c. Weiss, W. private communication, Freiburg, W. Germany, January 1985.

d. We fit the 1980's points with the formula  $-28.3 + 0.564xY$ , where Y is the year of the 20th century (e.g. 80).

e. The prediction obtained from a fit to the 1964 and 1972 as well as 1980's points:  $-27.6 + 0.553xY$ .

f. W. Weiss, priv. communication, Freiburg, W. Germany, 29 January 1985. The North Hemisphere values were obtained by extracting a background level from the measurements taken at an observation station near Freiburg. Weiss suggested that the ratio between the Northern Hemisphere tropospheric average concentration and the Freiburg baseline should be about 0.95. We find a factor of 0.915 for the ratio of the N. Hemisphere ocean surface average and the Freiburg baseline. Weiss also suggested a ratio of 0.90 between the South and North Hemisphere tropospheric averages. We find an average value of about 0.85 (see Table 5-4).

(Table 5-3b Notes, cont. 1)

g. Grossman, Frank R. and Robert W. Holloway (EPA, Las Vegas), "Concentrations of Krypton-85 Near the Nevada Test Site," Environmental Science and Technology 19 (1985), pp. 1128-1131. The linear interpolation suggested for the average of the offsite network measurements for the period 1972-83 is  $14.88 + 0.83*(Y - 79)$ . The onsite measurements were about  $0.5 \text{ pCi/m}^3$  higher on average. The difference may be due to leakage from underground tests but the difference may not be statistically significant.

h. Janssens, A. "Analysis of the  $^{85}\text{Kr}$  Concentrations Measured in Ghent," (Draft, Jan. 1985). The best fit to the background extracted from measurements during the period 1979-84 is  $16.4*\exp[0.05*(Y-79)]$ .

Table 5-4. Average Concentrations of Kr-85 in Various Compartments of the Atmosphere

Name	Boundaries <sup>a</sup>		Mass <sup>a</sup> Atmosphere (percent)	Kr-85 Relative Concentration				
	Latitude	Altitude (km)		1955.5	-58.5	-64.9	-72.5	-80-83
N Trop.	0-30N 30-90N	0-16 ) 0-10 )>	41.1	1	1	1	1	1
S Trop.	0-30S 30-90S	0-16 ) 0-10 )>	41.1	0.67 <sup>c</sup>	0.74 <sup>c</sup>	.87 <sup>b</sup> (0.80	.71 <sup>b</sup> 0.83	.82-.88 <sup>b</sup> 0.85) <sup>c</sup>
N. Lower Strat.	0-30N 30-90N	16-20 10-20	6.2	<----- 0.83 <sup>a</sup> ----->				
S. Lower Strat.	0-30S 30-90S	16-20 10-20	6.2	<----- 0.78 <sup>a</sup> ----->				
Upper Strat.	90S-90N	20+	5.4	<----- 0.51 <sup>a</sup> ----->				
WEIGHTED AVERAGE				0.81 <sup>c</sup>	0.84 <sup>c</sup>	0.90	0.83	0.88-.90 (0.87 0.88 0.89) <sup>c</sup>

a. Compartment definitions and 1973 values for relative concentrations in the stratosphere from Telegdas and Ferber. <sup>5-3b</sup>

b. See Table 5-3.

c. Based on the model in Table 5-5.

Table 5-5. Values for the Ratio Between Southern and Northern Hemisphere Kr-85 Concentrations Obtained from a Simple Two-Box Model in the Approximation of A Linearly-Increasing Total Atmospheric Inventory of Kr-85 (starting in 1950)<sup>a</sup>

t	1950	1955	1960	1965	1970	1975	1980
S/N	---	0.65	0.76	0.80	0.83	0.84	0.85.

a. Obtained from the following equations (see Appendix 5-A):

$$r^{-/+} = \frac{N^-}{N^+} = (T'/T_d) * \{1 + (1/t) * (T_d - T') * [1 - \exp(-t/T')]\}$$

$$S/N = (1 - r^{-/+}) / (1 + r^{-/+}).$$

Table 5-6. The Atmosphere's Increasing Inventory of Kr-85  
(uncertainties in parenthesis)

<u>Year</u>	<u>Global Average Concentration</u> <sup>a</sup> (pCi/SCM-Air)	<u>Inventory</u> <sup>b</sup>	<u>Decay During Preceding Period</u> <sup>c</sup> (MCI)	<u>Cumulative Additions</u> <sup>d</sup>
1945.0	0.0	0.0	0.0	
1955.5	0.4(0.2)	1.6(0.8)	0.5(0.3)	2.1(0.9)
1958.5	1.6(0.8)	6.4(3.2)	0.8(0.3)	7.7(3.2)
1964.9	8.0(1.2)	32.2(4.8)	8.0(1.2)	41.5(5.0)
1972.5	11.6(1.7)	44.6(7.0)	19.3(2.0)	75.2(7.4)
1981.0	16.5(0.8)	66.2(3.3)[67.2] <sup>f</sup>	30.9(2.1)	125.7(4.6)
1984.0	17.9(0.9)	71.9(3.6)[73.2] <sup>f</sup>	13.4(0.5)	144.8(4.8)

Change in the Interval 1981.0 - 1984.0:  
 1.4(0.4)<sup>e</sup>      5.6(1.6)      13.4(0.5)      19.0(1.7)

a. The values for 1955.5 and 1958.5 are based on values of 0.5 and 1.9 pCi/SCM obtained from the curve in (Report of the United Nations Scientific Committee on the Effects of Atomic Radiation, [New York, NY: United Nations, 1964], p. 24. showing interpolated North Hemisphere surface level concentrations. We assume a 50 percent measurement uncertainty and multiply by the factors in Table 5-4 to convert to average global concentrations. The values for 1964.9 and 1972.5 are obtained by multiplying the Northern Hemisphere tropospheric averages in Table 5-3 by the reduction factors calculated in Table 5-4. The values for 1981.0 and 1984.0 are obtained in the same way from a projected values of 18.5 and 20.1 pCi/SCM respectively from a linear fit to the N. Hemisphere surface-level concentrations for 1980.8-83.8. We assume 15%, 15%, 5% and 5% uncertainties for the last four points respectively, reflecting the uncertainties on the individual measurements quoted by the authors convoluted with the uncertainty in our extrapolation from the Northern Hemisphere surface-level to the global average concentration.

b. We assume a mass for the earth's atmosphere of  $5.2 \times 10^{18}$  kg (Handbook of Chemistry and Physics,<sup>3</sup> 1983-'84, p. F-154) and a density under standard conditions of 1.2929 kg/m<sup>3</sup>. An estimate of the global atmospheric inventory of Kr-85 (in MCI) can then be obtained by multiplying the average global concentration (in pCi/m<sup>3</sup> at standard pressure and temperature) by 4.02.



