38573/
DPST-86-483

TECHNICAL DIVISION
SAVANNAH RIVER LABORATORY

CC: R. P. Whitfield, DOE
S. R. Wright, 703-A
W. E. Wisenbaker, 703-A
J. M. Gaver, 703-A
J. A. Porter, SRP, 703-A
W. C. Reinig, 703-A
J. S. Roberts, 703-A
A. J. Garrett, 735-A
J. T. Lowe, SRL, 773-A
R. J. Guschl, 773-A
J. D. Spencer, 773-A
T. V. Crawford, 773-A
J. F. Proctor, 773-A
A. L. Boni, 773-A
W. E. Jule, 773-A
C. E. Ahlfeld, 773-A
R. M. Satterfield, 773-A
W. L. Marter, 773-42A
R. W. Taylor, 735-A
M. V. Kantelo, 735-A
W. G. Winn, 735-A
R. L. Kump, 735-A
C. C. Zeigler, 735-A
E. A. Anderson, 735-A
L. H. Spradley, 735-A
W. D. Dalton, 735-A
SRL Publications (4)
E/T Chron File

June 9, 1986

TO: R. W. BENJAMIN

FROM: R. A. SIGG RAS

TRAC LABORATORY MONITORING OF CHERNOBYL RADIOACTIVE DEBRIS

SUMMARY

A severe accident occurred at the Chernobyl nuclear power plant number 4 in the Soviet Union on April 25, 1986. An explosion released large amounts of radioactive debris, primarily fission products, to the atmosphere. As winds carried debris from the Soviet Union, scientists in Europe and the United States reported detecting fission product activities in air samples. Monitoring by the Tracking Radioactive Atmospheric Contaminants (TRAC) mobile laboratory showed concentrations in the Southeastern United States were well below those considered hazardous.

INTRODUCTION

The Savannah River Laboratory (SRL) developed TRAC (Ref. 1) to support emergency response and environmental research programs at the Savannah River site. In the event of a radiological emergency at the Savannah River Plant (SRP) the TRAC laboratory would collect and analyze for radionuclides in air in nearby population centers. Observed data would be provided to the Emergency Operations Center (EOC) in near real-time to enable timely emergency-management decisions.

Several of TRAC's analytical systems collected Chernobyl radioactive debris from air in population centers in the vicinity of the SRP. Gamma-ray emitting aerosol and volatile radioiodine samples were collected in Augusta, Barnwell, Columbia, Greenville, Madison, Savannah, and along roads leading to these cities. Samples were collected both before Chernobyl debris arrived in this region and during its passage through the region. Our objectives in applying TRAC to monitoring Chernobyl radioactivity included

- Measuring atmospheric radionuclide concentrations in population centers to permit dose estimates

- Testing and evaluating the mobile laboratory's radionuclide analysis systems. Only natural radionuclides and extremely low levels of man made radionuclides have been detected in previous tests. The debris allowed a more realistic test of capabilities than is normally possible.

Offsite population centers were monitored to simulate TRAC activites in the unlikely event of a radiological emergency at SRP. We also wanted to avoid duplicating monitoring efforts of the SRL Environmental Technology Division (ETD) and the SRP Health Protection Department (HP) at the Aiken airport and the SRP site, respectively.

TRAC monitors detected very low concentrations of Chernobyl fission products beginning on May 9, 1986. Our monitoring continued through May 14. Concentrations and doses through this period were far below levels considered hazardous. The observed radionuclides would give an effective dose equivalent of < 0.005 mrem and < 0.07 mrem to the thyroid to an adult individual exposed to the debris through the six day period. Monitoring by ETD and HP shows the debris persisted beyond May 14; therefore, the dose estimated from a six day exposure is a somewhat lower value than will be reported eventually.

The TRAC collection and spectrometry systems performed reliably through this period of extended operations. Some improvements to software are recommended.

