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ADDENDUM TO FMPC-2082 HISTORY OF FMPC RADIONUCLIDE DISCHARGES MARCH 1989

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Addendum to FMPC-2082

HISTORY OF FMPC RADIONUCLIDE DISCHARGES

Revised Estimates of Uranium and
Thorium Air Emissions
from
1951 - 1987

by
T. R. Clark
L. Elikan
C. A. Hill
B. L. Speicher

March 1989

FEED MATERIALS PRODUCTION CENTER
Westinghouse Materials Company of Ohio
P. O. BOX 398704
CINCINNATI, OHIO 45239-8704

PREPARED FOR THE
U.S. Department of Energy
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Approved: W. A. Weinreich
Vice President and Technical Director

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INTRODUCTION

Air emissions of radionuclides from the Feed Materials Production Center (FMPC) have been estimated on several occasions in the past. The latest and most comprehensive estimate is contained in FMPC-2082 "History of FMPC Radionuclide Discharges" which was issued in May 1987[1]. This information has been augmented by the publication of the Annual Environmental Monitoring Report for 1987 which was issued on April 30, 1988[9].

This addendum has been prepared following the determination in June, 1988, of higher than expected airborne concentrations of uranium at air monitoring stations in the northeast quadrant of the plant. These air concentrations were traced to the operation of the Plant 2/3 denitration gulping system which was a previously unmonitored source of uranium emissions. In addition, the question was raised as to what other unmonitored and potentially unreported air emission sources existed now or in the past. The scope of this addendum was to revise the Plant 2/3 emission estimate and identify any additional radionuclide air sources which were not included in FMPC-2082. The estimate covers the period from 1951 to 1987. Air emissions for 1988 will be reported in the Environmental Monitoring Report for 1988 to be published at a later date.

The review for additional radionuclide air sources required a re-creation of the history of operations at the FMPC. This was done by reviewing plant records and interviewing long term employees. While these efforts enabled the authors to prepare a revised estimate, the 37-year history of FMPC radionuclide air emissions can not be prepared without some degree of uncertainty. The factors which introduce uncertainty into the historic air emission estimate include:

- Documentation of all operations and events did not exist or could not be found.

- Personal recall of events was useful, but is incomplete and was not always consistent.

- Some measurements necessary to calculate emissions were not recorded or archived.

In preparing this addendum, the revised estimate of uranium and thorium emissions was developed as a conservatively based best estimate of the air emissions from 1951 through 1987. In addition, an estimate of the uncertainty of this value has been provided.

SUMMARY

This study results in estimated uranium air emissions of 179,000 kg and an estimated thorium air emission of 6500 kg for the years of 1951 through 1987.

These revised estimates are conservative and differ from the estimate reported in FMPC-2082 by the addition of emissions from the following:

- Uranium trioxide gulping - Plant 2/3
- Other unmonitored processes
- Building exhausts
- Laboratory vents
- Non-routine events
- Fugitive emissions from waste pits
- Additional years of thorium processing from which emissions had not previously been reported.

Table 1 provides an annual summary of these emissions. The uranium estimate shows the value reported in FMPC-2082 with the additional uranium emissions listed by category. The thorium estimate is a revised estimate that is to be used instead of the value reported in FMPC-2082.

Figure 1 shows graphically the relationship between the yearly estimates from FMPC-2082 and the new revised estimates for uranium. The figure also shows the distribution of the thorium emissions by year.