(Table 5-6, cont.)

c We approximate the rate of increase of the atmospheric inventory from a value of  $I(t_0)$  at time  $t_0$  to a value of  $I(t_1)$  at time  $t_1$  as linear during this interval:

$$I(t) = I(t_0) + (t - t_0) * [I(t_1) - I(t_0)] / [t_1 - t_0].$$

$I(t)$  is related to the rate of addition  $[A(t)]$  of Kr-85 to the atmosphere by the equation:

$$I(t) = I(t_0) * \exp[-(t - t_0)/T_d] + \int_{t_0}^t A(t') * \exp[(t' - t)/T_d] dt'$$

where  $T_d$  = the exponential lifetime of Kr-85 = 15.5 years.

The form of  $A(t)$  that gives the linear form of  $I(t)$  is

$$A(t) = I(t)/T_d + [I(t_1) - I(t_0)] / [t_1 - t_0].$$

The second term is the rate of Kr-85 that would be required in the absence of decay. The first term therefore represents the additional amount that must be added to offset decay. Integrating this term from  $t_0$  to  $t_1$  in the linear approximation gives

$$\text{Kr-85 Decay During Interval} = 0.5 * [I(t_1) + I(t_0)] * (t_1 - t_0) / T_d.$$

The uncertainty in this quantity is obtained by taking the uncertainty in the sum of  $I(t_1) + I(t_0)$  to be the square root of the sum of the squares of the individual uncertainties.

d. The uncertainties are calculated by taking the square root of the sums of the squares of the uncertainties of all the contributing quantities (the inventory plus the cumulative decay).

e. The uncertainties shown on the concentration differences are smaller than the uncertainties shown on the numbers which have been subtracted to obtain them because the uncertainties are correlated. The contribution of the measurement errors are only about 1 percent. The remainder of the 5 percent uncertainty is assumed to come from uncertainty in the projection of the Northern Hemisphere sea-level average concentrations to hemispheric average concentrations. We assume this factor is primarily a function of the time of year so that the uncertainty of the differences are five percent of the differences in the concentrations plus 2 percent of the average value of the concentrations whose differences are being taken.

f. Values estimated in Daniel J. Jacob et al, "Global Distribution of  $^{85}\text{Kr}$  in the Troposphere," [DRAFT, Center for Earth and Planetary Physics, Harvard University, June 1986].

Table 5-7a. Power History of Savannah River Production Reactors

<u>Fiscal Year</u> <sup>a</sup>	<u>Kr-85 Releases (MCi)</u>		<u>Total US</u> <sup>c</sup>	<u>Total Energy Output</u> <sup>b</sup> (TWtD)
	<u>Savannah River</u> [reported <sup>d</sup> ]	<u>(est<sup>e</sup>)</u>		
1955				0.51
1956				1.23
1957				1.84/3.58
1958				2.08
SUBCY58.5		(1.33)		
1959				2.92
1960				3.15
1961				3.25
1962				3.15
1963				3.12
1964				3.25/24.50
1965				2.14
SUBCY65.0		(8.53)		
1966				2.21
1967				2.63
1968				2.51
1969				1.75
1970				1.49
1971	0.64	(0.56)		1.42/38.65
1972	0.60	(0.53)		1.75
SUBCY72.5	[13.96	(13.81)		
CY72			1.060	(1.14)
1973	0.77	(0.65)		1.87
CY73			0.754	(0.64)
1974	0.50	(0.70)		1.90
CY74			0.774	(0.77)
1975	0.52	(0.71)		1.42
CY75			0.567	(0.58)
1976 <sup>a</sup>	0.74	(0.65)		1.64
CY76			0.779	(0.59)
1977	0.44	(0.49)		1.30
CY77			0.560	(0.57)
1978	0.53	(0.49)		1.23
CY78			0.545	(0.62)
1979	0.48	(0.46)		1.19
CY79			0.495	(0.51)
1980	0.58	(0.44)		1.46
CY80			0.682	(0.74)
SUB(CY72-80)			[6.216	(6.16)]
1981	0.84	(0.55)		1.38
CY81			0.905	(0.58)
SUB(FY71-81)	[6.64	(6.23)]		
1982		(0.51)		1.87
CY82			0.531	(0.57)
1983		(0.70)		1.87/57.53(tot.)
CY83			0.598	(0.70)/[8.25(8.01)--tot]
1984		(0.70)		
SUB(CY84.0)	[(20.75)	20.34]		

NOTES: TABLE 5-7a

- a. In 1976, the end of the fiscal year was changed from June 30 to September 30.
- b. A graph of the historical thermal energy output of the Savannah River reactors through fiscal year 1983 (Sept. 31 1983) is shown in Department of Energy National Security and Military Applications of Nuclear Energy Authorization Act of 1985, Hearings before the Procurement and Military Nuclear Systems Subcommittee of the House Committee on Armed Services, Feb.-Mar. 1984, p. 333.
- c. Data for total US releases for calendar years 1972-83 are given in Grossman, R. Frank and Robert W. Holloway, "Concentrations of Krypton-85 Near the Nevada Test Site," Environmental Science and Technology 19 (1985), pp. 1128-1131. In the parentheses, we give the values for the summed reported releases from Savannah River (interpolated from fiscal years to calendar years), Hanford (1972 only -- see Table 5-7b) and the Idaho Chemical Processing Plant (see Table 5-9).
- d. Krypton-85 releases for the fiscal years 1971-81 are given in Ashley, C. and Zeigler, C.C.: Releases of Radioactivity at the Savannah River Plant, 1954 through 1978, DPSPU 75-25-1 (1980); Environmental Monitoring at the Savannah River Plant, Annual Reports, DPSU 72-302 (1971), DPSPU 75-302 (1974); and Environmental Monitoring in the Vicinity of the Savannah River Plant, Annual Reports, DPSPU 74-30-1 (1973), DPSU 75-30-1 (1974), DPSU 76-30-1 (1975), DPSPU 77-30-1 (1976), DPSPU 78-30-1 (1977), DPSPU 79-30-1 (1978), DPSPU 80-30-1 (1979), DPSPU 81-30-1 (1980), DPSPU 82-30-1 (1981). [Data compiled by Bernd Franke and Robert Alvarez, Environmental Policy Institute, Washington, DC. in Franke, B., and Alvarez, R.: Analysis of External [Gamma]-Radiation Monitoring Around the Savannah River Plant, (Washington, D.C.: Environmental Policy Institute, Draft Report, 1983).]
- e. Calculated assuming the production of 0.86 grams of weapon-grade plutonium and the fission of 0.936 grams of U-235 and 0.103 grams of Pu-239 per MwtD in a production reactor producing weapon-grade plutonium (1.09 grams U-235 and 0.12 grams Pu-239 fissioned per gram of plutonium produced - see Table 3-2). On this basis and the heat and Kr-85 fission yields given in Table 5-1, we find that, with one year decay between fissioning and release, 0.373 MCi of Kr-85 would be released to the atmosphere per TwtD of fission. We include in the national totals estimated releases from the Idaho Chemical Processing Plant.