MONITORING RESULTS

Collections

TRAC began monitoring May 6 and continued through May 14, 1986. Table 1 lists vehicle locations, collection start and stop times, and the volume of air collected for each sample. As noted, some samples were collected while the vehicle was in transit while others were collected while stationary. Locations monitored were upwind of SRP except May 13 and May 14, so any observed fission product activity would not be attributed to SRP by offplant personnel. Numerous small thunderstorms occurred in the region through this period. Wind directions and the occurrence of rain are also indicated in the table.

Sample collection and counting durations were longer in this effort than would be used following a release from SRP. The longer times were used to improve sensitivity for debris diluted in transport much of the distance around the globe. Ten minute collections followed by ten minute counts would be typical of TRAC operations following an inadvertent release of gamma-ray emitting aerosols or volatile radioiodine from SRP.

Concentrations

Radionuclide concentration data are presented in Table 2 for each sample. Data are included for gamma-ray emitting aerosol activities and for volatile radioiodine. Only natural activities were observed from May 6 to May 8. Beginning May 9 trace levels of Ru-103, I-131. Cs-134 and Cs-137 were observed. Concentrations rose rapidly reaching maximum values on May 10 and then declining somewhat erratically over the next several days. Additional radionuclides were detected in samples having higher activities. TRAC sampling efforts were stopped as concentrations fell, with the intent of resuming if either ETD or HP observed a significant rise in concentrations.

Rainstorms are responsible for some of the concentration changes in successive aerosol samples. Since the local storms did not cover broad areas uniformly, the washout of aerosol activities was non-homogeneous; it varied from city to city, day by day.

Radioiodine

Volatile radioiodine concentrations did not change as dramatically as the aerosols, since rain does not scavenge the volatile chemical species significantly. The fraction of I-131 in aerosol form changed daily. An average of TRAC's results gives 56% of the radioiodine in the aerosol samples and 44% in the volatile form. Analyses of filter and charcoal samples collected at Three Mile Island (ref. 2) found 27% of the I-131 on filter samples and 71% as volatile radioiodine. Differences in containment, reactor materials and transport phenomena probably account for differences in iodine distribution for the two reactor accidents.

Dose

Table 3 gives the average concentration for each of the observed radionuclides for May 9 to May 14. The average Cs-137 concentration of 0.092 pCi/cubic meter is about 800 times greater than the value observed in day-long collections at the Aiken airport before the Chernobyl accident. The pre-accident Cs-137 value is typical of worldwide fallout from old weapons tests. Other fission products observed in the TRAC Chernobyl monitoring were not observed in samples collected before the accident due to their short half-lives. Average TRAC concentration values were compared to concentrations and durations of exposure in ICRP-30 (ref. 3) to calculate the dose to an adult individual exposed for six days. The total inhalation dose from all radionuclides is less than 0.005 mrem effective dose equivalent and less than 0.07 mrem to the thyroid. Since airborne concentrations have persisted beyond May 14, this is a preliminary value based on data collected while TRAC supported Chernobyl monitoring. This preliminary dose value is several orders of magnitude lower than the average annual background of 92 mrem in this region.

MONITOR PERFORMANCE

System Review

The TRAC environmental monitoring systems used in this effort were described previously (eg: ref. 1). As a brief review, the gamma-ray aerosol monitor draws air through a filter paper at 20 cubic meters/minute. The filter is compressed to improve geometry and is counted on a high resolution germanium gamma-ray spectrometer. Volatile iodine is adsorbed as prefiltered air is drawn at 1 cubic meter/min through a charcoal (5% TEDA loaded) canister. The canister is also counted by germanium gamma-ray spectrometry. Both collection systems are monitored in real time by NaI(Tl) gamma-ray detectives for higher levels of activity if present. Spectra are analyzed by an on-board computer. Analysis

software finds and integrates peaks in spectra and compares peak energies to those in a library to make radionuclide activity assignments.

Gamma-Ray Spectra

Figures Ia and Ib present gamma-ray spectra from a volatile radioiodine and a gamma-ray aerosol sample respectively. Analysis of radioiodine spectra is relatively simple, but analysis of more complex gamma-ray aerosol spectra poses a greater challenge. Some of the aerosol spectra contain over one hundred identified peaks. Many of the peaks are due to natural activities (primarily radon daughters) though more are from the fission-products. The analysis code performed reasonably well, after a few modifications, in analysis of spectra collected while TRAC was stationary. It also performed well on radioiodine spectra collected when the vehicle was in transit.

