UNCLASSIFIED
(CLASSIFICATION)

COPY

DOCUMENT NO.
HW-77345

GENERAL ELECTRIC
HANFORD ATOMIC PRODUCTS OPERATION - RICHLAND, WASHINGTON

DATE
May 23, 1962

COPY NO.
[redacted]

ISSUING FILE

TITLE

RECUPLEX INCIDENT, APRIL 7, 1962

EMISSION OF FISSION PRODUCTS FROM THE 291-Z STACK

AUTHOR

J. K. Soldat
Radiation Protection Operation
Hanford Laboratories

DISTRIBUTION

	NAME	BUILDING	AREA
1.	G. E. Backman	3746	300
2.	L. A. Carter	3701-L	300
3.	J. P. Corley	3746	300
4.	R. F. Foster	3746	300
5.	J. J. Fuquay - C. L. Simpson	622-W	200-W
6.	C. C. Gamertsfelder	326	300
7.	R. B. Hall	3746	300
8.	R. L. Junkins	309	300
9.	A. R. Keene	3746	300
10.	L. M. Knights - D. C. Bowden	234-5	200-W
11.	H. V. Larson	3705	300
12.	R. W. Meisinger - J. R. Bovingdon	329	300
13.	C. E. Newton - J. M. Selby	3705	300
14.	J. M. Nielsen - R. W. Perkins	325	300
15.	L. C. Rouse	3705	300
16.	L. C. Schwendiman	325	300
17.	J. K. Soldat	3746	300
18.	A. J. Stevens	3701-L	300
19.	F. Swanberg	747	700
20.	C. M. Unruh	3706	300
21.	E. C. Watson	3701-L	300
22.	R. H. Wilson	3746	300
23.	Record Center		
24.	300 Files		
25. - 35.	Extras		

[redacted]

THIS DOCUMENT IS PUBLICLY AVAILABLE

ROUTE TO	PAYROLL NO.	LOCATION	FILES ROUTE DATE	SIGNATURE AND DATE

RECUPLEX INCIDENT, APRIL 7, 1962

EMISSION OF FISSION PRODUCTS FROM THE 291-Z STACK

By

J. K. Soldat
Environmental Studies and Evaluation
Radiation Protection Operation

I. INTRODUCTION

During and after the Recuplex Incident at Hanford on April 7, 1962, filter samples of the 234-5 Building exhaust gases were collected at increased frequencies from the 291-Z stack. The filters were analyzed using gamma spectroscopy by personnel of Radiological Chemistry and the Radiological Chemical Analysis Laboratory. Results of the laboratory analyses have been evaluated in terms of the emission rates of various fission products from the 291-Z stack.

Trajectory of a cloud of radioactive materials discharged from the 291-Z stack at 1100 on April 7, 1962, was calculated by personnel of Atmospheric Physics. Cloud trajectory and concentration data were used to estimate possible environmental consequences.

II. SUMMARY

Based on laboratory analysis of stack filter samples collected at the 291-Z stack and the trajectory followed by the emitted radionuclides, the following conclusions were obtained:

(1) About 1,000 curies of noble gases and daughter products were released from the 291-Z stack at about 1100 AM on April 7, 1962, followed by an additional total of 200 curies of similar composition over the next two days.

(2) A minimum of about 10^{17} fissions in the K-9 vessel at approximately 1100 on April 7, 1962, followed by another approximately 10^{16} fissions over the next two days, would have been required to produce an emission of this magnitude.

(3) Radioactive decay rapidly reduced the quantities of radionuclides present in the environs so that of the first 1,000 curies emitted, 400 remained at the calculated time of maximum ground level exposure (five minutes later), and about 50 remained at the time calculated for the off-plant exposure (105 minutes from the stack).

(4) The maximum ground level exposure of released materials was calculated to have occurred near the 2704-W Building and could have resulted in a total external radiation exposure of 0.3 mr to anyone standing at that point during cloud passage.

(5) Maximum ground level exposure off-plant was calculated to have occurred five miles north of 300 Area. The resulting total radiation exposure to a person at that point would have been less than one micro-roentgen.

