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A DIFFUSION-DEPOSITION TRACER SYSTEM

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April 1964

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A DIFFUSION-DEPOSITION TRACER SYSTEM

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ABSTRACT

A dual tracer technique has been developed for use in experiments in low-level atmospheric dispersion and deposition. The system is considered unique in two respects. First, it permits definition of the airborne concentrations of a cloud of small particles representative of a true aerosol and simultaneously the deposition of larger particles on the soil and vegetation beneath. Second, the deposition tracer consisting of Cu^{64} particles in the 1 to 10-micron range can be detected directly on the surface without the intervention of any artificial collecting mechanism or the necessity of inferring deposition by mass balance techniques.

Techniques, instruments, and facilities for the preparation, activation, emission, sampling, and measurement of both tracers are described in sufficient detail to permit reproduction. The report is also intended as a background reference for future analytical studies resulting from field experiments.

A DIFFUSION-DEPOSITION TRACER SYSTEM

INTRODUCTION

The Meteorology Group at Brookhaven National Laboratory has developed a system with which many parameters relating to two diverse tracers can be defined. The basic purpose of the development has been to permit accurate measurements of particulate deposition directly on ground and vegetation surfaces, as well as simultaneous definition of the diffusion pattern of a tracer approximating a true aerosol over the same sampling grid. In this report the techniques are described in sufficient detail to permit reproduction of portions of the system and to serve as a basic reference for subsequent analytical papers.

Insofar as the authors are aware, all previous deposition tracer techniques used in field studies have required either collection of particles on devices simulating natural surfaces or estimation of the deposition by inference from the decrease in the mass of airborne material. Both techniques have serious drawbacks. Whenever an artificial surface is substituted for natural ground or vegetation, the dangerous assumption is made that the deposition on each is similar. As far as the mass balance techniques are concerned, the assumption is made that the distribution of the airborne material is accurately known, and this also is difficult to establish.

The initial investigation of a tracer system for the Brookhaven work resulted in the selection of uranine dye as the diffusion tracer, since particles one micron and less in diameter can be produced. This material was first used as an atmospheric tracer by Robinson et al.,[1] and his methods have been adopted with relatively little change by the Meteorology Group. Major deficiencies of the uranine system (decomposition under heat or prolonged exposure to sunlight) are not problems under current test conditions.

Despite certain obvious disadvantages in handling and emission procedures, it was decided that a radioactive tracer offered the only possibility for reliable measurements of true deposition. Development of such a tracer presented considerable difficulty, since rigid specifications had to be met. For the initial experiments, a tracer was desired that was reasonably monodisperse, would not react chemically or physically with other constituents of the atmosphere, had a half-life ranging from 6 to 40 hr, and would leave almost no residual long-lived activity. Particles that were spherical in nature were also desired to simplify certain mathematical aspects of the analysis.

Extensive effort was devoted to the development of a particle-system using montmorillonite clay containing La[140], an isotope having a 40-hr half-life. All development problems were solved except that impurities in the clay left an excessive percentage of long-lived isotopes in the final tracer. It was apparent that 20 tests in one area would render the section unsuitable for further low-level studies because of high background.

COPPER PARTICLE TRACER

Cu^{64} was finally selected as the deposition tracer and has proved to be acceptable. The particles of naturally occurring copper (Cu^{63} and Cu^{65}) are obtained from the New Jersey Zinc Company as type 1003 copper powder. This consists of a polydisperse mixture of nearly spherical copper particles, ranging in diameter from <1 to $>100 \mu$. They are almost pure copper, and the very small amounts of remaining impurities do not readily become radioactive.

The decay scheme of Cu^{64}, which has a half-life of 12.8 hr, is as follows: 39% goes to the stable isotope Zn^{64} with the emission of a beta particle having a maximum energy of 0.57 MeV; 60% goes to Ni^{64}, which is also stable, by the emission of a positron of 0.66 MeV or by electron capture. There is a subsequent emission of two 0.511-MeV gammas accompanying the annihilation of each positron. Finally, there is a second electron capture phenomenon (0.59%) resulting in the 2+ state of Ni^{64}, with a 1.34-MeV gamma being released when the nickel returns to its ground state.[2]

The measuring techniques used by the Meteorology Group circumvent the necessity of determining precisely the counting efficiencies and geometries for the various emissions by calibration of
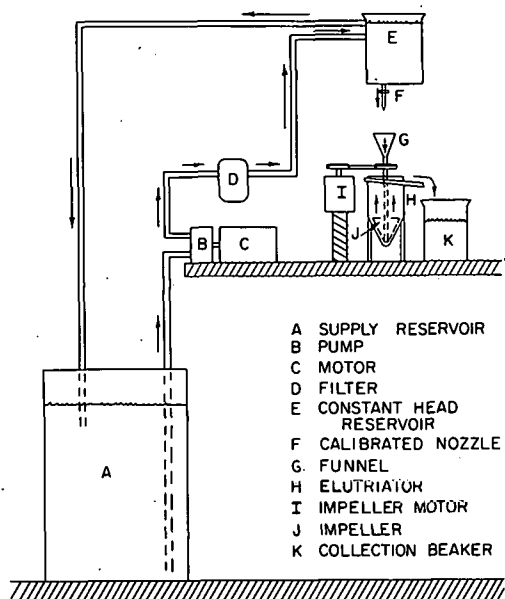
A	SUPPLY RESERVOIR
B	PUMP
C	MOTOR
D	FILTER
E	CONSTANT HEAD RESERVOIR
F	CALIBRATED NOZZLE
G	FUNNEL
H	ELUTRIATOR
I	IMPELLER MOTOR
J	IMPELLER
K	COLLECTION BEAKER

Figure 1. Diagram of elutriator system
showing directions of water flow.

the detection tubes and shielding assemblies with known amounts of Cu^{64} in configurations identical to those of the field samples.

Since the particles as received from the manufacturer are polydisperse, a sizing process is necessary prior to irradiation and emission. The method selected is a water elutriation system utilizing a device developed by Cooke[3] and evaluated by Wilder.[4] This instrument, diagrammed in Figure 1, utilizes the differential settling rates of the particles in water to accomplish the separation. Distilled water, containing suitable wetting and antifoam agents,* is fed into the base of the elutriator (H) by a metered nozzle (F) under a constant head of pressure. The particles, gently stirred by the two-bladed rotor, rise with the water flow whose velocity decreases as the funnel diameter increases toward the top. Thus, only those particles having a settling rate less than the final velocity of the water are carried off into the collector beaker (K). Successive runs with increasing flow rates separate the particles into the desired size groups.

Although elutriation is a slow and tedious method of producing sized particles, no better alternative has been found. Each complete separa-

*Aerosol C-61, American Cyanamid Co., and Antifoam B, Dow Corning Corp.

Table 1

Flow Rates and Times Required
for Elutriation of Copper Spheres

Size class	Flow rate, cc/min	Duration
Runoff 1*	15.2	114 hr, 45 min
Class I	21.5	8 hr, 15 min
Runoff 2	90	40 hr
Class II	114	2 hr, 30 min
Runoff 3	178	4 hr, 48 min
Class III	222	47 min

*Runoff refers to the discarded fractions beyond or between size classes desired.

Table 2

Copper Particle Characteristics

Size class	Mean mass diameter, μ	Mean mass, g	Mean settling velocity in air, cm/sec
I	2.7	9.17×10^{-11}	0.191
II	4.6	5.12×10^{-10}	0.57
III	9.45	3.92×10^{-9}	2.385

tion takes about a week and uses 115 gal of distilled water. At the beginning of the run, 200 g of copper powder are mixed with 700 cc of distilled water, ½ cc of Aerosol C-61, ½ cc of Antifoam B, and 8 cc of ethanol. This mixture is stirred vigorously until well mixed and is then placed in the elutriator. The purpose of the ethanol is to dissolve the wetting agent. The antifoam is necessary to eliminate the copious foam that would otherwise form.

Each gallon of distilled water for the supply reservoir (a 30-gal plastic-lined drum) is mixed with 3 cc of Aerosol C-61, 3 cc of Antifoam B, and 47 cc of ethanol before being added to the elutriator supply through a gauze filter. The water is then pumped through a Fiberglas filter to the constant head reservoir, from which it flows through a calibrated nozzle into the elutriator stem.

The overflow from the elutriator is caught in 4000-ml beakers. After all particles have settled, most of the water is decanted and the rest evaporated. The particles are then brushed into a vial for storage. From the initial 200 g of powder, <10 g of usable material are obtained, consisting of 1 g or less of the smallest particles (class I), 2 to 3 g of the medium size (class II), and 5 to 6 g of the largest (class III).

