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MONITORING FOR AIR-BORNE RADIOACTIVE MATERIALS AT HANFORD ATOMIC PRODUCTS OPERATION

(Presented at the Air Pollution Control Association Meeting Los Angeles, California, June 21-26, 1959)

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MONITORING FOR AIR-BORNE RADIOACTIVE MATERIALS

AT HANFORD ATOMIC PRODUCTS OPERATION

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RICHLAND, WASHINGTON

Manuscript to be presented at the
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ABSTRACT

Methods employed at Hanford Atomic Products Operation to monitor for air-borne radioactive materials are described. Criteria used to establish the atmospheric monitoring program include recommendations of the National Committee on Radiation Protection, H.A.P.O. Radiation Protection Standards, amounts and types of radioisotopes discharged to the atmosphere, and the meteorology and population density of the plant environs. Fixed and portable instrumentation measures and records external radiation dosage rates, and/or collects various types of samples. The samples are analyzed by radiochemical techniques for both gross radioactivity and specific radioisotopes.

MONITORING FOR AIR-BORNE RADIOACTIVE MATERIALS AT HANFORD ATOMIC PRODUCTS OPERATION

by
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INTRODUCTION

The Hanford project is a complex of fuel fabrication plants, nuclear reactors, chemical separations plants, and research laboratories designed for the production of plutonium. It is operated, under government contract, by the General Electric Company through its Hanford Atomic Products Operation.

The project is located in a semi-arid region in southeastern Washington, with an annual rainfall of eight inches. Natural vegetation in the region is sparse, primarily suited for grazing, although considerable areas are being converted to irrigation. The plant site, shown in Figure 1, comprises an area of about five hundred square miles(1).

The Columbia River flows through the area and forms the eastern boundary. The populated areas of primary interest are Richland, Pasco, and Kennewick. Other communities in the vicinity are Benton City, Mesa, and Othello. Altogether, about 80,000 people live in the region around the plant perimeter. The meteorology of the region(2)(3) is typical of a desert area with frequent strong inversions occurring at night, and breaking during the day to provide unstable and turbulent conditions. The prevailing winds are from the northwest in the plant areas with strong drainage winds causing distorted flow patterns.

Monitoring for air-borne radioactive materials is one important phase of the radiation protection program at Hanford. For purposes of discussion, the air monitoring effort can be divided into three main sections:

1. Air monitoring at work locations,
2. Monitoring of gaseous wastes discharged to the atmosphere, and
3. Atmospheric monitoring in the project environs.

The methods and equipment employed under all three headings are very similar. Some differences arise from variances in concentrations being measured.

AIR MONITORING AT WORK LOCATIONS

Control of air-borne radioactive contamination in work locations is accomplished by careful regulation of air flow patterns. Incoming ventilation air is directed first to areas where no radioactive contamination is permitted, and then to areas

of progressively higher contamination potential. A typical air flow pattern might be designed to ventilate work locations in the following order: lunchrooms, offices, locker rooms, maintenance shops, laboratories, cells containing process equipment, air clean-up facilities, ventilation fans, and finally discharge from a high stack. This type of air flow regulation ensures maximum breathing safety from a minimum of air moving equipment. In addition to this over-all flow scheme, certain individual pieces of equipment have separate exhaust and/or air clean-up arrangements.

Sampling and monitoring of working atmospheres is performed by the radiation monitoring group assigned to the plant facility involved. The results of this air monitoring are used to:

1. Ensure that air in "uncontaminated" zones is free of detectable radioactivity,

2. Determine what respiratory protection is needed for entry into "contaminated" zones,

3. Establish trends in concentrations of air-borne contamination,

4. Locate sources of air-borne contamination, and

5. Record air concentrations for permanent legal records of employee exposures.

Some factors affecting the magnitude of the air monitoring program include: identity and state of radioisotopes present, layout of working space, degree of ventilation provided, and probability of generating air-borne contamination during the progress of work. In addition to these four factors, the need to collect a sufficient sample for convenient measurement of extremely low concentrations frequently must be balanced against the need to detect short-term fluctuations in concentrations.

In a discussion of methods for sampling air-borne radioactivity, presented before the APCA in November 1954, Heslep and Bellamy mentioned that concentration by filtering was the most widely used sampling method for particulates[4]. This statement still holds true today. Recent developments in other methods have not displaced filter sampling from its number one position.

At Hanford, Hollingsworth-Vose H-70 filter paper (18 mils thick) is used with a variety of sampling equipment. Tests of the collection efficiency of this and other filter papers have been conducted at Hanford[5] and at Knolls Atomic Power Laboratory (KAPL)[6]. In the tests performed at KAPL, particles whose mean size ranged from 0.2 to 1.2 microns were drawn through H-70 filter paper at face velocities of 0.5 to 250 cm/sec. The maximum penetration observed was three percent. More recently, tests at Hanford employed K_2CrO_4 particles of 0.18 microns (mass mean size). Penetration of these particles through H-70 filter paper (18 mils thick) was 0.02, 0.2, and 0.5 percent, at face velocities of 5, 25, and 51 cm/sec, respectively[7]. The equipment used to draw the air sample through filters varies from centrally exhausted systems of fixed piping to portable, electrically-operated air pumps. Filter holders can be attached directly to the vacuum source or to flexible hoses which permit sampling at less accessible places. Standardization of filter holder design and uniformity of filter media permit the calibration of air flow rates in terms of pressure drop across the filter paper. A simple vacuum gauge is used to measure this pressure drop.

Normally air-borne concentrations of naturally radioactive alpha particle emitters are several orders of magnitude higher than concentrations of Pu and U being sampled. To calculate the amounts of Pu and U present, it was necessary to make two or three measurements of the alpha radioactivity on the filter sample over a period of a few days. The radioactive half-life of the alpha emitters was then used as a measure of their composition. This extended period of analysis prevented immediate determination of air-borne Pu and U concentrations.

One recent solution of this difficulty at Hanford has been the development of a portable impactor air sampler. With this sampler, it is possible to collect Pu and U particulates without collecting naturally occurring radon and thoron daughter products. In the impactor, air is drawn at ~200 feet per second through a sharp 180° turn. The velocity is high enough to collect the relatively large, high density Pu and U particles by impaction, while the fine, light dust particles containing radon and thoron daughter products pass on through(8).

Analysis of gross radioactivity collected by a filter or impactor sample is made with standard alpha, beta, and gamma counting equipment. Alpha counters employ either a parallel plate condenser or a zinc sulphide scintillation screen for a detector. End window G.M. tubes and beta sensitive scintillation crystals are employed for beta counting. Measurements of specific gamma emitting radioisotopes are performed on gamma energy analysis equipment. Single or 256-channel spectrometers are used to make qualitative and quantitative measurements of gamma emitters present in certain samples.

