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PLUTONIUM AND AMERICIUM CONTRIBUTIONS TO  
TOTAL RADIOACTIVITY OF AIR NEAR ROCKY FLATS

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Environmental Analysis

Distribution

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## INTRODUCTION

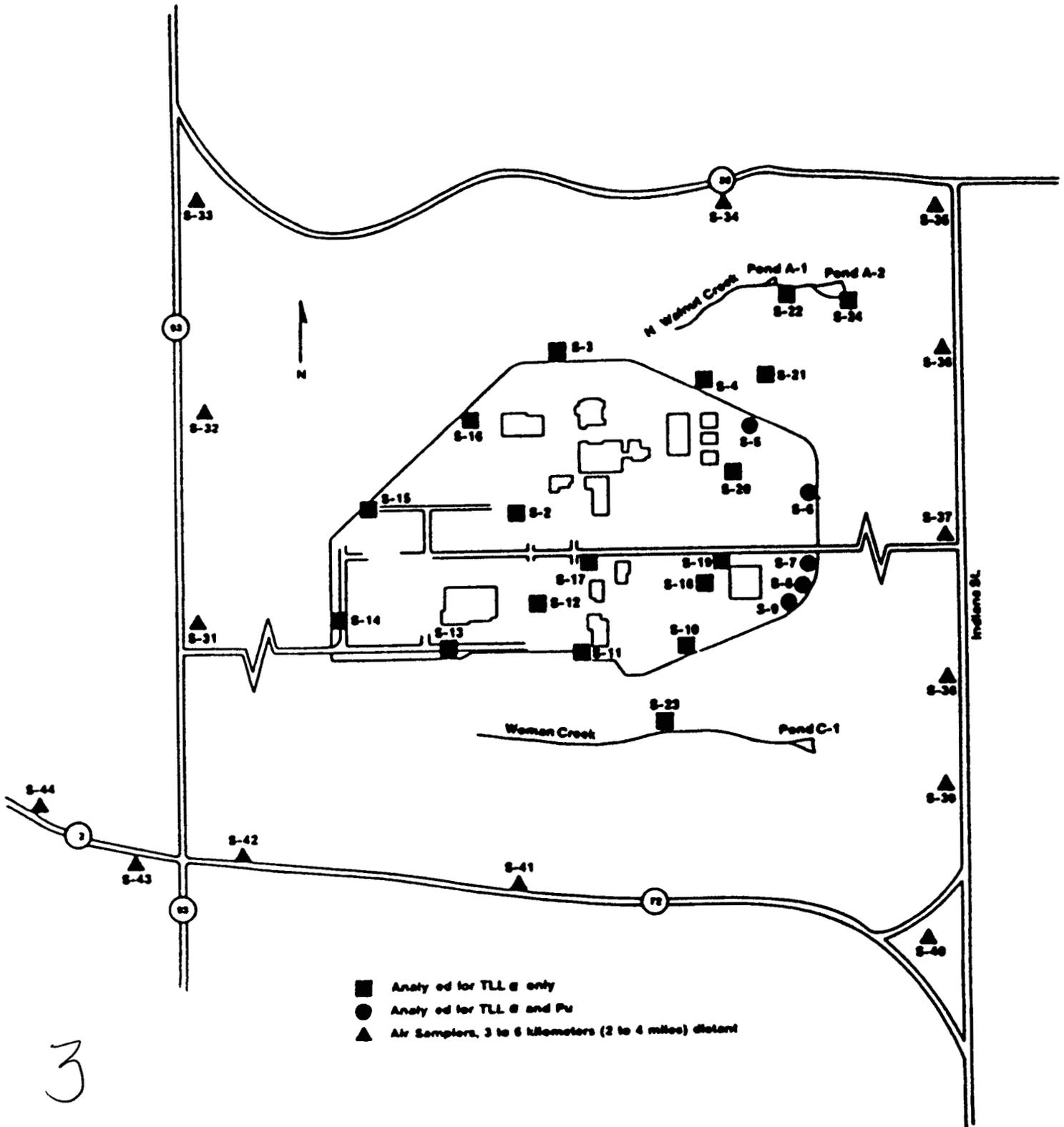
Ambient air is monitored for airborne particulate matter by a network of 49 air sampling stations located both on and off the Rocky Flats Plant site. Each of the air sampling stations contains a vacuum pump that draws a known volume of air through a standard filter. The air samplers are of a Rocky Flats design which is described in Rockwell Engineering Drawings 27261 1 through 27261 6. The high volume samplers operate continuously at a volume flow rate of approximately 19 l/sec (40 ft<sup>3</sup>/min) and particulates are collected on a 20 X 25 centimeter (8 X 10 inch) Delbag Microsorban<sup>®</sup> filter medium.