TABLE 1

FMPC EMISSIONS SUMMARY IN KILOGRAMS

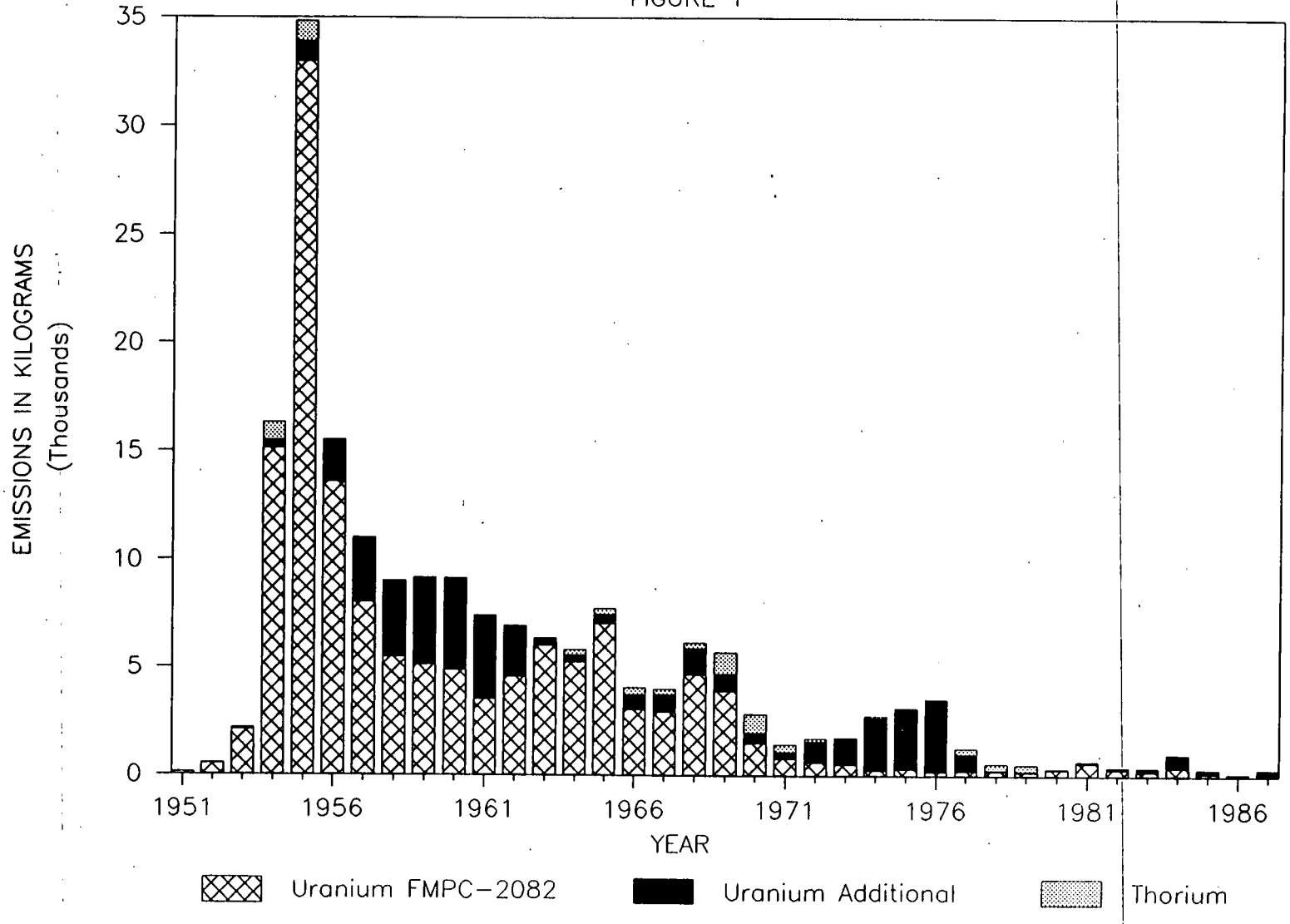
Year	1 Uranium FMPC-2082 Totals	2 Uranium Gulping Emissions	3 Other Uranium Process Emissions	4 Uranium Emissions Building Exhausts	5 Uranium Estimated Laboratory Emissions	6 Uranium Fugitive Emissions Waste Pits	7 Uranium Emissions Non-Routine Events	8 Total Uranium Emissions	9 Thorium Process Emissions	10 Thorium Fugitive Emissions Waste Pits	11 Total Thorium Emissions
1951	123.0	0	0	0	2	0	0	125	0	0	0
1952	499.0	0	0	0	2	0	42	543	0	0	0
1953	2,077.8	0	3	1	2	0	99	2,183	0	0	0
1954	15,119.2	210	8	5	2	0	142	15,486	815	0	815
1955	32,976.2	750	11	11	2	1	142	33,893	933	0	933
1956	13,595.4	1,750	12	16	2	2	142	15,519	0	0	0
1957	8,045.2	2,750	12	26	2	48	142	11,025	0	0	0
1958	5,513.4	3,300	14	31	2	95	100	9,055	0	0	0
1959	5,127.4	3,810	15	28	2	95	100	9,177	0	0	0
1960	4,872.8	4,020	17	33	2	108	100	9,153	0	0	0
1961	3,516.4	3,640	18	30	2	121	100	7,427	0	1	1
1962	4,568.0	2,070	19	26	2	125	132	6,942	0	2	2
1963	6,036.4	0	19	25	2	125	168	6,375	0	2	2
1964	5,253.4	0	22	20	2	129	157	5,583	246	2	248
1965	7,044.8	180	22	19	2	71	155	7,494	246	2	248
1966	3,048.5	460	12	16	2	49	143	3,731	335	2	337
1967	2,924.7	620	11	16	2	11	141	3,726	246	2	248
1968	4,655.2	1,110	7	14	2	12	85	5,885	246	2	248
1969	3,898.1	690	7	8	2	15	88	4,708	1030	2	1,032
1970	1,487.8	310	6	6	2	16	155	1,983	869	2	871
1971	772.0	280	5	2	2	16	15	1,092	356	2	358
1972	614.4	950	4	3	2	15	13	1,601	106	2	108
1973	496.0	1,210	5	3	2	15	8	1,739	38	2	40
1974	234.8	2,430	4	6	2	14	6	2,697	75	3	78
1975	318.0	2,780	4	7	2	18	9	3,138	2	3	5
1976	169.1	3,330	5	7	2	20	6	3,539	0	4	4
1977	191.9	750	5	2	2	20	7	978	292	4	296
1978	222.0	0	4	2	2	22	9	261	291	4	295
1979	154.7	0	4	1	2	31	7	200	291	4	295
1980	266.5	0	4	2	2	34	8	317	0	4	4
1981	587.2	30	5	2	2	42	9	677	0	4	4
1982	279.8	50	6	2	2	41	14	395	0	4	4
1983	181.2	130	6	4	2	40	13	376	0	4	4
1984	377.5	570	6	5	2	40	13	1,014	0	4	4
1985	121.3 *	130	6	4	2	40	12	315	0	4	4
1986	62.2 *	0	6	4	2	40	16	130	0	4	4
1987	42.3 *	200	5	2	2	41	10	302	0	4	4
Total	135,473.6	38,510	319	389	74	1,512	2,780 **		6417	79	
						TOTAL URANIUM		179,058 kg		TOTAL THORIUM	6,496 kg