A recent trend has been toward the development of equipment which combines the functions of sampling and analysis in one unit. Such instruments are finding wide use in many fields. Applications of this principle to air monitoring have produced such instruments as continuous strip-filter monitors, counter-current flow air scrubbers, and large volume internal-flow ionization chambers. Careful selection of monitor and associated counting equipment usually permits calibration of the recorded results directly in units of air-borne concentrations. These instruments will be discussed further in the section on stack effluent monitoring.

Respiratory protection required at Hanford for entry into "contaminated" zones can be summarized in the following rule. If the air-borne concentrations of radioisotopes are less than the MPC*, no respiratory protection is required. For concentrations up to 20 times the MPC, an army assault mask is required. For concentrations over 20 times the MPC, a fresh air or "chemox" mask is needed. There are exceptions to this general rule, of course. Radioactive gases normally require the use of fresh air or chemox masks at any concentrations over the MPC.

The actual air-borne concentrations encountered at work locations at Hanford vary widely and cover the range from the lower limits of essentially contamination-free air to several times the recommended maximum permissible limit. The specifications for respiratory protection equipment, its use, and the limited amount of time spent by employees in regions of high air-borne concentrations combine to maintain personnel exposures to within a small fraction of the established maximum permissible limits.

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* The Maximum Permissible Concentration for continuous occupational exposure recommended by the International Commission on Radiological Protection(9) and the National Committee on Radiation Protection and Measurement(10).

MONITORING OF SEPARATIONS PLANTS EFFLUENT GASES

Monitoring of radioactive wastes released to the project environs is performed by an environmental radiation monitoring group. This group is responsible for measuring, recording, and evaluating radiation exposures in the project environs and in the communities surrounding the project. As part of this responsibility, this group maintains inventories of radioactive wastes discharged to the environs, by means of direct monitoring or through compilation of monitoring results obtained by others.

Ventilation air and process effluent gases are potential contributors to atmospheric contamination both on and off the project. Ventilation air from reactor buildings normally contains negligible amounts of radioactive materials. The separations plant effluent gases discharged to the atmosphere contain small quantities of fission products. These gases are released through 200-foot high ventilation stacks, after treatment to remove some 99 percent of the radioactive materials present(11). The rates of discharge of various radioisotopes are summarized in Table I:

TABLE I

EMISSION RATES FROM SEPARATIONS PLANT STACKS
1958

Isotope	Average Curie/day
I-131	1.2
RE+Y	0.02
Nb-95	0.01
Zr-95	0.004
Ru-103	0.004
Ru-106	0.002
Sr-89	0.0035
Sr-90	0.0005

Iodine-131 is emitted in the gaseous form, while the remaining isotopes are normally associated with particulate material. Local limits restrict the emission to ten curies per week of I^{131} from the separations plant stacks.

A counter-current flow scrubber sampler is used to monitor and record I^{131} emission rates (Figure 2). The scrubbing liquid, 0.2 normal NaOH, is pumped from a storage reservoir to the top of the packed scrubbing column at a constant rate of 5 cc per minute. Approximately 0.5 cfm of gas sample passes through a flow recording device and then into the bottom of the column. As the gas rises through the column, the caustic solution collects the I^{131}. The liquid then drains from the bottom of the column, through a "counting cell" and into a collection bottle. A static pressure balancing arrangement and a liquid level regulator maintain a constant liquid head in the column. At the "counting cell", a variety of counting

systems can be used. If only gross radioactivity is to be monitored, then a simple system consisting of a G.M. tube, a count-rate-meter, and a recorder is sufficient. Monitoring for mixtures of radioisotopes may require the use of separate beta and gamma radiation detectors with gamma energy discrimination devices.

The spent caustic solution accumulated in the collection bottle can be taken to the radiochemical analysis laboratory for supplemental radioisotopic analyses or for calibration checks on the monitoring system.

For radioactive particulate materials, two types of filter monitors are employed. One type of filter sampler uses a large area (4" diameter circle) piece of H-70 filter paper, 18 mils thick, through which about 1.0 cfm of stack gas sample is continuously drawn. These samplers are operated for 24 hours and then the filter paper is replaced. The exposed filter is taken to the radiochemical analysis laboratory for measurements of gross activity and radioactive ruthenium. Spot checks for other specific radioisotopes are made on selected filters.

The second type of filter sampler is used for continuous monitoring and recording of emission rates of radioactive particulates. This type of sampler is shown in Figure 3. In it, a strip of filter paper is drawn past a sampler head, either continuously or in a step-wise fashion. The exposed filter paper then passes by a counting device, such as a G.M. tube, or scintillation crystal connected to a count-rate-meter and recorder. Here again, more complex counting and recording systems can be employed to measure emission rates of individual radioisotopes.

One system combining a strip filter monitor and an air scrubber has been developed at Hanford by the Radiation Instrument Development Operation[12]. In this monitor (Figures 4 and 5), a metered flow of stack gas sample passes first through a continuously moving strip filter and then through a counter-current flow caustic scrubbing column. Both the exposed filter paper and the spent scrubber liquid are monitored by separate beta and gamma scintillation crystals, and radiation energy discrimination devices. The counting rates from the four scintillation crystals are integrated electronically to yield recorded information on the individual emission rates of I^{131}, Ru^{103}, and Ru^{106}. Enough electronic equipment has been incorporated to provide space for measuring and recording some fourth radioisotope in the future.

This unit and the other continuous sampling and monitoring devices described are necessary aids to maintaining control limits on stack emission rates. They also allow correlation of emission rates with process operating conditions, and prediction of environmental concentrations of emitted isotopes.

ATMOSPHERIC MONITORING

One of the purposes of environmental air monitoring is to measure potential radiation exposure to the general public from the radioisotopes discharged from the separations plant stacks. In addition, two sources of radiation exposure not of Hanford origin are measured. These two are natural background radiation and radioactive fallout from nuclear test explosions.

Under present recommendations of the National Committee on Radiation Protection and Measurement (NCRP), natural background radiation can be subtracted from total radiation and the maximum permissible exposure criteria can then be applied to only the "man-made" contributions[9][10][13]. To perform this subtraction, however, the natural background radiation either must be evaluated separately, or must be eliminated directly from the measured radiation exposure. Separate identification of fallout and of plant contributions is a necessary aid to maintaining plant limits designed to control the release of radioactive wastes to the environs.