At the beginning of the run, all particles smaller than class I are removed by using a very low flow rate. The flow is then increased and the class I particles are removed, followed by successive classes as required. Table 1 gives the flow rate and time required for each group. Runoff refers to the discarded fractions between size classes. Table 2 gives the characteristics of each size class in current use.

While the elutriated particles are not monodisperse, they are restricted to a fairly narrow range of particle size. The standard deviation for each group is $<18\%$ of the mean diameter. A typical distribution is shown in Figure 2, and Figure 3 is a photomicrograph of particles in the class III size group.

It is apparent from Table 1 that preparation of a group of particles smaller than $1\ \mu$ would take too long to be practical. Experiments already completed indicate that both copper acetate and copper nitrate can be used in the same fashion as the uranine to provide adequate quantities of submicron particles. They are not spherical in shape, but this is less important in the smaller particles.

Activation

Brookhaven National Laboratory is well equipped to provide irradiation facilities permitting the preparation and immediate use of the copper particles. The Brookhaven Graphite Research Reactor contains pneumatic tubes designed for the insertion of samples in the core by remote control. At the normal operating power of 20 megawatts, the neutron fluxes in the two sample positions used are $\sim 6 \times 12^{12}$ and 1×10^{13} neutrons/cm²-sec, respectively.

The sample to be activated is weighed and folded tightly within a 3-in. square of pure aluminum foil. This package is then placed in an aluminum isotope can (Figure 4). Aluminum is used for the container, since its isotopes have very short half-lives, and after a brief "cooling" period the container offers no likelihood of contamination of the copper. In handling the sample, precautions are taken to avoid contamination from foreign materials that might have long-lived activity, such as sodium from perspiration.

The sample is inserted in one of the isotope tubes and left for periods ranging from 15 min to 16 hr, depending on the size of the sample, the irradiation tube flux characteristics, and the activity desired. Samples vary from $\approx$0.8 to 6 g, the amount increasing with particle size. Typical activities are
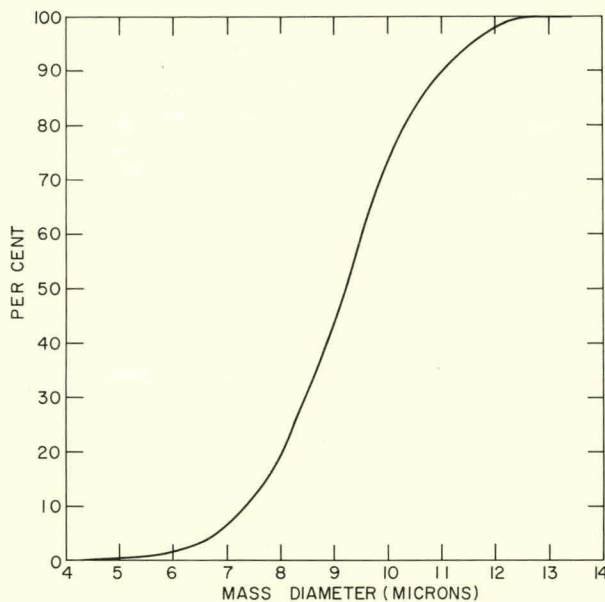

Figure 2. Cumulative mass distribution of class III copper particles.
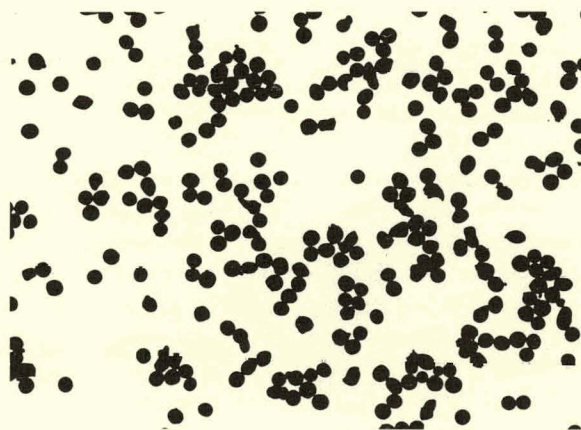

Figure 3. Photomicrograph of class III copper particles.

from 0.05 to 2.0 C/g with total activities of 0.3 to 2.0 C at field emission time. Attempts are made to obtain equal activity per particle regardless of size by using longer irradiation times for the smaller particles. After withdrawal from the reactor, each sample is stored for an hour or more to allow for decay of the activity in the aluminum can and foil. The sample is transported from the reactor in a lead "pig" and removed at the emission point with appropriate tools.

The activity of the sample is determined by two methods. The maximum specific activity of an iso-

tope exposed to a given flux is a constant and is reached when the rate of decay equals the rate of induced activity. At a constant flux, the activity after a time t is given by

$$A = A_{\max}(1 - e^{-0.69t/T})$$

where

$A =$ specific activity (C/g),
$A_{\max} =$ equilibrium specific activity (C/g) (a constant for a given isotope and neutron flux),
$t =$ irradiation time (sec), and
$T =$ half-life (sec).

Because the reactor flux is known only approximately, another procedure is used for a more precise determination. A second, identical sample is irradiated in the same can with the sample for emission. The Analytical Radiochemical Group of the Hot Laboratory Division then determines the specific activity of the second sample by reliable analytical procedures. This value is then applied to the sample used in the field. As a third check, a weighed piece of copper foil is included in the isotope can and analyzed in a similar fashion. The three determinations are usually rather close.

Suspension

The very small amounts of copper used in a given operation make dispersion of the dry powder at a uniform rate over an extended period virtually impossible. Therefore the particles are introduced into a much larger volume of liquid which is eventually sprayed into the atmosphere. Transferring the particles from the aluminum foil to the liquid without loss or undue exposure to radiation is accomplished by dropping the foil packet into a solution of HCl. This quickly dissolves the aluminum and forms aluminum chloride, which is water soluble. Since aluminum chloride is hygroscopic, there is a possibility that some of the material with associated water adheres to the copper particles and thus increases their effective diameter. However, calculations indicate that an increase in diameter is nearly counteracted by a reduction in effective mass; hence there is little change in settling rate. After the aluminum is dissolved, additional liquid with the same wetting and antifoam agents used in the elutriator is added to reduce the acidity of the solution and increase its volume. The liquid is then stirred vigorously for at least 20 min before emission. Tests indicate that mixing is uni-
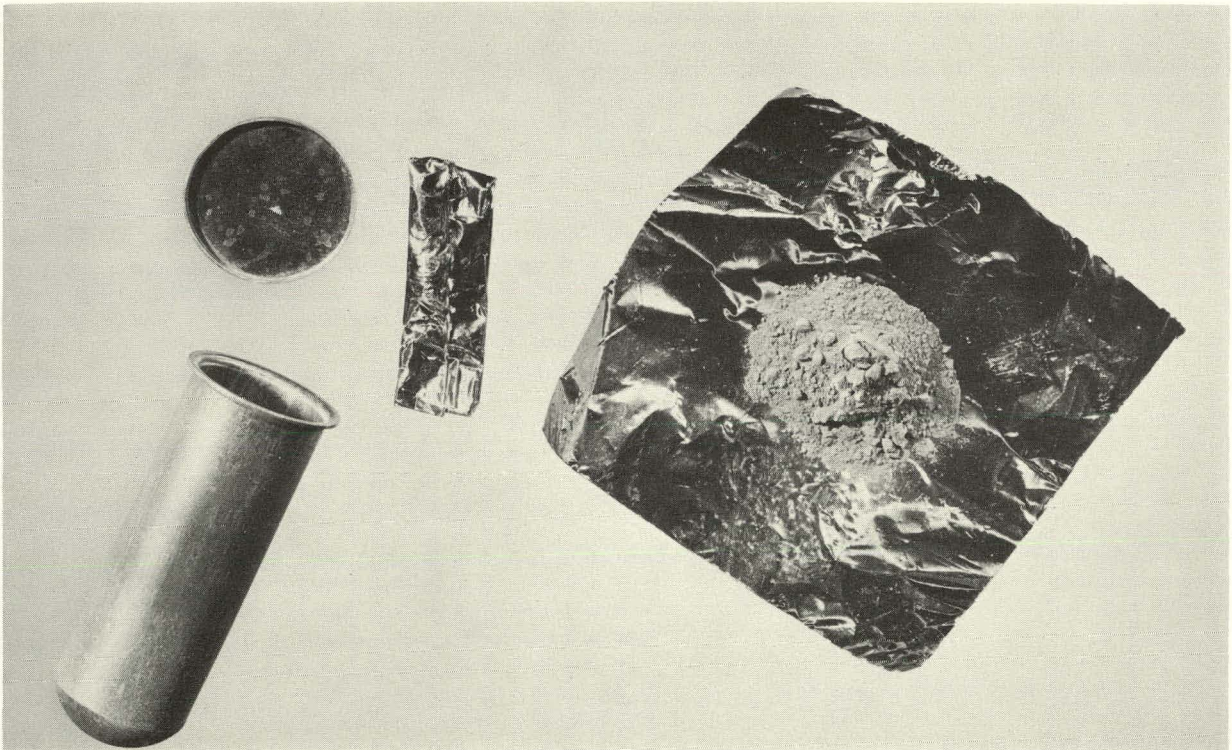

