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I-129: A NEW ATMOSPHERIC TRACER

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by

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Summary Report to:

Division of Reactor Development
United States Atomic Energy Commission

Contract No. AT(30-1)-3201

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The work described in this report was performed under contract with the Division of Reactor Development of the United States Atomic Energy Commission. The authors wish to acknowledge the continuing interest and assistance of Mr. Harold Bernard and Dr. Isaac Van der Hoven of that division. The field tests held in Idaho could not have been performed without the full cooperation of Dr. Norman Islitzer, Mr. C. R. Dickson and the other members of the U. S. Weather Bureau staff at the NRTS and of the staff of the USAEC Health Physics Group in Idaho.

ABSTRACT

The requirements for tracers of atmospheric motions are reviewed, particularly with respect to their utility over distances greater than ten miles. The development of tracers incorporating the long-lived nuclide I-129 is discussed. The two I-129-labeled tracers, diiodofluorescein and trifluoromethyl iodide, were developed for use as an aerosol and a gaseous tracer, respectively. Detailed descriptions of the methodology for the synthesis, dissemination, collection, and analysis of these tracers are given.

The use of I-129 as the tracer can result in the extremely high sensitivity and low background required for tracers useful over long distances. High sensitivity is achieved by using specially developed neutron-activation analysis methods. Low background concentrations result from the fact that there is virtually no naturally occurring I-129.

The results of a series of field tests of these tracers showed that the method is basically sound. Results, essentially in agreement with theoretical predictions, were obtained to distances of 64 kilometers for the aerosol tracer. However, the results for the gaseous tracer were inconclusive due to the inadvertent use of charcoal contaminated with fission-product-I-129 as the gas sampling media.

Calculations, based on assumptions of somewhat improved background levels and reasonable increases in the quantities of I-129 released, show that the I-129 tracer method can be effective over ranges up to 800 kilometers.

I. INTRODUCTION

Since April, 1963, NSEC has conducted a research and development effort with the objective of developing tracers and methods for performing atmospheric diffusion studies over ranges beyond those for which existing methods are already adequate. This report reviews the history, progress, and results of the investigations performed through June, 1965. The program, which culminated in a group of field tests to determine the feasibility of the methods, is being carried out for the Division of Reactor Development of the United States Atomic Energy Commission under Contract AT(30-1)-3201.

The problem of conservation of unpolluted air has received considerable attention in recent years from the viewpoint of conventional industrial air pollution. However, the increasing development and projected civilian utilization of nuclear energy is expected to create a new type of potential air-pollution problem. This problem, with its implications for management of airborne nuclear wastes, is analogous to that of conventional air pollution(1). A need exists for meteorological research to investigate the meso-scale and large-scale diffusion or transport of both types of pollutants in the atmosphere. Valid methods of tracing atmospheric motions over these long distances are required to obtain the desired experimental measurements.

The tracer method may be described as a sequence of operations involving the introduction of the tracer element, in a specific chemical or physical form, into the system to be studied, and the determination of its behavior in that system by measurement of its concentrations in predetermined portions of that system. In measurements of atmospheric diffusion, the tracer is typically disseminated at a known rate, and the concentration in the atmosphere is determined by sensitive analytical measurements on appropriate samples collected at selected positions and times down-wind from the disseminator. Therefore, an evaluation of potential tracer methods must take into account four operations: (1) the preparation and characterization of the tracer material, (2) the method of dissemination, (3) the type of sample collector, and (4) the analytical method. Although the detailed specifications for each of these operations may vary according to the technical objectives of specific experiments, certain general requirements can be set forth for typical atmospheric tracer applications.

In most previous work, it has been assumed that particulates having diameters on the order of microns behave in the atmosphere in a manner which accurately simulates the behavior of gases. Some evidence exists that this assumption is not valid (2). Therefore, for the purpose of this study, a basic premise has been established; viz., particulate tracers are valid indices of aerosol behavior only, and true gas tracers must be used as indices of the behavior of gases in the atmosphere. This premise explicitly requires that potential tracer methods be evaluated separately for gas tracers and aerosol tracers, and that the development of a general tracer method should provide a tracer entity of each type.

The specific objectives of a particular tracer application will dictate the choice of a gaseous or aerosol tracer. Typically, gas tracers would be used for basic research (e.g. the study of atmospheric motions) or engineering tests in the field (e.g. determination of the dispersion and fate of volatile effluents from nuclear installations). Similarly, aerosol tracers would be used for general studies of the transport of particulates in the atmosphere or for the evaluation of potential hazards resulting from the release of radioactive particulates during credible nuclear incidents. The physical state of the tracer will generally establish the criteria for dissemination and collection devices. The technical objectives will also dictate the method of dissemination (continuous or batch) and the scale of the experiment. This latter factor will define the overall sensitivity required for the tracer method which, in turn, may be strongly dependent on the analytical methods used to measure the tracer in collected samples.

Comparative evaluations of alternative methods for an experimental measurement generally take into account estimates of accuracy, sensitivity, and cost. Frequently, an inverse relationship is encountered between cost and sensitivity. Since the sensitivity requirements for an atmospheric tracer experiment would normally be defined by the scale of the experiment, it appears reasonable to define three broad categories of applications and to compare the potential of the respective methods for each category. These categories have been arbitrarily defined as small-scale (<5 miles), meso-scale (5-50 miles), and large-scale (>50 miles). Applicable "conventional" and nuclear methods for gas tracers and aerosol tracers were considered separately for each category.

Typical conventional tracer methods used in the past involve sulfur dioxide, fluorescent particles (such as zinc cadmium sulfide), lycopodium spores, and dyes (such as uranine). Since the latter three methods all involve use of a tracer in the particulate form, they are excluded from consideration in this evaluation for use in measurements of gas motions in the atmosphere. However, the demonstrated use of these methods in the field(3) and the favorable cost of the tracer materials and analytical measurements(4) indicate qualitatively that these latter tracers may prove to be most advantageous for experiments having scales compatible with the overall sensitivities of these methods. The successful use of fluorescent pigments for ranges up to 22 miles has been reported (5). However, the general use of such tracers appears to be most advantageous for small-scale experiments. A short, general review of the characteristics of the common tracers is given by Dumbauld(4). A table from that paper is reproduced here (Table I) for convenience.

The nuclear tracer methods that were considered in this work include the use of radioisotopes, stable isotopes, and the quasi-stable isotope, I-129. The use of radioisotopes involves direct radiometric analyses for the tracer in the collected samples, while in the latter two methods, neutron-activation analysis appears to be the optimum method. These three methods have intrinsically better sensitivity than the "conventional" methods cited previously. However, the cost of the tracer materials and analytical measurements is normally higher. On the basis of this qualitative comparison alone, it is expected that these tracers would be more advantageous for meso-scale or large-scale aerosol-tracer experiments where their increased sensitivity would be required. There is, in principle, a wide selection of tracer elements for both aerosol and gas tracing using these methods. Therefore, one or more of these methods may be of substantial interest for small-scale applications using gas tracers. The radioisotope tracer method has been used successfully (2, 6) for such applications.

The extension of these qualitative observations to a quantitative comparison of the respective methods for small-scale, meso-scale, or large-scale experiments must take into account numerous other considerations peculiar to the methods, the desired properties of the optimum tracer element, and a comparison of candidate tracer elements for each method. For example, the use of a radioisotope tracer would require evaluation of the potential radiological hazards resulting from its release to the environment. For a short-lived tracer (e.g., 1.83-hr Ar-41) in a small-scale application, some restriction on personnel access to the experimental area would probably be required. However, the use of a longer-lived radioisotope (e.g., 5770-yr C-14) in large-scale experiments would require consideration not only of the short-term potential hazards to the population, but also of the long-term implications of its release to the environment.

The estimation of the ultimate sensitivity which can be achieved practically using stable isotopes, I-129, or certain radioisotopes depends in part on the intrinsic analytical sensitivities for the specific tracer elements and partly on the existing concentrations of these tracers in the atmosphere. The intrinsic sensitivities are readily estimated from the nuclear properties of the respective tracer elements. However, the basis for estimates of background concentrations are much less precise, especially for I-129 and for stable isotopes of naturally-occurring elements.

The quantitative comparison of tracers for the respective methods considered the optimum physical and chemical properties for both gaseous and aerosol tracer entities. In each case, the tracer had to be stable with respect to credible chemical or photochemical interactions in the atmosphere or biosphere for the duration of the experiment. Hence, the criteria are somewhat more stringent for large-scale than for small-scale experiments. Water-soluble or hygroscopic substances were avoided. For aerosol tracers, control of particle size during dissemination is also important.

The choice of a chemically inert stable gaseous tracer to be analyzed by activation analysis and with a relatively low natural background is severely limited. This is largely due to the fact that most rare elements do not exist in chemical forms which are gases at ordinary environmental temperatures. A notable exception is the "noble" gas, xenon, but the xenon content of the atmosphere is reported to be $\sim 6 \times 10^{-6}$ weight percent (7).

The choice of a particulate tracer is not limited in this way. However, the background concentrations may vary with locale. Therefore, on-site sampling is required to determine the extent to which such concentrations may limit the selection.

The use of an extinct nuclide, I-129, provides a tracer for which the background concentration is, in principle, nearly zero. In practice, anthropogenic material generated in nuclear detonations and released from nuclear fuel processing plants is present in the environment. Therefore, small background concentrations are expected. The concentrations will normally vary with the locale of any field experiments. Contamination of equipment, materials, and chemicals used in the tracer tests may also result.

The element iodine exists in diverse chemical forms, many of which represent suitable particulate materials for tracers. A dye, diiodofluorescein (DIF), was selected for use in this program. This decision was made, in

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part, because of the chemical similarity of DIF to uranine (the sodium salt of fluorescein), because it is non-toxic, and because the technology for the dissemination of uranine was already developed (8). Furthermore, the economical fluorimetric procedure already in use for uranine analyses could be applied to DIF or to mixtures of DIF and fluorescein for measurements of samples collected at close range. The use of DIF and fluorescein is preferable to that of their sodium salts, because the former compounds are insoluble in water. Thus, the use of DIF permits exploitation of the economic advantages of a fluorescent dye tracer at short ranges and the exceptional sensitivity of the I-129-activation-analysis method for long ranges.

Inert, stable compounds of iodine which are true gases at ordinary temperatures, are somewhat less abundant. Elemental iodine and methyl iodide are volatile at ordinary ambient temperatures but are really a solid and a liquid, respectively, and their vapors tend to condense and/or adsorb on solid surfaces. In addition, these compounds are somewhat reactive. Hydrogen iodide, iodine pentafluoride and iodine heptafluoride are all true gases at ordinary temperatures but are chemically very reactive. The 'freon'-like material, trifluoromethyl iodide (CF_3I), however, is a true gas above -22°C at one atmosphere pressure (9) and, like other compounds of its class, is chemically and biologically inert. CF_3I, therefore, is the obvious choice for an iodine-bearing gaseous tracer.

The experimental effort was therefore directed to the development of methods to utilize the two tracers, DIF and CF_3I, labeled with I-129. The remainder of this report describes the experimental effort and the results obtained in the laboratory and in the field, and discusses the results in terms of the practical utilization of these tracers.

TABLE I

PROPERTIES OF CERTAIN METEOROLOGICAL TRACERS*

	Oil-Fog	Sulfur dioxide	Zinc-cadmium sulfide	Uranine dye	Rhodamine B	Amino G Acid
Conservative properties	Fluorescent, dense smoke. Limited downwind travel in temperature lapse conditions	Acid gas. Limited downwind travel in temperature lapse conditions	Fluorescent pigment. Down wind travel to great distances	Fluorescent dye. Downwind travel to 10-20 km	Fluorescent dye. Downwind travel to 10-20 km	Fluorescent material. Downwind travel to 10 km
Dispersive mechanism	Smoke generator. Source strength can be calibrated on mass basis	Compressed gas cylinder. Source strength easily determined on mass basis	Particle dispersion. Source strength difficult to determine on mass basis	Pneumatic nozzle, liquid dispersion. Source strength easily determined on mass basis	Pneumatic nozzle, liquid dispersion. Source strength easily determined on mass basis	Pneumatic nozzle, liquid dispersion. Source strength easily determined on mass basis
Ease of analysis	Instantaneous analysis with densitometer. Filter analysis and photographic techniques	300 samples per hour using 2 analysts	200-250 samples per day - automatic particle irradiation technique, 40-60 samples per day -microscopic technique	240-260 samples per day using 2 analysts	240-260 samples per day using 2 analysts	240-260 samples per day using 2 analysts
Tracer material cost	$0.53/gal	$0.22/lb	$5.50-6.00/lb	$2.34/lb	$4.76/lb	$0.98/lb
Detection sensitivity	3×10^{-6} gm per sample for filter paper analysis	1 part per 100 million	1×10^{-8} gm per sample using automatic technique. 1×10^{-10} - 10^{-11} gm per sample using microscopic analysis	1×10^{-10} gm/ml	1.7×10^{-11} gm/ml	2.5×10^{-9} gm/ml
Approximate Particle Size	0.3 μ	Gas	2.5 μ	88% less than 2.8 μ	Unknown, but should be similar to uranine	Unknown, but should be similar to uranine
Decomposition as a Function of Temperature				26 percent at 620F for 10 min.		2.5 percent at 600 F for 10 min.
Temperature Sensitivity				0.1 percent decrease in fluorescence per degree C increase in temperature	2.0 percent decrease in fluorescence per degree C increase in temperature	
Relative decrease in sensitivity due to presence of water soluble background material				Average	Least	Greatest
Color of fluorescent tracers in solution				Green	Red	Colorless

* [illegible]

II. EXPERIMENTAL

A. The Particulate Tracer

1. Preparation of Labeled Diiodofluorescein (DIF).

Two methods of preparing DIF labeled with I-129 were investigated using 58-day I-125 as a tracer. One method [(10)] using a mixture of dichloramine-T, potassium iodide (labeled), fluorescein, and acetic acid, gave apparently excellent chemical yields (>93%). However, analysis of the reaction product by thin-layer chromatography revealed that the product was impure in that it contained a mixture of iodinated fluoresceins along with some chlorinated fluoresceins. Since purification of the desired product from the objectional components would involve a tedious procedure, emphasis was placed on an alternative preparative method.

The second method [(11)] involves reaction of a buffered solution of sodium fluorescein with a chloroform solution of (labeled) elemental iodine for approximately thirty minutes. This method gave acceptable chemical yields of approximately 80 percent of theoretical. Although this method intrinsically utilizes only one-half of the initial tracer, the unused material was readily recoverable. In the original experiments, the reaction product consisted of at least 70 percent DIF with the remainder almost entirely triiodofluorescein (TIF). Further experiments determined that, by careful control of the temperature during the synthesis, DIF having satisfactory purity could be prepared reproducibly. The product contained only a very small fraction of TIF, which had no significant undesired effect, since the mixture could be readily calibrated for both fluorimetric and activation analyses. Such a calibration would also be required for pure DIF. The developed preparatory procedure was used to label DIF with I-129. The details of the procedure are given in Appendix I.

Characterization of the DIF prepared in this manner (as well as the I-129 labeled product prepared for the field tests) was accomplished

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using paper chromatography. The method is typical of those described in the literature (12) using a descending solvent system composed of 200 parts of n-butanol, 88 parts of water, 2 parts of ammonium hydroxide, and 40 parts of ethanol. The presence of DIF and any impurities was easily discernible by visual inspection and could be measured accurately when radioiodine was used in the synthesis.

2. Dissemination of DIF. A pneumatic nozzle, essentially identical to those described elsewhere (4, 8, 13) was assembled and connected to a tracer reservoir and a source of compressed air, as shown in Figures 1 and 2. Tests using this apparatus were performed using either fluorescein or DIF dissolved in denatured ethyl alcohol or a mixture of that solvent and acetone. In these tests, the apparatus was operated at an air pressure of 80 psig and with an air flow rate of approximately 150 scfm. The dye solution was disseminated in a series of 20-second bursts at rates of from 0.03 g to 0.2 g dye per second. The aerosols produced were collected on a millipore filter as close to the disseminator as possible (~2 meters) without interference from unevaporated solvents. The masses of the collected samples ranged from 4 mg to 12 mg.

Microscopic examination of the dry particulates indicated that a useful size range was easily obtained. The particles were generally spherical, and the diameter ranged from a maximum of approximately 7-10 μ to sizes below the resolution of the microscope (a few tenths of a micron). Subsequently, samples were submitted to Particle Data Laboratories of Elmhurst, Illinois, for more quantitative particle-size analysis. The results of these analyses showed acceptable particle-size distributions for various conditions of dissemination. Apparently normal size distributions were observed. Complete elucidation of the dissemination parameters which define the sizes of particulates has not been obtained. However, the results indicate that the reproducibility with which the aerosol is produced under various dissemination conditions is satisfactory.

Under a variety of weather conditions (clear, overcast, windy), temperatures (11°C - 31°C), tracer solution concentrations (2.9 mg/ml to 23.6 mg/ml), and using fluorescein or DIF, mean-weight diameters were obtained ranging from ~0.2 μ to 2.3 μ. Appendix III describes the experimental conditions and presents the data for the respective experiments.

3. Chemical Stability of DIF. As a check on the long term chemical stability of DIF, a small quantity of material labeled with I-125 was set aside for a period of ~6 months. Comparison of paper chromatographs taken before and after the storage period revealed no observable difference. Since no significant chemical decomposition had occurred, the stability of this material was judged to be satisfactory.

4. Sampling of DIF Aerosols in the Field. Because of its chemical inertness, Gelman Type E glass fiber filter material was selected as the sampling medium for the aerosol tracer. This material is reported to have an efficiency of 99.8% for particles larger than 0.2μ in diameter and greater than 98% for particles as small as 0.05μ. Pairs of these filters were tested in a Staplex Hi-Vol air sampler two meters from the aerosol generator. No visible deposit appeared on the second (downstream) filter.

The contribution of these filters to any I-129 background concentration was determined by subjecting blank filters to the complete analytical procedure described below. The results of measurements on three four-inch diameter filters were $(8 \pm 6)\ 10^{-12}$ g, $(9 \pm 5)\ 10^{-12}$ g, and $(8 \pm 1)\ 10^{-12}$ g of I-129 per filter. This level of I-129 was judged satisfactory.

5. Analytical Method. A general method for determining I-129 by neutron-activation analysis had been developed previously at NSEC and refined so that levels approaching $\sim 6 \times 10^{-14}$ gram per sample had been measured (14, 15). A review of this general method for applications when very high sensitivity is required is presented in Appendix IV.

The procedures for post-collection treatment of DIF samples and for determination of their I-129 content must provide for quantitative recovery of the DIF from the collection device, pre-irradiation chemical treat-

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ment and post-irradiation radiochemical purification. A rigorous pre-irradiation procedure was developed which would be appropriate for samples requiring analytical sensitivities approaching the limits of the method. In summary, this method involves recovery of the I-129 labeled DIF from the glass-fiber filter by elution with methanol, addition of unlabeled DIF as a carrier, purification of the DIF by solvent extraction, decomposition of the organic material with nitric acid, purification and recovery of the iodine fraction with a series of extractions and encapsulation of the sample for irradiation. After irradiation, radiochemical purification involves repeated extraction cycles and recovery of the iodine fraction in a form suitable for chemical yield determinations and radioactivity. measurements. This procedure is presented in detail in Appendix V.

A simplified variant of this procedure, also presented in detail in Appendix V, proved to be satisfactory for samples collected at short-ranges (generally up to ~3200 meters). The simplified procedure is normally adequate for samples containing more than 10^{-9} gram I-129.

In the development of these procedures, I-125 was used as a tracer to check for completeness of recovery of the iodine fractions. Thus, for example, it was found that quantities of DIF (labeled with I-125) of less than 1 mg were recovered quantitatively from filters with three successive 5-ml methanol washes. (Four portions of ~25 ml each are used for the final procedure). Trial analyses involving the entire preparation and purification procedure from filter wash to final precipitation of silver iodide gave material balances in excess of 95% after making the appropriate corrections for chemical yields.

B. The Gaseous Tracer

1. Preparation of Labeled Trifluoromethyl Iodide (CF_3I)

Two practical methods appear to be available for this preparation, direct synthesis and labeling by exchange. The initial effort was directed to the use of the Hunsdieckers reaction (9) for direct synthesis. This reaction is shown in equation II-1.

$$CF_3COOAg + I_2 \longrightarrow CF_3I + CO_2 + AgI \qquad (II-1)$$

Although this method utilizes only 50 percent of the labeled iodine, the unused iodine is recoverable. However, poor yields (less than ~16%) were obtained when the reaction system was scaled down in size from those described in the literature. Therefore, refinement was deferred pending investigation of an alternative procedure involving the exchange reaction shown in equation II-2.

$$CF_3I + I_2^* \rightleftarrows CF_3I^* + I_2 \qquad (II-2)$$

In this equation, the asterisks denote the tracer iodine.