Air-borne concentrations of radioactive materials and radiation dosage rates are measured by equipment located in 31 atmospheric monitoring stations, and by portable ionization chambers placed at selected field locations. Twenty-seven of the monitoring stations are within project territory, while the remainder are situated in the four towns south and southeast of the project. Each station (Figure 6) is a 6' x 6' x 8' high building with a louvered cupola in the roof to permit air circulation around the monitoring equipment.

Iodine-131 scrubber samplers are operated in a dozen of the monitoring stations concentrated around the separations plants. These samplers consist of a calibrated, electrically-driven vacuum pump which draws 2.0 cfm (3.4 m^3/hr) of air through one liter of 0.1 normal NaOH solution. A balancing platform and siphon arrangement permits introduction of distilled water into the scrubber at a rate equal to the rate of evaporation. This water feeder helps maintain constant liquid head, air flow rate, and scrubber efficiency.

After one week of operation, the scrubber bottle is replaced and taken to the radiochemical analysis laboratory for determination of the I^{131} content. The analytical procedure used provides for the addition of an iodine carrier and $Ag\ NO_3$ to the scrubber solution, followed by filtration of the resulting silver iodide precipitate. The radiation from the I^{131} on the filter is measured by an end-window G.M. tube connected to a scale-of-64 scaler. Atmospheric concentrations of I^{131} are then calculated from these counting rates by applying factors for counter calibration, chemical recovery of the I^{131}, scrubber efficiency and the volume of air sampled. The detection limit for I^{131} in the atmosphere using the above methods is 10^{-14} µc per cc of air.

Air-borne radioactive particulates are also sampled at selected monitoring stations. These samples are collected by drawing 2.0 cfm of air through small filters, 1-11/16 inches in diameter, for a period of one week. The collected radioactive materials are measured in the laboratory for gross beta activity by counting with an end-window G.M. tube - scaler arrangement. The filters are also counted for alpha particle emitters using alpha sensitive zinc sulphide scintillation equipment.

A delay period of 2-3 days between filter changing and alpha counting is used to permit the relatively short half-life, naturally radioactive, alpha particle emitters to decay to insignificence. The detection limits for these filter sampling and counting techniques are: 10^{-14} µc of gross beta emitters per cc of air, and 10^{-15} µc of gross alpha emitters per cc of air. The filters may also be spot-checked for specific radioisotopes by conventional radiochemical means or by use of a 256-channel gamma-ray spectrometer.

Measurements for concentrations of radioactive particulates in the atmosphere are made with 2 inch by 4 inch H-70 filter paper. Twenty-four of these filter samplers are operated within the project, four are operated in nearby towns, and nine others are located in remote locations scattered throughout the Pacific Northwest. The filters are changed on either a daily or a weekly schedule and then are autoradiographed using Eastman Kodak, Type-K, X-ray film. The filters are placed next to the film for one week, the filter is removed, and the film is developed. The developed film is viewed on a standard X-ray viewer and each darkened spot counted as one radioactive particle. Air-borne concentrations of radioactive particles are calculated by dividing the number of darkened spots obtained per filter by the total volume of air sampled. The detection limit is one particle per filter, of 20 disintegrations per minute or higher activity. For weekly filter operation, this amounts to a detection limit of 2×10^{-3} particles per cubic meter of air (2×10^{-9} particles per cc).

Table II is a summary of the results obtained from operation of these filter and scrubber samplers during 1958:

TABLE II

ATMOSPHERIC CONCENTRATIONS
1958

Location	10^{-13} µc per cc			Radioactive Particles per m^3
	I-131	Alpha Emitters	Beta Emitters	
On Plant	5.0	< 0.02	14	0.04
Plant Perimeter	2.4	< 0.02	12	0.04
40 - 100 Miles	---	----	12	0.06
Over 100 Miles	---	----	14	0.08

The perimeter locations are primarily in the Richland-Pasco-Kennewick area, while those over 100 miles from the plant are in Washington, Southern Oregon, Southern Idaho, and Western Montana. The influence of debris from nuclear detonations is evident in these data. Examination of the results from individual monitoring stations indicates that locations upwind and cross-wind from the project have atmospheric concentrations of radioactive particulates equal to or greater than those locations immediately downwind of the project.

External radiation dosage rates in the project environs result from radioactive materials suspended in the atmosphere or deposited on the ground. Measurement of the low dosage rates encountered is accomplished through the use of small portable ionization chambers and a novel chamber reader developed at Hanford(14).

Measurements made with these chambers vary with atmospheric concentrations of naturally radioactive radon, thoron, and their decay products, and with the amount of freshly deposited radioactive fallout from nuclear detonations. In the absence of fresh fallout, the results average about 0.3 to 0.4 milliroentgen (mr) of gamma radiation per day. This figure includes about 0.2 - 0.3 mr per day of natural background radiation, so that the Hanford contribution is on the order of 0.1 mr per day or less.

The isotopic composition of the deposited radioactive materials is determined by collecting and analyzing vegetation samples, principally grass and sagebrush, around the project and in Eastern Washington and Northern Oregon. Locally, garden produce and milk are also purchased for analysis. A large part of the environmental air monitoring program involves the study of I^{131} because of its high volatility during dissolving of the irradiated uranium, and because of its strong tendency to deposit on vegetation and other surfaces.

The amount of radioactive contamination deposited on the vegetation is measured with a 256-channel gamma-ray spectrometer. Specific isotopic concentrations are calculated from counting rates obtained at their characteristic gamma energies, after applying corrections for compton scattering and natural background radiation. Strontium^{89} and Sr^{90} concentrations are determined by radiochemical separation and beta particle counting. The majority of radioisotopic measurements made on milk and produce samples are below their respective detection limits of about 10^{-7} μc per gram. Higher isotopic measurements are obtained for the grass samples because of the higher surface to weight ratio of grasses.

Figure 7 illustrates the annual average deposition pattern of I^{131} on grasses and sagebrush, based on an emission rate of one curie per day(1). This pattern shows the influence of the prevailing winds and the frequent inversions which tend to carry the plume to long distances before it reaches the ground.

Deposition of I^{131} and particulates from the atmosphere can be followed by performing radiological surveys of the ground. These surveys are made with portable transistorized equipment employing a gamma sensitive scintillation crystal, five inches in diameter by five inches thick. The equipment can be carried by car, boat, or airplane. In the event that unusual ground contamination is detected, more detailed surveys can be made with hand portable instruments and by collecting supplemental vegetation samples.

SUMMARY

The air monitoring program at Hanford employs a variety of equipment and techniques for measurement of air-borne and deposited radioactive materials. Some of the instruments used were developed at Hanford for specific applications. The measurements made are complicated by the presence of naturally occurring radioactive materials and of radioactive fission products from nuclear detonations. Present methods are adequate to ensure that exposures of humans and grazing animals to air-borne or deposited radioisotopes are well below applicable limits; however, refinements in air monitoring techniques are constantly being made.