Table 5-7b (also 3-4b). Power History of the Hanford Reactors

Original Eight Production Reactors

N-Reactor

<u>CY</u>	<u>Thermal Output</u> <sup>a</sup> (Twt-days)	<u>Kr-85 Releases</u> <sup>b</sup> (M Ci)	<u>Output</u> <sup>c</sup> (Twt-days)	<u>Kr-85 Releases</u> <sup>d</sup> (M Ci)
1944	0.021			
1945	0.076			
1946	0.194			
1947	0.146			
1948	0.185			
1949	0.234			
1950	0.307			
1951	0.50			
1952	1.00			
1953	1.25			
1954	1.50			
SUBCY54.5	(4.663)			
1955	2.25			
SUBCY55.5		(1.727)		
1956	2.50			
1957	3.40			
SUBCY57.5	(11.86)			
1958	2.90			
SUBCY58.5		(4.418)		
1959	4.25			
1960	4.75			
1961	4.30			
1962	4.50			
1963	4.90			
SUBCY64.0	(39.16)			
1964	4.02		0.234	
SUBCY65.0		(14.59)		
1965	3.14		0.660	(0.09)
1966	3.55		0.750	(0.24)
1967	2.40		0.600	(0.28)
1968	2.20		0.850	(0.22)
1969	1.95		0.800	(0.31)
1970	0.75		0.550	(0.29)
1971 (0.5 yr.)			0.250	(0.20)
SUBCY71.5	(57.17)		(4.69)	
1971 (0.5 yr.)			0.250	(0.09)
1972 (0.5 yr.)			0.350	0.41 <sup>e</sup> (0.09)
SUBCY72.5		(21.30)		[1.81]
PUREX (HANFORD NUCLEAR FUEL REPROCESSING PLANT) SHUT DOWN DURING 1972				
1972 (0.5 yr.)			0.350	
1973			0.845	
1974			0.799	
1975			0.679	
1976 (Jan - Sept.)			0.294	

(Table 5-7b, cont.)

Fiscal Years		
1977	0.792	
1978	0.768	
1979	0.751	
1980	0.575	
1981	0.257	
1982	0.581	
1983	0.727	
PUREX PLANT RESTARTED (11/83) <sup>b</sup>		
SUB84.0 (21.3)		(1.81)
1984	0.682	

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Table 5-7b, Notes and References

a. Values for 1951-71 from a letter from John L. Meinhardt, Director, Office of Nuclear Materials Production (Department of Energy) to Thomas B. Cochran, quoted in ref. b. Earlier values are estimates by Cochran, based on the assumption that the original reactors were rated at 250 MWt each and operated at an average of 80 percent of capacity.

b. 0.373 MCi Kr-85 released per Twt-day -- see Table 5-7a, note a.

c. Cochran, Thomas B.; Arkin, William M.; and Hoenig, Milton M.: Nuclear Weapons Databook, Vol. II: The Production Complex (Cambridge, MA: Ballinger, DRAFT to be published).

d. We assume 0.7 grams of plutonium containing 9 percent Pu-240 produced per Mwt-Day (see ref. c). This translates into 0.13 grams of Pu-239 fission and 0.91 grams of U-235 fission. Assuming one year between fission and Kr-85 release, there would be 0.368 MCi of Kr-85 released per Twt-day.

e. Draft Environmental Impact Statement, Operation of Purex and Uranium Oxide Plant Facilities, Hanford Site, Richland, Wash. (Washington DC: Department of Energy, May 1982 (DOE/EIS 0089D), p. 3.11). According to this reference, irradiated fuel containing 1013 tonnes heavy metal (HM) was processed in this year. Assuming 0.368 MCi per TWtD of fission, the total burnup of the fuel reprocessed at Hanford in 1971 would have been 1.11 TWtD and its average burnup would have been 2682 MWtD/tonne-HM. The average burnup of the N-reactor fuel reprocessed at the West Valley plant in 1971 was 2850 MWtD/tonne-HM (see Table 5-8).

Table 5-8. Uranium<sup>a</sup> Fuel Reprocessed by Nuclear Fuel Services<sup>b</sup>

<u>Uranium Content</u> tonnes(yr.-react.)	<u>Burnup</u> (GwD)	<u>Plutonium Fissioned<sup>c</sup></u> (kg)	<u>Plutonium</u> (kg)
48.5(1966-N)	4	0	4
46.7(1966-N)	60	9	60
238.1(1969-N)	665	214	473
15.8(1971-N)	45	14	45
SUB(N-REACTOR)	(774)	(237)	(582)
50.0(1966-BWR)	426	115	191
49.8(1967-PWR)	558	90	185
19.6(1969-PWR)	400	109	176
21.5(1969-BWR)	235	60	105
15.6(1969-PWR)	248	72	108
9.3(1969-PWR)	228	72	96
18.4(1970-BWR)	171	45	73
7.6(1971-PWR)	178	53	68
20.8(1971-BWR)	219	56	87
9.5(1971-PWR)	224	68	96
5.8(1971-BWR)	80	26	28
<u>17.2(MISC)</u>	<u>60</u> <sup>d</sup>	<u>?</u>	<u>23</u>
594.2	3801 <sup>d</sup>	1003	1818

Kr-85 Releases: (0.25 MCi from the N-Reactor fuel, and 0.71 MCi from all other. Since, it is assumed in Table 5-7b that all the thermal energy released at the N-reactor through 1971.5 was reprocessed at Hanford, we avoid double counting by not including the Kr-85 released from the N-reactor fuel in the West Valley total.)