Spectral Tailing

Figures IIa and IIb are expanded views of the 400 to 800 keV region (Figures IIa & IIb) in aerosol spectra collected when TRAC was stationary and when the vehicle was in transit. A worsening of peak shapes is clear. This effect is due to microphonics in the first stage of the germanium detector's preamplifier. Measures previously taken to minimize this effect include selecting a preamplifier designed to reduce microphonics, air bag suspension for the vehicle, housing the detector in a cabinet with shock absorbing mounts and use of an isolation pad. Without these measures tailing would likely be far more severe. Our analysis code missed peaks and misintegrated others in such complex spectral regions. Since these spectra are probably about as good as reasonably achievable for data collected in transit, an analysis code able to locate and accurately integrate such degraded Gaussian peaks is needed. Since peak shapes depend on the road surface, etc., the code should dynamically determine shape parameters based on shapes of several intense peaks through the spectrum. The RAGS code--used in the activation analysis facility--was being modified for use aboard TRAC at the time of the Chernobyl accident. It and other codes will be tested to benchmark performance against archived Chernobyl raw spectral data.

Timely Response

Peak integrations in more complex regions of the spectra were checked manually. These data were consistent with data obtained

when samples were recounted later in time when the vehicle was stationary. Spectra analyzed while in transit in this study had a higher peak density than any previously observed by TRAC. Although more sophisticated analysis codes were in development, they were not in usable form for this test. The consequence was a degraded time response for data analysis.

REFERENCES

1. Sigg, R. A. "A Mobile Laboratory for Near Real-Time Measurements of Very Low Level Radioactivity", CONF-84117-2, Procceedings of the Fifth DOE Environmental Protection Information Meeting, Albuquerque, NM, 1984.

2. Pelletier, C. A., et al, "Iodine-131 Behavior During the TMI-2 Accident", NSAL-30, Nuclear Safety Analysis Center, 1981.

3. ICRP-30 Publications, Part 1. "Limits for Intake of Radioiodine by Workers", International Commision on Radiation Protection, 1979 and ICRP-30 Publications, Part 2. "Limits On Intakes By Workers", International Commission on Radiation Protection, 1980.