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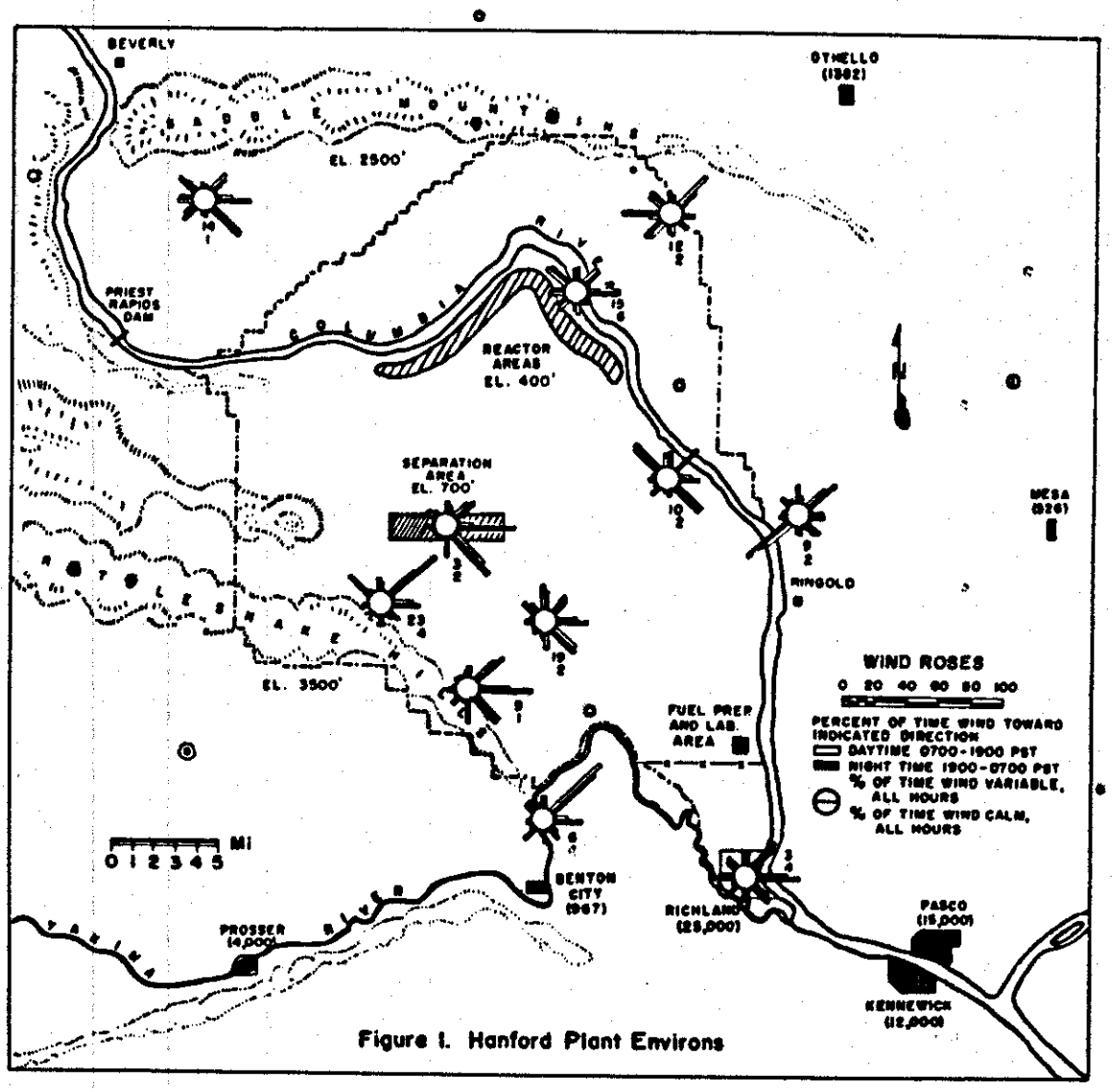