Figure 4. Preparation of copper particle sample for irradiation.

form and that the number of particles emitted is proportional to the volume of fluid emitted. The liquid mixture used contains the following:

20 cc 37.9% HCl
670 cc H_2O
10 cc isopropyl alcohol
2 drops Aerosol C-61
4 drops Antifoam B

Emission

The emission apparatus is mounted on a movable stand (Figure 5). A shelf whose height may be adjusted from ≈ 0.3 to 3.2 m supports two square open-topped boxes and two keyed rods. The boxes are composed of ½-in.-thick lead on three sides and the bottom and of transparent plastic on the front. A square glass container for the liquids to be emitted fits snugly into each box, one container
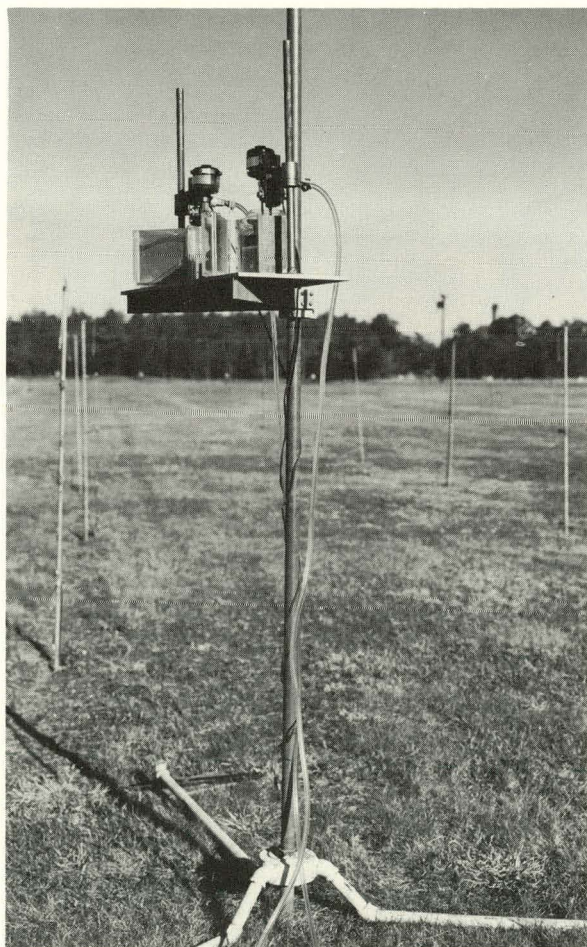

Figure 5. Emission stand in sampling grid.

holding the uranine and the other the copper suspension. A mirror arrangement permits inspection of the liquid levels from the rear. The atomizing nozzle and the stirrer comprise a unit. When slid down the keyed rods the stem of the nozzle and the stirrer blades are positioned properly in the glass containers, and the nozzle opening is 22 cm above the shelf. The stirrer speed is controlled by a Variac at least 10 ft in the rear, while the nozzles are connected by quick-disconnect fittings and tubing to tanks of compressed air at the same distance to the rear.

The system for producing the cloud of droplets is important. To minimize initial agglomeration and to insure rapid evaporation the drop size should be small. The ideal is a drop that cannot contain more than a single particle, but the compromise reached in this application merely makes the probability of >1 particle/drop low. For example, the spray from 700 cc of water contains $\approx 1.35 \times 10^{12}$ drops 10 μ in diameter or $\approx 4.9 \times 10^{10}$ 30-μ drops. Five grams of class I copper particles contain $\approx 1.28 \times 10^9$ particles. If these are sprayed out in 10-μ drops, there is ≈ 1 particle/1000 drops, but in 30-μ drops there is 1 particle/38 drops. Even here the probability of having 2 particles/drop is only $\approx 3.4 \times 10^{-4}$.

Several types of atomizing nozzles have been used and none is completely satisfactory. The desired characteristics of uniform flow rate and small uniform droplet size are difficult to attain. Initially, the glass Chicago atomizing nozzle (Figure 6) was used, but this gave unexplained variations in flow rate from one test to the next, and the nozzles dif-

Figure 6. Chicago atomizing nozzle.

fered greatly one from another. The droplet diameter averaged $\approx 40\,\mu$ with no excessively large drops. A Spraying Systems Company flat nozzle combination F1 was next tried. The output was somewhat greater and more uniform, but the main droplet size was much larger, $\approx 120\,\mu$, and a number of even larger drops were produced under some conditions. This was replaced by a similar round spray nozzle (Figure 7) which can be made to produce droplets with a mean diameter as small as $10\,\mu$, with no large drops, by increasing the air to liquid ratio. However, the flow rate is then so low that two nozzles are required to obtain the desired rate of output, and each nozzle must be individually adjusted before each use.

Emission rates have averaged between 10 and 20 cc/min for both uranine and copper tracers, with runs ranging from 20 to 40 min. Emission is normally continued until the liquid level in one of the containers reaches the bottom of the nozzle stem. The volume emitted is determined from the difference between the initial volume and the remainder.

With the smaller droplets (10 to 20 μ), evaporation takes place within a few feet of the nozzle at normal humidities, and with droplets $>100\,\mu$ in diameter, evaporation seemed to be complete within 5 to 6 m, except in one test under inversion conditions, when the relative humidity was near 100%, the temperature near 32°F, and the wind speed <0.2 m/sec. As a result, the liquid droplets remained visible for >50 m.

Cooling produced by expansion of the compressed air and evaporation of the liquid causes an initial downward displacement of the plume so that the effective source height may be as much as 30 cm less than the actual nozzle height. This effect is less pronounced at higher wind speeds.

Autoradiographs of particles deposited on sheets of x-ray film laid on the ground in light-tight envelopes and microscopic examination of slides also placed on the ground indicate that most particles are single and that agglomeration is not significant.

AEROSOL TRACER

Uranine

Uranine (fluorescein sodium) is a water and alcohol soluble compound which fluoresces in solution. For emission, 3 g are dissolved in 450 cc of methanol and 300 cc of H_2O. The solution is made shortly before use and is not exposed to light until emission to minimize any possible decomposition. The same type of spray nozzle is used for uranine as for copper. When sprayed from the atomizing nozzle, the liquid quickly evaporates and leaves crystalline particles of uranine. Since the size of these particles depends on the strength of the solution and the droplet size, a fairly wide range of sizes is possible with any given volume of liquid emitted. However, the desired length of the test, the nozzle output capacity, and the lower limit of detectability at the most distant sampling point must all be considered in selecting a concentration. With the amount listed above, most particles are $<1\,\mu$ in diameter and the quantity emitted is adequate for detection at the farthest points of the sampling grid.
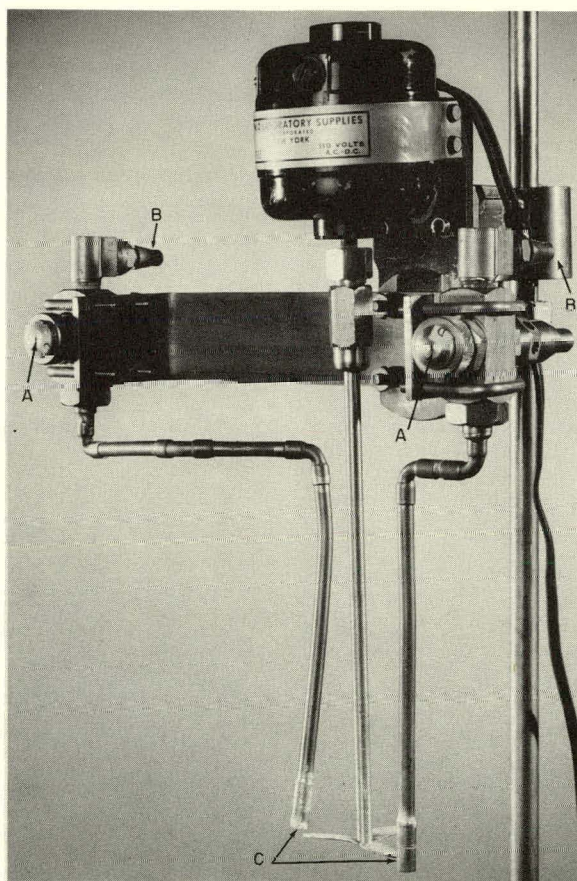

Figure 7. Dual round spray nozzle and stirrer unit. A, nozzles; B, compressed air connections; C, siphon stems.