FIGURE 1A

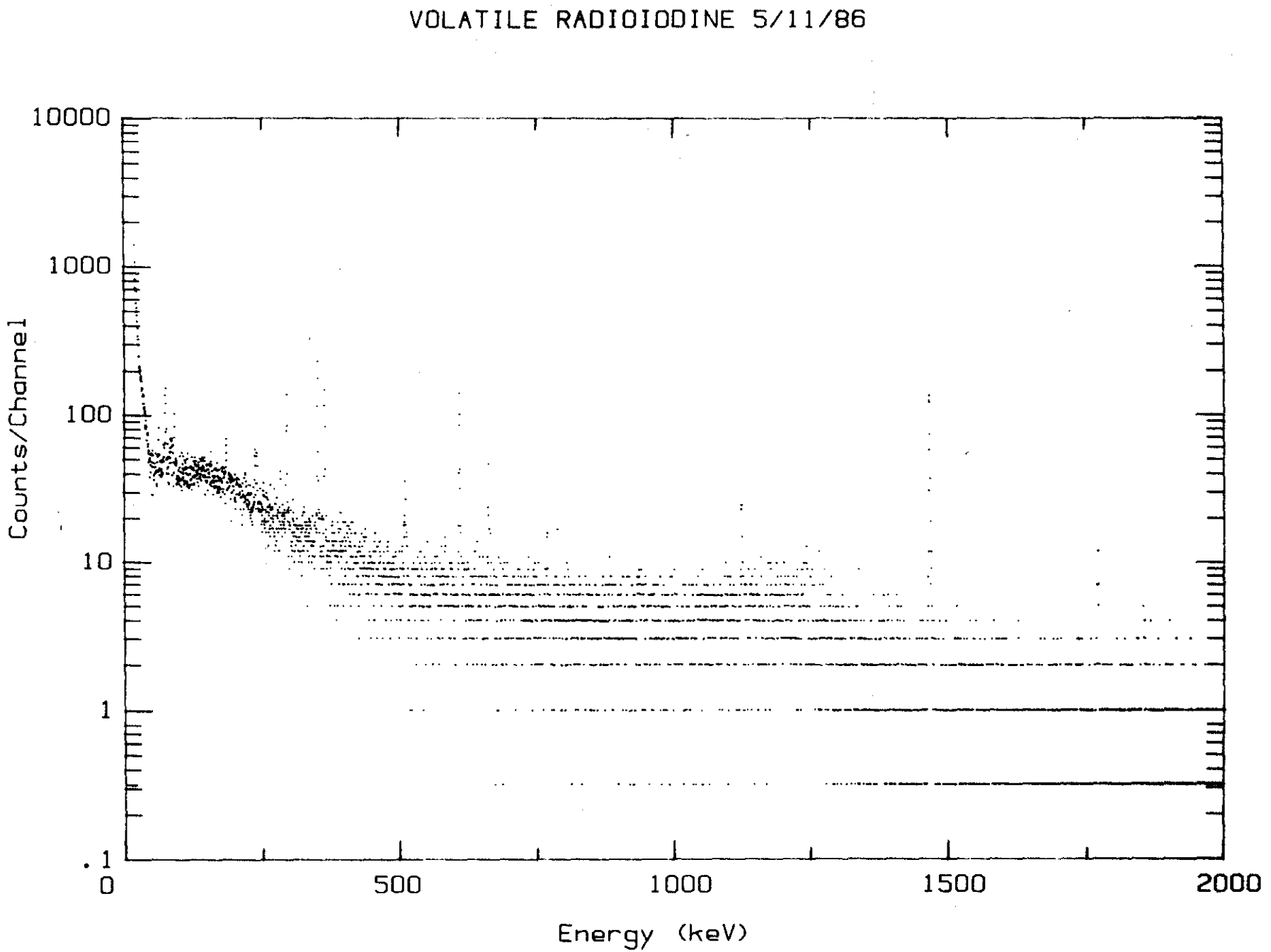
VOLATILE RADIOIODINE 5/11/86
10000
1000
100
10
1
.1
Counts/Channel
0
500
1000
1500
2000
Energy (keV)