Airborne particulates in ambient air are sampled continuously at 23 locations within and adjacent to the Rocky Flats exclusion area (Figure 1). The sample filters are collected weekly and analyzed for total long-lived alpha activity (TLL $\alpha$ ). If the TLL $\alpha$  concentration for an ambient air sample exceeds a Plant guide value of 0.01 pCi/m<sup>3</sup>, a specific plutonium analysis is performed. On a routine basis, filters from 9 of the 23 samplers are composited and analyzed biweekly for plutonium. Figure 1 identifies these samplers.

Since the TLL $\alpha$  concentrations have historically been considerably higher than the plutonium concentrations at the nine samplers analyzed for both parameters, it was decided to measure the contributions made by other alpha particle emitters. Although a total activity balance was not anticipated, it was assumed that activity from natural uranium, thorium and their daughter nuclides would comprise a major portion of the total alpha activity. The data might indicate the relative proportion of man-made radioactivity (plutonium and americium) to the total measured radioactivity.

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FIGURE 1 Location of Onsite Ambient Air Samplers



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## METHODS

Filters were selected from samplers S 3 S-14 S-23 and S-24 (Figure 1) during the months of April May and June for the years 1978 and 1979. This process yielded over 100 individual filter papers. In order to reduce the analytical costs filters were composited by each month to yield a total of 24 samples or 12 samples for each year. The composites were sealed in envelopes and given numbers relating to the sampler location i e 3 1 through 3-3 14-1 through 14-3 23-1 through 23-3 and 24-1 through 24 3. These numbers were then given blind numbers selected from a table of random numbers.

The samples were submitted for analysis to the Radiological and Environmental Sciences Laboratory (RESL) of the Department of Energy (DOE) and to the Los Alamos National Laboratory (LANL) contractor to the DOE. RESL employed a sequential radiochemical procedure for the analysis of  $^{226}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{234}\text{U}$ ,  $^{239+240}\text{Pu}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$ . Blank samples were also analyzed and the results were subtracted from the sample values. LANL used several different types of procedures for the analyses performed by that laboratory. Plutonium and americium were determined by alpha spectrometry following chemical separations. Uranium isotopes were measured by delayed neutron assay after reactor irradiation. The isotopic ratios of uranium isotopes were determined by using the  $^{239}\text{Np}$  from  $^{238}\text{U}$  and the  $^{143}\text{Ce}$  fission product from  $^{235}\text{U}$ . Thorium and radium isotopes were not measured by LANL.

## RESULTS

The results of the analyses of the sample filters were summarized and are presented in Table I. Data are reported as picroCuries of each nuclide per sample. The normal procedure of reporting concentrations in picro Curies per cubic meter was not followed because all comparisons are made between amounts of each nuclide present on each filter paper. The volume of air pulled through the filter is a common denominator for each filter sample.

Since LANL did not analyze the samples for radium or thorium, an attempt was made to estimate the amount of each nuclide present. Based on the uranium plus daughter product analyses by RESL, it was determined that  $^{226}\text{Ra}$ ,  $^{230}\text{Th}$  and  $^{234}\text{U}$  were present in almost equilibrium amounts to the parent  $^{238}\text{U}$ . Therefore, the LANL-analyzed samples were assigned daughter-nuclide quantities equal to the measured  $^{238}\text{U}$ . Evaluation of ratios of  $^{232}\text{Th}$  to  $^{238}\text{U}$  in samples analyzed by RESL show an average ratio of 1:15. This ratio was used to calculate the amounts of  $^{232}\text{Th}$  and  $^{228}\text{Th}$  in the LANL samples.