(6) Any possible environmental effects of the cloud were completely undetectable at off-project locations.

III. ASSUMPTIONS AND CALCULATIONS

Several broad assumptions were required during the calculation of the emission rates from the 291-Z stack and of the number of fissions which occurred. Different, but still reasonable, assumptions are possible. Different assumptions would probably not have affected the calculated emission rates by more than one order of magnitude and certainly would not have led to a different conclusion concerning the lack of detectable

environmental effects. The assumptions and bases of calculation actually used are all listed here for easy reference.

(1) Fission yields were taken from Nucleonics, Volume 18, Number 11, November, 1960.

(2) All noble gases (or their parents) escaped from the K-9 solution immediately.

(3) Travel time from K-9 to the absolute filter in the vacuum system is about one minute.

(4) One-hundred per cent of the noble gases passed through the absolute filter and the stack sample filter and all other types of nuclides were collected with one-hundred per cent efficiency on both of these filters.

(5) Travel time from the absolute filter to the sample filter is 0.8 minute as estimated by L. C. Schwendiman of Chemical Effluents Technology.

(6) The initial quantity of fission products released at about 1100 AM was of such magnitude that further activity released during the remainder of the first filter sampling period (1100 - 1418) was negligible in comparison.

(7) The fission product release rate was relatively uniform during each of the subsequent sampling periods.

IV. RESULTS AND CONCLUSIONS

A. Description of Ventilation System and Stack Sampler

Process effluent gases and building ventilation air from the 234-5 Building are exhausted to the atmosphere through a 200 foot high stack (291-Z) at a flow rate of 2.4×10^5 CFM. All the air

is filtered at least once through absolute filters before being discharged. A sample of the effluent gas is drawn from a sample line near the stack breaching and passed through HV-70 filter paper 4" x 4" square, at 5.0 CFM. Sample filters are normally run for a 24 hour period (except on week ends) before being collected and counted in the 234-5 counting room for alpha radioactivity.

B. Emission of Cs^{138}

Table I summarizes the results of the gamma spectroscopy measurements performed in the 329 Building counting room on filter samples collected April 7, 8, and 9, 1962, from the 291-Z stack. The data in Table I, coupled with assumptions 4, 5, 6, and 7 listed above, were used to calculate the emission rate of Cs^{138}. These emission rates are shown in Figure 1 and Table II. The first filter sample was actually put on at 0850 on April 6, 1962, but no fission products were being emitted and collected before about 1100 on April 7, 1962.

C. Emission of Xe^{138}

The estimated 0.8 minute travel time of the effluent gases between the absolute filter in the vacuum exhaust system and the stack sample filter, the data of Figure 1, and equations for radioactive parent-daughter relations were combined to yield an estimate of the emission rates of the Xe^{138} parent of Cs^{138}. These rates are also shown in Table II.

D. Characterization of the Reaction From the Stack Sample Analyses

The laboratory analyses of the filter samples collected at 291-Z stack on April 7, 8, and 9, 1962, were evaluated in terms of the minimum number of fissions required to create the quantities

of radionuclides measured. Three radionuclides were detected on these filters by the gamma spectrometric analysis used: Cs^{138}, Ba^{139}, and Ba^{140} (Table I). Essentially only the first daughter products (rubidium and cesium) of the noble gases (krypton and xenon) should be collected on the filter samples. At the time of filter analysis, however, four of the second daughters, Sr^{89}, Sr^{91}, Ba^{139}, and Ba^{140}, could have been built up to measurable quantities.

Sr^{89} has no gamma emission and would not be detected in the gamma energy analysis performed. Sr^{91} is the second daughter of the short half-life (10 seconds) Kr^{91}. Therefore, errors in the various assumptions related to the timing of events (Section III) would significantly affect the quantity of Sr^{91} estimated to be present in the gases released from the stack and on the sample filters.

The high ratios of both the Ba^{139} and Ba^{140} to the Cs^{138} found on the first filter sample (Table I) indicated that most of the cesium isotopes were collected near 1100 in order to provide sufficient time for the barium to build-up to the levels measured. In subsequent filter samples the ratios of Ba^{139} and Ba^{140} to Cs^{138} were more indicative of collection of cesium isotopes throughout the entire sampling period.