FIGURE 1B

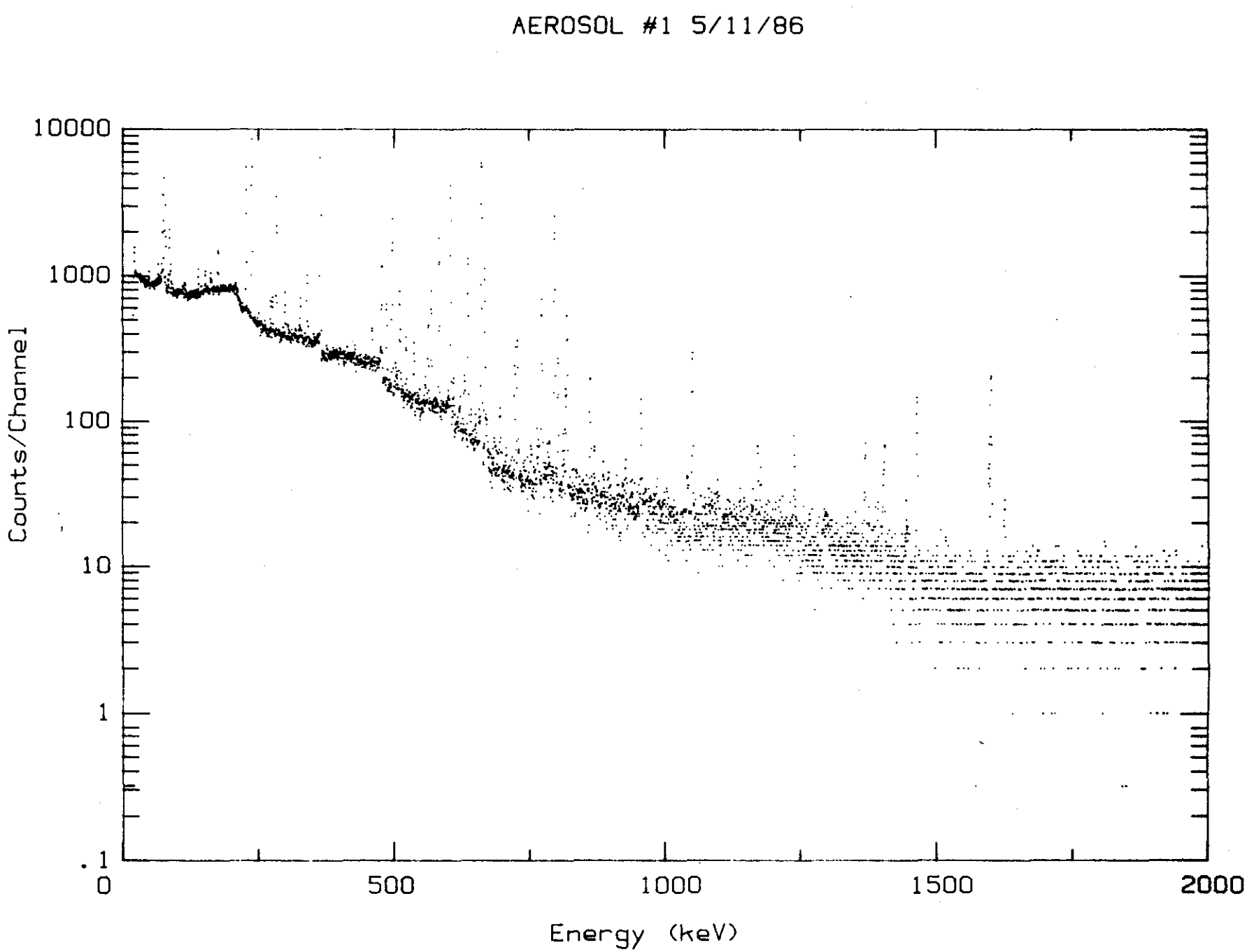
AEROSOL #1 5/11/86
10000
1000
100
10
1
.1
Counts/Channel
0
500
1000
1500
2000
Energy (keV)