Attempts were made, using I-125 as a tracer, to label CF_3I by exchange under a variety of experimental conditions. These attempts included direct reaction of the two compounds, reaction in the presence of various solvents and/or at various temperatures, and in vessels made of a variety of materials (including steel, aluminum, quartz, and Pyrex). It was found that the reaction cannot be conducted in a metallic vessel because chemical reaction between the metal and the labeled iodine leads to substantial loss of the tracer and inadequate labeling of the CF_3I.

One set of experimental conditions was devised which consistently resulted in nearly complete exchange using I-125. This procedure requires heating a mixture of labeled elemental iodine and unlabled CF_3I at 150 to 200°C in a quartz or Pyrex vessel for approximately one hour. The two components are then easily separated by distilling the CF_3I (B.P. = -22°C) away from the iodine. The detailed procedure is given in Appendix II.

This procedure does not permit complete utilization of the tracer in a single reaction. However, the quantity of remaining material

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can be minimized. Also, the residual tracer is present as elemental iodine and is available for immediate reuse. For example, the equilibrium ratio of tracer in each chemical form is equal to the molar ratio of all the iodine in each chemical form. Thus, if the initial molar ratio of the iodine in CF_3I to that in I_2 is 9 to 1, then 90% of the tracer will be in the chemical form CF_3I after the first exchange reaction. If, after separation of the constitutents, the residual labeled iodine is exposed to a second identical batch of CF_3I, only 1% of the original tracer would remain in the form of elemental iodine. Clearly, the specific activity of the second batch of labeled material would be reduced. However, for the present application, dilution of labeled CF_3I with ordinary CF_3I represents no serious disadvantage.

This exchange procedure was used successfully to prepare approximately 15-20 batches of I-125-labeled CF_3I. The labeled material was used in experiments for developing and testing CF_3I sampling devices and procedures and procedures for recovering and separating the iodine-labeled material in a form suitable for I-129 analysis. However, difficulties were encountered during the preparation of I-129-labeled CF_3I for the field tests (see below) which led to significant losses of I-129. In this connection, the direct synthesis method was re-examined.

Consultation with a manufacturer of (unlabeled) CF_3I led to the successful production of labeled CF_3I with useful yields. The Hunsdiecker reaction was used but with extra precautions taken during the preparation of the raw materials to ensure complete drying of the materials. Details of the procedure for labeling CF_3I by direct synthesis are given in Appendix I.

2. Dissemination of CF_3I Tracer. The controlled dissemination of a gas can be accomplished by several alternative methods. A frequently used procedure involves containment of the gas under pressure and its release into the atmosphere at a preselected, controlled rate. For this application, this method would require dilution of a small quantity of labeled

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CF_3I (~1.5 grams or less) with a much larger amount of ordinary CF_3I to be able to control and measure the flow rate of the tracer. The expense of this extra quantity of CF_3I suggests that an inert carrier gas, such as nitrogen, be used instead. However, because of the significant difference in vapor pressures, the CF_3I might be liquified at useful nitrogen pressures. Thus, uniform mixing of the CF_3I and the carrier gas might not be controllable.

The method selected involves storing the labeled CF_3I in metal cylinders of known volume at a pressure slightly less than one atmosphere. (See Figure 3). The pressure is adjusted for the exact barometric and temperature conditions immediately prior to dissemination by allowing water to enter the lower end of the cylinder until pressure equalization is obtained. A new gas volume is calculated by noting the quantity of water entering the cylinder. Dissemination takes place by pumping water into the lower end of the cylinder (both valves open) at a known rate adjusted to fill the remaining gas space in the cylinder in the desired dissemination period. (A suitable pump for this purpose is a solution metering pump such as the Beckman Model D which has a continuously adjustable range of flow up to 20 ml/min). The gas is displaced and escapes through a nozzle with a ~0.1-mm diameter orifice. This small orifice minimizes the effect of inadvertent back-diffusion of air into the cylinder.

The nozzle is equipped with a "T" connection so that a sample can be withdrawn into a previously evacuated vessel during the dissemination operation. In practice, the volumes of the metal cylinders used for the field tests described below are either ~25 ml or ~500 ml for normal releases of 0.1 g and 1 g of I-129, respectively. The volumes of the evacuated sampling devices are only ~0.01 ml. The volumes of the piping, valves, and nozzle are kept under 2 ml and are measured. Correction must be made in computation of the dissemination to take into account the fact that these spaces are filled with air initially. The dissemination is terminated when water is expelled from the nozzle. The elapsed time for dissemination is measured carefully.

3. <u>Collection of CF_3I Samples in the Field.</u> The fact that CF_3I is chemically inert is a desireable attribute for its use as an atmospheric tracer. The property, however, makes the design of a quantitative sampler rather difficult, especially if the collection must be made from a stream of high-velocity air for a significant period of time. To optimize the experimental sensitivity of the tracer method, it is necessary to sample a large volume of air. Therefore, it was apparent that a collector based on sorption of the CF_3I on an appropriate substrate was the only practical approach.

A number of potential collection methods were evaluated under a variety of operating conditions in laboratory-scale experiments using the simulated sampling system shown in Figure 4. The experimental system consisted of an air blower (Gelman Hurricane) operating at ~15 cfm with various filters or adsorbent beds (usually about 3-4 inches in diameter) installed at the intake of the blower. The intake of the system was open to the atmosphere and was fitted with an injection device for addition of I-125-labeled CF_3I to the air stream. This device consisted of a 3-in section of tubing with a glass-frit inlet mounted in the center. The tracer gas was injected with a Toepler pump. Injection normally was completed in about five minutes.

The tracer gas consisted of a known quantity (~1 gram) of I-125-labeled CF_3I diluted with approximately 10 volumes of air. The tracer was further diluted with the air flow (~100 ft^3) entering the sampler during the normal injection time. At the conclusion of the injection operation, the various filters or adsorbent beds were removed from the system and their I-125 contents were determined. These trial collectors were reinserted in the experimental system which was then operated for varying periods of time. The degree of retention of the CF_3I was determined by periodic measurements of the I-125 in the collectors. The more promising collectors were operated under flow rates typifying field conditions for

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periods equal to or exceeding the maximum requirement for sampling periods in the field.

The types of adsorbent materials tested included (1) activated charcoal of various particle sizes, (2) activated charcoal previously treated and/or continually treated with trimethylamine, (3) filters and columns loaded with various oils having high vapor pressure, (4) various chemicals reported to react with CF_3I such as phosphorous, mercury compounds, and arsenic, (5) silica gel, (6) paraffin chips, and (7) combinations of some of these materials. After screening tests for most of these materials, it became obvious that a good activated charcoal cartridge would prove to exhibit the optimum retention efficiency for CF_3I. However, the recovery of the CF_3I for subsequent I-129 analyses appeared to be troublesome. Also, gradual elution of the adsorbed CF_3I with continual air flow occurred with all types of charoal.

A reaction between CF_3I and $N(CH_3)_3$ (trimethylamine) was reported in the literature to yield a solid material. The possibility of "fixing" the CF_3I on the charcoal using this reaction was explored. In these experiments, the charcoal was carefully "activated" by heating in a vacuum and was then pre-treated with $N(CH_3)_3$. Figure 5 shows the results of a typical experiment in which two charcoal beds were operated in series. Their performance is shown separately and in sum. The performance was only slightly better than that obtained with activated charcoal not treated with $N(CH_3)_3$.

A search for the most efficient activated charcoal culminated in a special activated charcoal cartridge produced by Mine Safety Appliances, Inc. Ordinarily, this type of charcoal is doped with iodine for reaction with organic materials. Special charcoal which contained no iodine was prepared for this program. The charcoal was packaged in ~1-in long cartridges having diameters of ~4 inches.

A series of experiments was begun to demonstrate that the performance of these cartridges would be satisfactory under field test conditions. The test specifications required that no more than two of these cartridges operated in series would collect and retain at least 90% of the injected CF_3I for at least one hour at an air flow rate of 10-15 cfm. Two tests were made in which three cartridges were operated in series with a Staplex sampler identical to those to be used in the field tests. After addition in the standard manner of a known quantity of I-125-labeled CF_3I, air was drawn through the cartridges at a flow rate of 11 cfm for periods of one hour and 1.5 hours. The cartridges were disassembled and individually analyzed (by a method described below) for their I-125 content. The results showed that 98% of the injected CF_3I was retained in the first two cartridges after one hour and that 79% of the CF_3I was retained after 1.5 hours.

A third test was performed in which three such cartridges were operated for three hours after injection of the tracer gas in an air flow of 15 cfm. Measurements were made of the radioactivity content of each cartridge at 15 minute intervals throughout the test. Table II shows the results of this experiment. These data show that some loss of CF_3I occurs from the third cartridge after 75 minutes (or 1125 ft^3 air). However, these experiments indicate that adequate retention is obtained with two cartridges with elution volumes of approximately 700 ft^3 of air or for operation for approximately one hour at 10-12 cfm after an instantaneous injection of tracer. For a typical field experiment, the sampling device operates for approximately one hour and the tracer is collected over approximately one-half of the operating period. It is estimated that at least 95-97% of the tracer would be retained by these charcoal cartridges under such conditions.

4. Analytical Methods. The basic objectives of the analytical method is to remove CF_3I labeled with I-129 from the charcoal to convert the iodine in it to an inorganic form so that it could be subjected to the activation analysis procedure described in Appendix IV.

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Numerous attempts were made to remove adsorbed CF_3I quantitatively from charcoal by chemical methods. Elutions with water, various aqueous solutions, and organic solvents were unsuccessful, as was vacuum desorption, even at elevated temperatures. Therefore, a procedure based on complete combustion of the charcoal was attempted and was eventually adopted. Briefly, this procedure consists of the removal of the charcoal from the cartridges, the addition of a known amount of carrier in the form of unlabeled CF_3I, the combustion of the charcoal in an oxygen stream, and the collection, dissolution, and extraction of the resulting inorganic iodine. The detailed procedure is given in Appendix IV. A variety of carriers, including a solution of elemental iodine in an organic solvent, and liquid aliphatic and aromatic iodine compounds, were investigated. It was found that CF_3I is the only carrier material which gives a true indication of the chemical yield obtained during the combustion.

Satisfactory performance of the method was checked by analyzing charcoal cartridges to which a small quantity of CF_3I labeled with I-125 had been added. In a series of four successive experiments, recoveries were obtained ranging from 93% to 108%.

C. Field Testing the Feasibility of the Method.

1. General Objectives. The objectives for the field tests (which took place in August, 1964 at the NRTS in Idaho), were (a) to determine the general feasibility of the method for long-range particulate and gaseous phase tracing using I-129 compounds, (b) to explore any differences between these methods and the method now in use for short ranges based upon measurement of the fluorescence of dyes (4, 8, 12), (c) to determine the practical sensitivity of the I-129-based method and hence establish the range over which it can be effectively employed, and (d) to determine what, if any, further development is required to perfect the I-129 tracer method. It was decided jointly by personnel of NSEC, DRD and the USWB that a useful program of field tests would include:

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a. DIF-I-129 to a range of 10 miles

b. A duplication of (a)

c. CF_3I - I-129 to a range of 10 miles

d. A duplication of (c)

e. Both tracers simultaneously to a range of 25-40 miles

f. A duplication of (e)

The results of the DIF tests would be reinforced by fluorescence measurements on portions of the same samples collected up to a distance of 3200 meters. Full meteorological instrumentation of the tests was to be provided by U. S. Weather Bureau personnel.

2. Preparations.

a. Synthesis of I-129 labeled DIF. The method given in Appendix I was used to synthesize DIF labeled with I-129. The product consisted of a stock solution containing the DIF dissolved in 400 ml of ethyl acetate. The concentration of the I-129 was 5.55 mg/ml for a total of 2.22 grams of I-129. Paper chromatographic analysis of a sample of the solution showed only small quantities of fluorescein and tri-iodofluorescein to be present.

b. Solution for dissemination of DIF. During the preparations for the field tests it was found by U. S. Weather Bureau personnel that the fluorescence of DIF was much lower than that of fluorescein (or uranine) and that the sensitivity of the fluorescence measurements for the short range samples would be inadequate. Since fluorescein and DIF are chemically and physically compatible, the original plans to include a large quantity of unlabeled DIF in the solution to be disseminated were altered to include the use of fluorescein instead. Several five gallon drums were prepared containing ethanol solutions of fluorescein at a concentrations of ~24 mg/ml. Immediately prior to a field test dissemination the appropriate (small) volume of DIF - I-129 solution was to be mixed into the appropriate (large) volume of fluorescein solution.

c. Synthesis of CF_3I - I-129. Following two failures during attempts to label CF_3I with I-129 by means of the exchange method, the direct synthesis method was perfected as described above (p. 11-13), and in Appendix II.

Appropriate quantities of the labeled CF_3I were then synthesized, purified, and loaded into soldered brass cylinders with needle valves on each end. The volumes of the cylinders were such that the total gas pressure within each cylinder was less than atomospheric. See page 14 for the description of the use of these cylinders for CF_3I dissemination. The total quantity of CF_3I - I-129 prepared and loaded into the cylinders represented ~2 g of I-129 distributed as shown in Table III.

d. Compatibility Tests. Because the proposed tests included two runs during which I-129 was to be released in both forms (DIF and CF_3I), two experiments were performed on a laboratory scale which successfully demonstrated that no cross-interference would occur.

In the first experiment, two filter discs of the type to be used for the aerosol sampling were mounted in series at the intake of a Staplex high volume sampler. The sampler was operated in close proximity (two meters) to the aerosol generator until a heavy deposit was obtained on the outer filter. No dye was detectable either on the rear surface of the outer filter or on the inner (second) filter.

A second experiment consisted of the addition of I-125 labeled CF_3I to the intake of an operating Staplex sampler through a pair of charcoal cartridges preceded by filters of the type to be used for DIF sampling (Figure 9). There was no detectable I-125 activity (less than 1% of the total) found in the primary filter or in a secondary charcoal impregnated filter.

e. Pre-Field Test Background Measurement. A group of six charcoal cartridges were used for the collection of background samples at the test site by U. S. Weather Bureau personnel some 45-60 days prior to the field tests. For three of these, charcoal impregnated filter paper was included in front of the cartridges to remove from the air stream any "fallout" I-129 present in its most commonly occurring forms, aerosols or elemental iodine. Due to the press of time and subsequent developments in the analysis of field test samples only two of these background cartridges were analyzed. The results for a sampling period of two hours were 1.1×10^{-9} g I-129 (without charcoal pre-filter, 2060 ft^3 of air), and 1.4×10^{-9} g I-129 (with charcoal pre-filter, 2300 ft^3 of air). Although these results were higher than expected, they represented a volume of air nearly four times larger than that to be encountered in the actual field tests, and it was expected that a background of $\sim 2\text{-}4 \times 10^{-10}$ g would not be a serious interference for most of the tests.

Background measurements for the aerosol tracer were made by using some filters of the type to be used for the field tests, to sample $\sim 1.2 \times 10^5$ ft^3 of air. These measurements were made approximately one year before the field tests. The results indicated that, for the expected exposure, a maximum contribution from the air of $\sim 1 \times 10^{-11}$ g of I-129 per filter was to be expected in addition to $\sim 1 \times 10^{-11}$ g from the filter material itself (p 10). This background was considered to be satisfactory.

3. The Field Test Site and Sample Collection Network

a. The Sample Collection Network. The sampling network, positioned for the prevailing winds of the area, consisted of a number of Staplex Hi-volume samplers distributed in three groups. The first group consisted of the U. S. Weather Bureau's permanent test grid which included 60 degree segments of arcs at distances of 100, 200, 400, 800, 1600 and 3200 meters from a central release point. Samplers in this grid could be remotely operated from a trailer located near the release point.

The second group of samplers was located along two roads, 11 and 16 kilometers downwind of the release point, roughly forming arcs comparable to those of the main grid. Their positions, shown in Figure 7 were determined by radar measurements on a transponder placed at each point in succession. This group of samples was powered by "portable" generators placed nearby for this purpose and required manual operation at each sampler location. The positions of the third group of samples, shown in Figure 8, were located at scattered points ~40 to 64 kilometers from the release point. These were remotely operated by telemetered signals from the Central Facilities Area.

In addition to these sampling stations, sample collection filters and cartridges were exposed to the oncoming air from a moving airplane circling at various distances during some of the test runs.

b. Supporting Meteorological Facilities. During each tracer release, and for sometime before and after, meteorological information at a number of positions on a 61 meter tower was continuously measured and recorded. At heights of 4, 16, 32, and 61 meters, bivanes measured the vertical and horizonal wind direction fluctuations. At heights of 1, 2, 4, 8, 16, 32 and 61 meters, temperatures were measured with thermocouples protected from direct radiation by Beckman and Whitely shields. At heights of 4,. 6, and 61 meters, Beckman and Whitely 3-cup anemometers were used to measure wind speeds. All data were recorded on a 10-channel digital system with a speed such that all inputs were sampled every 2 seconds. In addition, tetroons carrying transponders were released at various times and followed by radar during the tracer release and for an appropriate period afterwards.

4. The Field Tests

Only five of the proposed set of six tests were performed because unfavorable weather conditions prevailed for a period of time such that the test grids were not available for an additional test. The test which was eliminated was that of a duplicate, short-range, CF_3I release. Since the other tests were smoothly executed under favorable meteorological conditions, the cancellation of this test was not considered serious.

a. Test A. On August 13, 1964, a total of 12.6 liters of ethanol containing 111 mg of I-129 in the form of DIF and approximately 280 grams of fluorescein was disseminated from an aerosol generating nozzle (of the type described above) over a period of 29.5 minutes, beginning at 1202 hours. A total of 147 samplers fitted with fiber glass filters were distributed in arcs at 100, 200, 400, 800, 1600, 3200, 11,000, and 16,000 meters.

b. Test B. On August 14, 1964, a total of 11.9 liters of ethanol containing 104 mg of I-129 in the form of DIF and approximately 270 grams of fluorescein was disseminated from the aerosol generator over a period of 30.0 minutes beginning at 1258 hours. The sampling station distribution was essentially the same as for Test A.

c. Test C. On August 17, 1964, a total of 0.085 g of I-129 in the form of CF_3I gas was disseminated at a uniform rate in a manner described above, over a period of approximately 32 minutes, beginning at 1315 hours. A total of 106 samplers fitted with double charcoal cartridges preceded by a charcoal impregnated fiberglass filter was distributed approximately as for Test A. The exceptions were that the 100 and 200 meter arcs were eliminated as were also a few samplers on the edges of the grid at the 400, 800, 1600, and 3200 meter arcs.

d. Test D. On August 24, 1964, a total of 12.9 liters of ethanol containing 1.08 grams of I-129 in the form of DIF and approximately 280 grams of fluorescein was disseminated as in Tests A and B over a period of 30 minutes, beginning at 1455 hours. Concurrently, a total of 1.07 grams of I-129 in the form of CF_3I gas was disseminated at a uniform rate as in Test C. The sampling network consisted of 109 samplers equipped with fiber glass filters in addition to double charcoal cartridges preceded

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by a charcoal impregnated fiberglass filter. This arrangement is shown in Figure 9. These samplers were distributed over distances from 400 meters to ~64 kilometers. In addition, five samples were collected by exposing a similar filter arrangement to the slipstream of an airplane circling at discreet distances ranging from 1600 meters to ~40 kilometers.

e. Test E. On August 25, 1964, the same arrangement was used for sampling and for dissemination as for Test D, with the following quantities of material released: 0.87 g I-129 in the form of DIF, ~280 grams of Fluorescein, and 0.71 g I-129 in the form of CF_3I gas. The release took place over a period of 29.5 minutes, beginning at 1342 hours.

f. Grass Samples. Samples of grass were exposed to the plume during Test D and background samples of grass were also provided.