NOTE: Numbers may not add due to round-off.

* The 1985, 1986 and 1987 totals are those reported in the Environmental Monitoring Annual Reports as corrected for dust collector round-off error.
** Includes 272 kgs U from estimated emissions not distributed over production years - see Table 3

FMPC EMISSIONS SUMMARY

FIGURE 1



METHODS USED FOR ESTIMATING EMISSIONS

The methods utilized to estimate the emissions listed in columns 2-11 of Table 1 are outlined in this section. Documents are referenced which contain information relative to the estimating techniques.

Uranium Gulping (Column 2)

The quantity of uranium emitted from the Plant 2/3 gulping system was estimated based on uranium production records, recent measurements of the uranium content of mists re-entrained from the scrubbing system, combined measurements of mist and particulate emissions that occur when the contents of a denitration pot are gulped, and the collection efficiency expected from the particle control train. This method is explained in detail in FMPC/Sub-019, "Uranium Emissions from Gulping of Uranium Trioxide."[2]

Unmonitored Uranium Processes Other Than Gulping (Column 3)

A systematic evaluation was made of all unmonitored process stacks at FMPC. Estimates of radioactive emissions were made by engineering calculations or by actual stack emission measurements.[3]

In general, a detailed analysis was made of the number of operating hours, the number of tons processed, and the probable emissions for each unmonitored process for the year 1987. This information was utilized to estimate an emission factor per ton of uranium processed for each unmonitored process. These emission factors per ton were then utilized together with the plant-by-plant production data (Table 2) to estimate the emissions from the unmonitored processes in each year.

TABLE 2

PRODUCTION OPERATIONS DATA FROM COST STATEMENT ANNUAL REPORTS AND ESTIMATES
Production Quantities in metric tons.