Table 5.8, Notes and References

- a. Small amounts of fast-breeder and thorium-based fuel were also reprocessed.
- b. Resnikoff, Marvin, 1977: Sierra Club Testimony Related to Section IV E, Reprocessing, Final GESMO I, (US NRC Docket No. RM-50-5). Resnikoff cites as his sources Nuclear Fuel Services Quarterly Reports to the Nuclear Regulatory Commission, Docket No. 50-201. Table D-1.
- c. Heavy-Element Concentrations in Power Reactors (Clearwater Florida: NUS Corporation, 1977, Report# SND-120-2).
- d. The Safety of Nuclear Power Reactors and Related Facilities (Washington DC: US Atomic Energy Commission, Report # WASH-1250, 1973), Table 4-13 estimates the total burnup of the fuel reprocessed at West Valley as 4.220 TwtD. It also estimates total Kr-85 releases from Nuclear Fuel Services at 1.3 MCi. The difference between this estimate and our own is the cooling time assumed. A one year cooling time was assumed in the official estimate.
- e. Kr-85 yields calculated using the assumptions in Table 5-1. We have assumed 2 years delay between fission and the reprocessing of the N-reactor fuel. For the remainder we have assumed 5 years, on the basis of the initial full-power operating dates of the reactors which accounted for most of the civilian fuel (Dresden 1, 1960; Yankee Rowe, 1961; Indian Point 1, 1962; Big Rock Point, 1963 -- see "Power Reactors, 1982", Nuclear Engineering International, August 1982, Supplement).

Table 5-9. Releases of Kr-85 from the Idaho Chemical Processing Plant<sup>a</sup>

<u>Year(s)</u>	<u>Annual Releases</u> (M Ci)
1953	0.015
1954	0.037
1955	0.052
SUB55.5	(0.078)
1956	0.083
1957-1963	(0.05-0.10/yr assumed <sup>b</sup> )
SUB58.5	(0.26-0.34)
1964	0.085
SUB65.0	(0.62-0.97)
1965	0.046
1966	0.053
1967	0.021
1968	0.086
1969	0.111
1970	0.148
1971	0.137
1972	0.046
SUB72.5	(1.25-1.60)
1973	0.005
1974	0.260
1975	0.024
1976	0.033
1977	0.111
1978	0.101
1979	0.000
1980	0.092
1981	0.059
1982	0.009
1983	0.003
TOTAL84.0	(1.97-2.32)

a. Data for 1953-'56 and 1964-'72 from National Reactor Testing Station Radioactive Waste Management Information, 1972 Summary and Record-to-Date (US Atomic Energy Commission, Idaho Operations Office, IDO-10054(72); data for 1973-'82 from Radioactive Waste Management Information, 1982 Summary and Record-to-Date (EG&G Idaho, Inc., IDO-10054(82); for 1983 from 1983 Environmental Monitoring Program Report for Idaho National Engineering Laboratory Site [US Department of Energy, Idaho Operations Office, 1984, DOE/ID-12082-83)], p.17.

b. Hot fuel was reprocessed to recover a short-lived isotope. Kr-85 releases were swamped by shorter lived gaseous isotopes [Draft Environmental Statement, Waste Management Operations, Idaho National Engineering Laboratory (US Energy Research and Development Administration, Report # ERDA-1536, 1976), Table II-XVII).



Table 5-10a. Releases of Kr-85 by Nuclear Explosions in the Atmosphere

<u>Years</u>	<u>Fission Yield</u> <sup>a</sup> (Mt)	<u>Kr-85 Releases</u> <sup>b</sup> (Mci)
1945.0-55.5	38.6	0.8
1955.5-58.5	38.5	
SUB58.5	(77.1)	1.5
1958.5-64.9	116.6	
SUB64.9	(193.7)	3.9
1964.9-72.5	17.2	
SUB72.5	(210.9)	4.2
1972.5-81.0	6.2	
SUB81.0	(217.1)	4.3

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a. United Nations Scientific Committee on the Effects of Atomic Radiation, Ionizing Radiation: Sources and Biological Effects (New York, NY: United Nations, 1982), p. 227.

b. Prorated to the total of 4.3 Mci estimated in ibid, p. 216, para. 33.

Table 5-10b. Releases of Kr-85 By Underground Nuclear Explosions

<u>Period</u>	<u>Total Yield</u> (Mt)		<u>Kr-85 Release</u> <sup>c</sup> (MCI)
	<u>US</u> <sup>a</sup>	<u>Soviet</u> <sup>b</sup>	
1957	0.0017		
1958.0-1958.5	negl.		
SUBTOTAL			neg.
1958.5-1959.0	0.027		
1961	0.06		
1962	0.6	one small test	
1963	0.6		
1964	1.0	0.2	
SUBTOTAL			2.4 0.02
1965	0.6	1.	
1966	2.2	2.3	
1967	1.2	2.	
1968	4.7	1.	
1969	2.8	1.5	
1970	3.0	5	
1971	4.8	4	
1972	0.3	4	
SUBTOTAL			42.8 0.3
1973	1.0	8	
1974	0.7	9	
1975	4.0	6	
1976	4.5	1	
1977	0.4	1	
1978	0.5	1.5	
1979	0.5	1.5	
1980	0.4	1	
1981	0.4	1	
1982	0.6	1.5	
<u>1983</u>	<u>0.3</u>	<u>1.5</u>	
TOTALS	35.1	54	89.1 0.7

<sup>a</sup>. Robert S. Norris, Thomas B. Cochran and William M. Arkin, Known U.S. Nuclear Tests, July 1945 to 31 December 1985 (Washington, DC: Natural Resources Defense Council Working Paper NWD 86-2, 1986).

<sup>b</sup>. Jeffrey I. Sands, Robert S. Norris and Thomas B. Cochran, Known Soviet Nuclear Explosions, 1949-1985, Preliminary List (Washington, DC: Natural Resources Defense Council Working Paper NWD 86-2, 1986).

<sup>c</sup>. 0.008 MCI/Mt (assuming 50 percent fission yield and 0.015 MCI/Mt-fission (ref. 5-5)).