TABLE II

RETENTION OF CF_3I ON CHARCOAL CARTRIDGES

Elution Time (Hrs)*	Distribution (Percent)			Total Retention (Percent)
	#1	#2	#3	
0	100	--	--	100
0.25	90	10	--	100
0.50	72	26	2	100
0.75	49	42	9	100
1.00	38	46	16	100
1.25	27	46	22	95
1.50	19	46	25	90
1.75	15	39	30	84
2.00	10	34	34	78

*** - Air Flow Rate = 15 cfm**

TABLE III

I-129 - CF_3I PREPARED FOR FIELD TESTS

Cylinder No.	I-129 CF_3I (g)	Volume (ml)	Pressure at 30°C (mm Hg)
1	0.0905	23	625
2	0.085	23	633
3	0.087	24	624
4	1.07	514	359
5	0.71	514	239

III. RESULTS AND DISCUSSION

A. General

Tables IV through VIII list the samples collected, originally scheduled for analysis, and analyzed for the three tests A, D, and E. During the course of the analyses it became apparent that the validity of the results of all the gaseous tracer studies was in jeopardy due to background considerations. Also, the accuracy of the results obtained at the more distant points during the particulate tracer studies was somewhat affected by similar considerations of a lower order of magnitude. The original program of analysis was then suspended in order to ascertain the causes of these problems. The results obtained, the problems encountered, and the progress made toward solving them are presented and discussed below.

B. Particulate Tracer Studies - Tests A, D, and E

Tables IX, X, and XI give the results obtained for the three tests. The data given include the available results of the fluorescence measurements as provided by U. S. Weather Bureau personnel. All the calculated data includes a value for the flow rate of the samplers as actually calibrated by U. S. Weather Bureau personnel, but in some cases, when these calibration data were not available, an average figure was used. The presentation is self-explanatory except for the last two columns. These data are given in terms of the mass of tracer collected by a sampler per cubic meter of sampled air divided by the total mass of tracer released. The sampling time is assumed to be identical to the release time.

The meteorological data furnished by U. S. Weather Bureau personnel for these three tests are given in Table XII. The tracing data for the three tests are also presented graphically in Figures 10, 11, and 12. For the I-129 tracer, individual values are plotted, while for the fluorescein tracer, only the maximum values are plotted for each sampling distance. The slopes of the curves in these figures are shown in Table XIII. A description of the method of calculating the predicted values shown in the figures follows.

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Islitzer (8) gives the equation for continuous point-source emanation:

$$\chi = \frac{Q}{2\pi \bar{U} \sigma_y \sigma_z} e^{-\left\{\frac{y^2}{2(\sigma_y)^2} + \frac{h^2}{2(\sigma_z)^2}\right\}} \qquad \text{(III-1)}$$

where χ is the concentration of tracer sampled at ground level (g/m^3), Q is the source strength (g/sec), $\bar{U}$ is the mean wind speed (m/sec), h is the height of the source, y and z are the crosswind and vertical coordinates (x would be the downwind coordinate), and σ_y and σ_z are standard particle deviations in the crosswind and vertical directions. These standard deviations are functions of downwind distance and of the standard deviations of the horizontal and vertical wind direction fluctuations (σ_θ and σ_ϕ, respectively) as

$$\sigma_y(x) = a\,\sigma_\theta\, x^q \qquad \text{(III-2)}$$

$$\sigma_z(x) = b\,\sigma_\phi\, x^p \qquad \text{(III-3)}$$

For normal conditions, a, b, and p are taken as unity, and the value of q is somewhat less than unity. Islitzer (8), however, has experimentally determined σ_y as a function of x. With equation (III-3) substituted in equation (III-1), for a source at ground level, the latter becomes:

$$\chi_{(y=0)} = \frac{Q}{2\pi \bar{U} \sigma_y \sigma_\phi x} \qquad \text{(III-4)}$$

where $\chi_{(y=0)}$ indicates that the sample collection is at the center of the plume. The predicted curves in Figures 10 through 12 are based on this equation using the meteorological parameters σ_ϕ and $\bar{U}$ as measured at ground level during the tests and σ_y as taken from Islitzer's results (8) with the necessary linear extrapolation for the distances beyond 2000 meters.

It is readily seen that the results of the fluorescense measurements do not agree with those of the I-129 measurements. The latter agree more closely with the predicted curves.

For rectilinear flow, p = q = 1, and the predicted slope for the plots shown would be 2.0, which presumably represents a maximum value (8). The slopes obtained from the I-129 data were 1.59, 1.52, and 1.92, with an average of ~1.7. The slopes obtained from the fluorescense data were 2.69, 2.32, and 2.14 with an average of ~2.4. The value of the predicted slope obtained using the methods described above was 1.85. In general, it would appear that there is either a preferential loss of fluorescent material or of the fluorescense of the material with distance. Although it is a priori impossible for the former to occur without a corresponding loss of I-129 content, a loss of fluorescense is conceivable. An equivalent order of magnitude in the degradation of the fluorescent properties of uranine (sodium fluorescein) has not been observed in previous tests (8). However, the use of fluorescein itself in an organic solution may result in an aerosol having different properties and having an increased sensitivity to sunlight. The mean particle size obtained in pre-field test experimentation was generally smaller than that reported for aerosols produced using aqueous solutions or uranine. The smaller size may enhance the known sensitivity of the dye to degradation by exposure to sunlight. Furthermore, the sodium salt could conceivably be more stable than the acid form. Such differences in stability could, of course, be investigated in the future.

The "crossover" of the fluorescein and I-129 concentration curves, as shown in Figure 10, remains unexplained. It is difficult to conceive of a mechanism by which the concentration of the fluorescent tracer is higher than the I-129 concentration at close range and becomes lower at greater distances.

Some of the I-129 concentration data is adversely affected by background problems. This is most easily observed in the results for Test A, when only 0.1 g of I-129 was released. For the 1600 and 3200-meter samples, blank sample values for background concentrations exceeded 50 percent of the gross values for the experimental samples. The results obtained for samples collected at 11,000 and 16,000 meters in Test A are somewhat better because the rigorous method of analysis was used resulting in lower blank values. The general problem of background limitations, although still present, was less acute for Tests D and E because of the greater quantity of I-129 released in these tests. Thus, for the samples collected at 40 to 65 kilometers, the results shown represented approximately two or three times the background value.

C. Gaseous Tracer Studies

The results of the analyses of the charcoal cartridges used in the field tests were completely ambiguous. Random I-129 concentrations ranging from $\sim 4 \times 10^{-9}$ to $\sim 8 \times 10^{-8}$ grams per sample were obtained. Blank samples exhibited I-129 contents of comparable orders of magnitude. The gas samples collected at 400 meters during Test D were the only ones definitely showing excesses over the range of blank values encountered. A listing of values obtained for samples and blanks is shown in Table XIV. The results definitely indicated that there was a contamination problem associated with these samples. The balance of the experimental effort was immediately replanned in an effort to ascertain the source or sources of the problem.

D. Background and Contamination Problems

A review of the data obtained indicates that there are essentially two problems, a high-level background or contamination problem which seriously jeopardizes the results of all of the gaseous tracing tests, and a lower-level background problem, which affects the accuracy of the results from the long-range particulate studies. Since the latter did not interfere significantly with the achievement of the objectives of the experiments, full attention has been given to the high-level background concentrations in the gas samples.

As a first step, the history of the charcoal cartridges, from manufacture to analysis, was reviewed. The following facts were ascertained.

The source of supply of the activated charcoal cartridges used in the field tests was Mine Safety Appliances (MSA). They had also supplied a small quantity of similar cartridges during the preliminary experimental program (used, for example, to ascertain background values) prior to the field tests. MSA, in turn, obtains raw material, coconut charcoal, from a bulk supplier. The detailed procedure used by MSA to process the raw material is a trade secret. Details in the general handling of the raw material have not been determinable.

A large (3000 cartridge) shipment was made from MSA in early July, 1964. The shipping cartons, each containing 36 cartridges individually wrapped in heat-sealed plastic bags, were shipped by NSEC directly to the field test site in Idaho shortly thereafter. A few days before the tests, the required number of cartridges were removed from their bags, taped together in pairs, and immediately resealed in new plastic bags. The paired cartridges were removed from these bags only for loading into the air-sampling devices immediately prior to each test. Following collection they were again resealed in new plastic bags as quickly as possible. Several boxes of cartridges remained unopened or remained unused after opening, pairing, and resealing. Following return of the samples to the NSEC laboratory, the samples were kept sealed until immediately before analysis. Samples collected at long range (11,000 meters or more) were opened and analyzed in a building separate from that used for analysis of the short-range (3200 meters or less) samples. The unused cartridges and unopened packages were retained in a separate storeroom building.

A series of analyses was performed to ascertain whether the high levels of I-129 encountered could have been the results of contamination introduced in the NSEC laboratories. Samples of an iodine compound known to have a low I-129 concentration and various samples of the CF_3I and DIF carriers used in the analyses were processed. Only low levels of I-129 (10^{-10} to 10^{-12} g of I-129 per sample or less) were observed. Since these samples were processed in the same laboratory as the experimental samples, it was tentatively decided that laboratory reagents and apparatus were not responsible for the high-level contamination problem. It was then postulated that the I-129 observed in the field test samples was a contaminant of the charcoal as received by NSEC.

Several of the cartridges from the unopened boxes and from those not used at the field tests were then analyzed for I-129 concurrently with some new samples of charcoal obtained from Fisher Scientific Co. and from MSA. Several samples of the iodine compound with very low I-129 content served as blanks. The material from Fisher Scientific Co. included activated coconut charcoal of various mesh sizes (6-14 and 50-200) including one sample which had been at NSEC for ~5 years. The new material from MSA included charcoal of the same type as that used in the cartridges as well as a different type of coconut charcoal (8-14 mesh) and a 10-20 mesh coal-base charcoal.

The results of these analyses are shown in Table XV. All of the cartridges, whether opened or unopened, exhibited an I-129 content of between 1.1 and 5.7×10^{-8} grams; levels similar to those found in the cartridges used during the field tests. The new samples of charcoal, regardless of source, contained approximately two orders of magnitude less I-129 The blanks showed considerably less I-129, but the values obtained were more than an order of magnitude above typical results obtained previously. These values may be due to cross-contamination of these samples. arising from the simultaneous processing of the cartridges which contained at least 100,000 times more I-129.

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These results are convincing proof that the charcoal was contaminated prior to its use. This conclusion immediately implies that the field tests of the gaseous tracer yielded no directly useful experimental data.

TABLE IV

TEST A - SAMPLING SCHEDULE

Distance (meters)	Number of Samples Collected	Originally Scheduled for Analysis	Analyzed
100	10	4	4
200	10	4	4
400	21	5	5
800	21	5	5
1600	31	5	5
3200	25	5	5
11000	15	12	10
16000	15	12	6

TABLE V

TEST D - SAMPLING SCHEDULE

(Particulate Tracer)

Distance (meters)	Number of Samples Collected	Originally Scheduled for Analysis	Analyzed
400	5	5	5
800	18	5	5
1600	27	5	5
3200	21	5	5
11000	14	13	--
16000	15	10	--
40000-64000	8	7	7
Airplane	5	--	--

TABLE VI

TEST E - SAMPLING SCHEDULE
(Particulate Tracer)

Distance (Meters)	Number of Samples		
	Collected	Originally Scheduled for Analysis	Analyzed
400	5	5	5
800	17	5	5
1600	27	5	5
3200	21	5	5
11000	15	12	--
16000	15	12	--
40000-64000	7	7	--
Airplane	5	--	--

TABLE VII

TEST D - SAMPLING SCHEDULE
(Gaseous Tracer)

Distance (Meters)	Number of Samples		
	Collected	Originally Scheduled for Analysis	Analyzed
400	5	5	5
800	18	5	5
1600	27	5	5
3200	21	5	5
11000	14	13	3
16000	15	10	4
40000-64000	8	7	6
Airplane	5	--	--

TABLE VIII

TEST E - SAMPLING SCHEDULE
(Gaseous Tracer)

Distance (Meters)	Number of Samples		
	Collected	Originally Scheduled for Analysis	Analyzed
400	5	5	4
800	17	5	5
1600	27	5	2
3200	21	5	5
11000	15	12	--
16000	15	12	--
40000-64000	7	7	--
Airplane	5	--	--

TABLE IX

RESULTS OF TEST A (PARTICULATE TRACER STUDY)

Sample Distance (meters)	Sample (a) Position	$\frac{\text{I-129 Collected}}{\text{I-129 Released}}$ $\frac{g/m^3}{g} \times 10^{10}$	$\frac{\text{Fluorescein Collected}}{\text{Fluorescein Released}}$ $\frac{g/m^3}{g} \times 10^{10}$
100	2	160	390
100	3	230	850
100	4	600	1900
100	7	310	630
200	4	140	180
200	5	150	310
200	6	150	430
200	7	100	200
400	8	53	45
400	9	60	58
400	10	53	45
400	11	57	38
400	12	47	35
800	7	32	9.2
800	8	22	9.3
800	9	37	7.2
800	10	22	13
800	11	15	8.4
1600	11	10	1.0
1600	12	11	1.5
1600	13	24	1.8
1600	14	11	1.3
1600	15	16	0.9
3200	12	9.3	0.2
3200	13	31	---
3200	14	13	0.2
3200	15	28	---
11000	5	1.1	---
11000	6	0.83	---
11000	7	0.7	---

(Continued)

TABLE IX (Continued

RESULTS OF TEST A (PARTICULATE TRACER STUDY

Sample Distance (meters)	Sample (a) Position	I-129 Collected / I-129 Released $\left\{\frac{g/m^3}{g} \times 10^{10}\right\}$	Fluorescein Collected / Fluorescein Released $\left\{\frac{g/m^3}{g} \times 10^{10}\right\}$
11000	8	0.5	---
11000	9	0.8	---
11000	10	0.3	---
11000	11	0.3	---
11000	12	0.5	---
11000	14	1.0	---
11000	15	1.2	---
16000	6	0.3	---
16000	7	0.3	---
16000	8	0.03	---
16000	9	0.3	
16000	10	0.03	
16000	11	0.1	

(a) See Figures 6 and 7

TABLE X

RESULTS OF TEST D (PARTICULATE TRACER STUDY)

Sample Distance (meters)	Sample (a) Position	I-129 Collected / I-129 Released $\left\{\frac{g/m^3}{g} \times 10^{10}\right\}$	Fluorescein Collected / Fluorescein Released $\left\{\frac{g/m^3}{g} \times 10^{10}\right\}$
400	3	43	10
400	5	66	44
400	7	58	26
400	9	61	7.6
400	11	42	3.0
800	2	38	4.4
800	3	35	7.1
800	4	39	6.6
800	5	33	6.9
800	6	28	6.6
1600	3	7.9	1.7
1600	4	3.1	1.6
1600	5	7.2	2.2
1600	6	9.1	1.7
1600	7	12	1.2
3200	3	6.1	0.34
3200	4	3.1	0.33
3200	5	3.9	0.34
3200	6	0	0.35
3200	7	2.1	0.28
40000	1	0.039	---
40000	5	0.036	---
47000	8	0.044	---
48000	13	0.027	---
48000	19	0.029	---
64000	W-1	0.033	---
53000	W-2	0.045	---

(a) - See Figures 6 and 8

TABLE XI

RESULTS OF TEST E (PARTICULATE TRACER STUDY)

Sample Distance (meters)	Sample (a) Position	I-129 Collected / I-129 Released $\left\{\frac{g/m^3}{g} \times 10^{10}\right\}$	Fluorescein Collected / Fluorescein Released $\left\{\frac{g/m^3}{g} \times 10^{10}\right\}$
400	3	33	1.7
400	5	67	25
400	7	140	21
400	9	36	48
400	11	69	41
800	9	90	11
800	10	59	13
800	11	50	11
800	14	48	16
800	15	72	9.3
1600	15	12	2.0
1600	16	7.2	2.4
1600	17	7.8	1.3
1600	20	11	3.1
1600	21	11	2.1
3200	16	0.26	0.41
3200	17	2.3	0.48
3200	18	1.6	0.44
3200	19	3.1	0.51
3200	20	3.8	0.55

(a) See Figure 6

TABLE XII

METEOROLOGICAL PARAMETERS

	Height (meters)	Average Temperature (°C)	σ_θ (degrees)	σ_ϕ (degrees)	Wind Speed (mps)	Wind Direction (degrees)
Test A	4	25.6	18.0	5.4	6.0	238
	16	24.4	---	6.2	7.2	---
	32	23.9	12.9	7.6	---	231
	61	24.8	11.9	8.3	8.2	229
Test D	4	29.1	19.2	5.8	4.8	214
	16	28.0	---	7.0	5.8	211
	32	27.5	12.5	7.8	---	---
	61	---	14.1	9.3	6.5	210
Test E	4	29.2	14.0	4.0	9.0	232
	16	27.8	---	4.4	11.1	---
	32	27.2	11.9	4.5	---	231
	61	---	13.9	4.7	12.6	231

σ_θ - Horizontal wind direction, standard deviation

σ_ϕ - Vertical wind direction, standard deviation

TABLE XIII

SLOPES OF LINES OF FIGURES 10, 11, 12

Test	Slope		
	I-129	Fluorescein	Predicted
A (Figure 10)	1.59	2.82	1.85
D (Figure 11)	1.52	2.32	1.85
E (Figure 12)	1.92	2.14	1.85
Average	~1.7	~2.4	1.85

TABLE XIV

RESULTS OF GASEOUS SAMPLE AND BLANK ANALYSES

Sample Identity		I-129 Content* (grams x 10^{10})
Test D	400 meters	300
	400	400
	400	990
	400	840
	400	430
	800 meters	120
	800	130
	800	290
	800	200
	800	120
	1600 meters	200
	1600	140
	1600	240
	1600	70
	3200 meters	100
	3200	200
	3200	300
	3200	200
	3200	220
	11,000 meters	340
	11,000	67
	11,000	100
	16,000 meters	85
	16,000	50
	16,000	140
	40,000 meters	1200
	40,000	170
	43,000	100
	47,000	53
	48,000	230
	48,000	240

Sample Identity		I-129 Content* (grams x 10^{10})
Test E	400 meters	140
	400	270
	400	160
	400	200
	800 meters	100
	800	310
	800	420
	800	86
	800	96
	1600 meters	160
	1600	81
	3200 meters	170
	3200	63
	3200	160
	3200	78
	3200	38
Blanks		260
		140
		490
		470
		450
		130
		150

* - Blank values not subtracted

TABLE XV

I-129 CONTENT OF VARIOUS CHARCOAL SAMPLES

Type of Sample	I-129 Content (a) (g I-129/sample x 10^{10})
Unopened Cartridges	360, 350
	510
	570, 560
Cartridges Opened in Idaho, Taped and Resealed but Unused	320, 340
	440, 390
	110, 420
	330
	370, 130
	290
Activated Coconut Charcoal	
Fisher (6-14 mesh)	2.1, 2.1
Fisher (6-14 mesh, 5 yrs old)	8.3, 7.5
Fisher (50-200 mesh)	6.1, 3.8
MSA (Same material as cartridges)	1.5, 2.5
MSA (8-14 mesh)	<1, 1.9
Coal-base Charcoal	
MSA (10-20 mesh)	2.3, 1.7
Blanks (Analyzed simultaneously with various groups of samples)	<0.035, 0.032, 0.008

(a) Two numbers on a line are duplicates, for cartridges this means the samples are from the same box.

IV. CONCLUSION

A. General Practicability of the Method

1. Particulate Tracer. The experimental results indicate that the use of I-129 incorporated into an aerosol as an atmospheric tracer is definitely feasible. Apparently valid data were obtained to distances of 40-65 kilometers. Unfortunately, the lack of agreement between the fluorescence and the I-129 tracer methods at shorter distances was disappointing. The use of the combined methods is extremely desireable since it would significantly reduce the costs of long-range atmospheric diffusion studies by exploiting the advantages of the two methods. However, additional experimental work is required to resolve the discrepancies which were observed.

2. Gaseous Tracer. Because of the demonstrated contamination of the charcoal cartridges used for sampling the gaseous tracer, completely ambiguous results were obtained. Therefore, the feasibility of the method was not demonstrated. Potential approaches to resolution of this problem are discussed below. Assuming that I-129-free charcoal can be obtained, it is believed that results comparable to those achieved with the particulate tracer would be forthcoming.

B. Theoretical Sensitivity of the Method

The limit of detection of I-129 by current methods may be deduced from the following considerations. The counting equipment typically registers one count per minute for 6 x 10^{-13} gram of I-129 irradiated and processed in the standard manner. The normal background of the counter is approximately 0.02 count per minute. For practical counting periods, a count rate of approximately five times this background can be detected reliably. This corresponds to approximately 6 x 10^{-14} gram of I-129 per counting sample. Assuming a reasonable overall chemical yield (~50 percent), the result corresponds to the detection of ~10^{-13} gram of I-129 in a sample as collected during an actual atmospheric tracing study.