Fiscal Year	Plant 2/3 Evap. Calciner	Plant 2/3 UO3 Prod.	Pilot Plant UF4	Plant 4 UF4	Plant 5 Derby Prod.	Plant 5 Metal Prod.	Plant 8 Total Production	Plant 9 Metal Prod.	Plant 6 Machining	Plant 9 Briquet	Plant 9 Pickling	Plant 6 Briquet	Plant 6 Pickling	Plant 9 Zirnlo
1952 *	0	0	10 #	0	0	0	0	0	0	0	0	0	0	0
1953 *	0	0	6 #	0	45	90	0	0	1608	0	0	170	10	0
1954 *	0	642	7 #	1568	2099	3976	266	0	3581	0	0	756	462	0
1955 *	0	3288	21 #	3314	5824	9528	1160	0	6752	0	0	1628	1281	0
1956 *	0	5329	31 #	5029	8459	12037	1764	0	8086	0	0	2012	1861	0
1957 *	0	8370	27 #	9358	6113	12680	1927	0	8629	0	0	2131	1345	0
1958 *	0	10039	0	12117	6749	12727	2018	732	8378	476	527	2184	1485	0
1959 *	0	11540	0	9454	7759	13365	2568	1251	7320	813	901	2194	1707	0
1960 *	0	12187	0	11388	10586	16708	3188	1388	9131	902	999	2723	2329	0
1961 *	0	11039	0	10642	8470	12691	2902	2364	7552	1537	1702	2261	1863	0
1962 *	0	6288	1342	9468	8563	12865	2820	2663	8211	1731	1917	2374	1884	0
1963 *	0	0	1197	10482	10243	14285	2657	3660	9232	2379	2635	2718	2253	144
1964 *	0	0	1468	7203	7648	11655	3505	5297	9279	3443	3814	2623	1683	144
1965 *	0	543	658	6797	6432	10234	2134	5361	8674	3485	3860	2427	1415	144
1966	0	1347	478	6174	5166	11239	1617	1197	6987	778	980	1945	1834	210
1967	0	1835	13	6263	7172	10969	1837	1258	5837	818	843	1865	1674	176
1968	0	3251	0	4809	5339	10144	2222	691	5105	0	0	1351	1333	133
1969	0	2028	0	2827	4017	6638	1036	778	3227	0	0	1395	1434	96
1970	0	880	0	1927	2885	5425	649	499	2882	0	0	999	1363	104
1971	0	809	0	0	1344	2375	307	422	1413	0	0	549	646	21
1972	0	2761	0	348	1217	1683	111	599	922	0	0	397	532	0
1973	0	3534	0	0	2139	3292	66	452	1881	0	0	715	733	0
1974	0	7114	0	342	1317	1711	3	1031	870	0	0	408	638	0
1975	0	8129	0	634	1121	1167	43	1189	797	0	0	297	509	0
1976	0	9752	0	0	1703	2142	51	304	1065	0	0	436	517	0
1977	0	2191	0	0	1780	2175	386	381	1110	0	0	445	446	20
1978	11	0	0	544	2139	1968	122	480	1172	0	0	418	851	78
1979	11	0	0	0	1618	1386	184	604	900	0	0	325	865	67
1980	0	0	0	479	2023	1989	118	380	999	0	0	360	957	80
1981	0	103	0	562	2608	2047	41	796	1127	0	0	461	1410	31
1982	0	203	0	366	4159	3732	237	974	1821	0	0	795	1667	76
1983	0	219	0	1145	4802	4569	376	1366	2191	0	0	880	1969	157
1984	0	937	0	1240	6290	3933	261	1516	1924	0	0	843	3657	250
1985	0	145	622	1146	5075	4559	188	1074	1860	0	0	763	2120	213
1986	0	0	462	1068	6205	4310	176	1640	1743	0	0	797	3629	344
1987	0	98	1642	388	2667	2365	261	394	8	0	0	159	1915	4049
TOTAL	22	114,601	7,984	127,082	161,776	232,659	37,201	40,741	142,274	16,362	18,178	42,804	50,277	6,537

* Cost statements were not available for 1953-1965. Production quantities were obtained from MC&A inventory records for Plant 2/3, Pilot Plant UF4, Plant 4 UF4, Plant 5 Derby and Ingot Production and Plant 6 Machining.