FIGURE 2A

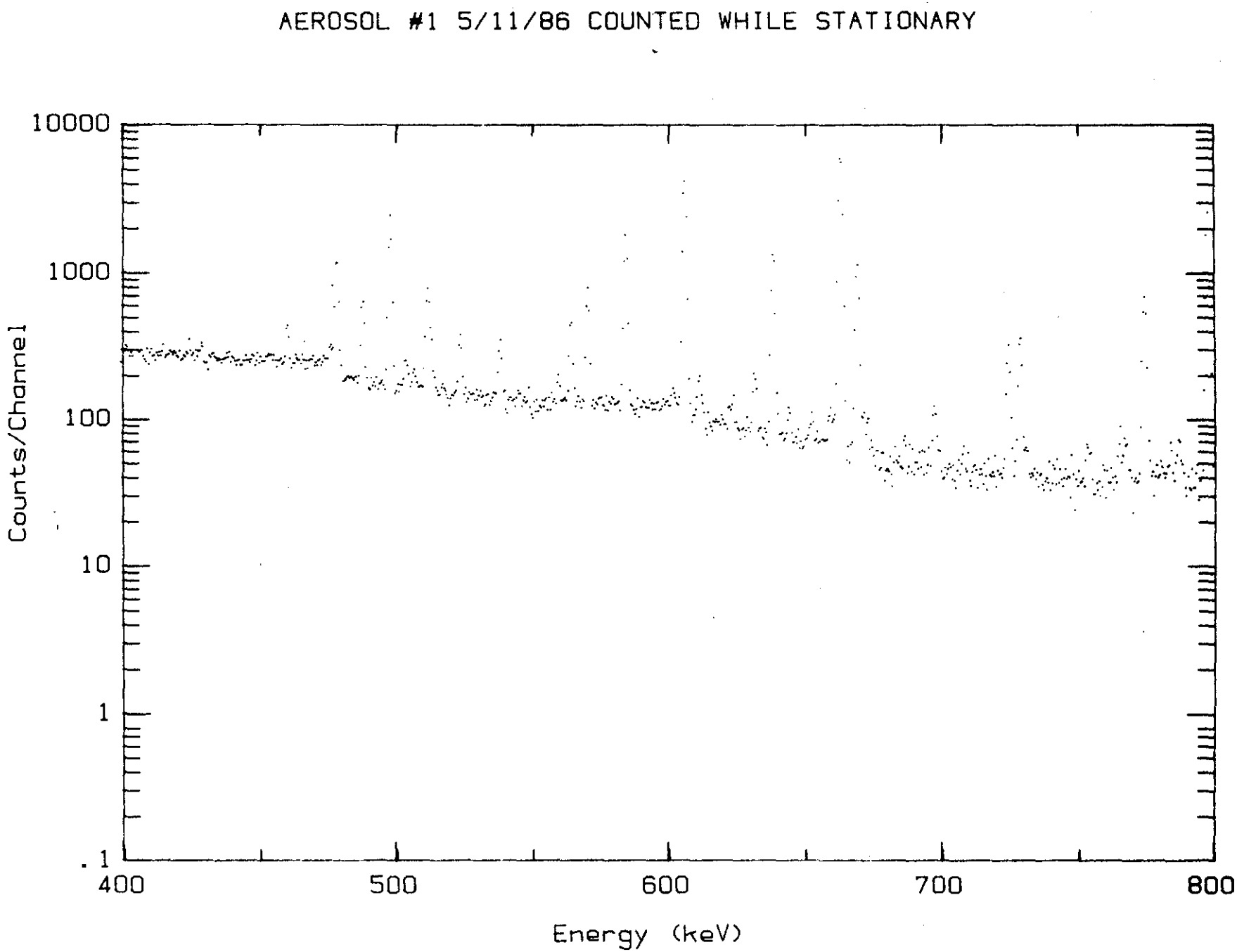
AEROSOL #1 5/11/86 COUNTED WHILE STATIONARY
10000
1000
100
10
1
.1
Counts/Channel
400
500
600
700
800
Energy (keV)