Figure 1. Hanford Plant Environs

FIGURE 1
Hanford Plant Environs

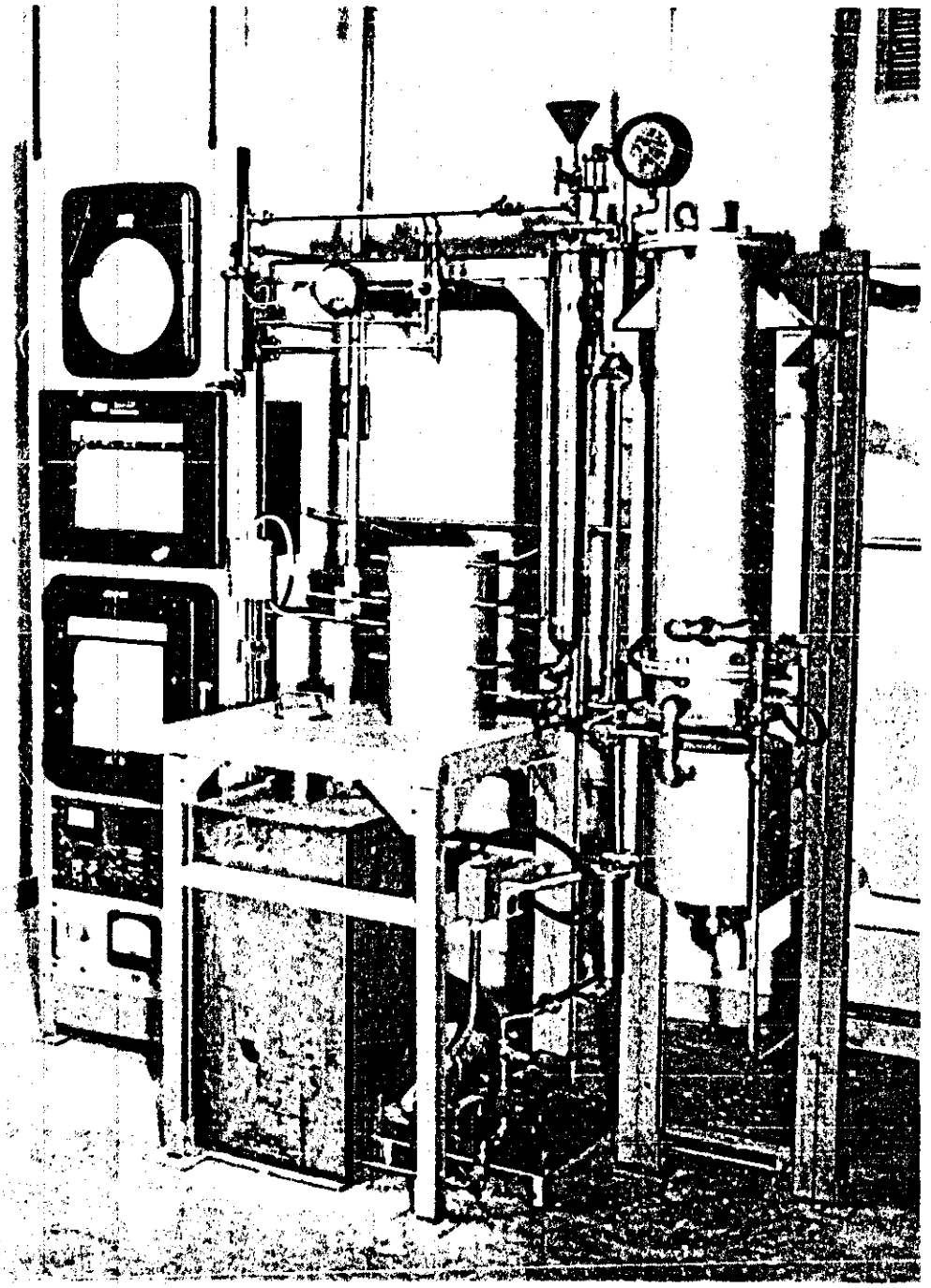


FIGURE 2