The Cs^{138} was the most reliable of the measured isotopes to use for estimating the reaction rates in the K-9 vessel for several reasons.

(1) Twenty-four of the filter samples contained detectable amounts of Cs^{138} compared to nine samples each for Ba^{139} and Ba^{140}.

(2) Barium isotopes are present on the filter samples principally as decay products of the cesium originally collected. As a result, the calculations of the number of fissions required to produce the measured amounts of barium involve an additional degree of uncertainty over those for the cesium.

(3) The half-lives of Cs^{138} (32 minutes) and its parent Xe^{138} (17 minutes) are long enough to minimize the errors introduced by the various assumptions on time sequences and emission rates listed in Section III.

The measured amounts of Cs^{138}, the plutonium fission yields for Xe^{138}, the equations for radioactive parent-daughter relationships, and the estimated 0.8 minute travel time from the absolute filter in the vacuum system to the sample filters were combined to yield estimates of the minimum number of fissions occurring in the K-9 vessel.

(1) A minimum of 1.2×10^{17} fissions at about 1100, April 7, was required to produce the quantity of Cs^{138} measured on the first filter sample.

(2) A minimum of 2×10^{16} fissions was required to create the total Cs^{138} measured over the next 19 hours, up to 0600 on April 8.

(3) A minimum of 3×10^{15} fissions was required to create the total Cs^{138} measured over the period from 0600, April 8 to 0700, April 9, 1962.

These values yield a lower total number of fissions than the value of 8×10^{17} obtained by more accurate means, such as analysis

of a sample of the K-9 vessel contents. However, for the purpose of estimating the amounts of radionuclides actually released from the 291-Z stack, the number of fissions postulated from the filter samples is believed to be more appropriate. Therefore, a reaction of 10^{17} fissions (rounded from the 1.2×10^{17} obtained from analysis of the first filter sample) was used to estimate the composition of the radioactive materials released from the stack at about 1100 on April 7. The results of this calculation are summarized in Table III.

The first two columns of this table show the composition of the approximately 2,200 curies of noble gases passing through the absolute filter in the vacuum system. The third column lists the composition of the approximately 1,300 curies of noble gases and their daughter products in the effluent gases 0.8 minutes downstream of the absolute filter, at the point when the stack gas sample is withdrawn from the stack breaching. This latter composition also approximates that which is discharged to the atmosphere from the 291-Z stack.

E. Cloud Trajectory and Environmental Effects

The trajectory of the cloud of radioactive materials emitted from the 291-Z stack at 1100 AM on April 7, 1962, was calculated by personnel of Atmospheric Physics. The probable cloud path and ground level exposures in units of μc/cc per curie/second of material released are shown in Figure 2 uncorrected for decay. The maximum cloud concentration was calculated to have occurred at a point 1500 meters (five minutes) downwind of the 291-Z stack near the 2704-W Administration Building. The centerline of the cloud then passed near the 2701-W Gate House about 0.3 mile farther east

(downwind). From this point the cloud traveled essentially southeast. The maximum off-plant exposure probably occurred where the cloud crossed the project boundary (105 minutes post-release) at a point about five miles north of 300 Area. The composition of fission products in the cloud corrected for decay to the points of on-plant and off-plant maxima are shown in columns 4 and 5, respectively of Table III.

The comparison of the exposure parameters of Figure 2 with the cloud compositions shown in Table III, yield the estimated maximum external radiation exposure which could have been received by someone on the ground during passage of the cloud. These values were 0.3 mr at 2704-W and 4×10^{-4} mr at the project boundary. These low values of external exposure precluded the possibility of any significant exposure to persons on or off the plant from the emitted materials by any other pathway conceivable, i.e., inhalation, or deposition on foodstuffs, etc.