FIGURE 2B

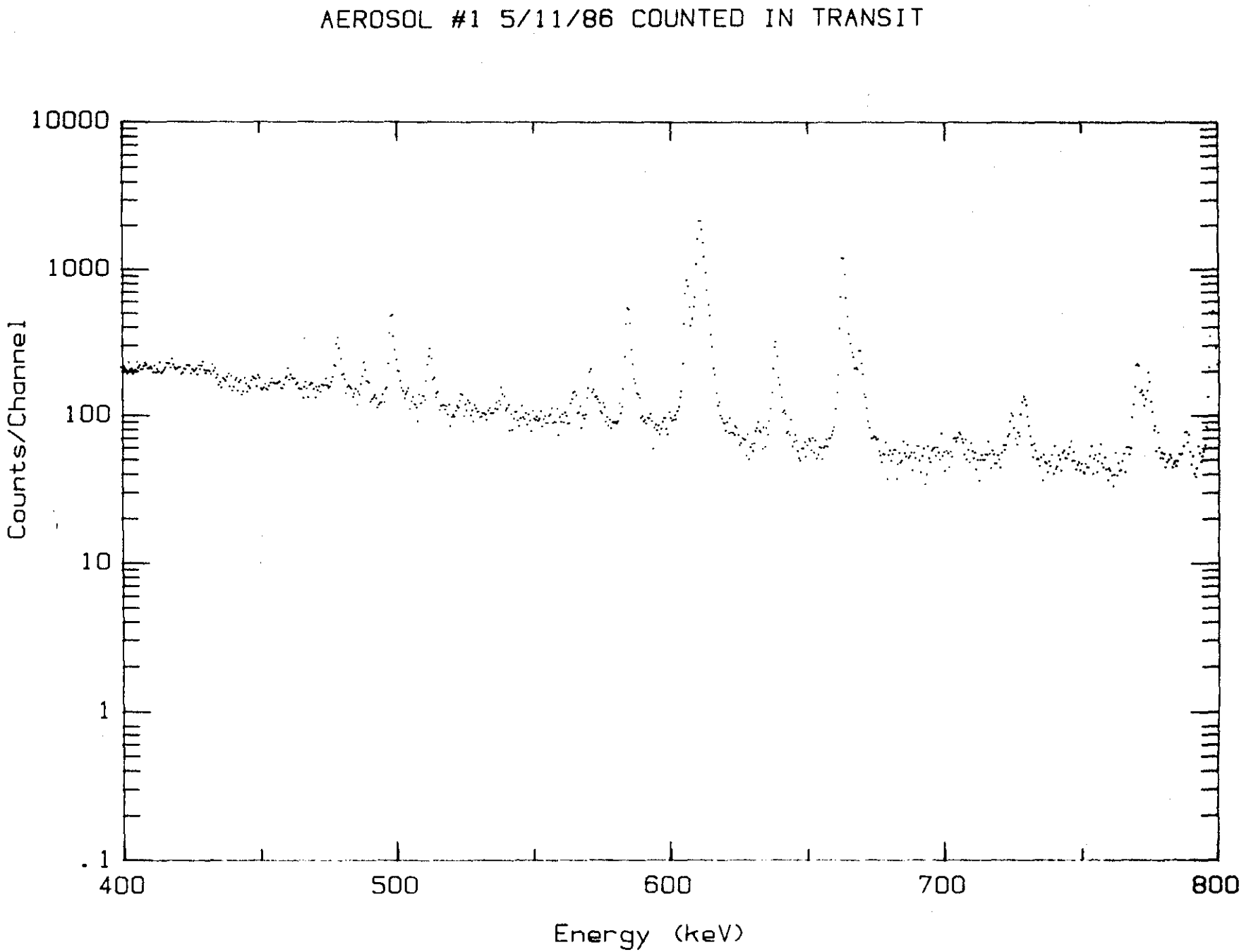
AEROSOL #1 5/11/86 COUNTED IN TRANSIT
10000
1000
100
10
1
.1
Counts/Channel
400
500
600
700
800
Energy (keV)

TABLE 1. TRAC SAMPLE COLLECTION DATA

DATE MAY 1986	COLLECTION START TIME	COLLECTION STOP TIME	LOCATION	VOLUME CUBIC METERS IODINE	VOLUME CUBIC METERS AEROSOL	WIND DIR. (deg)	RAIN PREVIOUS 24 HOURS (SRP)
6	11:56	12:58	Aiken, SC to Columbia, SC	123	911	247	NO
7	09:57	11:12	Hwy 19 to		1206	283	YES
	13:32	15:23	Anderson, SC	339	1293	264	
8	12:33	14:53	Augusta, GA to Clark Hill Dam	158	2541	250	YES
9	10:20	14:17	Augusta, GA to Madison, GA	277	3286	92	YES
10	10:36	13:45	Barnwell, SC to I-95	197	3749	179	NO
11	13:11	16:11	Savannah, GA		3448	276	NO
	16:19	19:14		347	3045	265	
12	13:12	16:08	Augusta, GA at I-20	171	3108	311	YES
13	11:07	14:07	Columbia, SC at I-20	182	3283	219	NO
14	12:33	15:35	Greenville, SC		938	115	YES
	15:40	16:40		242	1096	115	