Production quantities for Plant 9 Briquetting and Pickling and Plant 6 Briquetting and Pickling were estimated based on ratios of material pickled or briquetted to metal produced during the years for which information was available.

Plant 9 Zirnlo production was estimated for 1963-1965 as an average of material processed from 1966-1970.

\# Data obtained from 1956 report on Pilot Plant uranium hexafluoride operations. 1957 production was estimated.

In the few instances where a comparable process was not available, a methodology was utilized similar to that discussed on page 12 for thorium process emissions.

The summation of the unmonitored emissions from all plants for each year is indicated in column 3 of Table 1.

Uranium Emissions from Building Exhausts (Column 4)

Building ventilation emissions were calculated based on data collected by continuous air monitors located in each plant. Continuous air monitors are located throughout the plants near processes suspected to produce radionuclide emissions. The monitors are used to measure alpha and beta radiation in microcuries per cubic centimeter. In order to convert from microcuries to grams of uranium, the source of all radiation measured by the continuous air monitors was assumed to be normal uranium; this assumption is reasonable since the plants process large quantities of depleted uranium and lesser amounts of slightly enriched uranium. Data on the uranium concentration in each plant for each month was averaged to provide an annual concentration for 1987.

The total volumetric flow capacity of all the fans in each plant was determined from FMPC records or by estimation based on the size of the exhaust fan. To calculate yearly emissions for each plant, the average annual concentration reported from the plant continuous air monitors was multiplied by: 1) the estimated hours each fan is operated per year; and 2) by 0.1 to account for dilution by building make-up air (since the air concentration in the plants is only measured in the areas of highest suspected concentration, the actual plant average uranium concentration in air is significantly lower than the average calculated from continuous air monitor results).

It was also assumed that emissions from building exhausts would be proportional to production rates. Historical emission levels for each plant were estimated by multiplying its 1987 emission value by a ratio of production in the year in question to production in 1987.

Uranium Emissions from Laboratory (Column 5)

Estimates were based upon the average number of samples processed per year in each hood, the probable loss per sample and the estimated uranium concentration of the samples. Existing hoods and known former lab exhausts were evaluated and summarized[4].

Fugitive Uranium and Thorium Emissions from Waste Pits (Columns 6 and 10)

Fugitive dust generated from storage piles can generally be categorized as follows:

1. Load-in of material into the storage area,

2. Wind erosion of the stored material,

3. Vehicle movement in the storage area, and

4. Load-out of material from the storage area.

In the calculation of the fugitive dust emissions from the FMPC waste pits, only the first two categories were judged to contribute significantly to the overall generation of dust emissions. The calculations for vehicle movement were based on wheeled vehicle movement over paved or unpaved roadways. Since the amount of this type of traffic in the waste pits was minimal, this term was not considered. The load-out factor is not applicable since the material has not been removed from the waste pits. The load-in contribution to fugitive emissions from the waste pits that were operated in a wet mode was not calculated because the material was transferred to the pits in a slurried form, and therefore, would not contribute to the dust emissions.

The basic methodology used in determining the masses of contaminants (uranium and thorium) that potentially left the waste pits was first to determine a factor for each of the applicable categories contributing to fugitive emissions for each waste pit. Once this emission factor was determined, the overall mass of material that could become airborne was estimated. Finally, an average contaminant concentration was applied to the overall mass to estimate the mass of contaminant potentially leaving the waste pits.

Computations were performed in accordance with the method recommended by EPA.[5] Reference 6 details the calculations performed to arrive at the numbers listed in columns 6 and 10 of Table 1.

Uranium Emissions from Non-Routine Events (Column 7)

The estimate of emissions from non-routine events was based upon review of historical site documents and interviews with plant personnel to determine the types and sequences of events. Estimates of emissions from each type of event were based upon historical information, familiarity with plant operation, and engineering judgement, where applicable data was not available.

Four categories of non-routine events were identified that would have significant radioactive emissions: uranium fires, solid spills, UF_6 leaks, and releases of uranyl nitrate. There were previously reported non-routine events included in FMPC-2082, including a 1966 UF_6 release. These emissions are included in column 1 of Table 1 and not duplicated in column 7.