Table 5-11. Fission Energy Released in British Gas-Graphite Reactors

<u>Year</u>	<u>Military</u> <sup>a</sup> (Twt-days -->Mci)	<u>Burnup</u> <sup>b</sup> (Mwtd/tonne)	<u>Kr-85 "Age"</u> <sup>c</sup> (yrs.)	<u>Kr-85 Rel.</u> <sup>d</sup> (Ci/Mwt-Day)	<u>Civil</u> <sup>e</sup> (Twt-days -->Mci)
1951	0.057	350	1	0.39	
1952	0.057	"	1	"	
1953	0.057	"	1	"	
1954	0.079	"	1	"	
SUB54.5	(0.21 -->0.08 Mci Kr-85)				
1955	0.079	"	1	"	
1956	0.079	"	1	"	
1957	0.139	"	1	"	
SUB57.5	(0.48 -->0.19 Mci Kr-85)				
1958	0.228	"	1	"	
1959	0.264	"	1	"	
1960	0.455	"	1	"	
1961	0.502	"	1	"	
1962	0.502	"	1	"	(prev. yrs
1963	0.502	"	1	"	(included
SUB64.0	(3.00 -->1.17 Mci Kr-85)				
1964	0.502-->0.20	"	1	"	in 1965
1965	0.714-->0.24	1000	2	0.34	3.11-->1.06
1966	0.800-->0.27	1350	2	0.34	2.58-->0.88
1967	0.778-->0.26	1800	2	0.33	3.04-->1.00
1968	0.777-->0.24	2100	3	0.31	3.41-->1.04
1969	0.820-->0.25	2500	3	0.31	3.34-->1.06
SUB69.5	-- including military				(22.87-->7.02)
1970	0.875-->0.26	2800	3	0.30	3.00-->0.90
1971	0.898-->0.25	3200	4	0.28	3.29-->0.92
1972	0.914-->0.26	3300	4	0.28	3.50-->0.98
1973	0.903-->0.25	3400	4	0.28	3.29-->0.92
1974	0.900-->0.23	3600	5	0.26	4.08-->1.06
1975	0.781-->0.20	"	5	"	3.68-->0.96
1976	0.738-->0.19	"	5	"	4.23-->1.10
1977	0.648-->0.17	4100	5	"	4.24-->1.10
1978	0.720-->0.19	"	5	"	3.77-->0.98
SUB79.0	-- including military				(63.33-->17.94)
1979	0.694	"	5	"	3.73
1980	0.666	4600	5	0.26	3.16
1981	0.776	"	5	"	3.16

a. Prior to 1965, calculated from the assumptions given in Simpson, John, 1983: The Independent Nuclear State: the United States, Britain, and the Military Atom (London, McMillan, 1983), Appendix 4. From 1965 on, calculated from the numbers given for gross electricity production in Simpson, Appendix 4, assuming a ratio of gross electricity production to thermal output of 0.175. (See "Power Reactors, 1982", Nuclear Engineering International, August 1982, Supplement.)

(Table 5-11, Notes, cont.)

b. Unweighted averages of estimates made for the civil reactors by Barnham, K.W.J.; Hart, D.; and Stevens, R.A.: The Production and Destiny of UK Plutonium, Table 4 (address of Hart: School of Environmental Sciences, University of East Anglia, Norwich NR4 7TJ).

c. We assume 0.8 years between fission and discharge from the reactor for every 1000 MWD/tonne and add one year between discharge and reprocessing (based on Barnham et al, ibid).

d. Based on a curve in Barnham et al which shows the Pu-240 content of Magnox fuel increasingly quadratically with burnup to 0.067 kg/te at 1000 Mwt-days/tonne-U burnup, then to 0.19 at 2000, 0.35 at 3000, 0.52 at 4000, and 0.68 at 5000 Mwt-days/tonne-U. We assume two fissions of Pu-239 per Pu-240 formed. See also Table 3-1.

e. The terrawatt-days are back-calculated from the numbers given for gross electricity production in Simpson, Appendix 5, assuming a ratio of gross electricity production to thermal output of 0.30 (see "Power Reactors, 1982." A comparison of the electrical output for these reactors as given in Simpson (op cit) with the thermal power outputs given in Table 6 in Barnham, et al (op cit) also indicates that the figures that they were given correspond to an average efficiency of 30 percent.

Table 5-12. Kr-85 Releases from the British Nuclear Fuel Reprocessing Plant at Sellafield

<u>Year</u>	<u>Release (MCi)</u> Reported <sup>a</sup> (Estimated)
SUB55.5	(0.08)
SUB58.5	(0.19)
SUB65.0	(1.17)
SUB71.0	(5.08)
1971	1.2 (1.28)
1972	1.2 (1.31)
SUB72.5	[6.88 (7.02)]
1973	0.8 (1.16)
1974	0.8 (0.00)
1975	1.2 (1.17)
1976	1.2 (1.24)
1977	0.8 (1.17)
1978	0.7 (0.00)
1979	0.94(1.29)
1980	0.84(1.16)
1981	1.40(1.29)
1982	1.19(1.27)
1983	1.13(1.17)
SUB84.0	[18.48(18.59)]
SUB71-83	[13.40(13.51)]

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a. 1971-82: Letter from R.S. Edmonds, Senior UKAEA Liason Officer, British Embassy, to David Albright, 21 March 1984; 1983: G. Fraser, ECC Health and Safety Inspectorate, ECC Luxemburg, private communication, to Frank von Hippel, 14 May 1985.

Table 5-13. Nuclear Fuel Reprocessed in France

Year	Marcoule <sup>a</sup>		La Hague <sup>b</sup>	
	Tonnes (Av. Burnup -MwtD/t-U)	Burnup [Plutonium Fissioned] (TwtD) [tonnes]	Tonnes (Av. Burnup -MwtD/t-U)	Burnup [Plutonium Fissioned] (TwtD) [tonnes]
59	190(100)	0.02[0.00]		
60	130(100)	0.01[0.00]		
61	320(100)	0.03[0.00]		
62	620(200)	0.12[0.01]		
63	640(200)	0.13[0.01]		
64	760(300)	0.23[0.01]		
SUB65.0		<0.54[0.03]>		
65	890(300)	0.27[0.02]		
66	869(300)	0.26[0.02]		
67	960(400)	0.38[0.03]		
68	730(400)	0.29[0.02]	189(1170)M*	0.22[0.03]
69	890(450)	0.40[0.05]	158( 990)M	0.16[0.02]
70	530(450)	0.24[0.03]	245(1080)M	0.27[0.04]
71	571(450)	0.26[0.03]	126(2290)M	0.29[0.08]
72	462(500)	0.23[0.03]	250(2160)M	0.54[0.15]
SUB72.5		<2.76[0.25]		<1.21[0.25]>
73	480(600)	0.29[0.04]	213(2390)M	0.51[0.15]
74	240(700)	0.17[0.02]	635(2330)M	1.48[0.43]
75	280(800)	0.22[0.03]	443(3038)M	1.35[0.45]
76	260(1000)	0.26[0.04]	218(2783)M	0.61[0.20]
			15(15803)O*	0.24[0.05]
77	170(1200)	0.20[0.04]	355(2947)M	1.05[0.36]
SUB77		<4.02[0.43]>	18(28077)O	0.51[0.17]
78	250(3700) <sup>c</sup>	0.93[0.33]	371(3345)M	1.24[0.45]
			38(27271)O	1.04[0.34]
79	250(3700) <sup>c</sup>	0.93[0.33]	240(3590)M	0.86[0.32]
			79(20375)O	1.61[0.44]
80	250(3700) <sup>c</sup>	0.93[0.33]	252(3317)M	0.84[0.30]
			105(20980)O	2.21[0.62]
SUB81.0		<6.81[1.42]>		<15.03[4.60]> <5.61[1.62]> +<9.42[2.98]>M
81	250(3700) <sup>c</sup>	0.93[0.33]	250(3672)M	0.92[0.34]
			101(25420)O	2.57[0.81]
82	250(3700) <sup>c</sup>	0.93[0.33]	226(4000?)M <sup>d</sup>	0.90[0.36]
			154(21100)O <sup>e</sup>	3.25[0.93]
83	500(3700) <sup>E</sup>	1.86[0.66]	117(4000?)M <sup>f</sup>	0.47[0.19]
			221(23241)O <sup>h</sup>	5.14[1.55]
SUB84.0		<9.60[2.41]>		<28.94[9.04]> <16.55[4.90]> +<12.39[4.14]>M