The total of the analyzed radionuclides in each filter sample is given in Table I. The sum of plutonium plus americium activities was divided by the total activity and the percentages are shown in Table I. These data are summarized in Table II.

TABLE I Summary of Results for Rocky Flats Ambient Air Samples Special Analyses (pCi/Sample)

Sampler	Date	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>232</sup> Th	<sup>230</sup> Th	<sup>238</sup> U	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> Pu	<sup>239+240</sup> Pu	<sup>241</sup> Am	Total pCi	Percent Pu + Am/Total
S-3	04/78	0 38†	0 44*	0 44*	0 38†	0 38	0 38†	0 06	<0 006	1 13	0 46	4 06	39 2
S 3	05/78	3 93	6 39	6 21	4 11	4 34	4 33	0 15	0 020	2 83	0 39	32 7	9 9
S 3	06/78	2 99†	3 44*	3 44*	2 99†	2 99	2 99†	0 14	<0 006	0 47	0 84	20 3	6 5
S-3	04/79	2 90	5 29	5 00	3 41	3 53	3 42	0 15	0 008	1 54	0 28	25 5	7 2
S 3	05/79	1 53†	1 76	1 76*	1 53†	1 53†	1 53	0 08	<0 004	0 19	0 53	10 4	6 9
S 3	06/79	1 11†	1 28	1 28*	1 11†	1 11	1 11†	0 07	0 03	0 53	0 60	8 23	14 1
S-14	04/78	3 29	3 93	3 53	2 99	3 04	3 18	0 09	0 050	3 09	0 29	23 5	14 6
S-14	05/78	0 47†	0 54	0 54*	0 47†	0 47	0 47†	0 04	<0 006	0 83	0 18	4 02	25 4
S 14	06/78	2 17	2 23	1 83	1 92	2 17	2 26	0 08	0 017	1 92	0 14	14 8	14 1
S 14	04/79	1 25	1 88	1 69	1 23	1 28	1 43	0 02	0 018	1 21	0 13	10 2	13 3
S 14	05/79	1 05	1 70	1 50	1 04	1 18	1 31	0 03	0 009	0 82	0 08	8 73	10 4
S 14	06/79	0 88†	1 01	1 01*	0 88†	0 88	0 88†	0 04	<0 013	0 50	0 64	6 73	17 1
S 23	04/78	0 93†	1 07	1 07*	0 93†	0 93	0 93†	0 04	0 05	2 00	0 09	8 04	26 6
S-23	05/78	0 27†	0 31	0 31*	0 27†	0 27	0 27†	0 03	<0 005	0 86	0 10	2 70	35 8
S-23	06/78	0 50†	0 58	0 58*	0 50†	0 50	0 50†	0 03	0 01	0 51	0 16	3 87	17 6
S-23	04/79	0 52†	0 60	0 60*	0 52†	0 52	0 52†	0 03	0 083	0 75	0 67	4 81	31 2
S-23	05/79	0 80	0 81	0 64	0 52	0 80	0 72	0 01	0 003	0 77	0 10	5 18	16 8
S-23	06/79	1 00	1 01	0 89	0 87	1 06	1 12	0 06	0 005	0 72	0 09	6 82	11 9

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TABLE I (Continued)

Sampler	Date	$^{226}\text{Ra}$	$^{228}\text{Th}$	$^{232}\text{Th}$	$^{230}\text{Th}$	$^{238}\text{U}$	$^{234}\text{U}$	$^{235}\text{U}$	$^{238}\text{Pu}$	$^{239+240}\text{Pu}$	$^{241}\text{Am}$	Total pCi	Percent Pu + Am/Total
S 24	04/78	0 34+	0 39*	0 39*	0 34+	0 34	0 34+	0 02	<0 005	0 26	<0 09	2 52	14 1
S 24	05/78	0 82	0 89	0 70	0 89	1 10	1 13	0 03	0 023	2 67	0 22	8 47	34 4
S 24	06/78	0 63+	0 72*	0 72*	0 63+	0 63	0 63+	0 04	<0 007	1 40	0 10	5 51	27 4
S 24	04/79	0 87	1 38	1 10	0 92	1 15	1 30	0 02	0 011	1 19	0 11	8 05	16 3
S 24	05/79	0 39	0 47	0 41	0 04	0 40	0 41	0 00	0 005	0 54	0 09	2 76	23 0
S-24	06/79	1 18	1 34	1 17	1 11	1 45	1 49	0 03	0 004	1 29	0 13	9 19	15 5