Routine surveillance of the atmosphere in the 200 West Area includes a daily sample of air taken outside of the 2701-W Gate House, and weekly samples taken inside the Gate House and in the Administration Area. For two days after the incident all of the 200-W Area air filter samples were changed about twice per shift, but no evidence of concentrations above normal was found at any location in the 200 West Area. No deposition from the cloud was evident from analysis of vegetation samples collected every 0.1 to 0.2 mile around the perimeter of the 200 West Area and collected every mile along the road between Byers Landing and Ringold, in the afternoon of April 7, 1962.

Portable radiation detection instruments were used to survey main project roadways in a circumferential pattern around the 200 West Area on April 7. The road between the Prosser Barricade and Hanford, and the road between Byers Landing and Ringold were also surveyed since they crossed the trajectory of the cloud estimated by Atmospheric Physics personnel. Again no evidence of deposition from the cloud was found.

All sample results were in the range of values normally expected from the historical levels of fallout and normal stack emission. Although concentrations of I^{131} in the atmosphere in the vicinity of both 200 East and 200 West Areas were above normal at and following the incident, the abnormal values were related to above average emissions of I^{131} from the Purex facility in 200 East Area during the first two weeks of April.

F. Relationship of Instrument Readings and Laboratory Analysis of Samples

At the time each filter sample was removed from the sampling equipment at the 291-Z stack breaching, it was surveyed with portable radiation instruments. All readings made with an alpha scintillation meter were less than 500 d/m per filter. Positive CP meter readings were recorded for eight of the first ten samples and positive GM meter readings were recorded for all samples removed through 1900, on April 9, 1962. The recorded instrument readings were compared to the results subsequently obtained by radiochemical analyses of the filters in the laboratory. The relationship between the GM meter reading and the amount of Cs^{138} on the filter samples is illustrated in Figure 3. Included in this figure is an estimated

370,000 c/m for the first filter removed. This estimate is based on a CP meter reading of 50 mr/hr on this filter and the average ratio for seven pairs of GM meter and CP meter readings obtained on subsequent filter samples (7,350 c/m per mr/hr).

Figure 3 shows a reasonably consistent ratio of about one c/m on the GM meter for each 10 d/m of Cs^{138} on the filter. This approximation holds over four orders of magnitude and for two days after the incident. There are two points which seem to deviate from the pattern in Figure 3. Had the recorded value for the GM reading been ten-fold greater, these two points would have been consistent with the remainder of the data.

Evaluation of Figure 3 leads to the following conclusions, all of which are compatible with the diagnosis of the incident available from other means.

(1) A constant "geometry" (between the GM meter reading and the Cs^{138} present) of ten per cent was found over the two day period of sampling, allowing prediction of the Cs^{138} content of the sample filters in the absence of laboratory analysis.

(2) The relative composition of the nuclides collected on the filter must have been fairly constant because of the constant "geometry" for Cs^{138}.

(3) A fresh source of the collected nuclides must have been continuously generated to maintain the constant composition of relatively short-lived materials collected.

V. ACKNOWLEDGEMENTS

The laboratory analysis data was supplied by R. W. Perkins of Radiological Chemistry. The cloud trajectory was defined by J. J. Fuquay, and C. L. Simpson of Atmospheric Physics. L. C. Schwendiman of Chemical Effluents Technology supplied an estimate of travel time between the absolute filter and the sampling point. Valuable suggestions on calculation methods and format were received from R. F. Foster and G. E. Backman of Environmental Studies and Evaluation.