\* "M" - Magnox or metal fuel from graphite-moderated reactors. "O" - Oxide fuel from light-water moderated reactors.

Table 5-13, NOTES

a. Except where otherwise indicated, the source of estimated tonnages and average burnups is: Syndicat CFDT de l'Energie Atomique, Le Dossier Electronucleaire (Editions du Seuil, 1980), pp. 188, 190. Estimates of amounts of plutonium fissioned (per Mwt-day as a function of fuel burnup) from ref. Heavy-Element Concentrations in Power Reactors (Clearwater Florida: NUS Corporation, 1977, Report # SND-120-2).

b. Numbers for the tonnage of metal and oxide fuel reprocessed at La Hague for the period 1968-81 and the average burnup of each appear in the NRC Translation of Castaing, Raimond et al, Rapport du Groupe de Travail sur la Gestation des Combustibles Irradies (France, Ministere de la Recherche et de l'Industrie), (1982), Attachment 4 "Analysis of the Dosimetric Results of the External Exposure," Table VIII.

c. In a private communication to David Albright, Feb. 1983, an official in the Cogema office in Washington DC stated that Marcoule had reprocessed 1358 tonnes cumulatively as of the end of 1982. Also in Feb. 1983, the French Nuclear Attache in the French Embassy in Washington stated that Marcoule had reprocessed about 250 tonnes of commercial Magnox (metal) fuel per year. As of the end of 1983, the six gas-graphite power reactors operating in France and Spain had a total gross electrical (thermal) generating capacity of 2.8 Gwe (9.41 Gwt) and had produced 7.87 twt-days (26.4 twt-days) of output. Their cumulative weighted average load-factor was 0.557. [Nuclear Engineering International, May 1984, p. 36.] This would correspond to an average annual output of 1.91 twt-d. Subtracting an average of 0.99 Twt-days a year for the fuel reprocessed at La Hague, would leave an average of 0.92 Twt-days to be reprocessed at Marcoule -- or 3700 Mwt-days/tonne for an average annual throughput of 250 tonnes. In addition, France has operated 3 production reactors -- the second and third of which each had a rated thermal output of 0.26 Gwt -- and a dual-purpose power reactor with an output of 0.3 Gwt (all now shutdown).

d. Cogema official (Washington, DC office), private communication to David Albright, Feb. 1983.

e. J. Megg, "Reprocessing Spent Fuel in France," Nuclear Engineering International, March 1983, p. 8.

f. In a private communication to David Albright, May 1984, an official in the Cogema office in Washington DC stated that 4457 tonnes of Magnox fuel had been reprocessed at La Hague as of the end of 1983, 117 tonnes during 1983. We assume a burnup of 4000 Mwtd/tonne

Table 5-13, Notes, cont.

g. According to Nuclear Fuel (December 31, 1984), slightly under 7,000 tonnes of gas-graphite power reactor fuel were reprocessed at Marcoule and La Hague through the end of 1984. Subtracting the amount of gas-graphite fuel reprocessed at La Hague through 1984 (4,472 tonnes, for 1984 -- see Nuclear Fuel, February 25, 1985) and the amount reprocessed at Marcoule through 1982 (1,358 tonnes, see footnote c) leaves roughly 1,000 tonnes. The annual average for fuel reprocessed at Marcoule during 1983 and 1984 was therefore 500 tonnes per year.

h. "Cogema Exceeds Reprocessing Target by 20 %", Nuclear Fuel, November 21, 1983, p. 8.



Table 5-14. Reported and Estimated Kr-85 Releases from French Reprocessing Plants (MCI)

Year	Marcoule		La Hague	
	Reported <sup>a</sup>	Estimated <sup>b</sup> [no decay]	Reported <sup>a</sup>	Estimated <sup>b</sup> [no decay]
59		[0.01]		
60		[0.00]		
61		[0.01]		
62		[0.05]		
63		[0.05]		
64		[0.09]		
SUB-'65		<0.22>		
65		[0.11]		
66		[0.10]		
67		[0.15]		
68		[0.11]		[0.08]M*
69		[0.15]		[0.06]M
70		[0.09]		[0.10]M
71		[0.10]		[0.10]M
72(1/2 yr)	0.024	[0.045]	0.12	[0.09]M
SUB-'72.5		<1.08>		<[0.44]>
72(1/2 yr)	0.024	[0.045]	0.12	[0.09]M
73	0.13	[0.11]	0.23	[0.18]M
74	0.11	[0.07]	0.72	[0.51]M
75	0.10	[0.08]	0.66	[0.45]M
SUB72-'75	<0.39>	<0.35>	<1.85>	<[1.33]>
RATIO		1.11		1.39
76	0.09	[0.10]	0.35	[0.21]M+[0.09]O*
77	0.12	[0.07]	0.67	[0.35]M+[0.17]O
78	0.31	[0.31]	0.79	[0.41]M+[0.35]O
SUB72-'78			<3.66>	<[2.30]M+[0.61]O>
RATIO				1.26
79	0.28	[0.31]	0.64	[0.28]M+[0.56]O
80	0.54	[0.31]	0.83	[0.28]M+[0.77]O
SUB72-80	<1.73>	<[1.45]>	<5.13>	<[2.86]M+[1.94]O>
RATIO		1.18		1.07
SUB79-80			<1.47>	<[0.56]M+[1.33]>
RATIO				0.78
81		[0.31]		[0.30]M+[0.88]O
82		[0.31]		[0.29]M+[1.12]O
83		[0.62]		[0.15]M+[1.77]O
SUB84.0		<[3.73]>		<3.95]M+[5.72]O> -<[9.67]>
		2.64 if decayed 4 y.		7.64 if decayed 4 y.