SAMPLING AIRBORNE CONCENTRATIONS

Sampling Grid

The sampling grid at Brookhaven (Figure 8) is an area measuring 100×100 m, centered in a large, level, grassy field. The 100 basic sampling locations are arranged in a square array at 10-m intervals. At each location a pipe rises from an underground vacuum system. Filter samplers are installed on each pipe at 0.1, 1.0, and 2.0 m (Figure 9) by quick-disconnect fittings. At each position a mercury manometer and a control valve are available to set the flow to the desired value.

In addition to the 100 basic positions, 92 additional sampling positions are located at 5-m intervals in the north, west, and southwest portions of the field. These positions are equipped with short pipes from the vacuum system to ground level, capped when not in use. A portable stake is normally placed on each of the 12 stubs nearest the emission point to increase the density of coverage in this area. Two of these stakes have additional sampling levels at 0.5 and 1.5 m.
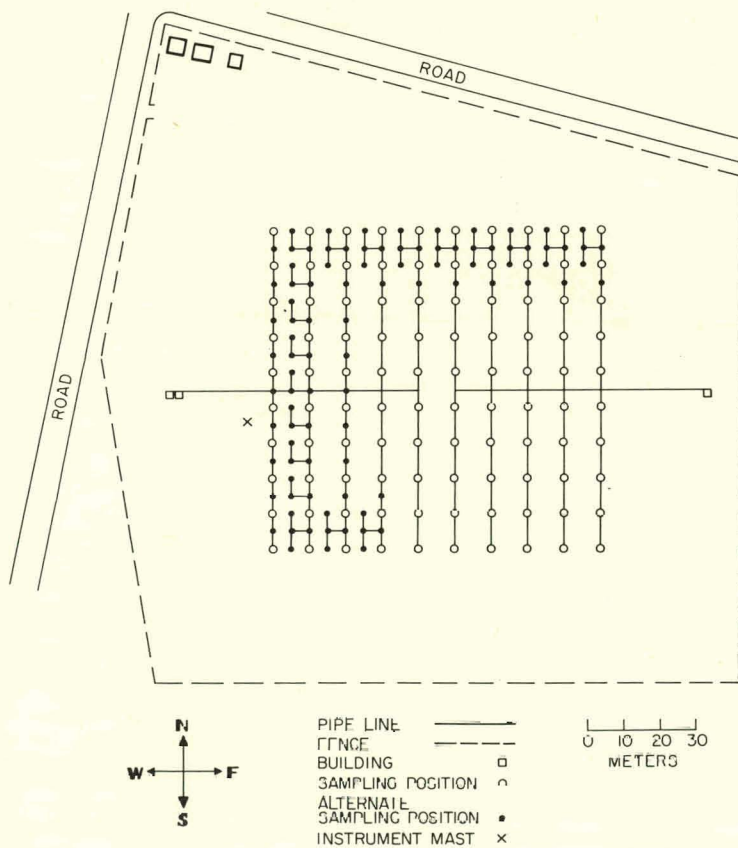
The two Kinney model DVD8-8-10 vacuum pumps have a combined theoretical displacement of 220 ft³/min. However, the filters used vary in their resistance to air flow, and it is necessary to select an operating flow that can be obtained through all filters. The flow normally used is 0.45 ft³/min, which gives a total flow of 153 ft³/min when all 340 filters are in use. Since this total flow is well below the capacity of the pumps, adjustments can be made to the filter flows without affecting positions previously adjusted.

Filters

Type AA Millipore filters 1 in. in diameter are used in Union Industrial Equipment Co. holders
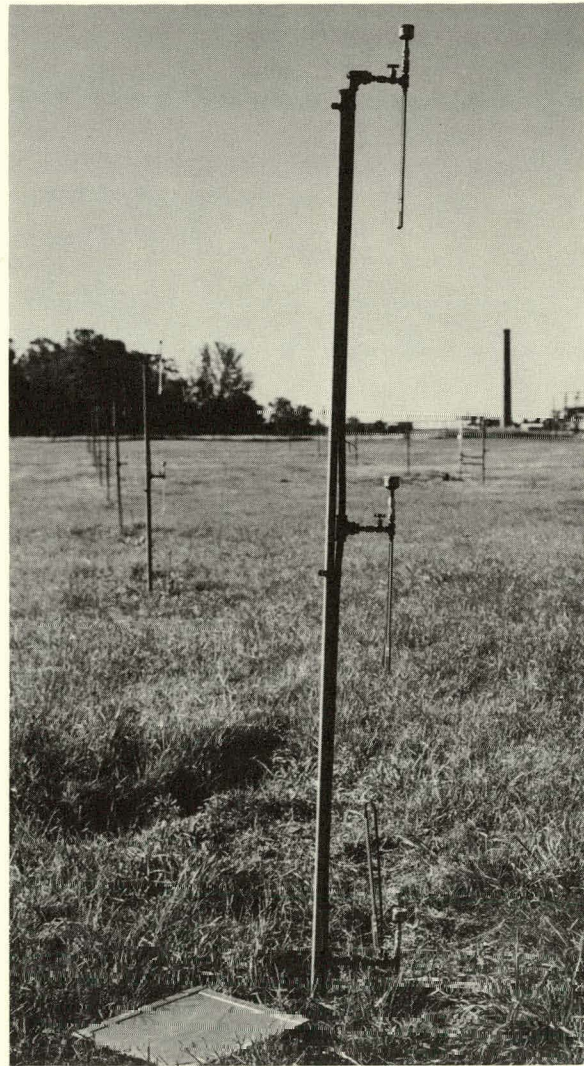

Figure 8. Map of sampling grid.

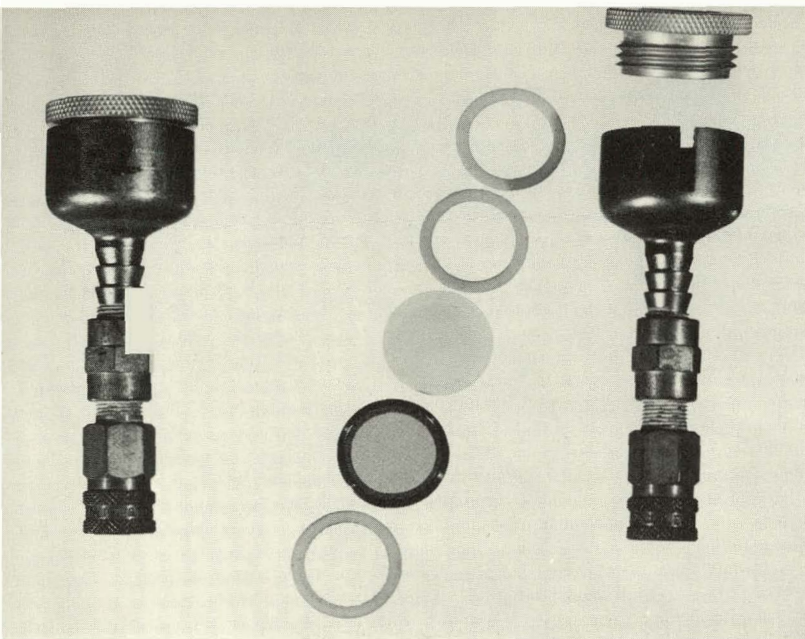Figure 9. Sampling position in the sampling grid.