For particulate tracing, this level of I-129 corresponds roughly to a single particle of DIF with a diameter of ~0.5μ, or to an equivalent number of smaller particles. By comparison, some of the most sensitive methods currently in use are reported to have sensitivities of 10^{-10} gram (uranine dye)(4, 8, 13) or of a single, ~0.5μ diameter particle (zinc cadmium sulphide)(16). In the latter case, however, detectability vanishes abruptly with any further decrease in particle size, independent of mass, whereas with I-129, the sensitivity is a function only of the total mass collected. In practice, the sensitivities for all methods may be considerably modified by other factors, particularly background considerations.

For gaseous tracing (using CF_3I) the theoretical sensitivity is the sensitivity required to detect the I-129 at a distance of approximately 200 kilometers for a one-gram release assuming experimental conditions comparable to those encountered in the present work.

C. Practical Limitations of Sensitivity

1. Particulate Tracer. There are a number of apparent sources of I-129 which may contribute to the overall I-129 background for particulate tracer studies. The following list shows the magnitude of these contributions as determined experimentally during the course of this work.

a. I-129 content of the filter media used: ~10^{-11} g/filter

b. Background concentration of atmospheric I-129: $\leq 10^{-11}$ g/sample

c. Extraneous sources of I-129 (including cross contamination): varies from negligible to ~8×10^{-11} g/ sample

The above values may be expressed in terms of the equivalent range for a one-gram release (assuming typical sampling and meteorological conditions):

a. Filter content: ~50 Km

b. Atmospheric content: ~50 Km or more

c. Other: ~16 Km or more

d. Combined effects: ~16 Km to ~50 Km

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Examination of the quality of the results obtained for various releases and at various distances confirms this evaluation.

Although, in principle, it is possible to obtain data for at least modest distances beyond the equivalent of the background, the reliability of the results becomes unacceptable at these distances if the background concentrations are variable. Thus, [illegible] careful assessment and control of background concentrations, the present method would be useful over a distance of ~50 kilometers for a release of one-gram of I-129 in the form of a labeled aerosol.

2. Gaseous Tracer. In the absence of meaningful experimental results for the gas-tracer tests, quantitative evaluation of the use of I-129 labeled CF_3I cannot be made. The background concentrations of I-129 in the charcoal cartridges limited the useful range of these tests to ~500 meters. If this factor can be eliminated, it appears reasonable that the useful range of this tracer would be at least comparable to that of the particulate tracer, (~50 Km for a one-gram release). Indeed, it is generally accepted that the use of a gas is preferable for tracing atmospheric motions over long distances. Whether or not the CF_3I labeled with I-129 will prove practical for use over such distances depends primarily upon substantial reductions of the background contamination of the collection media.

D. Background Reduction

1. Particulate Tracer. Significant improvements in various contributions to the background can be made relative to those observed in the program to date. Reduction of the background contributions of the filter media may be obtained by the use of alternative filter materials. There are a number of varieties of filter media available which are capable of collecting sub-micron-size particles efficiently. These could be surveyed to determine if a very low and constant I-129 background level can be determined.

At present, it is difficult to evaluate the significance of atmospheric I-129 because its contribution was comparable to that of the filter itself. If this contribution should eventually prove to limit the sensitivity, a more sophisticated pre-irradiation treatment of the samples may be required.

The levels of extraneous background occasionally experienced in the present program probably resulted from concurrent processing of the charcoal cartridges, containing unexpectedly high quantities of I-129. Therefore, the observed contributions of I-129 from this source are atypical. The results of the analysis of the 40-65 Km samples of Test D demonstrated that extraneous contamination can be held to negligible values. Additional analyses of selected long-range particulate samples from Test E could be performed to confirm the quality of the analytical method.

2. Gaseous Tracer Method. Experiments performed under the present program demonstrated that, by screening the material to be incorporated in the cartridges, one may readily obtain concentrations of I-129 of $\sim 10^{-10}$ gram per cartridge. A more extensive survey of conventional sources of supply for activated coconut charcoal may lead to the further reduction of I-129 background levels required for long-range tracer studies. The cooperation of the manufacturer of the charcoal cartridges (Mine Safety Appliances Co.) would be enlisted to ensure that the quality of the material with respect to its sorptive properties would be adequate.

Although the mechanism by which I-129 is introduced into the charcoal is indeterminate, it probably involves exposure of the charcoal to "fallout". There may be some significance in the fact that the highly-contaminated charcoal used for the field tests was produced in the spring, whereas the charcoal obtained before and after this period contained lesser quantities. In any case, if one assumes that the I-129 is derived from fallout, it may be worthwhile to obtain raw material from the southern hemisphere where fallout levels are generally lower. This is particularly true because most of the atmospheric I-129 is derived from reactor fuel reprocessing plant effluents (17). These effluents are not expected to reach the southern hemisphere.

The effects of background levels can be partially eliminated by increasing the source term. The dissemination of quantities of I-129 significantly larger than that considered previously is discussed below.

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E. Increasing the Source Term

Both economics and safety are factors to be considered when contemplating an increase in the source term.

The current prices for I-129 are tied to a low production rate. The I-129 presently available is derived from the silver-coated Berl saddles used for removing fission product iodine from the effluent of large reactor-fuel reprocessing plants. The technology is not difficult. Therefore, the price should eventually become comparable (or even less than) that of other long-lived fission products. Cesium-137, for example, costs $67/gram in large quantities. Thus, it should become economically attractive to increase the quantity of I-129 released for an atmospheric tracer study beyond that used in the present work. A factor of ten increase in the source term is equivalent to a factor of approximately three increase in the distance over which meaningful results may be achieved.

As to the safety considerations in releasing I-129 to unrestricted areas, the maximum permissible concentration averaged over a period of one year is 3×10^{-11} μc/ml for soluble material. Assuming a wind velocity of 5 meters/second and conservatively small horizontal and vertical standard deviations in the wind direction, one may calculate that at a distance of 10 meters the maximum concentration of I-129 resulting from a one-gram release averaged over one year is only $\sim 7 \times 10^{-13}$ μc/ml. This implies that by merely restricting the access of personnel to a distance of 10 meters downwind of the release point one may perform experiments releasing up to a total of more than 30 grams of I-129 per year without exceeding the permissible concentration limits. An increase of the restricted distance to ~30 meters gives an additional safety factor of 10. Actually both CF_3I and DIF are water insoluble materials and an additional factor of 100 is allowed in the permissible maximum concentration. Furthermore, CF_3I as in inert insoluble gas, may be considered still less hazardous.

Thus it may be seen that the increase of the source term by an order of magnitude or more is feasible from both an economic and safety viewpoint.

F. Summary of Required Additional Effort

The major requirements for achieving full operation capability of the I-129-tracer method are:

a. Reduction of the I-129 background concentrations for the CF_3I tracer methodology.

b. Improvement in the compatibility of I-129-DIF tracer method with fluorescent-dye tracer methods.

c. Reduction of the I-129 background concentrations for the DIF tracer methodology.

The following refinements in the methodology would improve the economy and reliability of the method and confidence therein.

a. Simplification of I-129 analysis.

b. Improvement in control of aerosol particle size.

c. Investigations of CF_3I and DIF stability under simulated field conditions.

G. Potential Capability of the Method

The general principles of I-129 tracing, using either an aerosol or a gas, are sound. The DIF-I-129 method has been proven feasible. The feasibility of the CF_3I-I-129 method hinges principally on one remediable difficulty. If it is assumed that the background effects associated with the method can be reduced to the order of $\sim 10^{-12}$ gram of I-129 per sample, and that a significant reduction in the price will occur, then it will be technically and economically feasible to extend the method to very long ranges using 100-gram releases. Using dissemination and collection times equivalent to those observed during some of the field tests, calculations (using equation III-4) show that tracer material would be detectable over a range of approximately 800 kilometers or 500 miles. The prospects of this powerful method are intriguing indeed.

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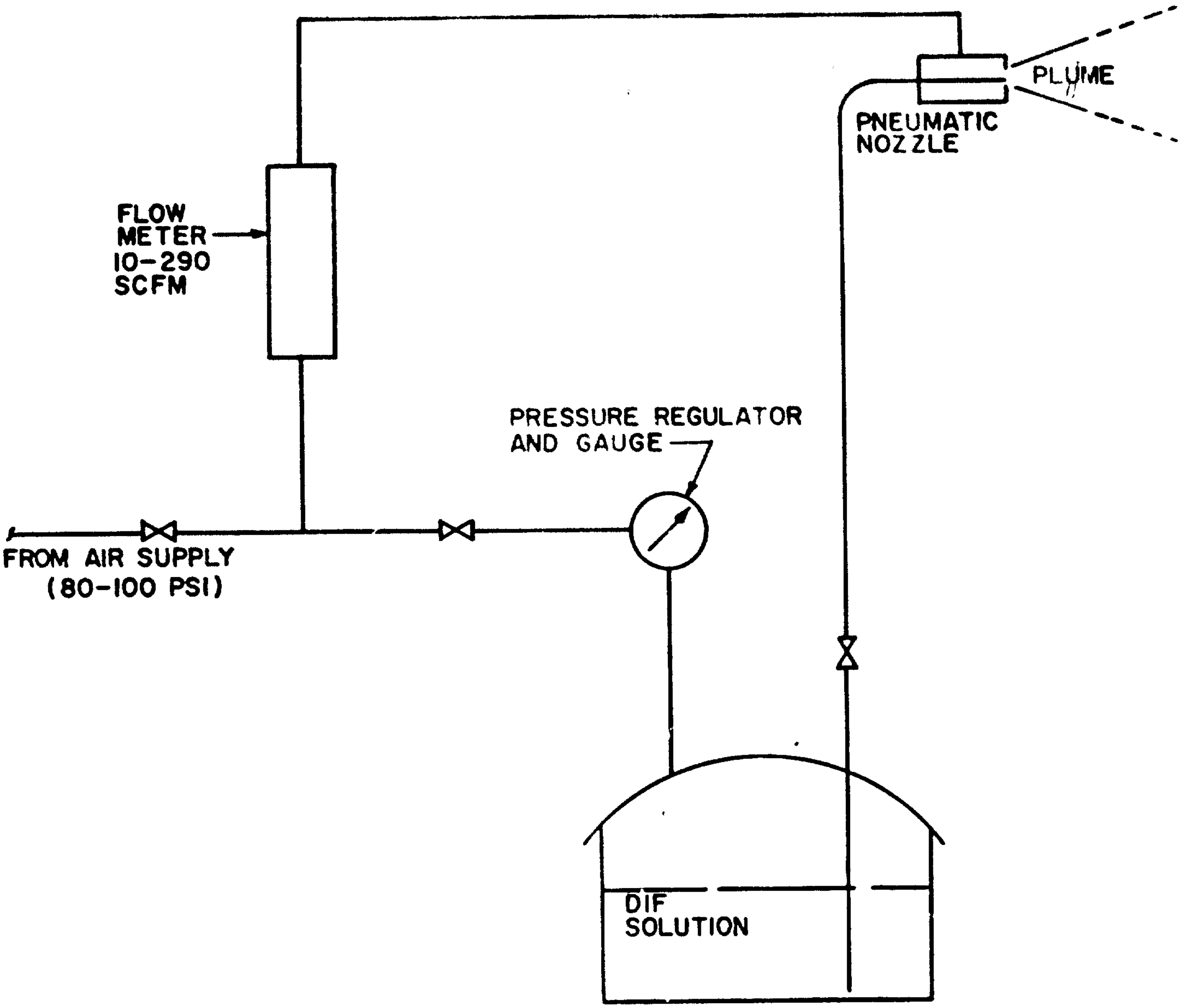
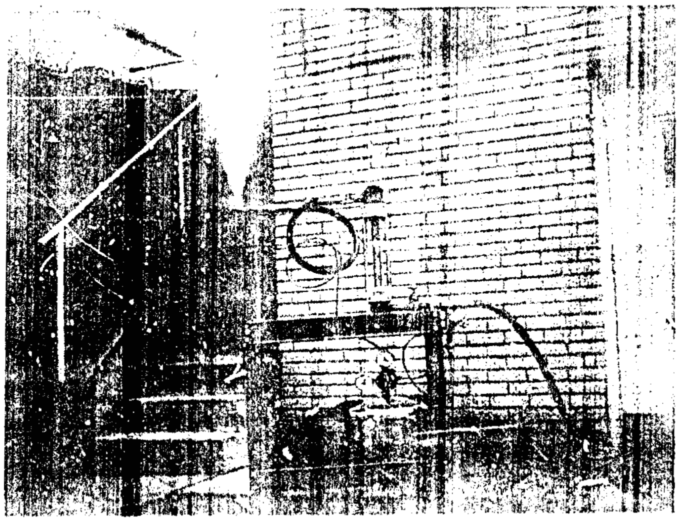


FIGURE 1

AEROSOL GENERATOR-SCHEMATIC



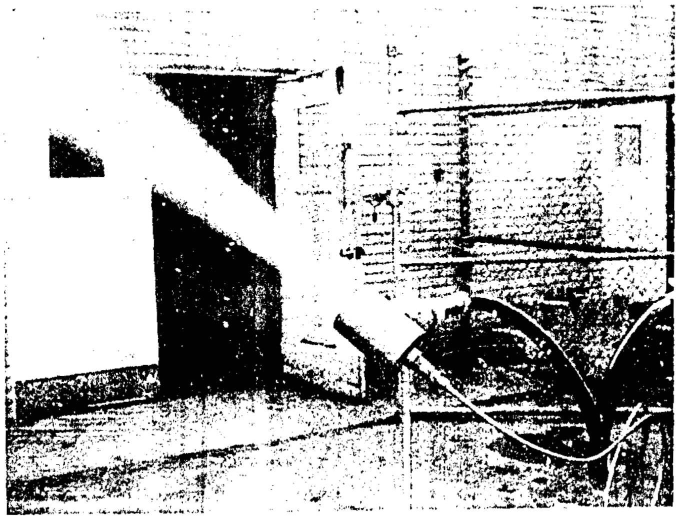


FIGURE 2

AEROSOL GENERATING EQUIPMENT

FIGURE 3

DISSEMINATION SYSTEM FOR CF_3I TRACER

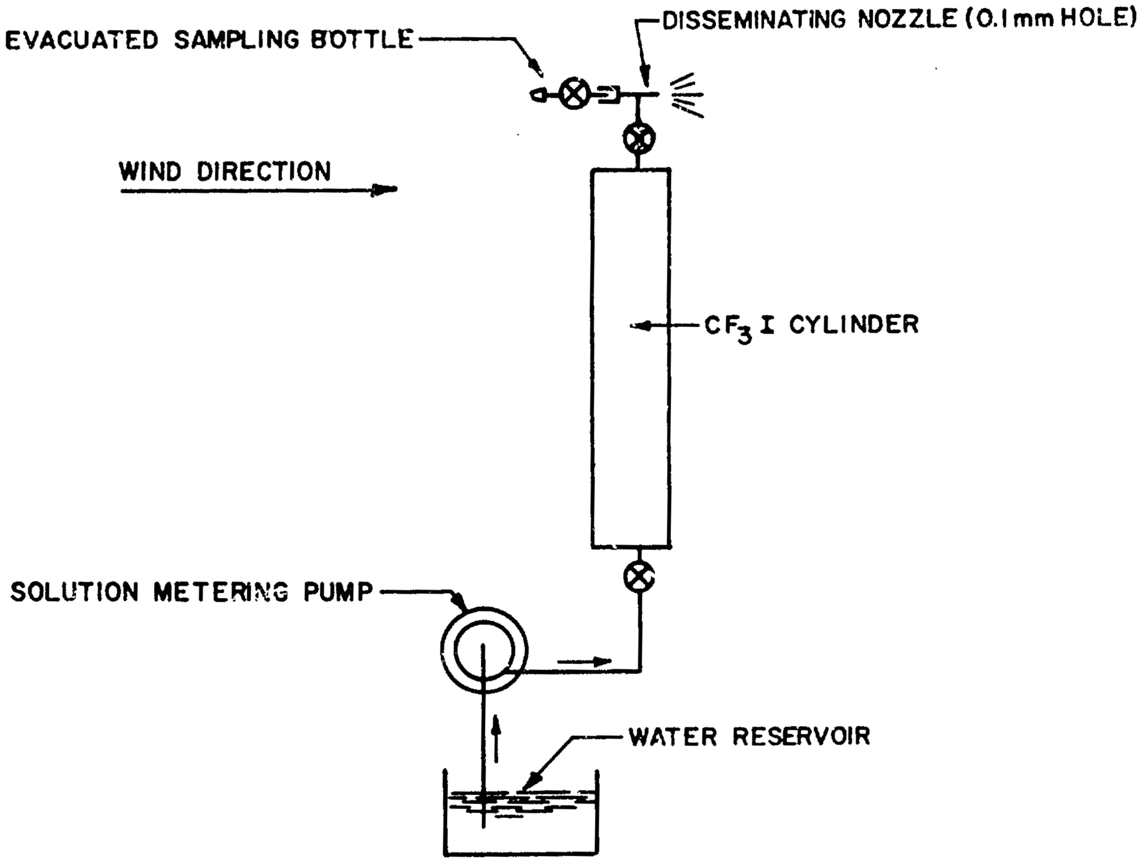
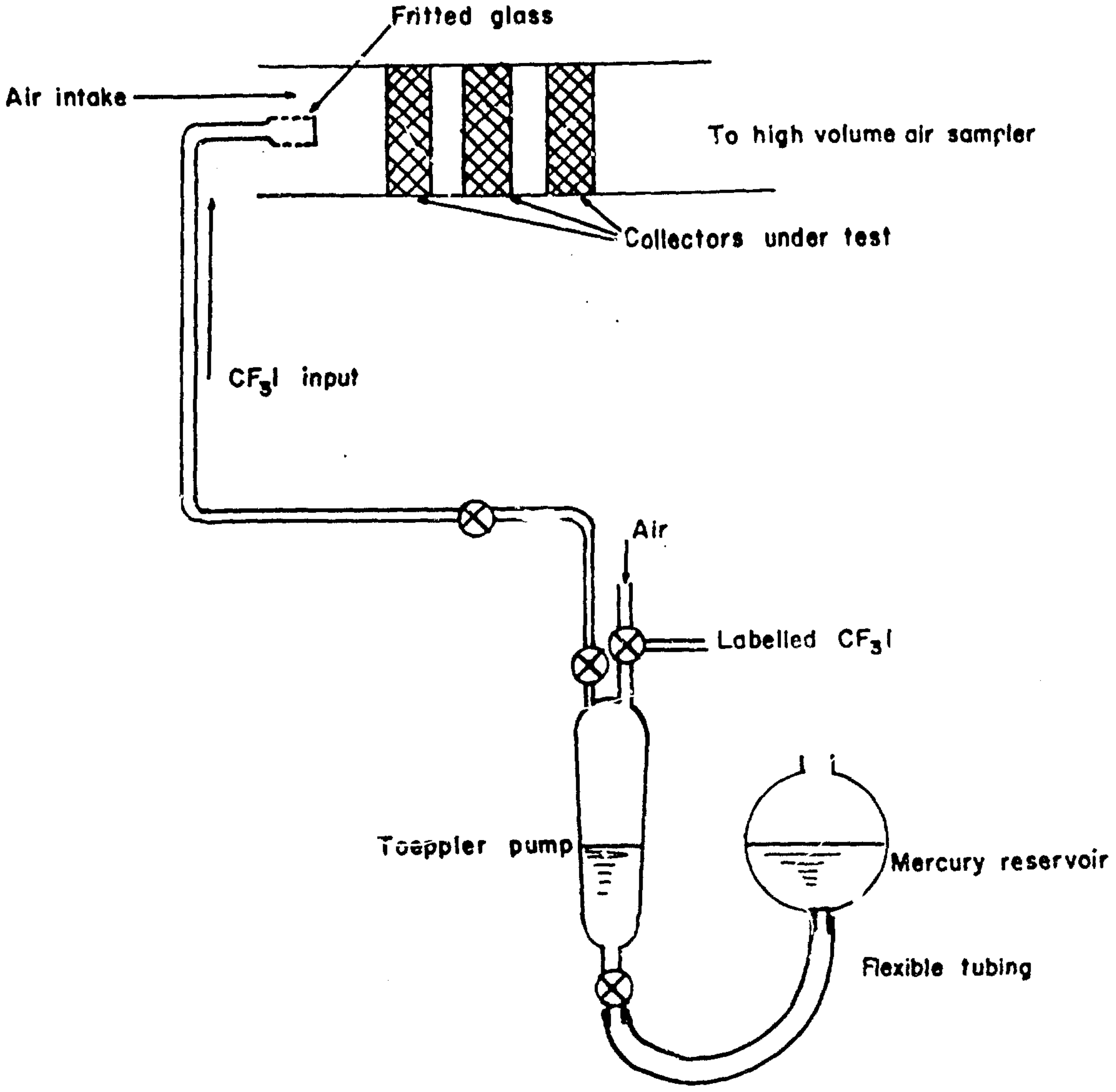