TABLE I

RESULTS OF GAMMA SPECTROSCOPY MEASUREMENTS

ON FILTER SAMPLES COLLECTED AT 291-Z STACK

APRIL 7, 8, and 9, 1962

			10^3 d/m per filter at time off		
Date on	Time on	Time off	Cs^{138}	Ba^{139}	Ba^{140}
4-7-62	~ 1100*	1418	1,424	1,780	52.3
	1418	1645	470	23.2	0.22
	1645	1805	712	-	0.14
	1805	1847	401	-	0.08
	2157	2302	726	-	0.07
4-7-62	2302	0003	564	-	0.03
4-8-62	0003	0055	445	-	0.03
	0145	0256	-	10.4	0.004
	0256	0357	-	6.7	0.01
	0357	0453	370	7.0	
	0453	0549	-	5.2	
	0549	0701	187	6.8	
	0951	1104	83.4	-	
	1104	1255	73.8	2.2	
	1255	1351	43.7	0.91	
	1351	1455	60.0		
	1455	1610	79.0		
	1610	1658	41.1		
	1658	1753	33.5		
	1753	1857	30.7		
	1857	2020	10.3		
	2020	2110	15.5		
	2110	2205	9.9		
	2205	2252	4.0		
4-8-62	2252	0032	4.5		
4-9-62	0032	0107	1.8		
	0107	0212	2.0		
	0212	0254	< 6.0		
	0254	0400	< 3.0		
	0400	0510	< 2.0		
	0510	0605	< 2.0		
	0605	0702**	< 2.0**		

* This filter was actually put on at 0850, April 6, 1962, but no fission products should have been emitted or collected before the initial nuclear reaction at about 1100 on April 7, 1962.

** All Cs^{138} analyses on subsequent filters were less than 10^3 d/m per filter.

TABLE II

EMISSION RATE OF Xe^{138} AND Cs^{138} FROM 291-Z STACK

APRIL 7, 8, and 9, 1962

Based on Analyses of Stack Filter Samples

Time Period	10^{-3} μc/ft.^{3}		10^{-3} Curies/Min.	
	Xe^{138}	Cs^{138}	Xe^{138}	Cs^{138}
April 7, 1962				
~ 1100*	**	**	**	**
1100 - 1418*	~ 50	~ 0.8	~ 10	~ 0.2
1418 - 2400	75	1.3	18	0.31
April 8, 1962				
0000 - 0701	52	0.88	12	0.21
0701 - 1351	12	0.20	2.8	0.05
1351 - 1857	7.8	0.13	1.9	0.03
1857 - 2400	1.3	0.02	0.31	0.005
April 9, 1962				
0000 - 0212	0.33	~ 0.006	0.08	~ 0.001
0212 - 0702	~ 0.2	~ 0.003	~ 0.05	~ 0.001
0702 - 2400	< 0.04	< 0.001	< 0.01	< 0.001
Total released over two day period			150 curies	2.5 curies

* This one filter sample actually ran from 0850, April 6, 1962, to 1418, April 7, 1962, but no fission products should have been emitted or collected before the initial nuclear reaction at about 1100, April 7, 1962.

** An estimated total of 130 curies of Xe^{138} and 2.2 curies of Cs^{138} was released at about 1100 on April 7, 1962.

TABLE III

COMPOSITION OF RADIOACTIVE MATERIALS

RELEASED FROM K-9 AT APPROXIMATELY 1100 AM

April 7, 1962

Total Curies Available Per 10^{17} Fissions

Isotope	Just Downstream of Absolute Filter	At Time of Release	Five Minutes Post Release*	105 Minutes Post Release**
Kr^{83m}	56	56	54	29
Kr^{87}	3	3	2.8	1.2
Kr^{88}	2.5	2.5	2.4	1.6
Rb^{88}	-	0.08	0.51	1.8
Kr^{89}	117	98	32	-
Rb^{89}	-	3.8	16	0.27
Sr^{89}	-	9×10^{-5}	5×10^{-3}	5×10^{-2}
Kr^{90}	320	116	0.17	-
Rb^{90}	-	37	18	-
Kr^{91}	58	2.1	-	-
Rb^{91}	-	55	0.29	-
Sr^{91}	-	4.5×10^{-3}	1.6×10^{-2}	1.5×10^{-2}
Xe^{137}	410	355	141	-
Xe^{138}	107	104	84	1.4
Cs^{138}	-	1.8	12	10
Xe^{139}	825	366	1.8	-
Cs^{139}	-	32	41	0.02
Ba^{139}	-	0.12	2.0	3.2
Xe^{140}	270	47	-	-
Cs^{140}	-	41	2.0	-
Ba^{140}	-	9×10^{-4}	4×10^{-3}	4×10^{-3}
Total	2,200	1,300	410	49

* Calculated time when the maximum concentration at ground level (near the 2704-W Building) was reached.