Values Assumed in Table 5-17:

3.3

9

\* "M" = Metal fuel from graphite-moderated reactors. "O" = Oxide fuel from light-water moderated reactors.

Table 5-14 Notes

a. Luykx, F.; and Fraser, G.: Radioactive Effluents from Nuclear Power Stations and Nuclear Fuel Reprocessing Plants in the European Community, 1972-'76 (Luxembourg: Commission of the European Communities, 1978), Table XIV. Luykx, F.; and Fraser, G.: Radioactive Effluents from Nuclear Power Stations and Nuclear Fuel Reprocessing Plants in the European Community, 1974-'78 and 1976-'80 (Luxembourg: Commission of the European Communities, 1980, 1983), Tables XIII.

b. Estimated from Table 5-13 using the formula (see Table 5-1):

$$\text{MCI Kr-85} = [\text{TwTD} - 0.98 * (\text{tonnes Pu fissioned})] * 0.405 / 0.96 + 0.177 * (\text{tonnes Pu-239 fissioned}) -$$

$$0.413 * \text{TwTD} - 0.228 * (\text{tonnes Pu-239 fissioned}).$$

Table 5-15. Releases of Kr-85 from Small Reprocessing Facilities in Europe and Japan (MCI)

<u>Year</u>	<u>Europe</u> <sup>a</sup>			<u>Japan</u> <sup>e</sup>
	<u>Eurochemie</u> <sup>b</sup>	<u>WAK</u> <sup>c</sup>	<u>Eurex</u> <sup>d</sup>	<u>Tokai-Mura</u>
1972	0.2	0.07	--	
SUB72.5	[0.1]	[0.035]		
1973	0.22	0.03	0.00	
1974	0.1	<0.01	0.00	
1975	*	0.00	*	
1976	*	0.01	*	
1977	*	0.12	*	
1978	*	0.00	*	0.06
1979	*	0.05	*	
1980	*	0.03	*	0.28
1981	*	*	*	0.11
1982	*	0.0	*	0.19
1983	*	0.08	*	0.09
SUB84.0	<0.53>	<0.40>	*	<0.73>

\* Shutdown.

a. i) Luykx, F.; and Fraser, G.: Radioactive Effluents from Nuclear Power Stations and Nuclear Fuel Reprocessing Plants in the European Community, 1972-'76 (Luxembourg: Commission of the European Communities, 1978), Table XIV. ii) Luykx, F.; and Fraser, G.: Radioactive Effluents from Nuclear Power Stations and Nuclear Fuel Reprocessing Plants in the European Community, 1974-'78 and 1976-'80 (Luxembourg: Commission of the European Communities, 1980, 1983), Tables XIII. For the years 1981-83, G. Fraser, private communication 14 May 1985. In addition, there is a reprocessing plant which began operation at Dounreay, Scotland in 1958. This facility has a nominal capacity for reprocessing 0.3 tonnes of research reactor fuel and/or 3 tonnes of fast breeder reactor fuel annually. A 60 Mwt fast-breeder went into operation at Dounreay in 1963 and a 600 Mwt breeder in 1976. However, according to the CEC report of 1983, all of the Kr-85 produced by the first reactor was released to the atmosphere at the reactor. The first fuel from the second reactor was reprocessed in 1980 and released an estimated 0.003 MCI to the atmosphere.

b. The Eurochemie plant began operation in 1966 (refs. a). During the period 1970-1974, it reprocessed:

<u>Fuel Type</u>	<u>Tonnes</u>	<u>Burnup</u> (MwtD/t-U)	<u>Fission</u>	
			<u>U-235</u>	<u>Pu-239</u>
			---(tonnes)-----	
Low-Burnup Gas-Graphite (Mgnox) Reactor	7.9	900-1500	0.01	0.00
Boiling Water Reactor	69.4	4000-6000	0.25-0.36	0.04-0.07
"	29.5	6000-17300	0.15-0.34	0.03-0.19
<u>Pressurized Water Reactor</u>	<u>71.4</u>	<u>12900-21000</u>	<u>0.75-1.12</u>	<u>0.21-0.43</u>
<u>TOTALS</u>			<u>1.16-1.83</u>	<u>0.28-0.69</u>

(Table 5-15, Notes, cont.)

[NRC Translation of Castaing, Raimond et al, Rapport du Groupe de Travail sur la Gestion des Combustibles Irradies (France, Ministere de la Recherche et de l'Industrie), (1982), Attachment 4 "Analysis of the Dosimetric Results of the External Exposure," pp. 36-42.] (According to H. Meyers, "Reprocessing," in Nuclear Europe (Dec. 1985, p. 23), during the period 1966-74, the Eurochemie plant reprocessed: 86 tonnes of uranium fuel from experimental reactors, 96 tonnes of fuel from power reactors, and 30 tonnes of MTR fuel (fuel initially enriched to greater than 90 percent U-235!).

The corresponding range for the total amount of Kr-85 originally produced in this fuel is 0.42-0.61 MCi. Assuming three years average decay before reprocessing, the total releases would lie in the range 0.35-0.50 MCi. This is consistent with the results reported in ref. a.

c. The WAK (Karlsruhe) reprocessing plant began operation in 1971. During the period 1971-'77, it reprocessed 66 tonnes of oxide fuel (ref. b, Castaing report). If we assume that this fuel had an average burnup of 20,000 MwtD/t-U, the corresponding amount of Kr-85 originally produced in the fuel would have been about 0.46 MCi. Three years of decay would have reduced this to 0.38 MCi. This is consistent with the results reported in ref. a.

d. Eurex is an Italian plant for reprocessing research reactor fuel. As of 1978, a total of 0.110 tonnes of highly-enriched uranium had been recovered from this fuel (ref. a, Table XIII). Even if an equal amount of U-235 had fissioned in the fuel, only 0.05 MCi of Kr-85 would have been produced.

e. Year	Tonnes	Average Burnup (Mwt-days/tonne-U)	Fission Energy [Pu-fissioned] (Twt-days [tonnes])
1977-'78	19.1 <sup>f</sup>	11,000 <sup>f</sup>	0.21[0.04]
09/79-12/80	60.5 <sup>f</sup>	17,000 <sup>f</sup>	1.03[0.25]
1/81-6/81	26.6 <sup>f</sup>	15,000 <sup>g</sup>	0.40[0.09]
6/81-8/82	45 <sup>h</sup>	15,000 <sup>g*</sup>	0.68[0.15]
8/82-2/83	21 <sup>h</sup>	15,000 <sup>g</sup>	0.32[0.07]
2/83-early '85 shutdown			

(We assume an average of 4 years between fission and reprocessing.)

f. PNC News and Reports, Nuclex Edition, Tokyo, Japan, July 1981.

g. Estimated.

h. Japanese embassy, personal communication to David Albright, 1984. Burnup estimates by Albright.