FIGURE 4
EXPERIMENTAL SYSTEM FOR CF_3I COLLECTION



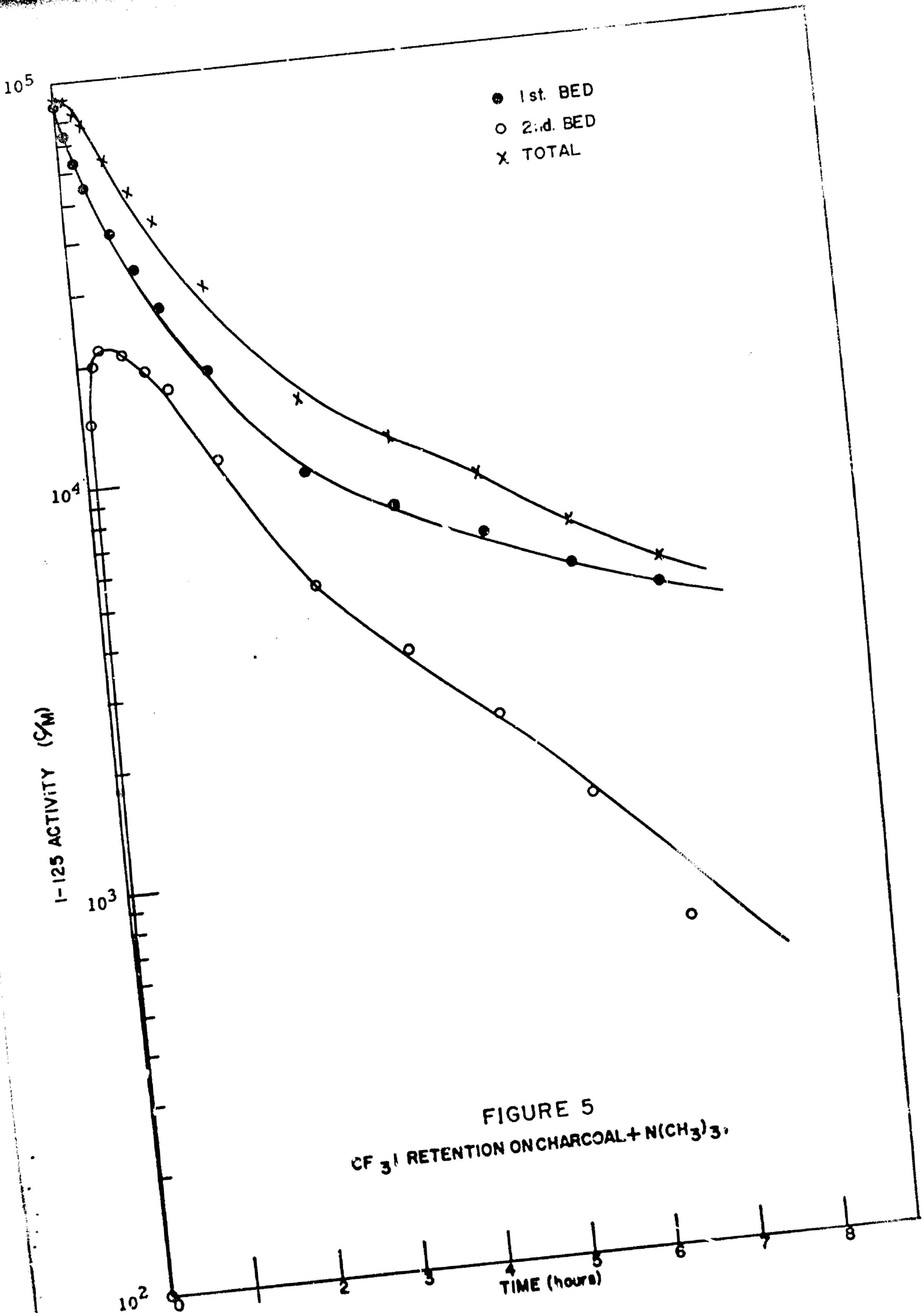


FIGURE 5

CF_3I RETENTION ON CHARCOAL + $N(CH_3)_3$.

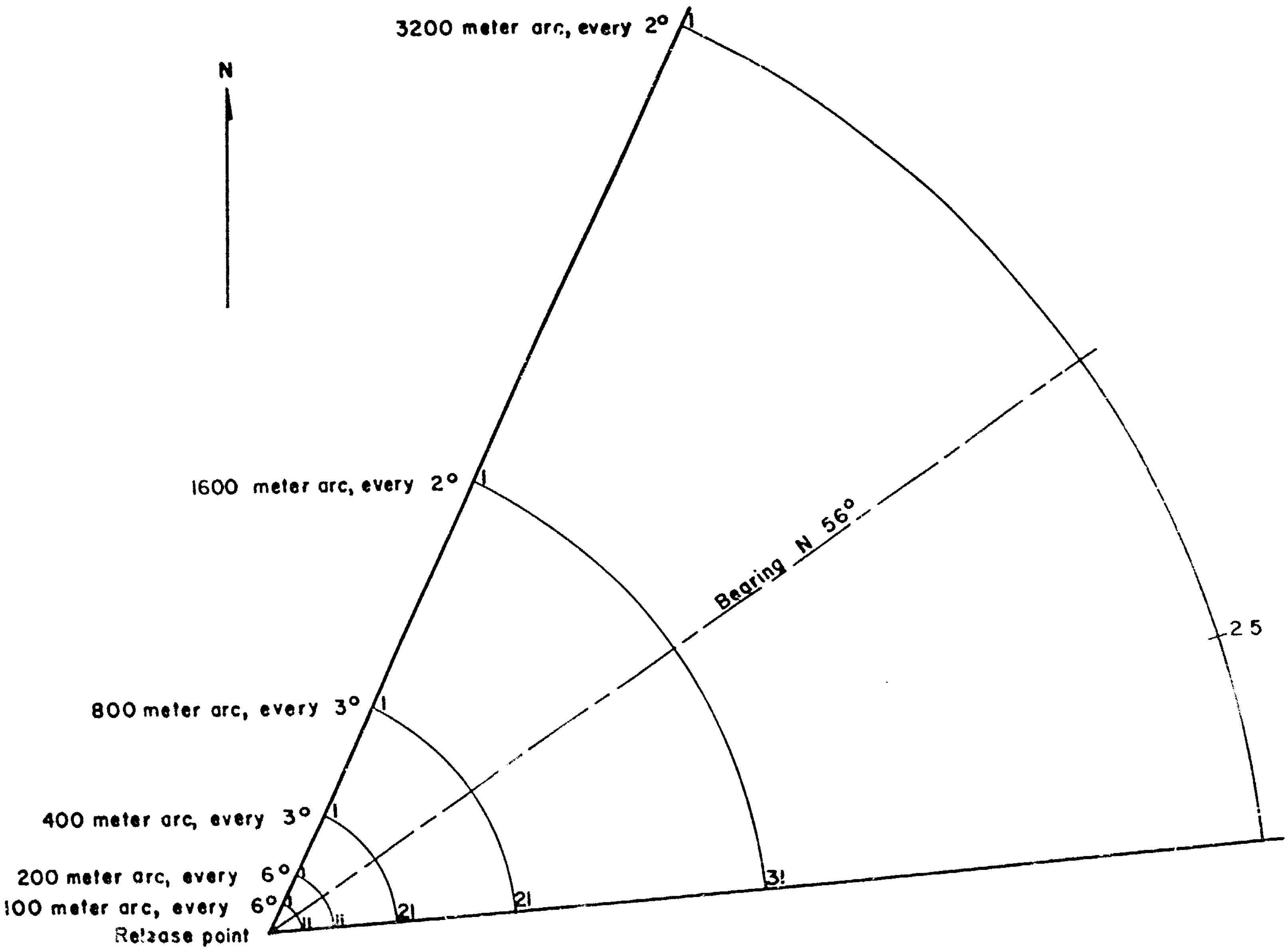


Figure 6, SAMPLING GRID, 100 TO 3200 METERS

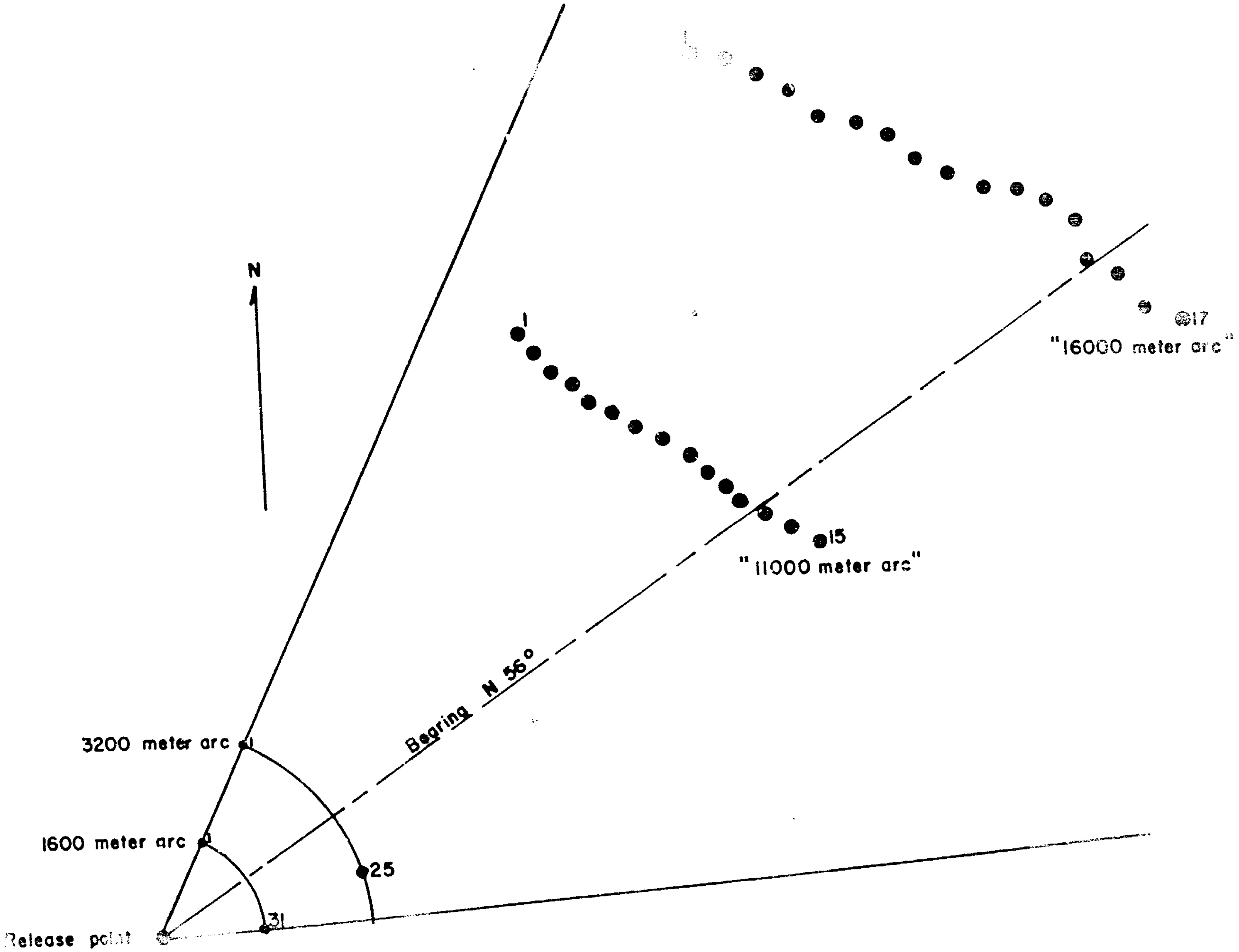


FIGURE 7, SAMPLING GRID, 11000 and 16000 METER ARCS

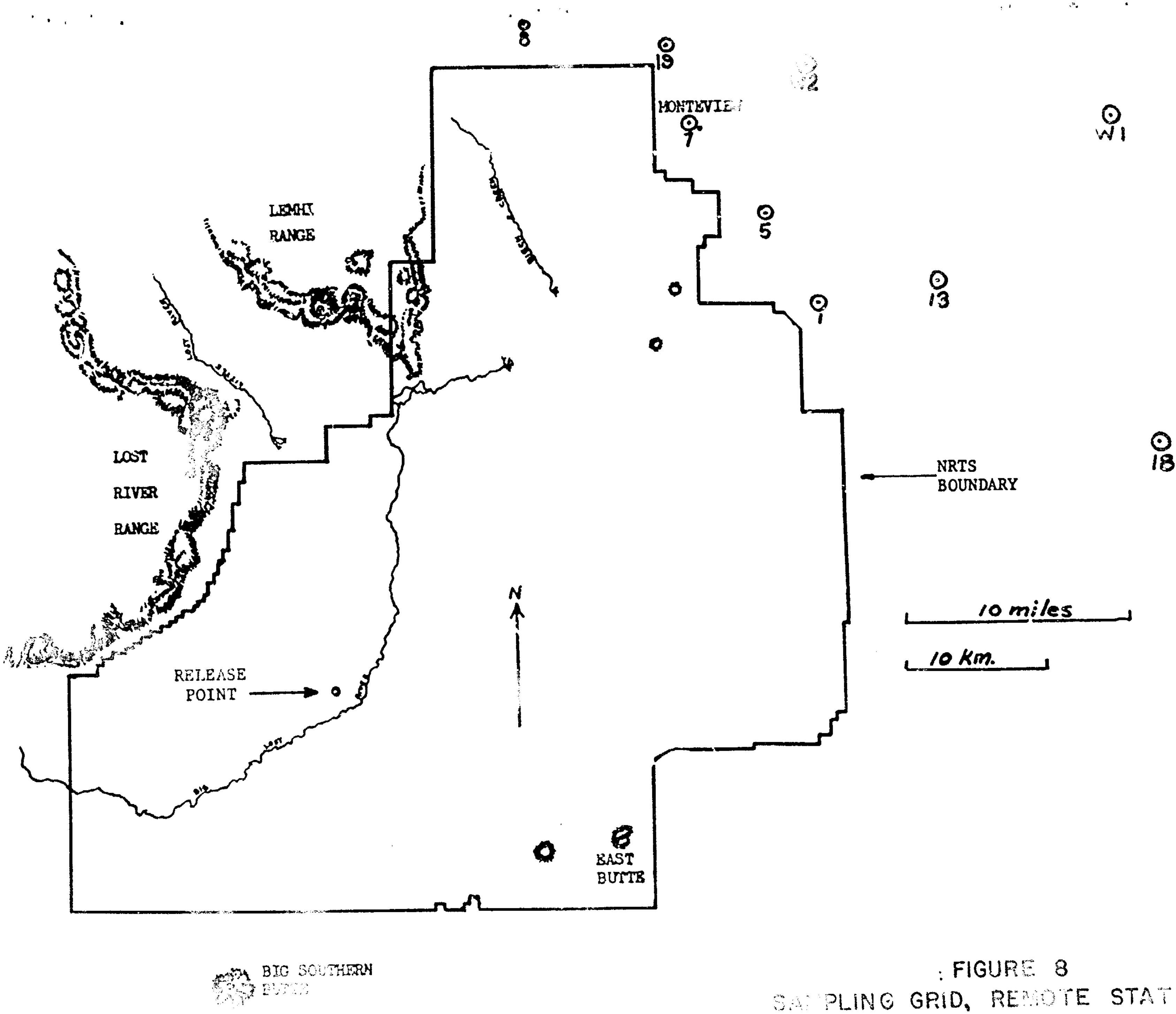


FIGURE 8
SAMPLING GRID, REMOTE STATIONS

FIGURE 9

FILTER AND CARTRIDGE ARRANGEMENT

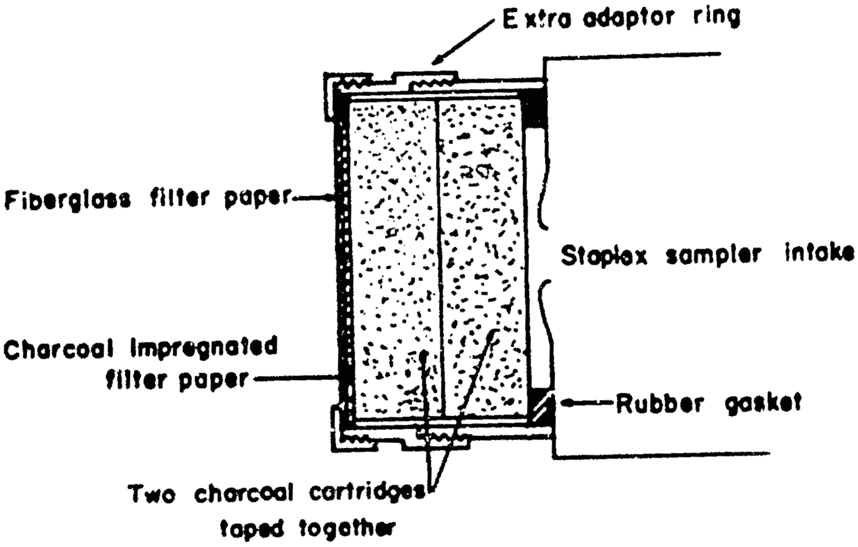


FIGURE 10, TEST A (Particulate Tracer Study)