Continuous I^{131} Monitor

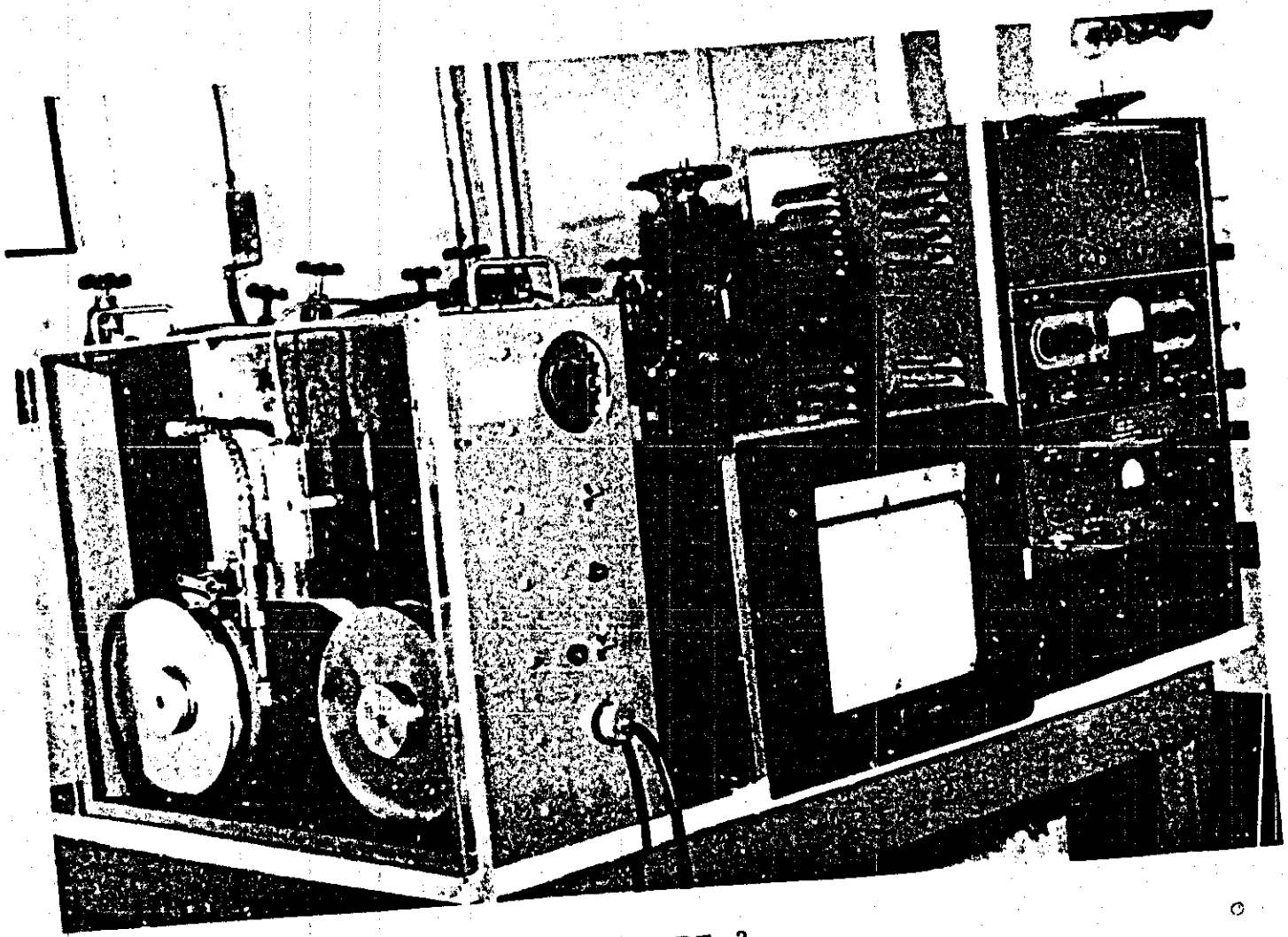
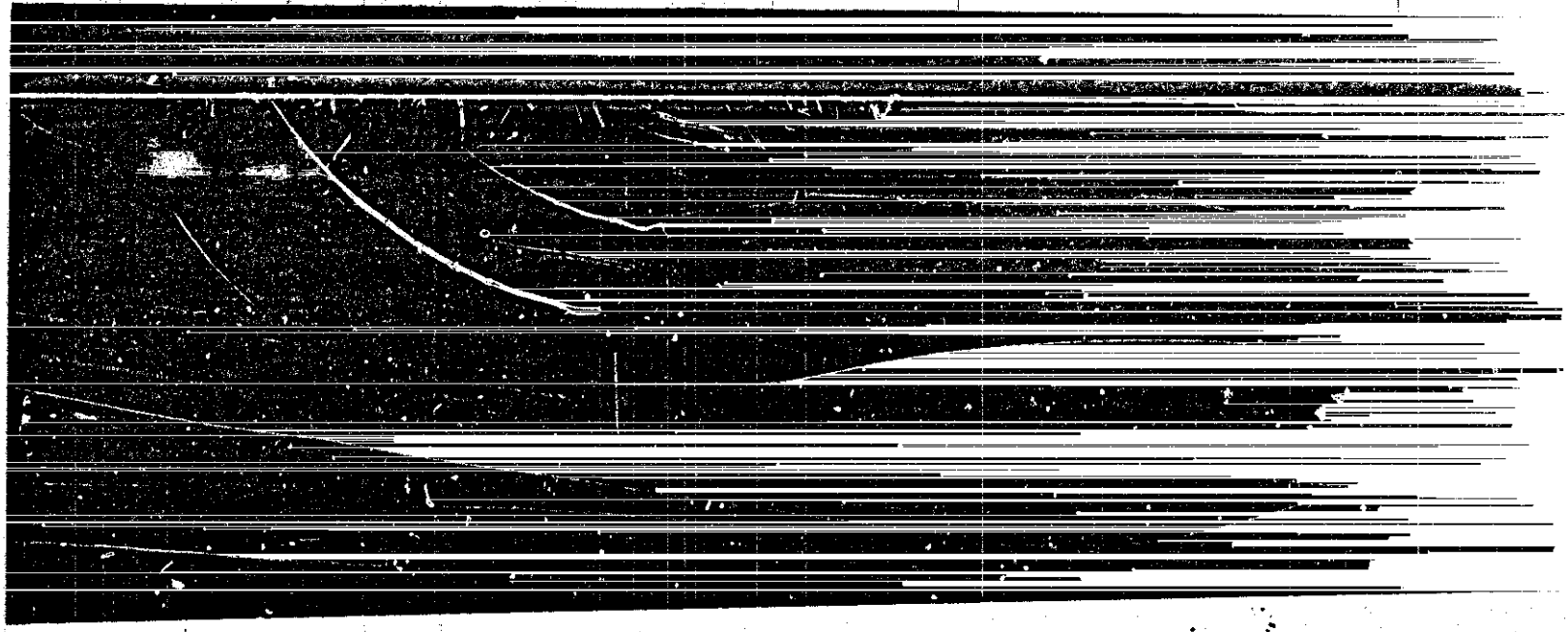