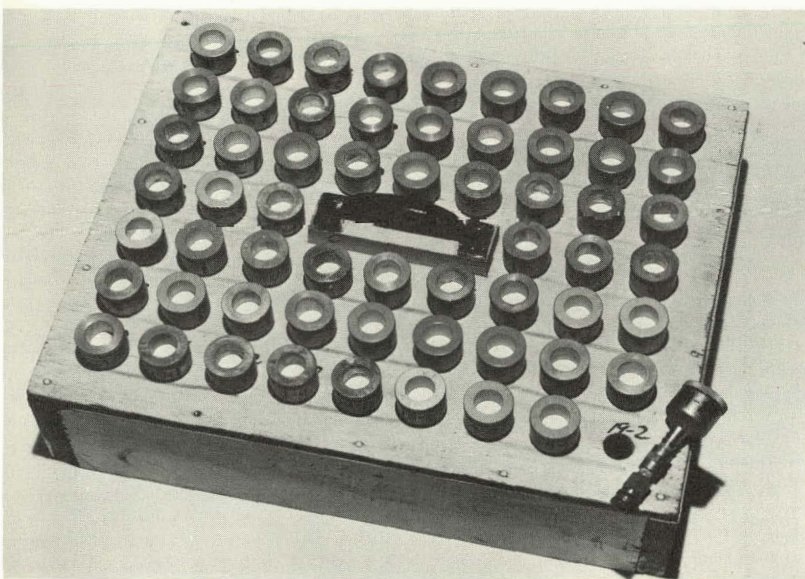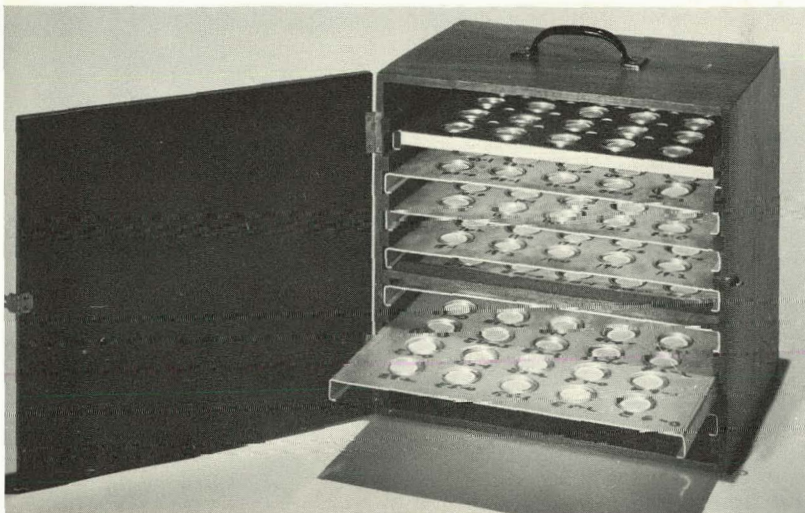Figure 10. Assembled and disassembled filter holders.

Figure 11. Filter holder carrier.

Figure 12. Filters in planchet box.

(Figure 10). The filters are taken to and from the sampling grid in numbered carriers (Figure 11), and after each run the individual filters are removed from the holders and transferred to clean aluminum planchets (Figure 12).

Analysis of Filters

The filters are analyzed immediately for radioactivity, and after a lapse of 1 to 2 weeks a fluorometric analysis is made to determine the uranine content.

Radiation on the filters is detected by two Anton type 1001T "pancake"-shaped tubes operating in the Geiger region and mounted in an Anton No. 6001 sample changer which gives nearly 100% geometry (Figure 13). These tubes are almost completely insensitive to gamma radiation but detect both the β^- and β^+ emissions from the Cu^{64}. Since there is little attenuation of radiation through the Millipore filter, use of two tubes doubles the count rate from a given sample. Conventional power supplies, timers, and scalers are included in the counting system. Two such systems are used to decrease the counting time. Counts range from 1 to 5 min in length, depending on the activity of the sample, and frequent background counts are taken. Because of the short half-life of Cu^{64}, the elapsed time from t_0, the start of emission, is recorded and the counts are corrected for decay. By using calibration procedures to be described later and the flow rate through the filters, counts are converted to C/m^3 of air sampled at each position. They are then normalized by division by the output rate in C/sec.

After the radiation has decayed, the filters are analyzed for their uranine content in a fluorometer designed and built at Brookhaven (Figure 14). Each sample is exposed to a G.E. type B-H6 ultraviolet light source beamed through a Corning No. 5860 optical filter which transmits light having wavelengths from 3200 to 3900 Å with a peak at 3650 Å. The light emitted from the uranine is filtered by a Corning type 3484 filter passing wavelengths above 5070 Å, and is then received by a type IP21 photomultiplier tube. The output of the tube is amplified and displayed on a meter. The fluorometer has 5 ranges and can read solutions from 1×10^{-9} to 5×10^{-5} g of uranine/ml. Because of the sensitivity of the instrument, extreme care is taken in the analytical procedure to avoid fluorescent contamination, since nearly all substances fluoresce to a minor degree. The small glass con-
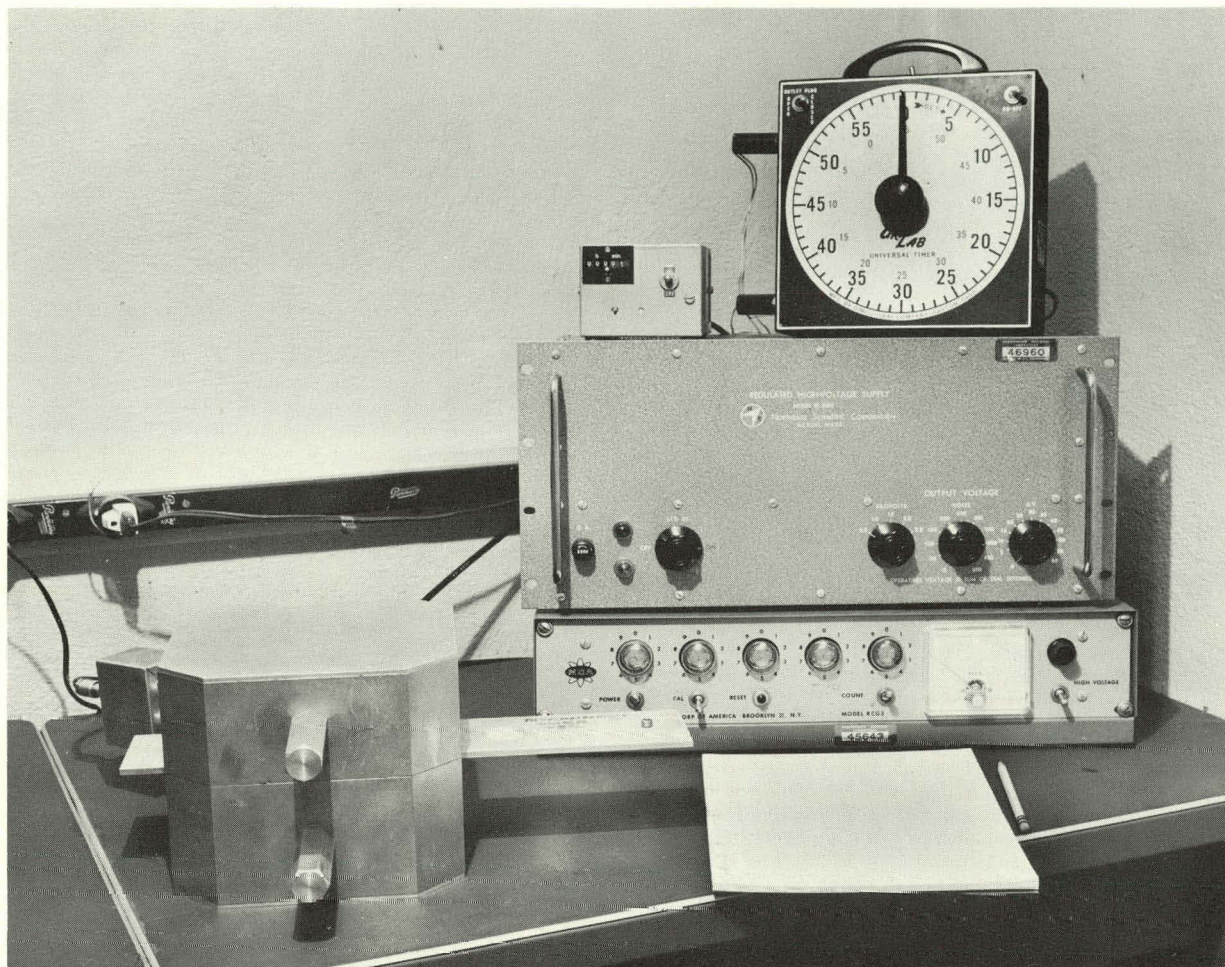
Figure 13. Equipment used for counting beta radiation on filters and ashed samples.
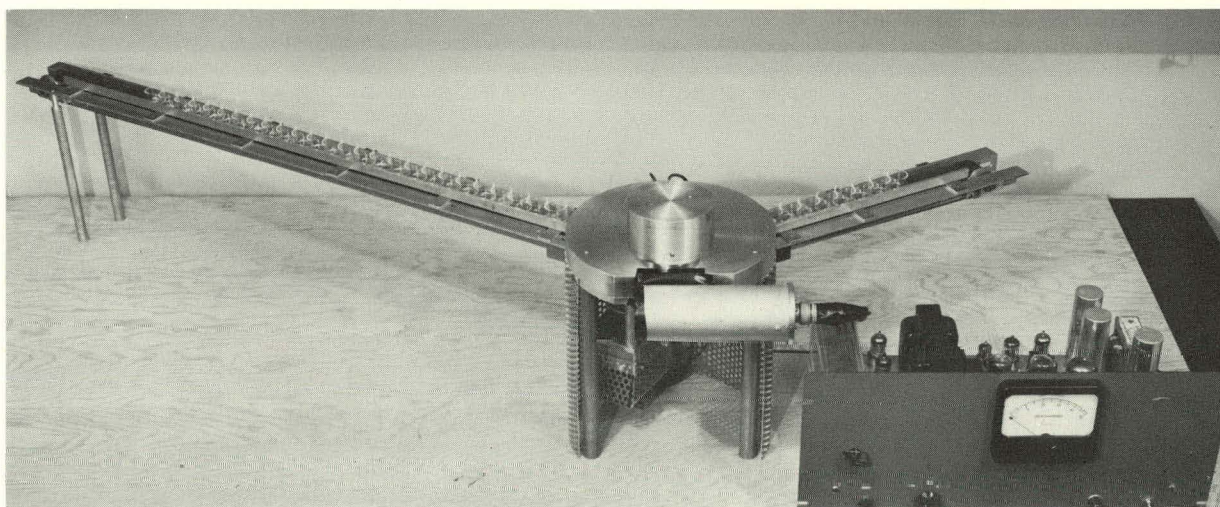