Detailed results of this work are contained in Reference 10, and the results summarized in Table 3.

TABLE 3

ESTIMATED EMISSIONS DUE TO NON-ROUTINE EVENTS REPORTED IN KILOGRAMS

Year	Uranium Fires	Solid Spills (Outdoor)	UF6 Leaks	Yearly Totals	Concentrated Liquid UNH Releases
1951	---	---	---		---
1952	---	---	42	42	---
1953	---	57	42	99	---
1954	43	57	42	142	---
1955	43	57	42	142	---
1956	43	57	42	142	---
1957	43	57	42	142	---
1958	43	57	0	100	---
1959	43	57	0	100	---
1960	43	57	0	100	---
1961	43	57	0	100	---
1962	33	57	42	132	---
1963	69	57	42	168	---
1964	58	57	42	157	---
1965	56	57	42	155	---
1966 ***	44	57	42	143	---
1967	42	57	42	141	---
1968	28	57	0	85	---
1969	31	57	0	88	---
1970	149	6	0	155	---
1971	9	6	0	15	---
1972	7	6	0	13	---
1973	2	6	0	8	---
1974	0	6	0	6	---
1975	3	6	0	9	---
1976	0	6	0	6	---
1977	1	6	0	7	---
1978	3	6	0	9	---
1979	1	6	0	7	---
1980	2	6	0	8	---
1981	3	6	0	9	---
1982	8	6	0	14	---
1983	7	6	0	13	---
1984	7	6	0	13	---
1985	9	1 *	2	12	---
1986	7	1 *	8	16	---
1987	7	1 *	2	10	---
SUBTOTALS	930	1,062	516	2,508	272 **
TOTAL					2,780

* Data for these years from Minor Event Reports and resulting emissions were rounded up to 1 Kg U.

** This estimate is based on an estimated 6 incidents (including 2 specifically documented), not on yearly averages, and is added to the sum of the yearly totals to provide the overall value.

*** Emissions from a 1966 UF6 leak are included in FMPC-2082.

Thorium Process Emissions (Column 9)

Since no thorium processes are currently in operation at the FMPC and because most of the equipment used to process thorium has been removed, emission factors could not be calculated based on sampling results. Additionally, the factors developed for uranium processes could not be applied to thorium processes due to differences in the processes. In order to develop an estimate of thorium emissions, thorium processing at the FMPC was reconstructed based on process flow descriptions and available production yields. Where production data was not available, estimates were made based on product volume and yield information.

Information regarding each of the individual processes was used to develop process flow sheets. As each process was broken into its components, locations of possible emission sources associated with each of the process operations were identified based on current process design, as well as information located during the file search. Digestion tanks, precipitation tanks, and furnaces were all considered to be possible unmonitored emission sources.

Because of the limited information available, the method developed used published handbook values to estimate the efficiencies of the emission control devices and the percentage of throughput which reached the emission control device, i.e., scrubber or dust collector. Both venturi-type scrubbers and packed-tower scrubbers were used in the thorium processes. An efficiency of 90 percent particle removal was chosen to represent the general "wet" scrubber used. The precedent for using this value was the Nuclear Regulatory Commission (NRC) which uses a 90 percent efficiency to estimate particulate emission reduction by a venturi-type scrubber. [11]

Ninety-five percent was chosen as the dust collector particle removal efficiency. Most dust collectors at the FMPC are currently rated at greater than 99 percent efficient; therefore, using an efficiency of 95 percent allows operational difficulties to be included in the estimate.[1] Most of the flow diagrams did not indicate which processing steps were vented to dust collectors. Dust collectors were assumed to be connected to dumping and packaging stations and all other sources of dry particulates, as they have historically been for all uranium production operations.

The quantity of total throughput which reaches each emission control device was estimated to be one percent of the production throughput (with respect to uranium or thorium). This is the quantity used by the NRC when estimating emissions from uranium milling processes.[11] One percent is a conservative estimate since most of the emission sources for thorium processes were oxidation furnaces and precipitation tanks which involve mixing of two solutions. Neither of these source types generate particulates in significant quantities.