FIGURE 3

Strip Filter Monitor



BLOCK DIAGRAM OF STACK ANALYZING MONITOR

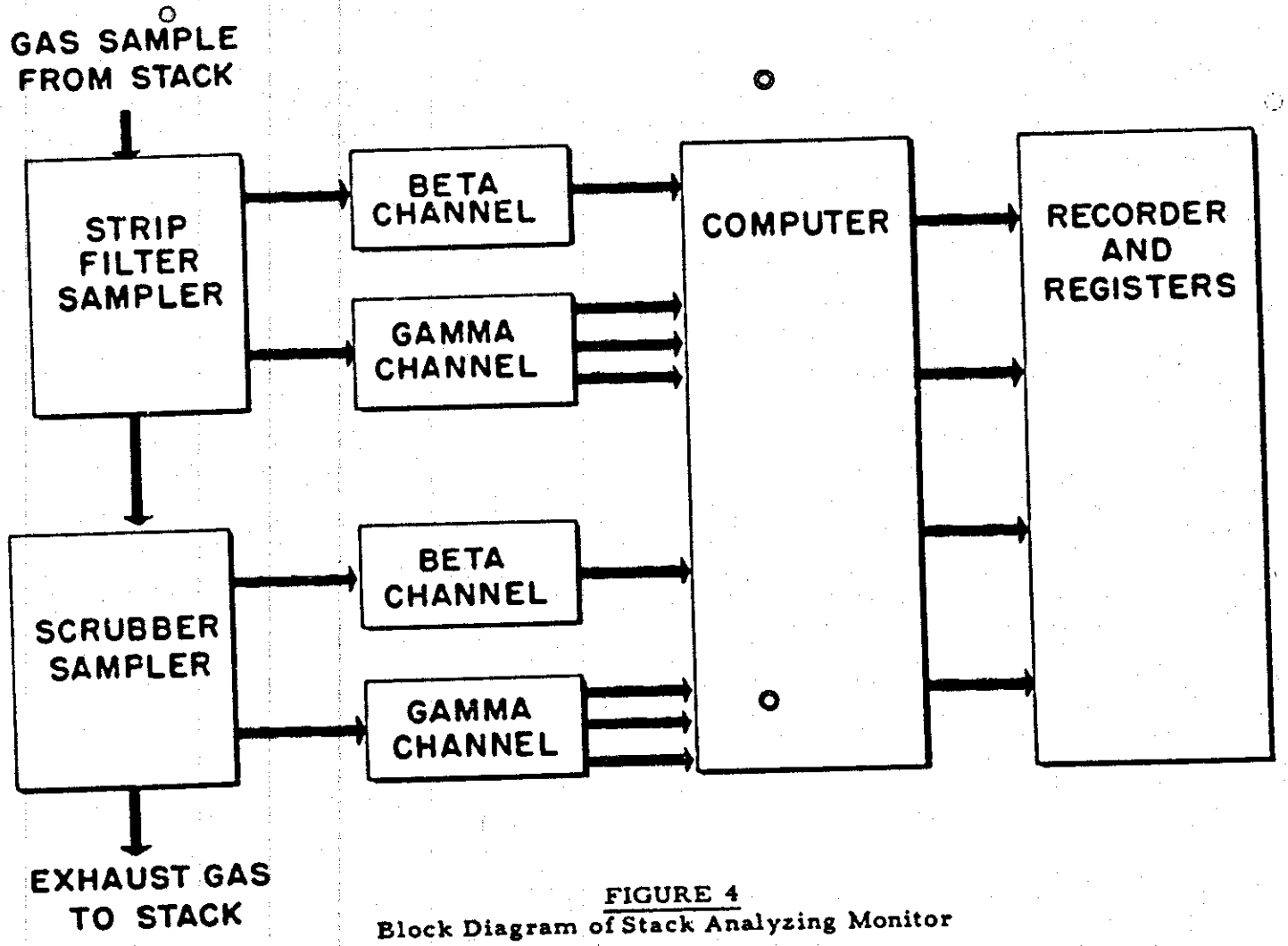


FIGURE 4
Block Diagram of Stack Analyzing Monitor

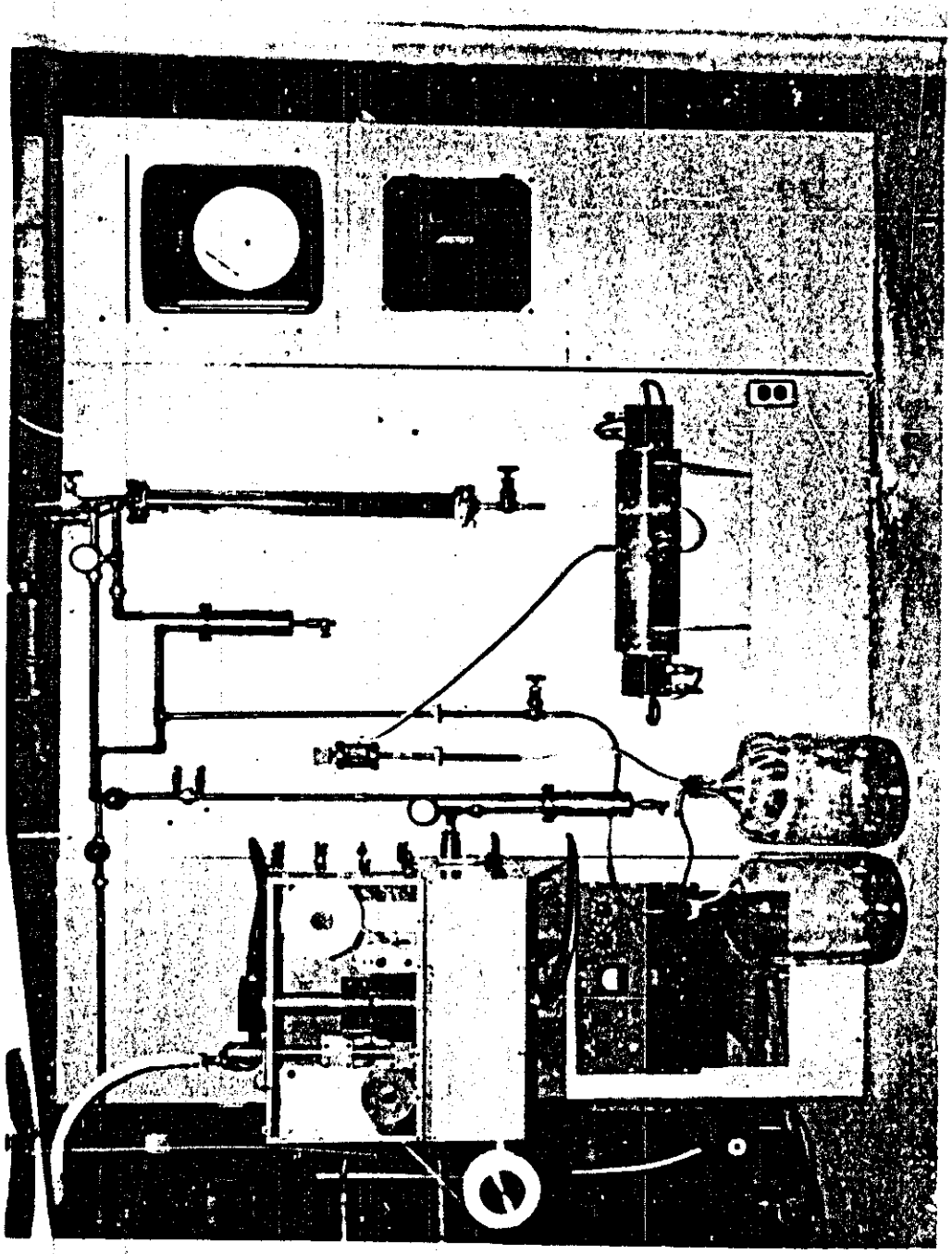


FIGURE 5

Stack Analyzing Monitor

AEC-GE RICHLAND, WASH.