Table 5-16. Nuclear-Electric Energy Generated Worldwide, 1960-1983

Year	Electric Energy Generated		Kr-85 Produced <sup>c</sup> (MCI)
	Non-Communist <sup>a</sup> [Twd(e) (Gwe)]	USSR <sup>b</sup>	
1960	0.13*		
1961	0.17		
1962	0.29*		
1963	0.46		
1964	0.58*	0.06 (0.21)	
SUBTOTAL	<1.63>	<0.06> <0.21>	1.91
1965	0.92	0.06 (0.21)	
1966	1.29*	0.06 (0.21)	
1967	1.46	0.08 (0.31)	
1968	2.00	0.08 (0.31)	
1969	2.13*	0.14 (0.51)	
1970	3.04	0.24 (0.88)	
1971	4.35*	0.24 (0.88)	
1972	5.83	0.36 (1.32)	
1973	7.86	0.60 (2.20)	
SUB72.5	<30.51>	<1.92>	36.65
1974	9.40*	0.88 (3.20)	
1975	13.94	1.00 (3.64)	
1976	16.20	1.27 (4.64)	
1977	19.62*	1.55 (5.64)	
1978	23.18	1.55 (5.64)	
1979	23.78	1.93 (7.04)	
1980	25.83	2.31 (8.45)	
1981	30.45	3.57 (13.05)	
1982	32.85	4.76 (17.37)	
1983	36.80*	<20.74>	
SUB84.0	<262.5>		325?

\* Doubling.

Table 5-16, Notes

a. Sources. 1960-'69: Energy Perspectives 2 (US Department of the Interior, 1976), p. 171; 1970-80: Annual Report to Congress, Vol. 2: Data (US Department of Energy, Energy Information Administration, 1981), p. 183; 1981-83, ibid (1984), p. 219.

b. B.A. Semenov, "Nuclear Power in the Soviet Union", IAEA Bulletin, June 1983, p. 47. We assume an average load factor of 75 percent.

c. We assume an average burnup of 25,000 Mwt-D/tonne, the fissioning of 8 kg Pu-239/tonne, and an average heat-to-electricity conversion ratio of 0.3. This results in the production of 1.13 MCI/Twt-day(e) (see assumptions in Table 5-1).

Table 5-17. Summary of Estimated Kr-85 Releases (MCI)

<u>Source</u>	<u>1955.5</u>	<u>1958.5</u>	<u>1964.9</u>	<u>1972.5</u>	<u>1981.0</u>	<u>1984.0</u>
<u>Nuclear Weapons Tests</u>						
<u>Atmospheric</u>	0.8	1.5	3.9	4.2	4.3	4.3
<u>Underground</u>				0.3	0.7	0.7
<u>Reprocessing in US</u>						
<u>Hanford</u>						
--Original 8	1.73	4.42	14.59	21.30	21.3	21.30
--N-reactor	0.00	0.00	0.00	1.81	1.81	1.81
<u>Savannah River</u>						
--Production React.	0.0	1.33	8.53	13.96	18.84	20.75
--Research React.	0	0	0	0.3	0.5	0.6
NFS (Civilian Fuel)	0	0	0	0.71	0.71	0.71
Idaho CPP	0.08	0.3	0.80	1.42	2.08	2.15
US SUBTOTAL	<u>1.81</u>	<u>6.05</u>	<u>23.92</u>	<u>39.50</u>	<u>45.24</u>	<u>47.42</u>
<u>Sellafield (Britain)</u>	0.08	0.19	1.17	6.88	14.76	18.48
<u>Reprocessing in France</u>						
La Hague	0.0	0.0	0.0	0.4	5.6	9.0
Marcoule	0.0	0.0	0.20	1.00	2.3	3.3
<u>Other Reprocessing</u>						
W. Europe	0.0	0.0	0.0	0.14	0.85	0.93
Japan	0.0	0.0	0.0	0.0	0.34	0.73
SUBTOTAL(W. Eur. & Japan)	<u>0.08</u>	<u>0.19</u>	<u>1.37</u>	<u>8.42</u>	<u>23.85</u>	<u>32.44</u>
<u>Leakage from Fuel (outside USSR)</u>	0.0	0.0	0.0	0.1	0.4	0.7
<u>Total From Tests and NonCommunist States<sup>a</sup></u>	<u>2.7(0.3)</u>	<u>7.7(0.8)</u>	<u>29.2(2.9)</u>	<u>52.5(5.2)</u>	<u>74.5(7.0)</u>	<u>85.5(9.0)</u>
<u>Total Released to Atmosphere (Uncertainty)</u>	2.1(0.9)	7.7(3.2)	41.5(5.0)	75.2(7.4)	125.7(4.6)	144.8(4.8)
<u>Residual From Reactor Fuel in Communist States<sup>b</sup></u>	<u>-0.6(1.0)</u>	<u>0.0(3.3)</u>	<u>12.3(5.8)</u>	<u>22.7(9.0)</u>	<u>51.2(8.4)</u>	<u>59.2(10.2)</u>
<u>Annual Average Releases From Communist Reactor Fuel<sup>c</sup></u>	--	--	1.9(0.9)	3.4(1.0)	2.7(1.7)	-----3.2(1.2)-----

Table 5-17, Notes

- a. We assume an uncertainty of 10 percent.
- b. The uncertainty is the square root of the sums of the squares of the uncertainties of the atmospheric inventory and the total from tests and nonCommunist reactors.
- c. We assume an uncertainty of 10 percent in the differences of the total releases from tests and nonCommunist states. For the 1981-84 period, we have used the uncertainty in the increase of the atmospheric inventory shown in Table 5-6.