** Calculated time when the material passed outside the Hanford Reservation about five miles north of 300 Area.

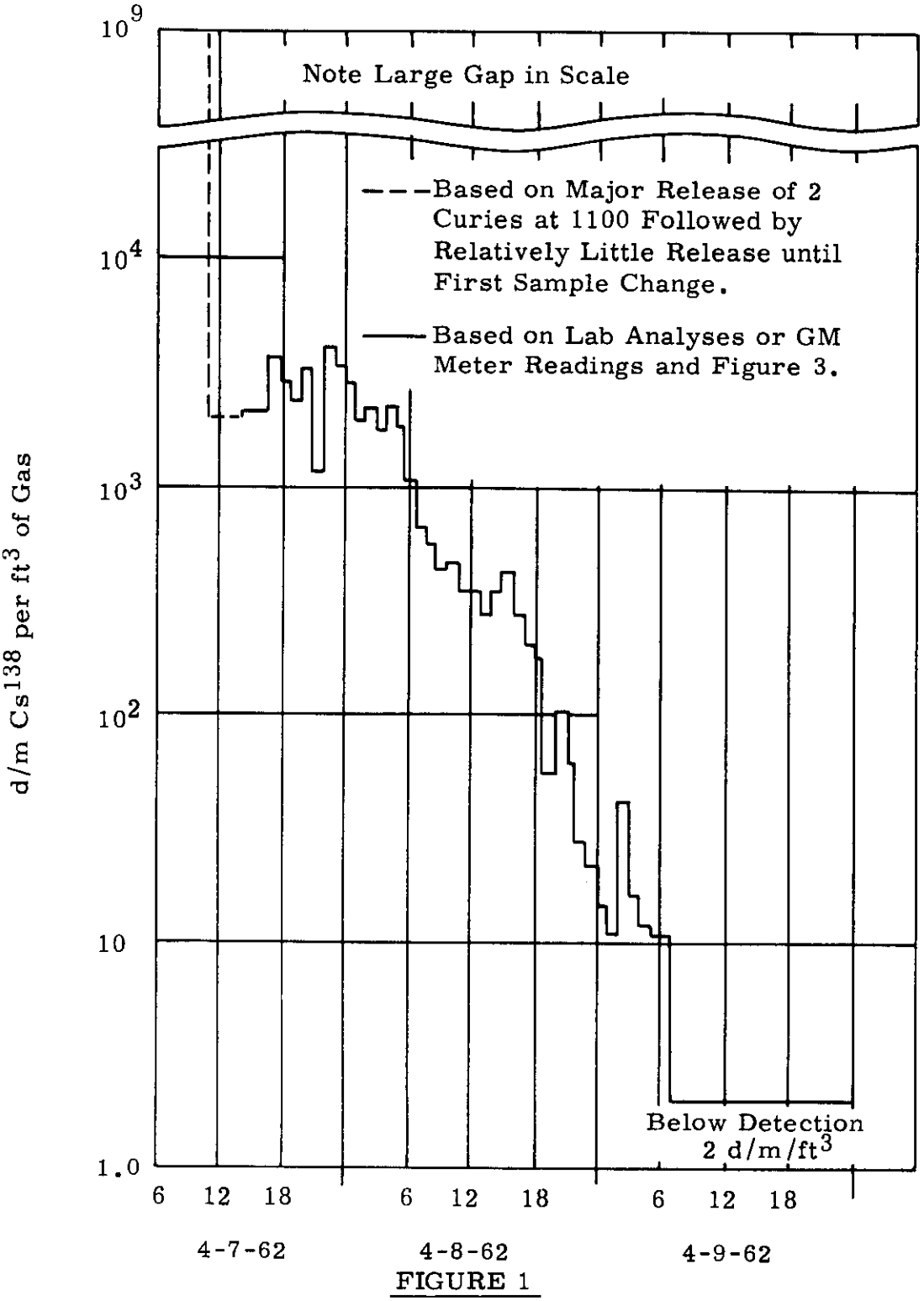

FIGURE 1

Emission Rate of Cs^{138} From 291-Z Stack April 7, 8, and 9 1962

AEC-GE RICHLAND, WASH.