Figure 14. The Brookhaven fluorometer.

tainers in which the uranine is washed out of the filters are cleaned ultrasonically and handled only with clean tweezers. The filters are also handled with clean tweezers, which touch only the unexposed edges. In preparation for analysis, 3.5 cc of distilled water are placed in a container by use of an automatic pipette. A background reading is taken on the glass and distilled water. The filter is then washed in this water and removed, and a reading is taken of the solution. The instrument background (the dark current of the photomultiplier) is taken at each reading and substracted from the sample or background reading. The background reading is then subtracted from the sample reading, and the true concentration is found by reference to a calibration curve. The fluorometer is calibrated by reading a series of standards of known dilution. To minimize decomposition, these standards are stored in the dark under refrigeration and are kept no longer than one month. The fluorometer readings are finally reduced to g/m³ of air sampled and normalized by the output rate.

Recent Changes in the Sampling Grid

The field vacuum system was constructed early in 1961 and was used during the summer and fall of 1961 and 1962. The plastic pipe used has proved very unsatisfactory. Because of expansion and contraction, leaks constantly developed in the system and much time was spent in repair. The field is being repiped with copper tubing. The basic plan is left unchanged, but the number of alternate positions has been increased to 136, each equipped with a quick-disconnect fitting. Each sampling position is fitted with a mount for a Roto-slide sampler[5] as well as a filter (Figure 15).

This last change is necessary to obtain reliable measurements of the airborne concentrations of the larger particles. It was recognized initially that the efficiency of the filter units in sampling such particles would be low and that it would be a function of wind speed. Unfortunately, even the detailed measurements of wind in the field coupled with wind tunnel studies do not adequately define the field collection efficiency.

In order to read copper impacted on slide edges, a new counting system has been designed and constructed (Figure 16). The slide is positioned between two 2-in.-diameter, thallium-activated, sodium iodide crystals mounted on Dumont type 6342-A multiplier tubes. The 0.511-MeV positron annihilation gammas, which are emitted in pairs
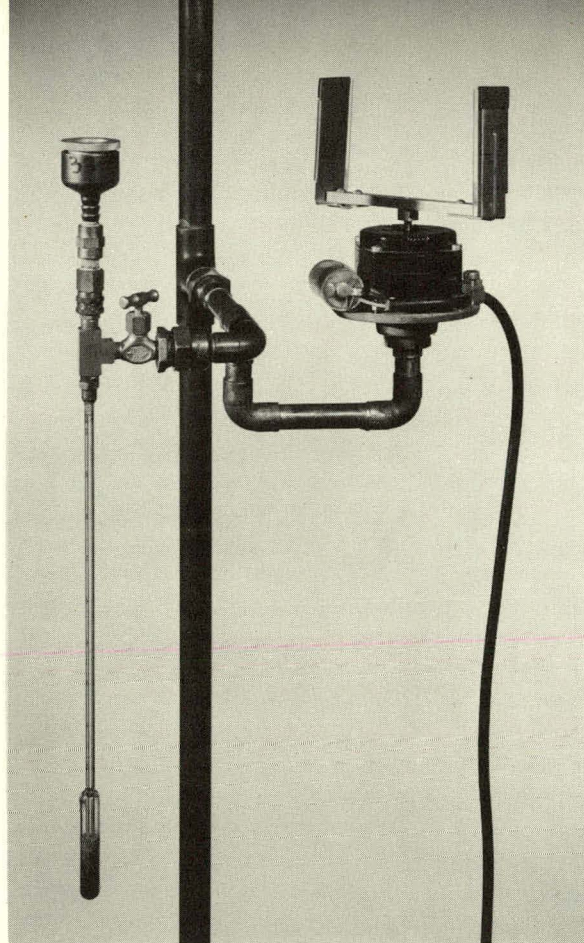

Figure 15. Sampling position in the sampling grid after reconstruction in 1964 showing both a filter and a Roto-slide sampler.

at 180° angles to each other, are counted by two separate circuits. The two counts are fed into a coincidence circuit having 1-μsec discrimination, so that only simultaneous counts on the two detectors are recorded as the output. This system eliminates most background interference, and the physical arrangement achieves nearly 4π geometry.

DEPOSITION MEASUREMENT

Copper deposition measurements are taken routinely by two methods, mobile radiation counters and sticky paper. Additional information is obtained by use of autoradiographs and other types of deposition surfaces. No attempt has been made to measure deposition of uranine routinely.