Results of the thorium emission estimates by process are summarized in Table 4. A report describing the historical thorium production processes and giving more detail on the sources of production data for thorium materials is provided[12].

TABLE 4

THORIUM EMISSION ESTIMATE

Process	Years of Production Operation	Years of Production Data	Total Production (Kilograms)	# of Scrubbers	# of Dust Collectors	Estimated Scrubber Efficiency	Estimated D.C. Efficiency	Estimated % of Material to Offgas	Estimated Thorium Emissions
Plant 9 Metal	1954-1955	estimated	380,000	2	4	90%	95%	1%	1,520
Plant 9 Briquetting	1954-1955	estimated	76,000	2	2	90%	95%	1%	228
Plant 4 ThF4	1954	none	unknown	2	2	90%	95%	1%	unknown
Pilot Plant Extraction	1964-1980	1966-1973	790,400	---	---	90%	95%	1%	negligible
Pilot Plant Gel	1964-1970	1966-1970	688,800	2	1	90%	95%	1%	1,722
Pilot Plant TNT Crystals	1966	1966	425	1	0	90%	95%	1%	negligible
Pilot Plant Metal	1969-1971	1969-1971	51,000	2	4	90%	95%	1%	204
Pilot Plant Oxalate	1971-1976	1971-1976	153,000	1	1	90%	95%	1%	230
Pilot Plant Gel	1977-1979	1977-1979	349,700	2	1	90%	95%	1%	874
Plant 2/3 Test	1968	none	negligible	---	---	90%	95%	1%	negligible
Plant 8 Hydroxide	1966	1966	59,000	1	1	90%	95%	1%	89
Plant 8 Oxalate	1969-1971	1969-1971	310,000	4	2	90%	95%	1%	1,550
								TOTAL	6,417 kilograms

Calculation method:

(# of Scrubbers)x(Total Production)x(0.01)x(1-0.90) + (# of Dust Collectors)x(Total Production)x(0.01)x(1-0.95)

ESTIMATES OF THE UNCERTAINTY

The estimation of historical air emissions over a period of 37 years involves numerous sources of uncertainty including: unavailable documentation, incomplete personal recall, and lack of measurements. In addition, the revised best estimate relies on terms which have a degree of variability.

Our analysis of the uncertainties indicates that there is a low probability that the cumulative air emissions for the years 1951 through 1987 will exceed 250,000 kg for uranium and 10,000 kg for thorium. This is based upon parametric analysis and subjective judgement and is not based on statistical analysis, since sufficient data for statistical analysis does not exist.

Considerations included in generating these upper bound values are:

1. Dust Collectors
   A dust collector emission bound was developed by analyzing for potential sources of bias in the methods used to sample the outlet stream and the chemical analyses performed on the samples. Sources for bias include sampler mechanical problems, analytical bias, representativeness of samples, and closeness to isokinetic condition. Each of these contributors was estimated to arrive at a cumulative potential bias of 12 percent for the dust collector emissions.

2. Plant 8 Scrubbers
   An uncertainty limit for uranium process emissions from scrubbers was based on a substantial group of data taken during the highest throughput period in Plant 8. This data indicates that scrubber efficiencies averaged 83 percent during the years of highest production. Other data indicates that scrubber efficiencies were generally higher when production was lower. Therefore, an upper bound for Plant 8 scrubbers was based upon achieving only an 83 percent efficiency, regardless of plant throughput.

3. Uranium Trioxide Gulping

Reference 2 examines the sensitivity of the calculational model used for estimating gulping emissions to various changes in parametric values utilized. In arriving at the estimated upper limit in this work, additional parametric variations were considered.

4. Non-routine, Laboratory, and Fugitive Sources

These sources were insignificant contributors due to their low percentage of the estimated emissions.

5. Thorium Processes

The upper bound for thorium was also developed by assuming the reduced scrubber efficiency of 83 percent instead of the 90 percent indicated on page 12.

In conclusion, the estimates of 179,000 kg uranium and 6,500 kg thorium are considered conservatively high. The values for the upper limits of uncertainty represent total air emissions from the site that are unlikely to be exceeded as a result of future source identification or improved estimating methods.

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