\* Estimated from RESL results  
 + Taken as equal to U-238

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TABLE II Comparison of Plutonium and Americium to Total Measured Activity Ratios (pCi/sample)

Station	Pu + Am		Total		Pu + Am/ Total X 100	
	Range	Avg	Range	Avg	Range	Avg
S 3	0 72 3 22	1 65	8 23-32 7	16 9	6 9-39 2	14 0
S-14	0 90 3 38	1 66	4 02 23 5	11 3	10 4-25 4	15 8
S 23	0 67-2 09	1 17	2 70 8 04	5 24	11 9 35 8	23 3
S 24	0 35 2 89	1 36	2 52- 9 19	6 08	14 1-34 4	21 8

Another evaluation of the analytical data was performed using the measured amounts of plutonium americium uranium thorium and the total amount of daughter products of the latter two. Some of the daughter products of uranium and thorium have relatively short half lives and some are gases. It is therefore impossible to maintain equilibrium in a sample collected on filter paper. These short lived and/or gaseous products are gone before the sample is processed in the laboratory. Although these daughter products could not be determined in the filters it is reasonable to assume that most if not all are present in ambient air. To determine the total activity of uranium plus daughters the uranium value was multiplied by eight and the thorium value was multiplied by six to calculate the total activity of thorium and its daughters. This accounts for all the alpha particle emitters in the two natural series.

A summary of these calculations plus plutonium and americium ratios similar to those in Table I is presented in Table III

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TABLE III Comparison of Plutonium Plus Americium to Total Calculated Ratios (pCi/Sample)

<u>Station</u>	<u>U Total*</u>	<u>Th Total†</u>	<u>Pu</u>	<u>Am</u>	<u>Total</u>	<u>Pu + Am Total X 100</u>
S 3	18 6	18 1	1 13	0 52	38 4	4 3
S 14	12 0	10 1	1 42	0 24	23 8	7 0
S 23	5 4	4 1	0 97	0 20	10 7	10 9
S 24	6 8	4 5	1 24	0 12	12 7	10 7

\* Calculated by multiplying <sup>238</sup>U activity by eight  
 † Calculated by multiplying <sup>232</sup>Th activity by six

DISCUSSION OF RESULTS

The results of analyses from the four air samplers taken for six months in 1978 and 1979 show that plutonium and americium comprise a small percentage of the total radioactivity in the air adjacent to the Rocky Flats Plant. The minimum contribution of plutonium and americium was 14 percent in samples from sampler S-3 located along the north security fence. The maximum contribution of 23 percent was from samples collected at sampler S-23 located south of the south security fence.

Although the results indicate slightly higher percentages of plutonium and americium in samples collected east and south of the Plant, it is not believed that contributions from the Plant are greater in these directions. The absolute amounts of these two radioisotopes appear to be almost the same at all four samplers with slightly higher amounts at the north and west samplers. The amounts of uranium and thorium, however, appear to be significantly higher west and north of the Plant. These conditions cause the percentages of plutonium and americium to be higher in the east and