Mobile Radiation Counters

Two mobile radiation counters (Figure 17) are used to survey the sampling grid after each run. An Anton 1001T Geiger tube is suspended from the cart axle $\simeq$20 cm above the surface. The tube
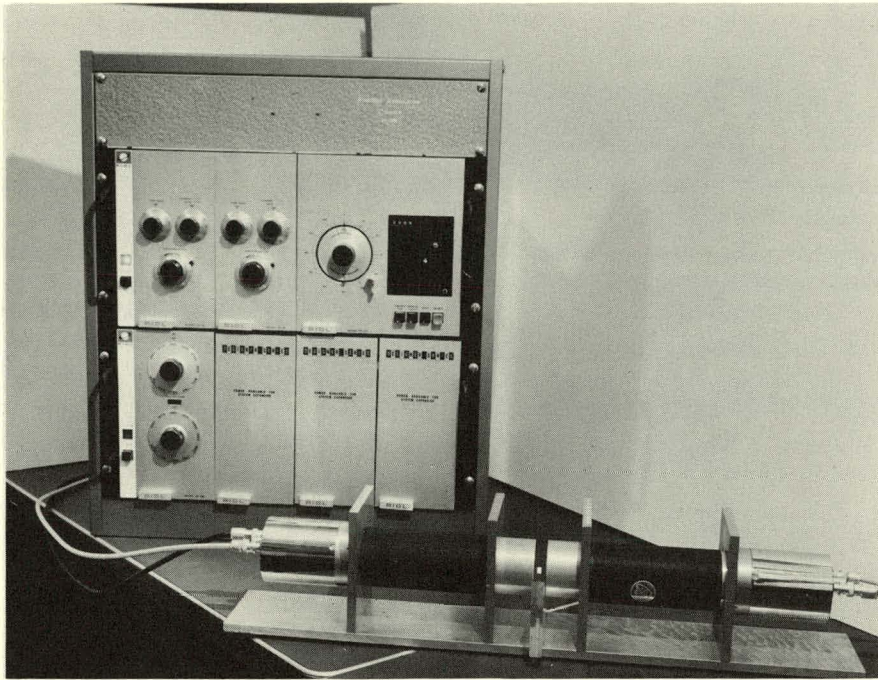
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Figure 16. The positron annihilation counter for counting radiation on slide edges. When the counter is in operation, the detection section is shielded with lead.
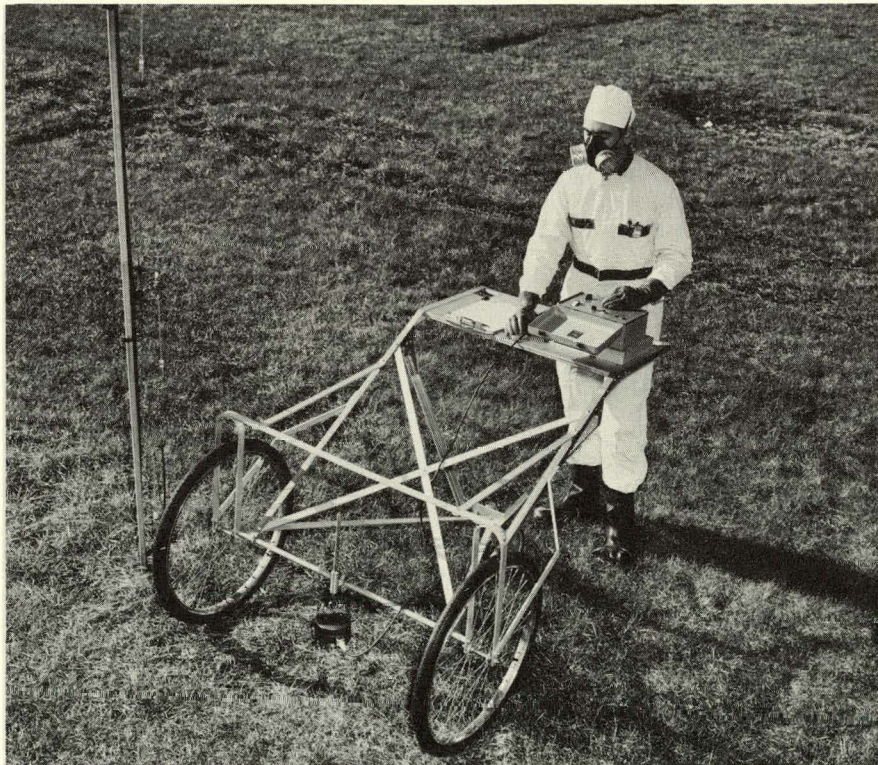

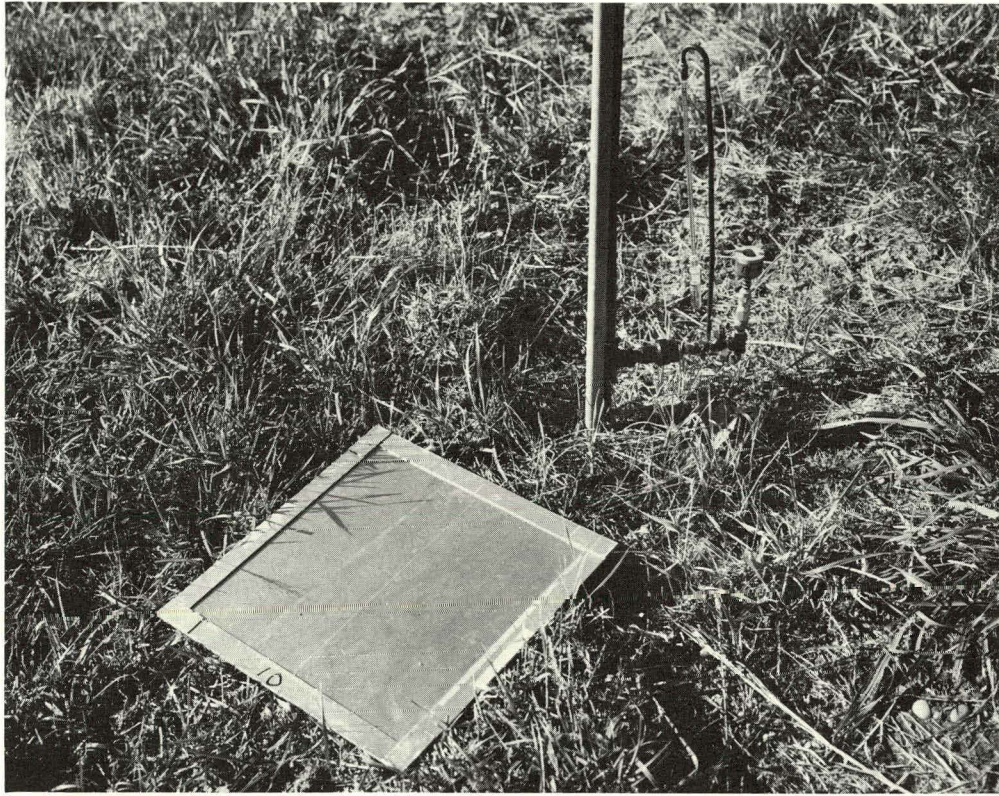Figure 17. Measuring deposition in the sampling grid with the mobile radiation counter.

Figure 18. A sticky paper deposition sampler in position in the sampling grid.
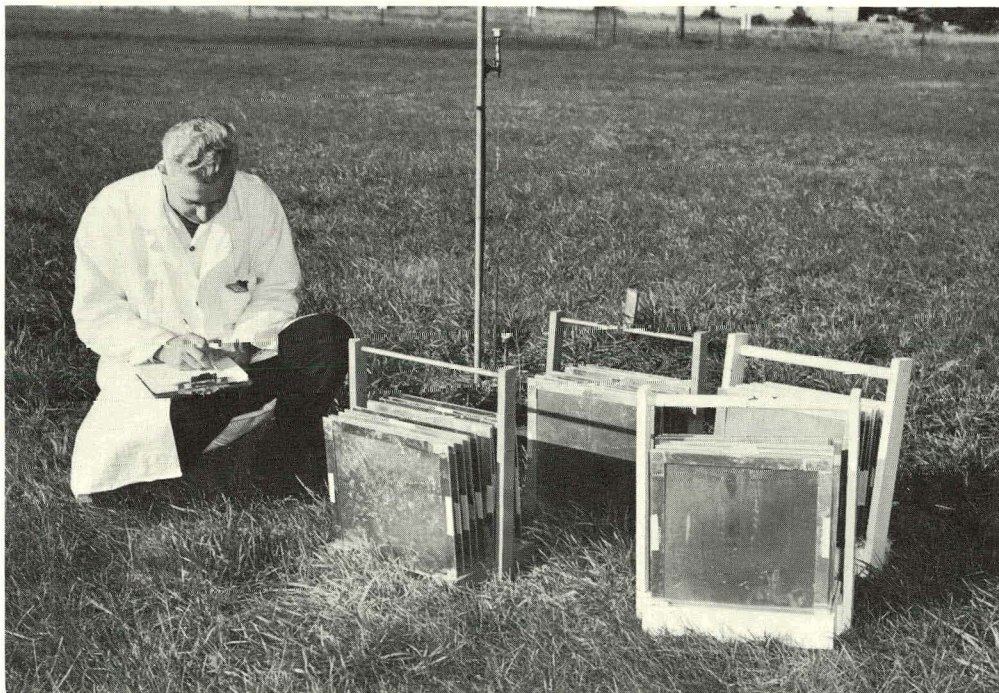

Figure 19. Carriers for transporting sticky paper samplers.

is shielded by lead in such a way that it is exposed to radiation only from a 1.52-m-diameter circle on the ground. The tube is connected to a battery-powered transistorized power supply and scaler mounted on the shelf of the cart. The operator pushes the cart to a designated location and takes a 1 to 5-min count, depending on the intensity of the radiation. Attempts are made to obtain counts at least 4 times background if this can be achieved in 5 min. Background measurements are made in the field before emission and in uncontaminated areas at frequent intervals during the measurement period. Counts are normally taken at each of the 100 basic sampling positions and in as many more locations as are necessary to define the deposition pattern. A complete survey of the field normally takes ≈ 4 hr with the 2 carts. If some areas are too radioactive to be counted immediately, they are surveyed later. Data obtained are corrected for decay, converted to C/m^2, and normalized by the output rate.