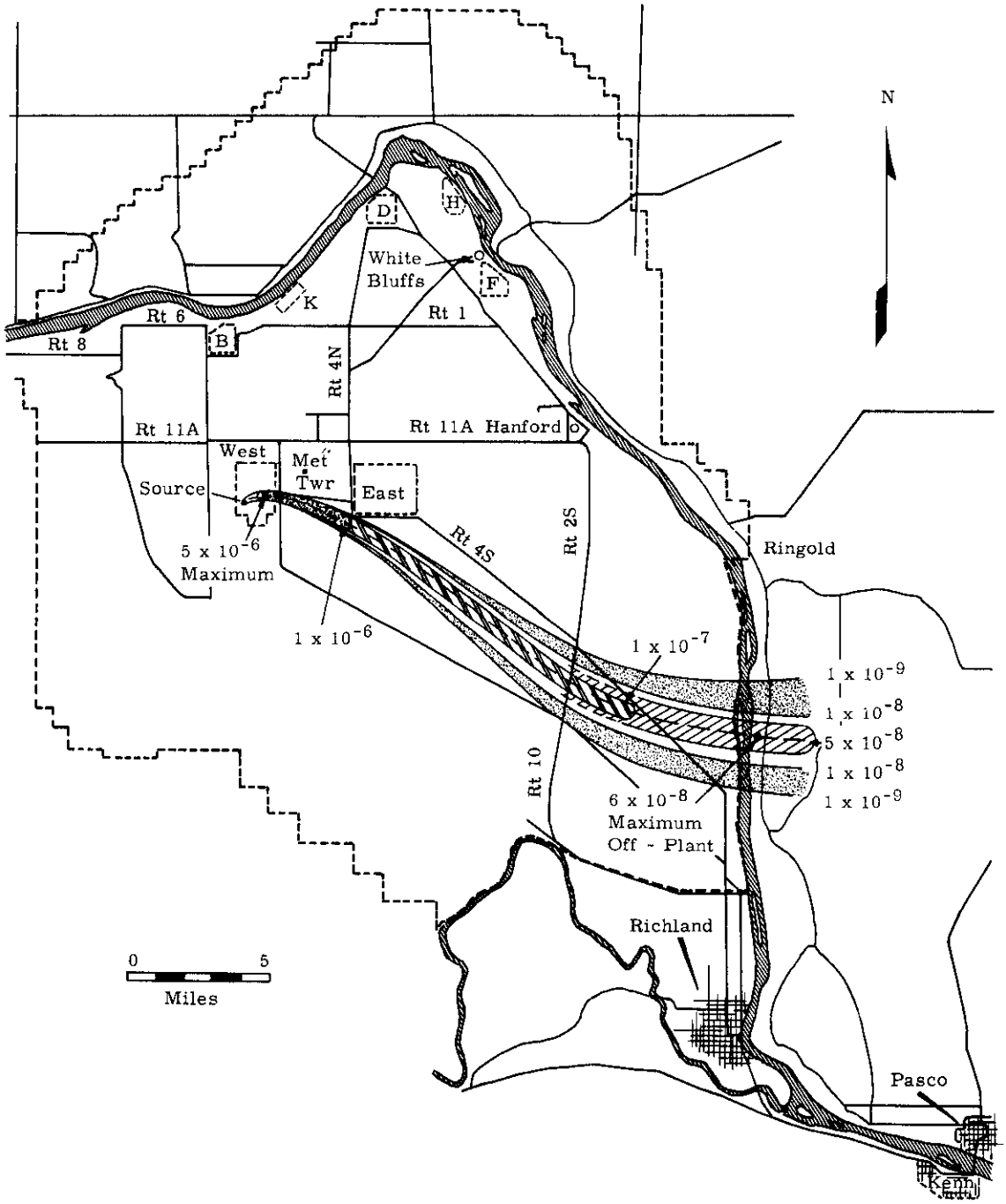

FIGURE 2

Trajectory of Materials Released from 291-Z Stack at 1100-1200 PST, April 7, 1962. Isopleths are in Units μc/cc per curie/sec (No Decay)

AEC-GE RICHLAND, WASH.