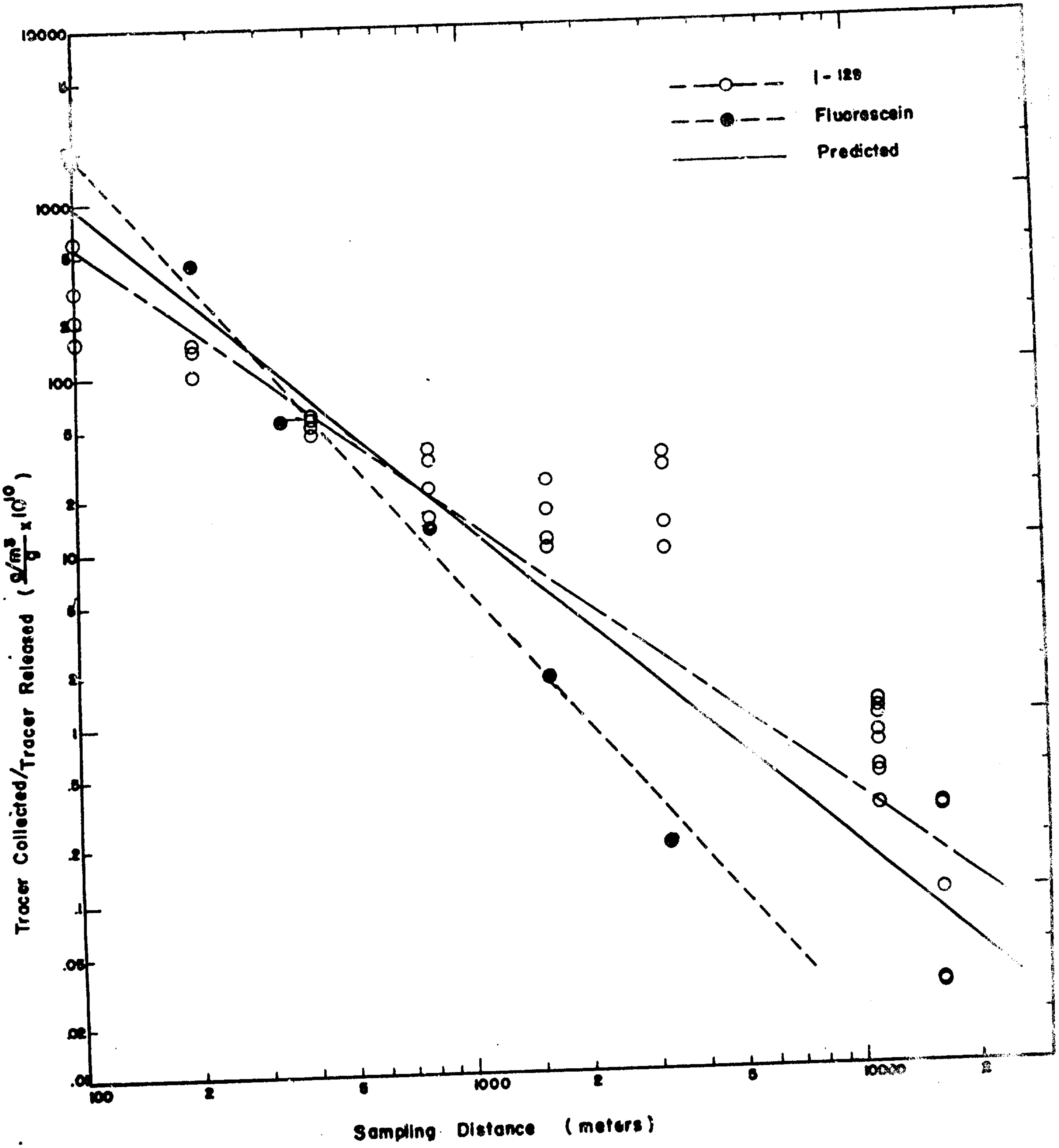


FIGURE 11. TEST D Particulate Tracer Study

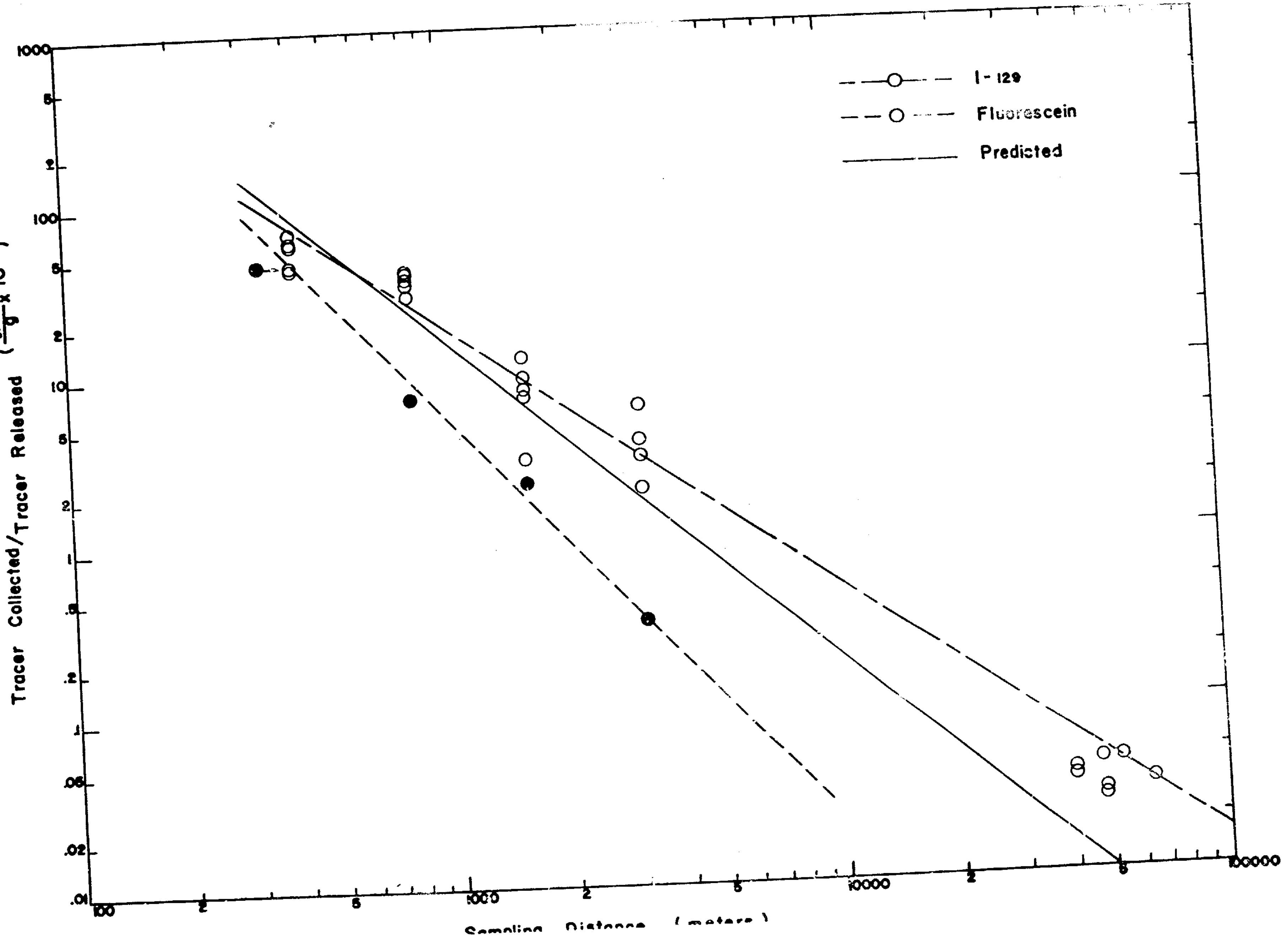
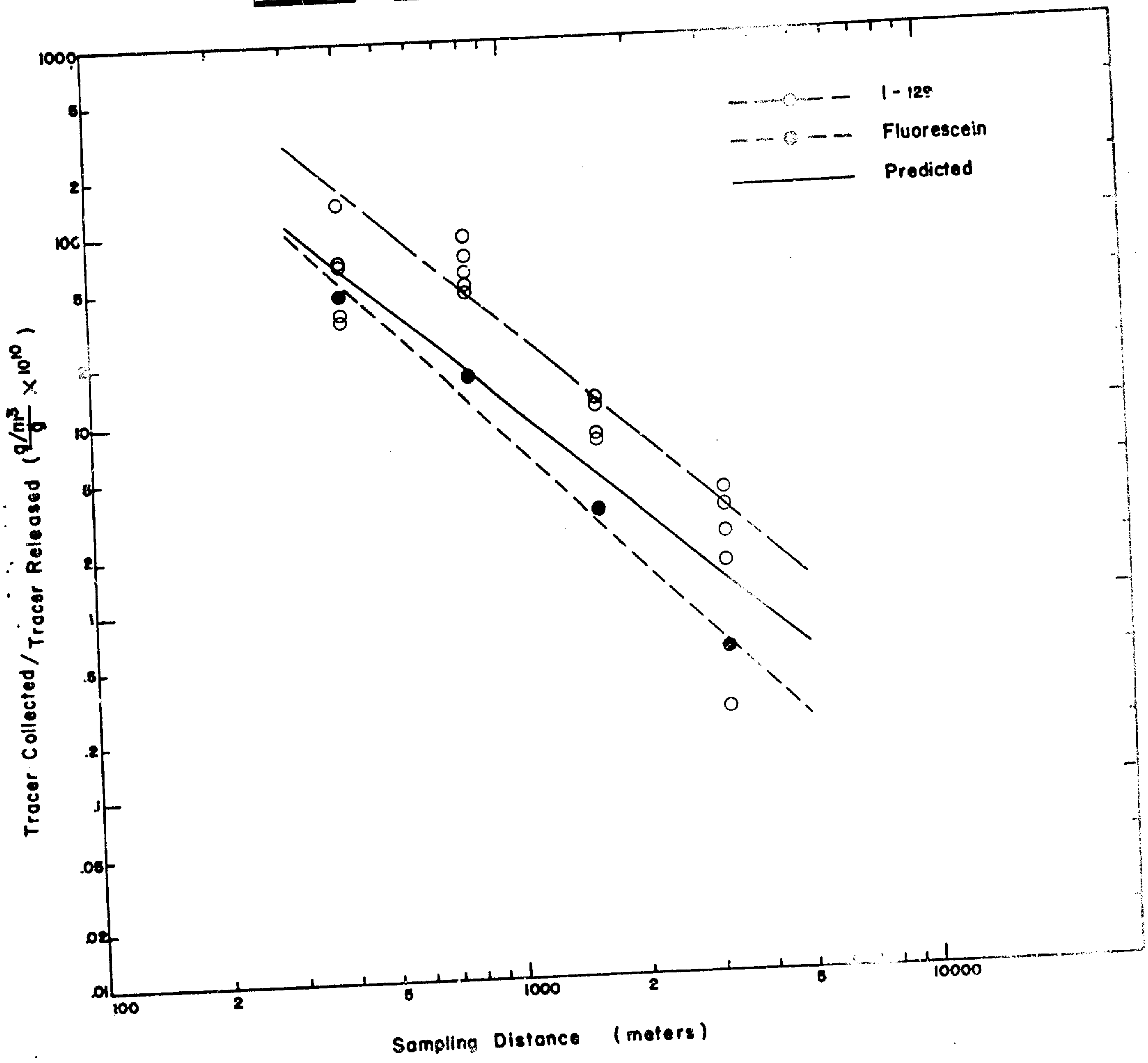


FIGURE 12. TEST E (Particulate Tracer Study)



APPENDIX I

PROCEDURE FOR THE SYNTHESIS OF LABELED DIIODOFLUORESCEIN

1. Prepare a solution (Solution A) by dissolving 23.05 grams of $Na_2HPO_4 \cdot 7H_2O$ in distilled water and diluting to 1000 ml.

2. Prepare another solution (Solution B) by dissolving 9.10 grams of KH_2PO_4 in distilled water and diluting to 1000 ml.

3. Prepare a $\frac{1}{15}$ N phosphate buffer solution (Solution C) by combining 517 ml of Solution A and 329 ml of Solution B. Adjust the resulting buffer solution to pH 7.0 (using a pH meter) by adding small quantities of Solution A or B, as required.

4. Dissolve 4.24 grams of sodium fluorescein in Solution C to make a 0.5% solution of sodium fluorescein in the buffer solution.

5. Dissolve 5.72 grams of I_2 (labeled with I-125 or I-129, as appropriate) in 1144 ml of $CHCl_3$ making a 0.5% solution of I_2 in $CHCl_3$. (Note 1)

6. Combine the solutions prepared in steps 4 and 5 in a two-liter separatory funnel and shake for 20 minutes.

7. Allow the phases to separate. Remove the organic phase and retain for further processing.

8. Wash the aqueous fraction from step 7 with two 200-ml portions of $CHCl_3$ for about 5 minutes each. (Following each wash cycle and phase separation, add the resulting organic phase to that obtained in step 7 and retain the combined organic fractions).

9. Acidify the washed aqueous phase and add ~100 ml of a 10% $KHSO_4$ solution to precipitate diiodofluorescein (DIF).

10. Extract the precipitated DIF into 150 ml of ethyl acetate. Transfer the aqueous phase to another separatory funnel, add ~25 ml 10% $KHSO_4$ solution and ~100 ml ethyl acetate and extract again for about 5 minutes.

11. Retain the aqueous fraction for later reprocessing. Combine the ethyl acetate fractions containing the product and retain.

12. Add concentrated HNO_3 to the aqueous fraction from step 11, until the solution is just acidic, treat with saturated $NaNO_2$ and extract I_2 into two or three successive 50-ml portions of $CHCl_3$. The $CHCl_3$ fractions are then combined with the $CHCl_3$ fractions obtained in steps 7 and 8.

13. Prepare a solution of 0.5% sodium fluorescein in the phosphate butter (pH = 7.0) containing one-half of the quantities of reactants specified in steps 3 and 4 (namely 258 ml Solution A, 165 ml Solution B, and 2.12 grams of sodium fluorescein).

14. Add the solution prepared in step 13 to the combined $CHCl_3$ portions obtained in step 12 and extract for twenty minutes. Following phase separation, retain the organic phase for further recycling (Note 2) if necessary. Wash the aqueous phase twice with ~100-ml fractions of $CHCl_3$. Add the organic washes to the other separated organic fractions.

15. Repeat steps 9, 10, and 11 and combine all ethyl acetate fractions.

16. Filter the combined ethyl acetate fractions through an anhydrous $NaSO_4$ column by gravity to remove traces of water.

17. Transfer the ethyl acetate solution to a large beaker, cover with a watch glass, and heat on a hot plate at low temperature to volatilize the ethyl acetate. After the beaker is cool, add absolute methyl alcohol or ethyl alcohol to dissolve the residue. Return the beaker to the hot plate and volatilize the alcohol. Cool the beaker again and redissolve the residue in alcohol with warming if necessary (Note 3).

NOTE 1: If a different quantity of I_2 than that specified in the procedure is to be used, the concentration ratio of I_2 to sodium fluorescein should remain fixed.

NOTE 2: If radioiodine is used the material should be assayed so that a negligible fraction of the amount originally present will be discarded. If concentrations are greater than an acceptable quantity, recycling should be repeated until an assay indicates further recovery to be uneconomical.

NOTE 3: This step is optional since the synthesized DIF may be retained as an ethyl acetate solution or an alcoholic solution or, may be prepared as a solid by crystallization from hot absolute ethanol by adding xylene and cooling in a refrigerator.

APPENDIX II

PREPARATION OF LABELED TRIFLUOROMETHYL IODIDE (CF_3I)

Details of two procedures for labeling CF_3I with either I-125 or I-129 are given below.

A. Labeling by Exchange

This method is well suited to labeling with small masses of tracer.

1. Assuming that the radioactive iodine is available in the form of an inorganic iodide in basic solution, convert the iodide to I_2 by one of the two following methods, depending upon the physical quantity of iodine involved.

a. For low masses (<500 mg) of iodine, slowly evaporate the basic iodide solution to dryness in one end of a tube using a heat lamp. Cover this material with an excess quantity of dry potassium dichromate. Seal the tube to a receiver such as that shown in Figure 2-1. Evacuate the system, and heat the chromate to oxidize the iodide to iodine. The iodine will distill into the cooled trap.

b. For larger masses (>500 mg) of iodine, evaporate the basic iodide solution to a minimum volume (without the formation of solids), oxidize the iodide to iodine in solution by acidifying with nitric acid and adding a small quantity of sodium nitrite, and remove the precipitated iodine from the solution by filtering through a fritted glass Gooch crucible. Place the iodine crystals in the bottom of a porcelain evaporating dish and add enough concentrated sulfuric acid to cover them. Cover the dish with a watch glass, heat the dish until the iodine melts under the acid. Cool the dish and contents as quickly as possible, pour off the acid, and dry the button of iodine with absorbent tissue.

2. Transfer the dry elemental iodine into the exchange vessel, shown in Figure 2-2, either mechanically or by distillation.

3. Seal the exchange vessel to the system shown in Figure 2-2. The exchange vessel should be constructed of well-annealed, heavy-wall Pyrex

tubing or of heavy-wall quartz tubing.

4. With the three-way stopcock open to the air, and the calibrated receiver dooled with a dry ice-acetone mixture, allow CF_3I gas to flow through the receiver until an appropriate quantity ([illegible] ml) (Note 1) is condensed in the receiver.

5. Close the valve and the stopcock between the receiver and the tank, cool the exchange vessel, and evacuate the system (~50μ).

6. Close the stopcock leading to the vacuum system and distill the CF_3I into the exchange vessel by allowing the receiver to warm to room temperature.

7. Break the seal at the point indicated in the figure and, while still cooling the exchange vessel, remove the latter from the system.

8. Immediately seal off the exchange vessel above the neck, retaining at least 5 cm of the smaller diameter tubing.

9. Place the exchange vessel inside a metallic shield, in a tube-type electric resistance furnace and hold at 150°C for one hour.

10. After the vessel is cooled to condense the contained material at the lower end of the vessel, break the entry tube open, seal onto an appropriate distillation system, and distill the labeled CF_3I away from the iodine. Repeat the distillation if further purification is necessary. By maintaining the iodine residue at 0°C, a colorless iodine-free product is obtained after two distillations.

NOTE 1: The ratio of radioactive iodine to stable iodine in the product at exchange equilibrium will be equal to the molar ratios of the iodine isotopes in the entire system. Thus, the ratio of the starting reactants and the concentration of tracer in the form of iodine will control the specific activity of the labeled CF_3I.

B. Labeling by Direct Synthesis

This method is only suitable for use when gram quantities of iodine tracer are to be used.

1. Prepare dry elemental iodine as given in Step A-1b above.

2. Place the button of iodine in the special distillation system shown in Figure 2-3 and evacuate the system.

3. Close the stopcock and with powdered dry ice in the cold finger, as indicated in Figure 2-3, heat the tube bottom with boiling water until all the iodine is condensed on the cold finger.

4. Remove the cold finger carefully and scrape the pure, dry iodine into a mortar. Pulverize and transfer the iodine onto a watch glass. Store in a vacuum dessicator over concentrated sulfuric acid until used.

5. Dry a quantity of CF_3COOAg (silver trifluoroacetate) by placing the pulverized material under a heat lamp for several hours with occasional stirring.

6. Dry all parts of the apparatus shown in Figure 2-4 in an oven at 120°C before use.

7. Add the powdered reagents to the round bottom reaction flask and mix by shaking together. Typical quantities to be used are 6 grams of iodine and 9 grams of CF_3COOAg.

8. With the system assembled, cool both receivers with dry ice-acetone and gently warm the reaction flask until the reaction begins. Thereafter, heat just enough to complete the reaction, at which time the residual mass will be the bright yellow color of silver iodide.

9. After the reaction is complete, evacuate the system (maintain cooling of both receivers).

10. Close the stopcock leading to the vacuum system and distill the CF_3I from the first receiver into the calibrated receiver by warming the former to ice-water temperature while maintaining the latter at dry ice temperature. Repeat this procedure, if necessary, to obtain a colorless, iodine-free product.

11. The yield of this synthesis should be >80% of theoretical (based upon 1/2 the iodine being converted to CF_3I). The iodine remaining in the form of silver iodide may be recovered using conventional chemical procedures if it is economically adviseable to do so.