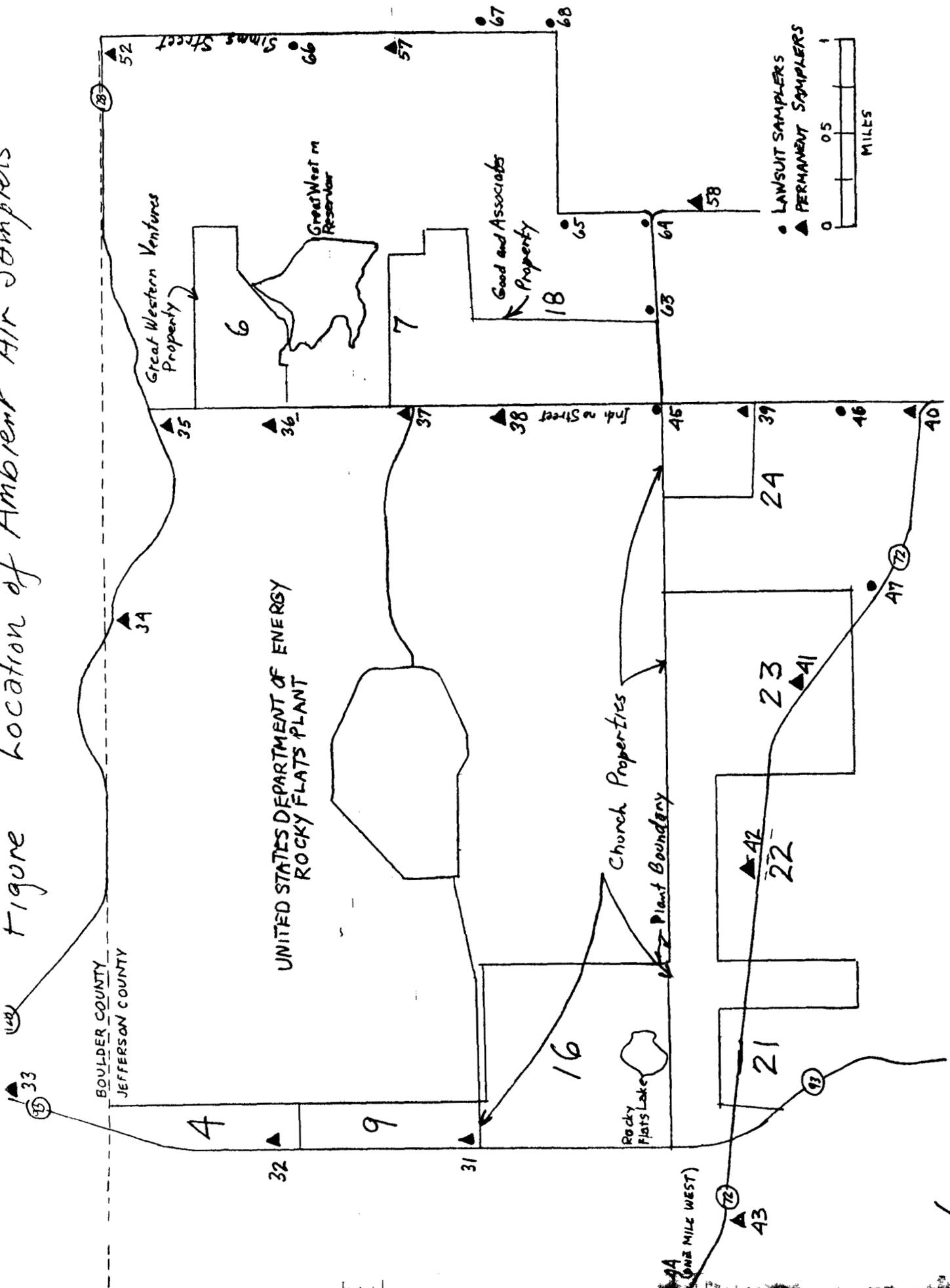
south samples. Similar but considerably lower percentages were determined by calculating the contributions made to the total radioactivity from all the daughter products of uranium and thorium. The same ranking of highest at S-23 and lowest at S-3 was found as a result of these calculations.

It is not unexpected that amounts of uranium and thorium are higher in the air samples collected north and west of the Plant. The air sampled at these locations has sources for particulates in the relatively undisturbed soils to the west. These soils are known to contain greater amounts of natural radioactivity. The mean concentration of uranium in soils in the area has been found to be equal to five parts per million which is more than twice background elsewhere and the thorium concentration averages 13 parts per million or three times normal<sup>1</sup>. The air sampled east and south of the Plant is lower in uranium and thorium because adjacent surface ground areas have been disturbed and covered with structures, roads and paved lots.

<sup>1</sup> Unpublished report C T Illsley

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Figure location of Ambient Air Samplers



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