TABLE 2a. TRAC CONCENTRATION DATA FROM CHERNOBYL ACCIDENT
CONCENTRATIONS IN PCI/M3

LOCATION	CLARK HILL	MADISON	BARNWELL TO I-95	SAVANNAH
DATE	5/8/86	5/9/86	5/10/86	5/11/86
NUCLIDE				
MO-99	<.0005	<.0004	<.015	0.0023
+-				0.0003
RU-103	<.0006	0.0069	0.0396	0.0490
+-		0.0006	0.0022	0.0030
RU-106	<.004	<.0056	<.014	0.0900
+-				0.0200
I-131	<.0006	0.0018	0.8080	0.5160
+-		0.0001	0.0350	0.0260
TE-129M	<.009	<.0085	<.035	0.0390
+-				0.0060
TE-132	<.0007	0.0023	0.0410	0.0360
+-		0.0003	0.0060	0.0020
CS-134	<.0006	0.0013	0.1070	0.1080
+-		0.0004	0.0060	0.0050
CS-136	<.0008	<.0008	0.0200	0.0190
+-			0.0020	0.0009
CS-137	<.0006	0.0027	0.2040	0.2080
+-		0.0005	0.0130	0.0090
LA-140	<.0017	<.0009	0.0254	0.0020
+-			0.0015	0.0009
BA-140	<.0022	<.0016	0.0227	0.0021
+-			0.0075	0.0009
CE-141	<.0007	<.0006	<.0024	0.0013
+-				0.0002
CE-144	<.0030	<.002	<.0008	<.0020
+-				
VOLATILE				
I-131	<.00087	<.00055	0.3110	0.3770
+-			0.0150	0.0180
TOTAL				
I-131	<.0015	<.0024	1.1190	0.6680
+-			0.0380	0.0220

TABLE 2b. TRAC CONCENTRATION DATA FROM CHERNOBYL ACCIDENT
CONCENTRATIONS IN PCI/M3

LOCATION	AUGUSTA	COLUMBIA	GREENVILLE
DATE	5/12/86	5/13/86	5/14/86
NUCLIDE			
MO-99	<.0034		<.00068
+-			
RU-103	0.0086	0.0337	0.0081
+-	0.0005	0.0015	0.0007
RU-106	<.0042	0.0092	<.008
+-		0.0017	
I-131	0.0279	0.1140	0.0350
+-	0.0012	0.0060	0.0020
TE-129M	<.0074	0.0217	<.015
+-		0.0022	
TE-132	0.0023	0.0119	0.0029
+-	0.0004	0.0005	0.0006
CS-134	0.0067	0.0564	0.0091
+-	0.0004	0.0025	0.0006
CS-136	0.0014	0.0081	<.0013
+-	0.0005	0.0005	
CS-137	0.0137	0.1080	0.0165
+-	0.0008	0.0060	0.0010
LA-140	<.0010	0.0090	<.0015
+-		0.0004	
BA-140	<.0009	0.0105	<.0035
+-		0.0009	
CE-141	<.0006	0.0008	<.0011
+-		0.0002	
CE-144	<.0024		<.0048
+-			
VOLATILE			
I-131	0.1860	0.1650	0.1410
+-	0.0090	0.0210	0.0250
TOTAL			
I-131	0.2140	0.2790	0.1760
+-	0.0150	0.0240	0.0250

TABLE 3. DOSE FROM 6 DAY EXPOSURE TO CHERNOBYL DEBRIS

NUCLIDE	pCi/m3 6 day average	+-	Eff. D.E. mrem	Thyroid mrem
MO-99	< 0.00363		< 0.000018	
RU-103	0.02431	0.00069	0.000001	
RU-106	< 0.02183		< 0.001325	
I-131	0.25045	0.00734	0.001105	0.03801
TE-129M	< 0.0211		< 0.000058	
TE-132	0.01606	0.00106	0.000017	0.00050
CS-134	0.04808	0.00137	0.000987	0.00027
CS-136	< 0.00843		< 0.000008	0.00000
CS-137	0.09215	0.00282	0.000406	0.00036
LA-140	< 0.00663		< 0.000004	
BA-140	< 0.00823		< 0.000004	
CE-141	< 0.00113		< 0.000001	
CE-144	< 0.002		< 0.000096	
VOLATILE I-131	0.1968	0.00686	0.000869	0.02987
TOTAL I-131	0.40933	0.00965	0.001807	0.06213
TOTAL DOSE			0.004904	0.06905