FIGURE 2-1

ELEMENTAL IODINE CONVERSION APPARATUS

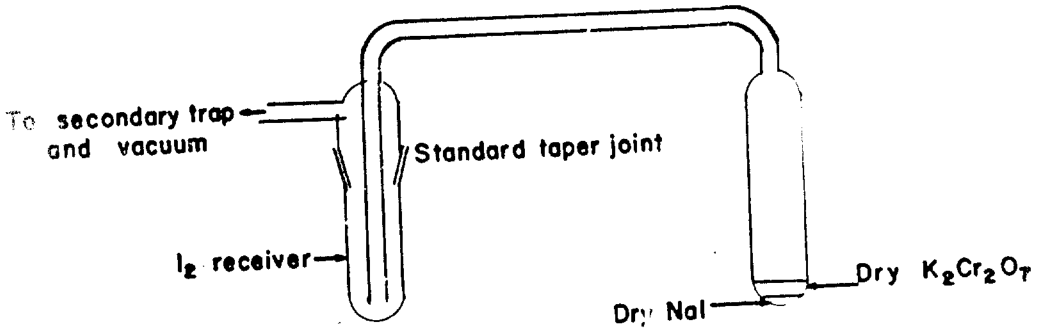


FIGURE 2-2
EXCHANGE VESSEL LOADING SYSTEM

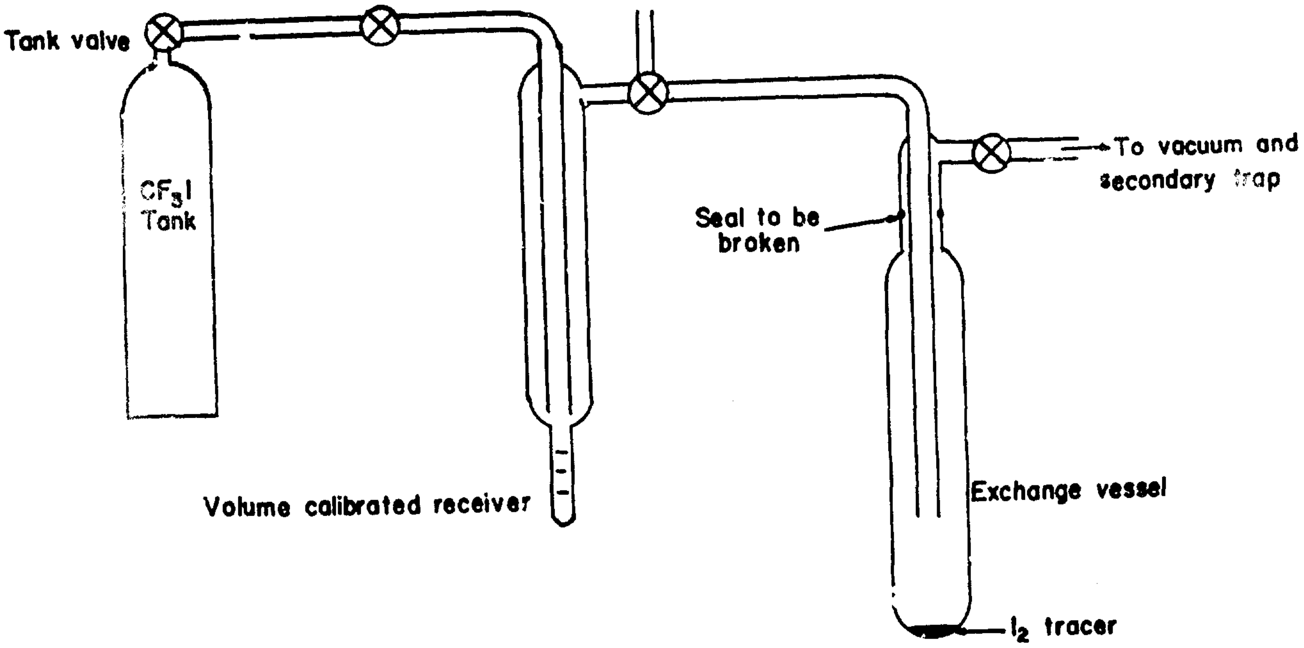


FIGURE 2-3
IODINE DRYING APPARATUS

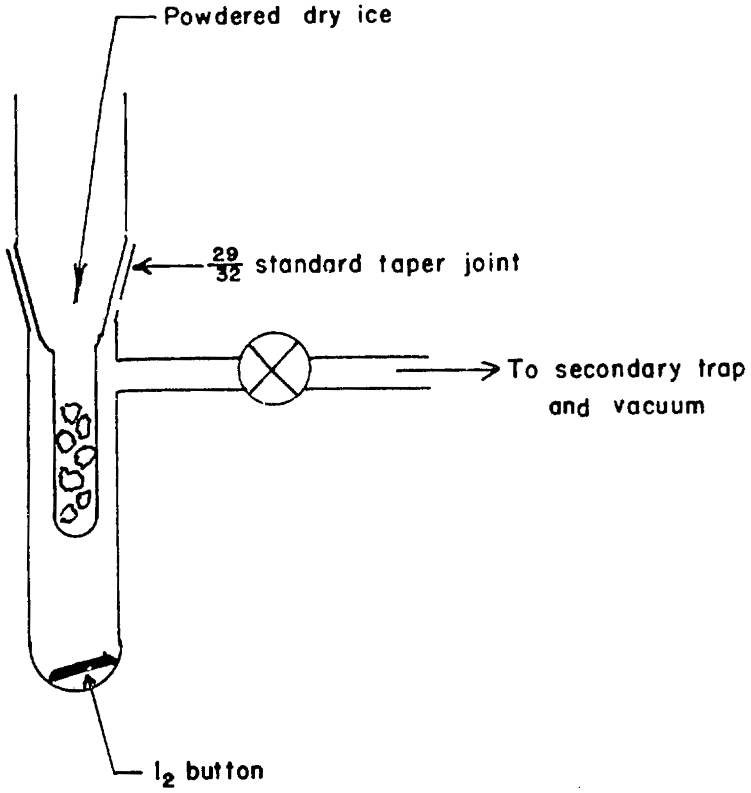
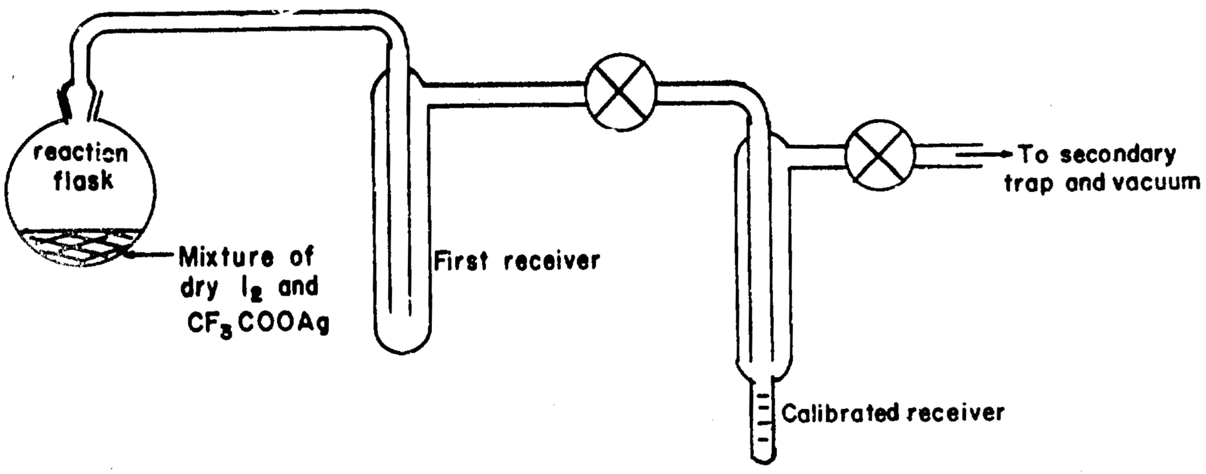


FIGURE 2-4
CF_3I SYNTHESIS APPARATUS



APPENDIX III

PARTICLE SIZES PRODUCED BY AEROSOL GENERATOR

Table III-I shows the results of 23 measurements of particle size distributions of fluorescein and DIF aerosols generated with the apparatus shown in Figure 1 of the text. All of these aerosols were produced under essentially the same conditions (air pressure 80 to 100 psi; air flow rate 145-155 (cfm) except where specified in the table. Sampling was accomplished with a 2-in diameter millipore filter paper (0.45-μ pore size) mounted on a 1-cfm sampling device located 2 meters from the generator.

The variations in the dissemination conditions and in the environmental conditions during the disseminations were sufficiently complex to preclude any practical basis for rigorous control of particle-sizes. However, the resulting range of particle sizes was judged to be satisfactory for use in the field tests. The development of a more rigorous control method was deferred pending resolution of more urgent questions.

TABLE III-I

SUMMARY OF PARTICLE SIZES PRODUCED BY AEROSOL GENERATOR

Experiment	Solution Composition[a]	Solution Pressure (psi)	Temperature (°C)	Weather	Median Particle Size (μ) Weight Basis
1	(F) 27 mg/ml	5	---	---	1.5
2	(F) 6.1 mg/ml	5	---	Into slight wind	1.2
3	(F) 18.1 mg/ml	5	---	"	0.65
4	(DIF) 6.1 mg/ml	5	---	With slight wind	0.5
5	(DIF) 18.3 mg/ml	5	---	"	0.3
6	(F) 9.3 mg/ml	0.5 - 1	11°	Overcast, Gusty winds	0.7
7	(F) 37.6 mg/ml	0.5 - 1	11°	"	0.58
8	(F) 37.6 mg/ml	0.5 - 1	11°	"	0.62
9	(F) 2.9 mg/ml	0.5 - 1	11°	"	0.2
10	(F) 5.9 mg/ml	0.5 - 1	11°	"	0.29
11	(F) 6.7 mg/ml	0.5 - 1	11°	"	0.18
12	(F) 7.3 mg/ml	0.5 - 1	13°	Sunny	0.45
13	(DIF) 5.4 mg/ml	0.5 - 1	14.5°	Overcast	0.53
14	(DIF) 18.3 mg/ml	0.5 - 1	14.5°	"	0.1
15	(DIF) 18.6 mg/ml	0.5 - 1	14.5°	"	0.44
16	(DIF) 2.7 mg/ml	0.5	19°	Clear	0.65
17	(DIF) 5.1 mg/ml	0.5	20°	"	0.84
18	(DIF) 8.2 mg/ml	0.5	23°	"	1.7

TABLE III-I (Continued)

Experiment	Solution Composition[a]	Solution Pressure (psi)	Temperature	Weather	Median Particle Size (μ) Weight Basis
19	(DIF) 10.7 mg/ml	0.5	24°	Clear	0.79
20	(DIF) 25 mg/ml	0.5	21°	Windy	0.98
21	(DIF) 24 mg/ml	0.5	22°	"	1.05
22	(DIF) 23 mg/ml	1.0	30°	"	2.3
23	(DIF) 22 mg/ml	2.0	31°	"	0.86

(a) - (F) = Fluorescein, (DIF) = Diiodofluorescein

APPENDIX IV

DETERMINATION OF I-129 BY NEUTRON-ACTIVATION ANALYSIS

Although I-129 is a radioactive nuclide, its extremely long half-life results in a specific radioactivity of only 0.16 μc/mg. Furthermore, the low energies of its radiations (150-Kev β and 38-Kev γ) also make its detection by ordinary radiometric methods very insensitive. A reasonably large cross-section (~27 barns) for thermal-neutron capture, leading to the formation of the activation product, 12.6-hr I-130, renders the determination of I-129 by neutron-activation analysis much more sensitive than by radiometric analysis.

The activation of material containing small quantities of I-129 for subsequent measurement of the I-130 produced leads to several interferences which cannot be circumvented except through the proper choice of the experimental method.

One type of interference involves nuclear reactions in constituents of the material, other than iodine, which contribute to the production of I-130; e.g., the reaction $\mathrm{Cs}^{133}\,(n, \alpha)\,\mathrm{I}^{130}$, or the neutron-induced fission of heavy elements. Another type of interference involves nuclear reactions in isotopes of iodine, other than I-129, which produce other iodine radioisotopes whose presence may compromise the accuracy of the I-130 measurements; e.g., the reactions $\mathrm{I}^{127}\,(n, \gamma)\,\mathrm{I}^{128}$ and $\mathrm{I}^{127}\,(n, 2n)\,\mathrm{I}^{126}$. Finally, multiple neutron-capture processes in I-127 can lead to production of I-130 and may result in an apparent large value of I-129.

The effects of phenomena of the first type must be compensated for by adequate pre-irradiation chemical procedures. However, the importance of interfering reactions in iodine isotopes is dependent, in part, on the I-129/I-127 ratio in a given sample. These effects can be reduced only by appropriate instrumental methods and/or by a judicious selection of irradiation conditions.

To circumvent or minimize the effects of these phenomena on the accuracy and sensitivity of the analyses, an experimental procedure is used which involves: (1) the chemical separation and purification of the iodine fraction in the sample prior to irradiation, (2) the irradiation of the samples under carefully controlled conditions, (3) the radiochemical purification of the radioiodine after irradiation, and (4) the measurement of the I-130 content in a beta-total-gamma coincidence counting system.

The details of the chemical procedures required for this program are presented in Appendices V and VII.

The necessity for special radiation measurement instrumentation arises from the following considerations. The normal occurrence of I-127 in the matrices being studied leads to the formation of 25-minute I-128 by the (n, γ) reaction and to the formation of 13-day I-126 by the (n, 2n) reaction. The interference due to the 25-minute I-128 may be reduced or eliminated by deferring measurement of the I-130 until the I-128 has decayed. However, the 13.2-day I-126 which may be present must be discriminated against instrumentally. An examination of the decay schemes of I-126, I-128, and I-130 reveals that the maximum total energies of the gamma cascades of the respective nuclides are 1.41 Mev, 0.99 Mev, and 2.34 Mev. Therefore, a 5-in diameter by 5-in long well-type NaI scintillation detector, with a proper choice of gamma-energy discrimination, will permit the measurement of I-130 with a minimum interference from the radiations from the other two iodine isotopes. However, without prohibitively excessive shielding, background count rates as large as hundreds of counts per minute are encountered with such a detector. To reduce the background and, thus, to increase the sensitivity of the measurement method, a β-γ coincidence counting system has been applied. A miniature end-window gas-flow proportional counter, which fits inside the well of the NaI detector, is used to measure the beta radiations. A storage battery is used as a power supply for preamplification stages, and a high efficiency line filter is installed on other power inputs. A background counting rate of 0.03 cpm is routinely achieved with this system.

With this low coincidence background, the limiting factor for detection of I-130 in very low-level samples was the contribution to the discriminated output of the NaI detector from accidentally coincident gamma rays from I-126.

These paired pulses were observed at all practical discrimination levels for samples having larger than average I-127 contents. The effects of such pulses have been minimized by imposing time-based discrimination on the coincidence circuit. This "paired-pulse rejector" eliminates the longer duration coincidence pulses from the output of the coincidence circuit and results in a three-fold reduction of accidental I-126 count rate. The complete counting arrangement is shown in Figure 4-1.

Further reduction of the I-126 interference and interference from triple neutron capture in I-127 requires optimization of the neutron irradiation conditions. The selection of optimum irradiation conditions therefore must take into account two important factors: (1) the enhancement of production of I-130 relative to that of I-126, and (2) the possible production of I-130 as a result of the successive capture of three neutrons by I-127.

With the improved instrumentation, the enhancement of I-130 relative to I-126 during irradiation is of substantial concern only when samples containing very small ratios (I-129/I-127) are involved. However, for many of the samples of particular interest in this investigation, the values of this ratio were such that the very small, but unavoidable, contribution of accidentally-coincident gamma rays from I-126 to the coincidence count rate represented a significant increment in the effective background above which it was necessary to detect the I-130. The use of the well-thermalized neutron flux in a heavy-water-moderated reactor (the MITR) provided adequate enhancement for most samples of interest.

The effect of neutron spectrum is shown qualitatively in a comparison of decay curves observed for similar samples irradiated under essentially the same flux-time conditions at the MITR and at a light-water moderated pool-type reactor at Battelle Memorial Institute (BRR). The upper set of curves in Figure 4-2 was obtained with samples containing approximately 10 mg of I-127, while the other set involved samples containing approximately

5 mg of I-127. It is immediately obvious that the interpretation of the decay curves obtained at the BRR involves uncertainties which lead to unacceptable errors in the determination of I-129. However, the samples irradiated in the well-thermalized neutron flux of the MITR exhibited decay properties amenable to accurate interpretation.

The experimental verification of the importance of the multiple neutron capture process has been described earlier. This phenomenon represents the ultimate limitation on the detection of I-130 in iodine-bearing materials. It was shown that the optimum sensitivity for the ratio measurement (I-129/I-127) is obtained at a thermal neutron flux of approximately 10^{13} n/cm^2-sec when sample mass is limited.

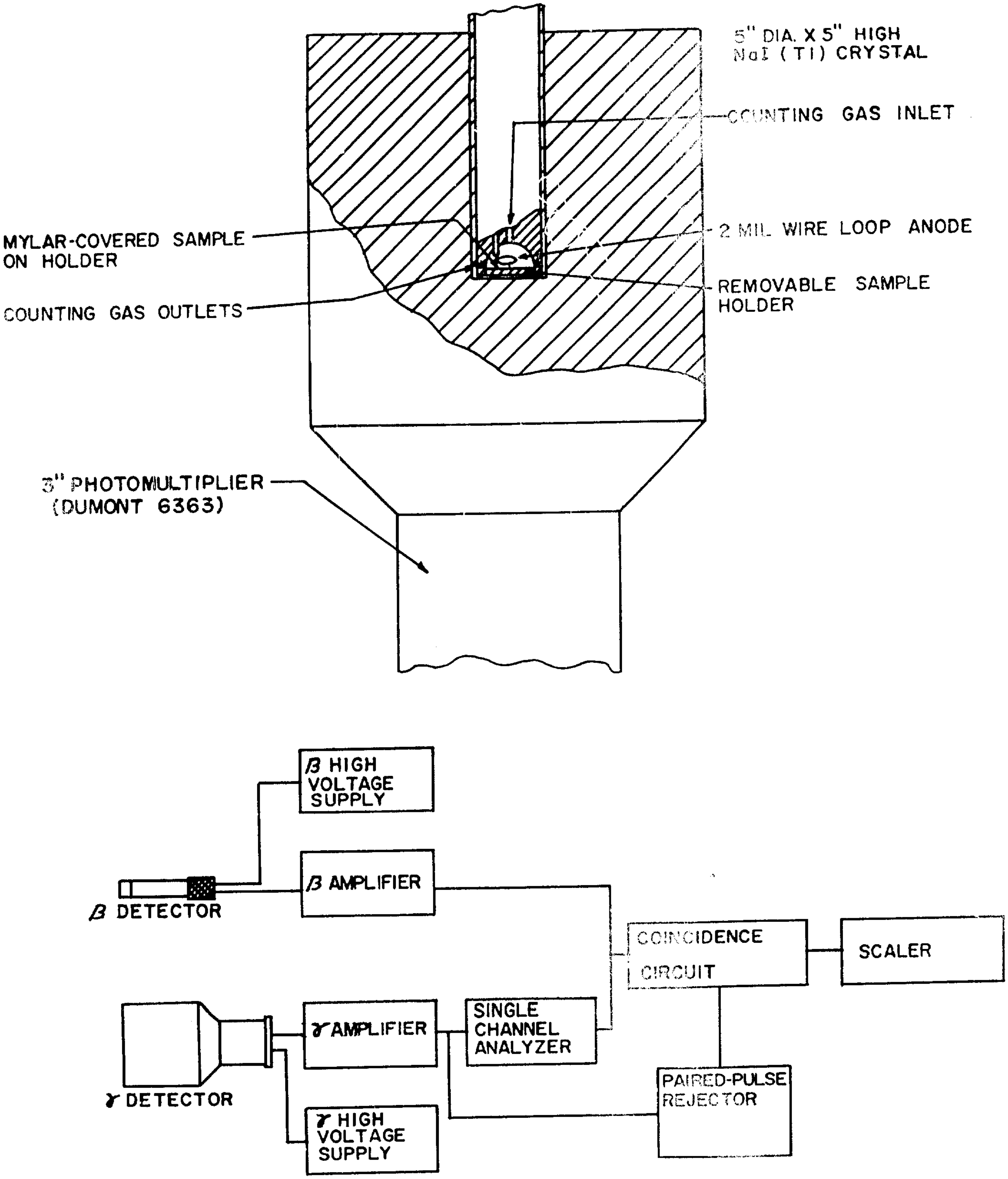
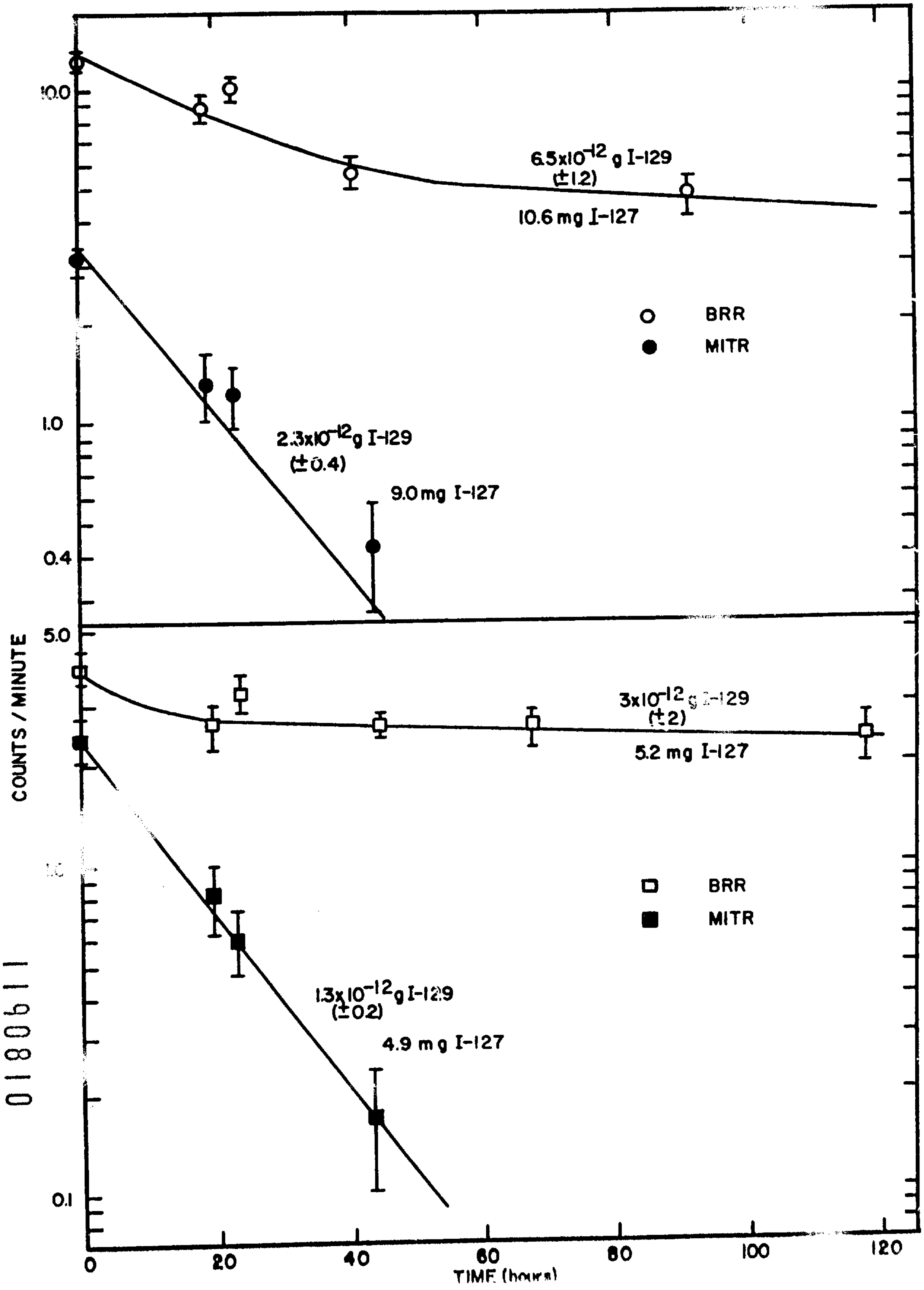


FIGURE 4-1

β -TOTAL γ COINCIDENCE COUNTING ARRANGEMENT

FIGURE 4-2
TYPICAL I-130 DECAY CURVES



APPENDIX V

ANALYTICAL PROCEDURES FOR DIIODOFLUORESCEIN COLLECTIONS

A. Procedure for Low-Level I-129 Measurements

"Low-level" samples are defined here to be samples containing insufficient I-129 (generally less than 10^{-9} gram) for accurate determination by γ-ray spectrometry of the I-130 produced by neutron activation. The detailed procedure is as follows:

1. Leach the filter paper containing the collected sample with four 25-ml portions of methanol using a Buchner funnel and applying suction.