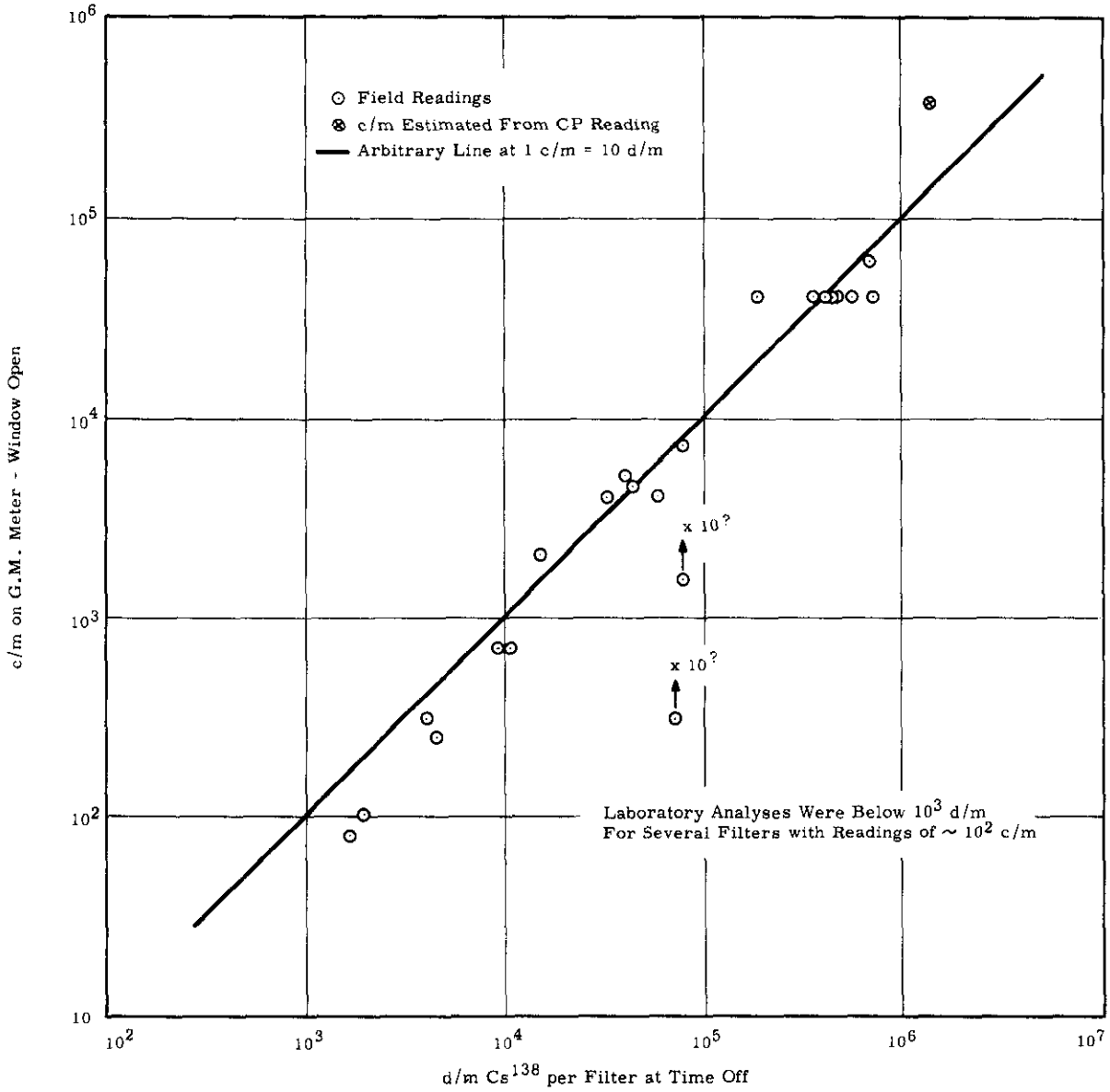

FIGURE 3

Filter Content vs Instrument Reading 291-Z Stack Samples,
April 7, 8, and 9, 1962

AEC-GE RICHLAND, WASH.