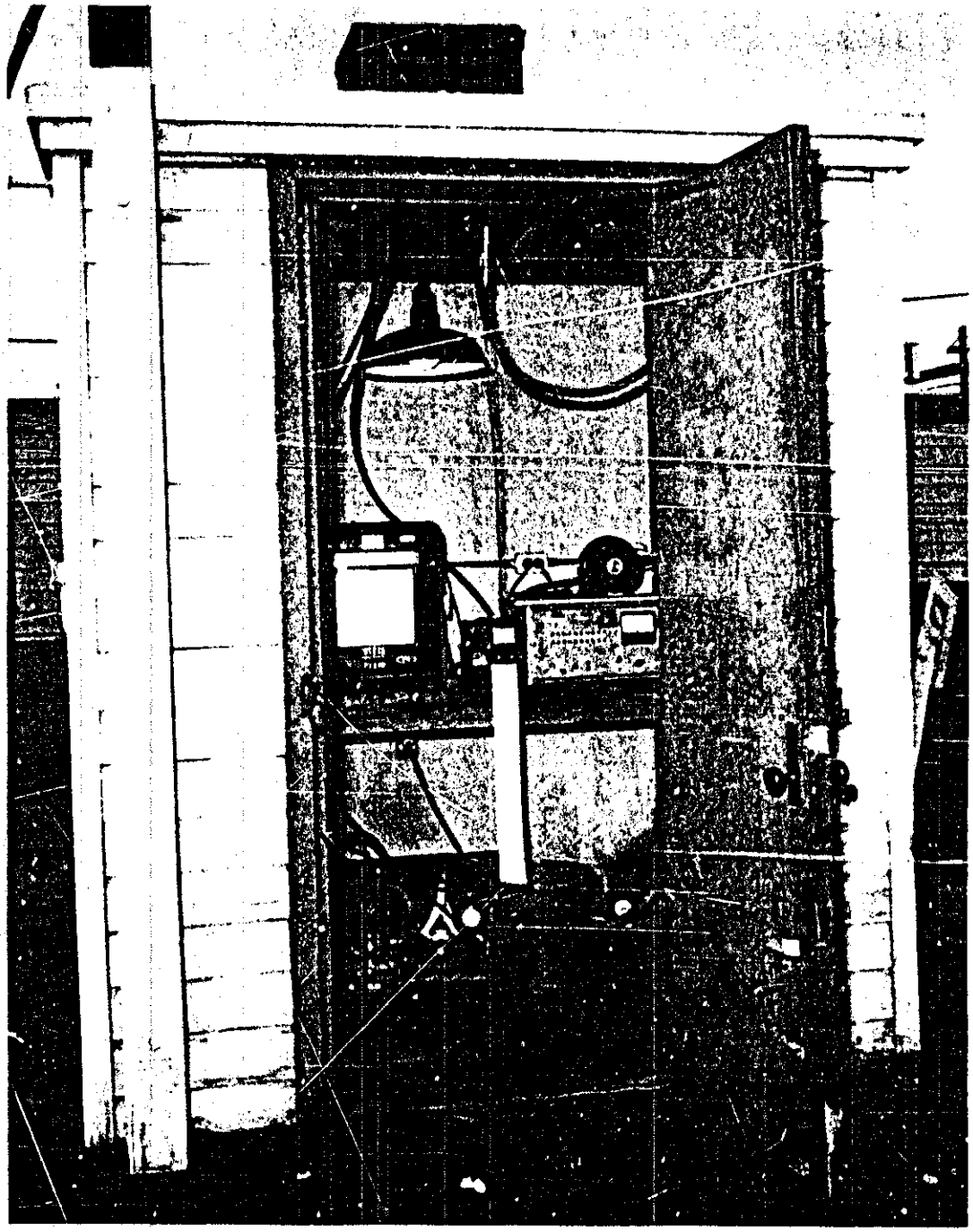
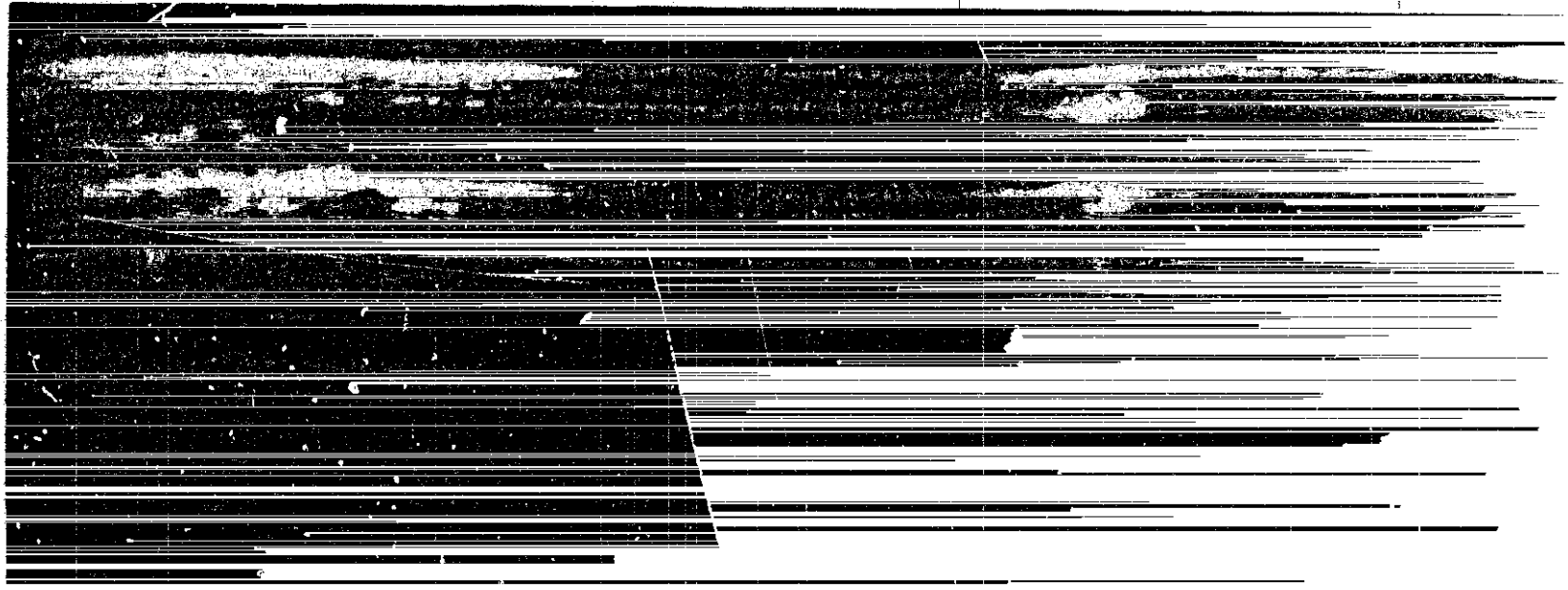
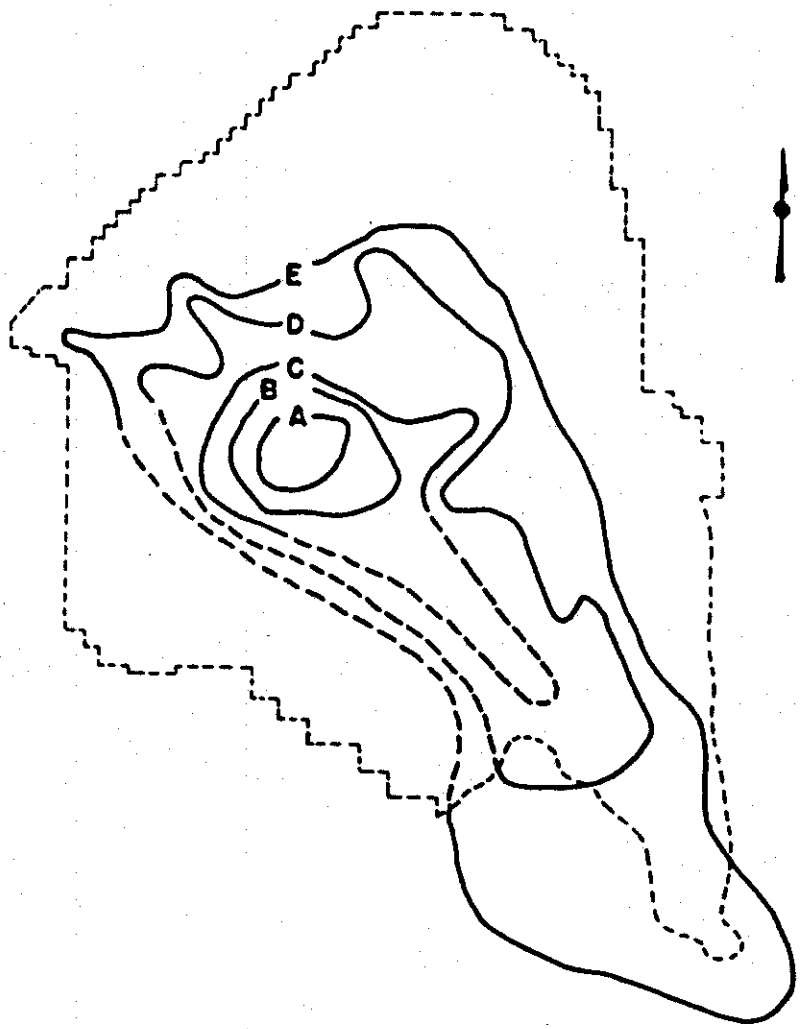


FIGURE 6

Atmospheric Monitoring Station



AVERAGE I^{131} DEPOSITION - μc/gm OF VEGETATION PER CURIE/DAY EMITTED



A - 3×10^{-5}
B - 1.5×10^{-5}
C - 9×10^{-6}
D - 6×10^{-6}
E - 5×10^{-6}

MILES
0 5

FIGURE 7
Average I^{131} Deposition - μc/gm of Vegetation per Curie/Day Emitted