2. Add a known amount (50-100 mg) of DIF in ethanol solution to the leachant. Slowly evaporate the resultant solution to dryness, allow the residue to cool, and dissolve it in 50 ml of n-butanol with gentle heating.

3. Wash the solution for about two minutes each with two successive portions of 10-15 ml of water containing 2-3 ml of H_2SO_3. Discard the aqueous washes.

4. Add 50 ml of CCl_4 to the organic phase and extract the DIF into 10 ml of 3N NaOH. Repeat this extraction three times.

5. Add ~25 ml of a 5% NaOCl solution of the combined aqueous fractions and heat moderately until a nearly clear solution is obtained.

6. After cooling, cautiously add concentrated nitric acid until the solution is barely acid. Add 10-15 ml of a 10% solution of $NH_2OH \cdot HCl$ and extract iodine with two successive 15-ml portions of CCl_4. Combine the organic fractions in a clean separatory funnel.

7. Re-extract the combined organic fraction into ~10 ml of water containing 2-3 ml of H_2SO_3.

8. Add several drops of saturated $NaNO_2$ solution to the aqueous phase to oxidize the iodide to iodine and repeat the extraction cycle.

9. Heat the aqueous phase to boiling, cool, and add 1-2 ml of a 5% solution of $PdCl_2$ to precipitate PdI_2.

10. Filter the PdI_2 with a weighed sintered glass crucible.

11. Wash the precipitate successively with H_2O and ethanol, and dry it in a vacuum dessicator overnight.

12. Clean a quartz U-tube by exposing it for 1-2 hours to concentrated nitric acid containing a few drops of hydrofluoric acid and then rinsing thoroughly with deionized (or distilled) water.

13. Transfer the PdI_2 to one end of the clean U-tube. Seal the tube at both ends and decompose the PdI_2 by heating. The resulting I_2 will distill to the cool end of the tube. Seal this end of the quartz tube to form the irradiation capsule. The samples are then irradiated under appropriate conditions in a nuclear reactor.

14. Decontaminate the surface of the irradiated capsule by etching it with boiling nitric acid.

15. Insert the capsule in a small section of vinyl tubing containing a few drops of 10 N NaOH, and seal the ends of the tube with screw clamps.

16. Break the quartz capsule inside the vinyl tubing.

17. Transfer the broken quartz capsule and its contents with water to a 150-ml beaker. Add a few drops of 10 N HaOH, 1 ml of NaBr solution (about 10 mg Br/ml) and 1 or 2 drops of H_2SO_3. If necessary, add iodine carrier (Note 1). Heat samples as necessary to promote dissolution. Add 16 N HNO_3 dropwise until the color of iodine appears. Immediately add H_2SO_3 dropwise to reduce the I_2 to iodide.

18. Transfer the sample with water to a specially prepared [illegible]-ml Nalgene wash bottle (Note 2).

19. Add sufficient saturated $NaNO_2$ solution to oxidize the iodide, and extract I_2 into 10 ml of CCl_4. Repeat until all of the iodine is extracted.

20. Combine the CCl_4 fractions in a clean wash bottle and wash twice with 15 ml of water. Each washing should be performed in a clean bottle.

21. Re-extract the iodine into 20 ml of water containing 4 drops of H_2SO_3 and 20 drops of NaBr solution. Discard the CCl_4 layer.

22. Repeat the extraction cycle twice. In the final cycle, back-extract the iodide into 20 ml of water containing only a <u>minimum</u> of H_2SO_3.

23. Heat the aqueous layer to boiling to remove traces of CCl_4, add 16 $\underline{N}$ HNO_3, until the brown iodine color appears, and immediately add 2 ml of 10% silver nitrate solution to precipitate AgI.

24. Filter the AgI precipitate as soon as possible onto a tared Whatman #542 filter paper. Wash with water and ethanol, dry under a heat lamp, weigh, and mount on a special assembly for counting (Note 3).

25. Measure the radioactivity of the sample as soon as possible with a β-total-γ coincidence counting system, and follow the decay for a sufficient period (at least two days) to define accurately the I-130 content of the sample.

NOTE 1: If a sample contains less than $\sim$10 mg of iodine, $\sim$20-30 mg of iodine carrier should be added to avoid excessive losses during purification steps. The original quantity of stable iodine in the sample can be determined by measuring the I-126 (after decay of I-130) and comparing with other samples in the run not requiring this additional iodine.

NOTE 2: Conventional 4-oz. Nalgene wash bottles having dispensing tubes set on the shoulders are used. After properly positioning the dispensing tube so that the bottle can be completely drained through the tube, the tube is sealed with "Duco" cement.

NOTE 3: The special assembly, designed to fit the counting system at NSEC consists of a 1-in. diameter plastic ring and disc. The weighed sample and filter paper are placed, sample side up, on the disc. A piece of 0.25-mil aluminized mylar is placed over the sample and clamped into place with the ring. Any excess mylar is then trimmed off.

B. Procedure for High-Level I-129 Measurements

"High-level" samples are defined here to be samples containing sufficient I-129 (generally greater than 10^{-9} gram) so that the I-130 produced by neutron activation can be determined accurately by γ-ray spectrometry. The detailed procedure is as follows:

1. Leach the filter paper containing the collected samples and add DIF carrier as described in steps A-1, 2 above, but do <u>not</u> add n-butanol.

2. Dissolve the residue by adding 5 ml of ~5% NaOCl solution, 1 ml of $1\underline{N}$ NaOH solution, and 5 ml of water and heating gently.

3. Perform one CCl_4 extraction cycle (See steps A-6, 7).

4. Boil the aqueous phase to reduce the volume to 10 ml or less. Add 1 ml of a 10% solution of $TlNO_3$ to the boiling solution. Continue boiling until a chrome-yellow color appears and the TlI precipitate coagulates. Filter the cooled solution through a Whatman #542 filter paper, wash the precipitate with absolute methanol and dry it at 110°C for about 30 minutes.

5. Transfer the precipitate to an appropriate irradiation package. In this work, a small (1-in x 1-in) polyethylene bag, sealed by fusion, constituted the irradiation package. The sample is then irradiated under appropriate conditions in a nuclear reactor.

6. Open the polyethylene bag containing the irradiated sample (thallium iodide) under a small volume of water.

7. Add approximately 2 ml of $16\ \underline{N}\ HNO_3$, and allow 10-15 minutes for the sample to dissolve.

8. Using successive portions of $16\ \underline{N}\ HNO_3$ and water, transfer the solution to a 125-ml Nalgene wash bottle. (See Note 2 of Part A above)

9. Extract the iodine into 10 ml of CCl_4. Repeat as required until all of the iodine is extracted.

10. Combine the CCl_4 fractions in a clean bottle and wash with 15 ml of water.

11. Transfer the CCl_4 fraction to another clean bottle, and back-extract

extract the iodine (as iodide) into 2 ml of 6% H_2SO_3.

12. Transfer the H_2SO_3 to a 4-ml screw-cap glass vial and measure the I-130 radioactivity in the sample with a scintillation detector connected to a multichannel pulse-height analyzer.

13. The iodine chemical yield can be determined by precipitating and weighing AgI after completion of the radioactivity measurements or by measurement of the I-126 content of the samples and of reference samples containing known quantities of iodine.

APPENDIX VI

ANALYTICAL PROCEDURE FOR CF_3I COLLECTIONS

The detailed procedure for conversion of CF_3I absorbed on charcoal to a solution of an inorganic iodide is presented here.

1. Remove the charcoal from the cartridges and place in the separatory funnel shown in Figure 6-1.

2. Fill the gas burette with water and then add more than 20 ml of CF_3I carrier gas to the burette by displacing the water.

3. Pump ~20 ml of the CF_3I from the burette through the charcoal.

4. Flush out the tubing leading to the charcoal by momentarily opening the three-way stopcock to the atmosphere. Then turn off the pump.

5. Read the burette to note the exact amount of carrier added.

6. Transfer the charcoal to the combustion tube shown in Figure 6-2. (Note 1)

7. With the receiver tube half full of water, allow oxygen to flow at a rate of approximately 3 bubbles per second. (Note 2)

8. Ignite the top of the charcoal by gentle heating of the tube.

9. When the charcoal has burned to within 3-5 cm of the quartz wool (several hours) add a 6<u>N</u> NaOH solution to the receiver to fill the tube to three-quarters full and allow the combustion to proceed to completion.

10. Allow the tube to cool and rinse (water) the contents of the receiver into a one-liter plastic bottle containing a small quantity of H_2SO_3.

11. Rinse the combustion tube into the plastic bottle with successive portions of 6<u>N</u> HNO_3, 6<u>N</u> NaOH, and water and repeat until the last nitric acid rinse shows no brown color. Discard the combustion tube.

12. Transfer the contents of the plastic bottle to a separatory funnel, add ~100 ml of CCl_4 and extract the iodine by acidifying the aqueous phase with nitric acid and adding a small quantity of sodium nitrite.

13. Wash the CCl_4-iodine phase with two small portions of water and discard the aqueous phases.

14. Back-extract the iodine (as iodide) into 50 ml of dilute sulfurous acid and discard the organic phase.

15. Proceed with Step A-8 or Step B-4, Appendix V, for "low-level" or "high-level" samples, respectively.

NOTE 1: The main body of the combustion has a 25-cm inside diameter and is 80 cm long, and is made of carefully annealed Pyrex. The receiver tube has a 3.2-cm outside diameter and is 30 cm long.

NOTE 2: During the combustion, the oxygen flow rate should be carefully maintained as high as possible without heating the Pyrex tube to the softening point.

FIGURE 6-1
CF_3I CARRIER ADDITION APPARATUS

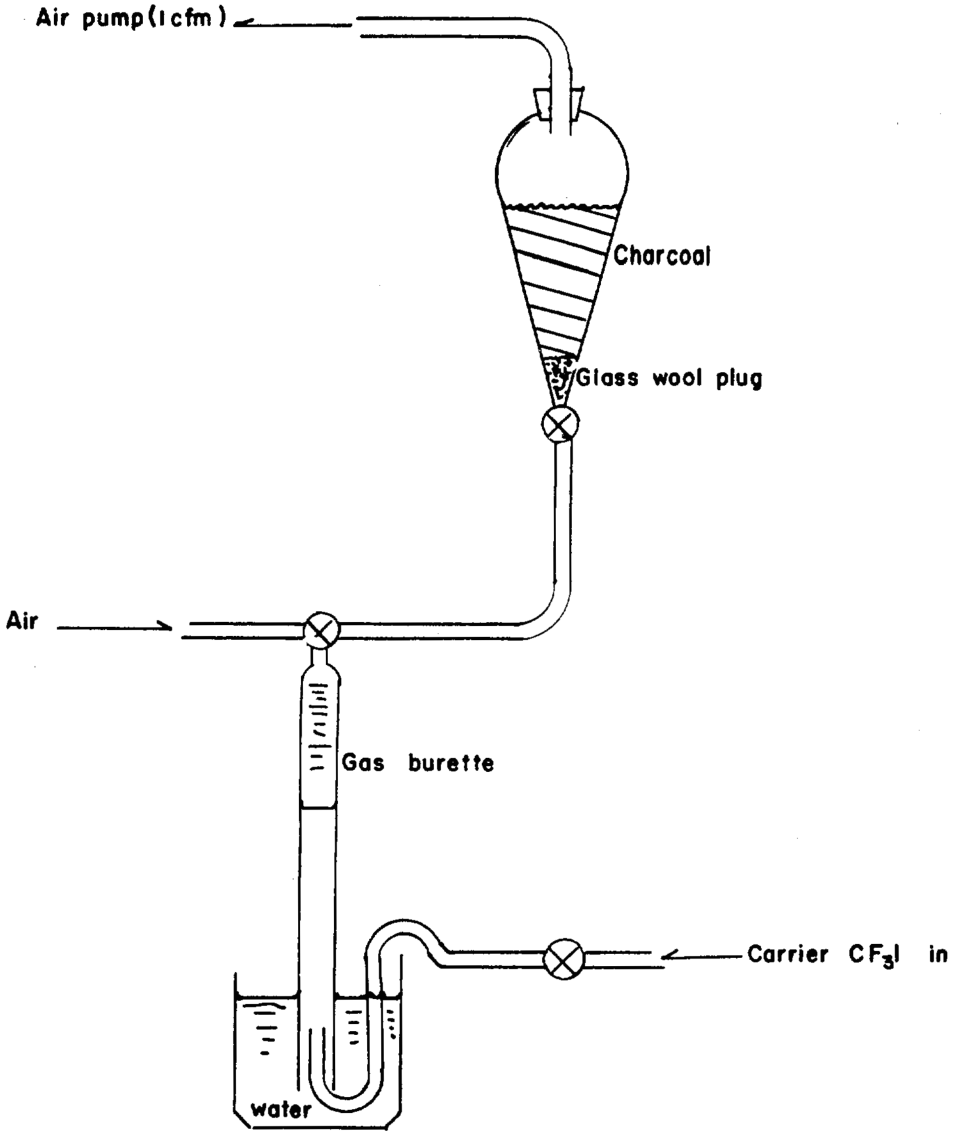
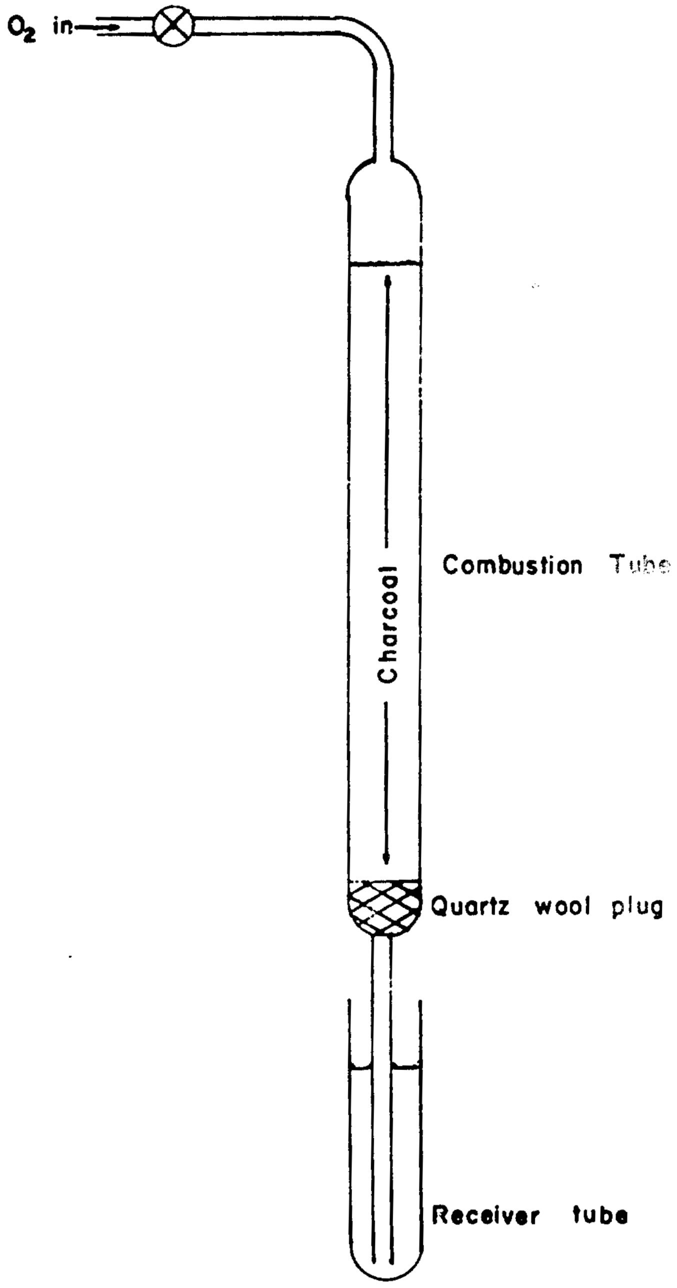


FIGURE 6-2
CHARCOAL COMBUSTION APPARATUS



APPENDIX VII

ESTIMATED COSTS OF ATMOSPHERIC TRACING WITH I-129 LABELED TRACERS

Experience derived from the experimental program is employed as a basis for making the following cost estimates for atmospheric tracing with I-129. The estimates provide for tracer synthesis, procurement of specially-required dissemination and collection devices, and measurement of I-129 in collected samples by the rather rigorous analytical procedures utilized in the present program. It is expected that additional developmental effort could yield significantly more simplified and rapid pre- and post-irradiation procedures and reduce the analytical costs.

The estimates assume the availability of (1) facilities and capital equipment for handling and processing gram quantities of iodine-129 and millicurie levels of other radioactive isotopes, (2) the necessary radioactivity measurement equipment for performing the activation analysis, and (3) a basic dissemination-collection grid with auxiliary meteorological instrumentation.

Costs associated with planning and carrying out the tracer tests (including labor devoted to actual dissemination and collection) and in evaluating the data may be assumed to be equivalent for alternative methods and are therefore excluded from this estimate.

The cost estimates are based upon the use of iodine-129 obtained in five-gram quantities at the current ORNL list price of $750/gram. Direct labor requirements are estimated in man-hours and then costed out at a rate of $17.50 per man-hour, which includes provision for indirect expenses. Miscellaneous supplies and expenses are stated at cost.

Estimates for the two tracer materials investigated are shown separately. In each case, it is assumed for illustration that five grams of iodine-129 would be processed and that five hundred samples would be analyzed. It is further assumed that half of the samples would require high sensitivity analyses owing to their anticipated low tracer concentrations.

A. Particulate Tracer Diiodofluorescein (DIF)

1. Synthesis		
I-129 5 grams	$3750	
Labor 32 man-hours	560	
Other Materials	200	
		4510
2. Collectors ~$10/100 units		500
3. Measurements		
a. Short-range ($>10^{-10}$g I-129/filter) 250 measurements		
Labor 400 man-hours	7000	
Materials and services	1875	
b. Long-range ($<10^{-10}$g I-129/filter) 250 measurements		
Labor 900 man-hours	15750	
Materials and services	3750	
		28375
Total		$33385

B. Gaseous Tracer Trifluoroiodomethane (CF_3I)

1. Synthesis		
I-129 5 grams	$3750	
Labor 40 man-hours	700	
Other Materials	400	
		4850
2. Collectors ~$300/100 units		1500
3. Special collection equipment (non-recurring)		400
4. Measurements		
a. Short-range ($>10^{-10}$g I-129/collector) 250 measurements		
Labor 500 man-hours	8750	
Materials and services	2500	
b. Long-range ($<10^{-10}$g I-129/collector) 250 measurements		
Labor 750 man-hours	13125	
Materials and services	5000	
		29375
Total		$36125

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