Sticky Paper Technique

Sheets of Eze-stick (1.5-mil acetate coated with 710 adhesive)* are inserted in aluminum frames exposing 1 ft² (930 cm²) of surface, and these are placed on the ground at each sampling location

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*Simon Adhesive Products Corp., 35-02 48th Ave., Long Island City, N.Y.
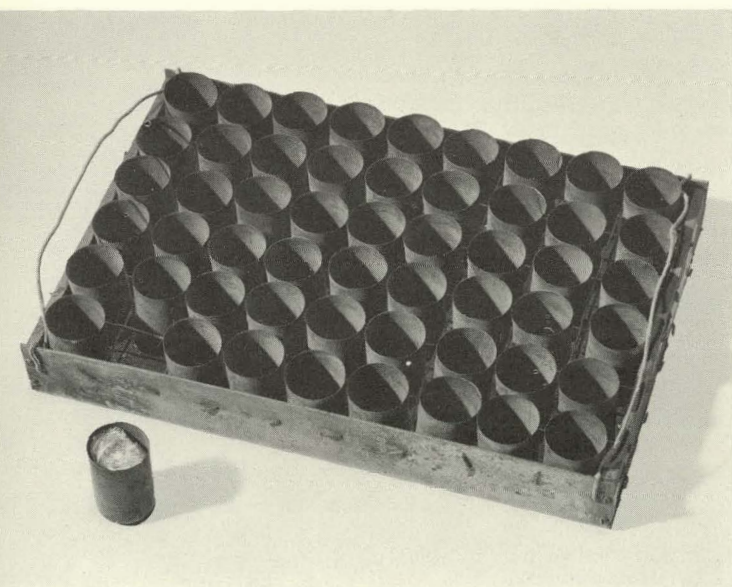
Figure 21. Ashes from a sheet of sticky paper sealed with tape in a 1-in.-diameter planchet.

and at ≈ 40 additional locations in the area of expected maximum deposition (Figure 18). In addition, several sheets are exposed upwind for background measurement. Special carriers (Figure 19) are used to transport the frames to and from the sampling positions. After the run the frames are taken to a worktable where the paper is cut from them by two-man teams working carefully to avoid cross contamination. The paper is folded sticky side in, refolded into a small bundle, and placed in a small steel can. These cans are placed in steel racks (Figure 20) and ashed at 600°C for ≈ 2 hr in a gas-fired oven furnace. After cooling, the ashes are poured into 1-in.-diameter aluminum planchets and covered with a layer of transparent tape (Figure 21). They are then analyzed in the same counting apparatus used for the filters except that only the upper tube is used. The counts are corrected for decay, converted to C/m^2, and normalized by the output rate.

The values obtained with the sticky paper technique are normally lower than those measured by the cart. This is to be expected, since deposition is greater on the rough grass surface than on the smooth paper. Attempts were made to relate the two methods. Two 3-ft² boards were covered with sticky paper. After exposure at different distances from the source, cart readings were taken over the boards. The paper was then ashed and analyzed. Because of various difficulties, these comparisons have not been successful, and further tests are required.

Evaluation of other deposition surfaces and deposition collectors is planned. These will include

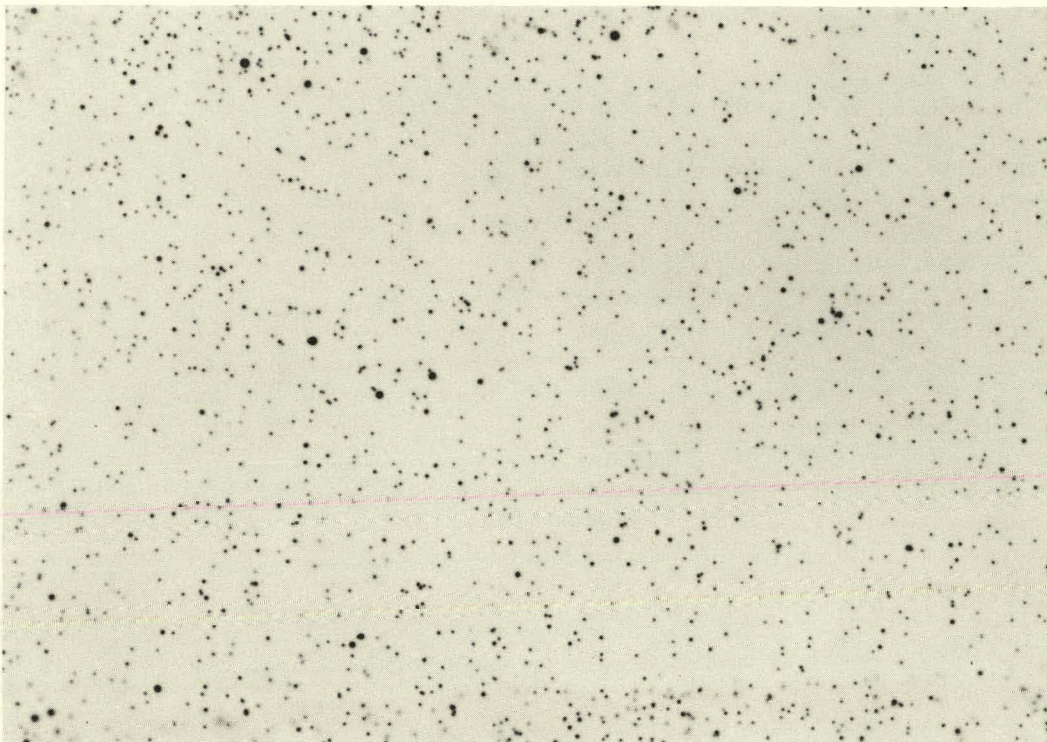F   20. Cans and racks used for ashing sticky paper samples.

Figure 22. Autoradiograph of radioactive copper particles deposited on an envelope containing x-ray film.

grass grown on filter paper and merged with the natural grass in the field, artificial grass, actual and simulated soil surfaces, glass plates, and various mechanical devices.

Autoradiographs

The distribution of particles deposited on the ground and the relation between number of particles and radiation reading are studied by use of autoradiographs. Two techniques are used. In the first, 8×10-in. sheets of Kodak type AA industrial x-ray film in light-tight envelopes are placed on the ground. After the particles have deposited, they are sealed against the envelope with a sheet of sticky paper. The film is then set aside and exposed until the Cu^{64} has decayed. It is then developed, and a dark spot is found on the negative corresponding to each particle. Tests show that the spot size is basically related to the beta range in the material and is not primarily a function of particle size or activity. Large, highly active particles give spots not significantly larger than small, lightly irradiated particles. The distribution of spots shows that deposition is uniform and is randomly distributed over the sampler areas (Figure 22).

The second technique involves placing microscope slides or larger plates of glass covered with an adhesive on the ground. After deposition has occurred, the glass is placed in contact with an envelope of film for exposure. With this method the actual particles can be examined and sized under a microscope and compared with the spots produced on the film.

When film is exposed in the field, one or more exposures are made upwind of the source to determine particulate background, which has been zero in all tests so far. Certain film used earlier, however, was found to have been sealed in envelopes that were not completely light-tight. In the presence of strong sunlight, numerous tiny holes produced spots on the film similar to those made by radioactive particles and caused considerable concern until the true source was discovered.

CALIBRATION PROCEDURES

The radiation detectors are calibrated in a simple manner which renders unnecessary the determination of individual efficiencies for each type of emission and for each component. Several filters

representing a wide range of activities are selected from those exposed in the field. Each is counted for at least 5 min in both counting systems, and the background is substracted. These filters are then sent to the Hot Laboratory where they are dissolved by the Analytical Radiochemical Group in an acid solution and diluted to a specific volume. An aliquot of the solution is counted for gamma radiation in a well-type counter using a NaI crystal previously calibrated for Cu^{64}. By comparison with the calibration, the total activity in disintegrations per minute is determined. The ratio of the counts/min on the counters used in these studies to the actual dis/min defines the counter efficiencies. The counters are calibrated for ashed samples in an identical manner.

The mobile radiation counter is calibrated by irradiating 6 strips of 1-mil copper foil 6 in. wide and 3 ft long. The foil is laid flat on the floor in a 3-ft square, and the cart-borne detector is centered above it (Figure 23). Previous tests with a point source of radiation show that an area of this size contributes 96% as much to the detector as an area of infinite size. Several 5 to 10-min counts are taken with both carts to provide appropriate background counts at a location in the same room but beyond the influence of the copper. After subtraction of background the counts are divided by 0.96, which gives a calibration for an infinite area of uniform activity.

Six 1-in. squares of copper foil are irradiated in the container with the large strips. Their specific activity is determined by the Analytical Radiochemical Group as for the field samples. With the activity per square inch of foil known, a correction factor to convert the net field counts to C/m^2 is given by the equation

$$F = 6.98 \times 10^{-10} \frac{A}{n/0.96},$$

Figure 23. Calibration of the mobile radiation counter.
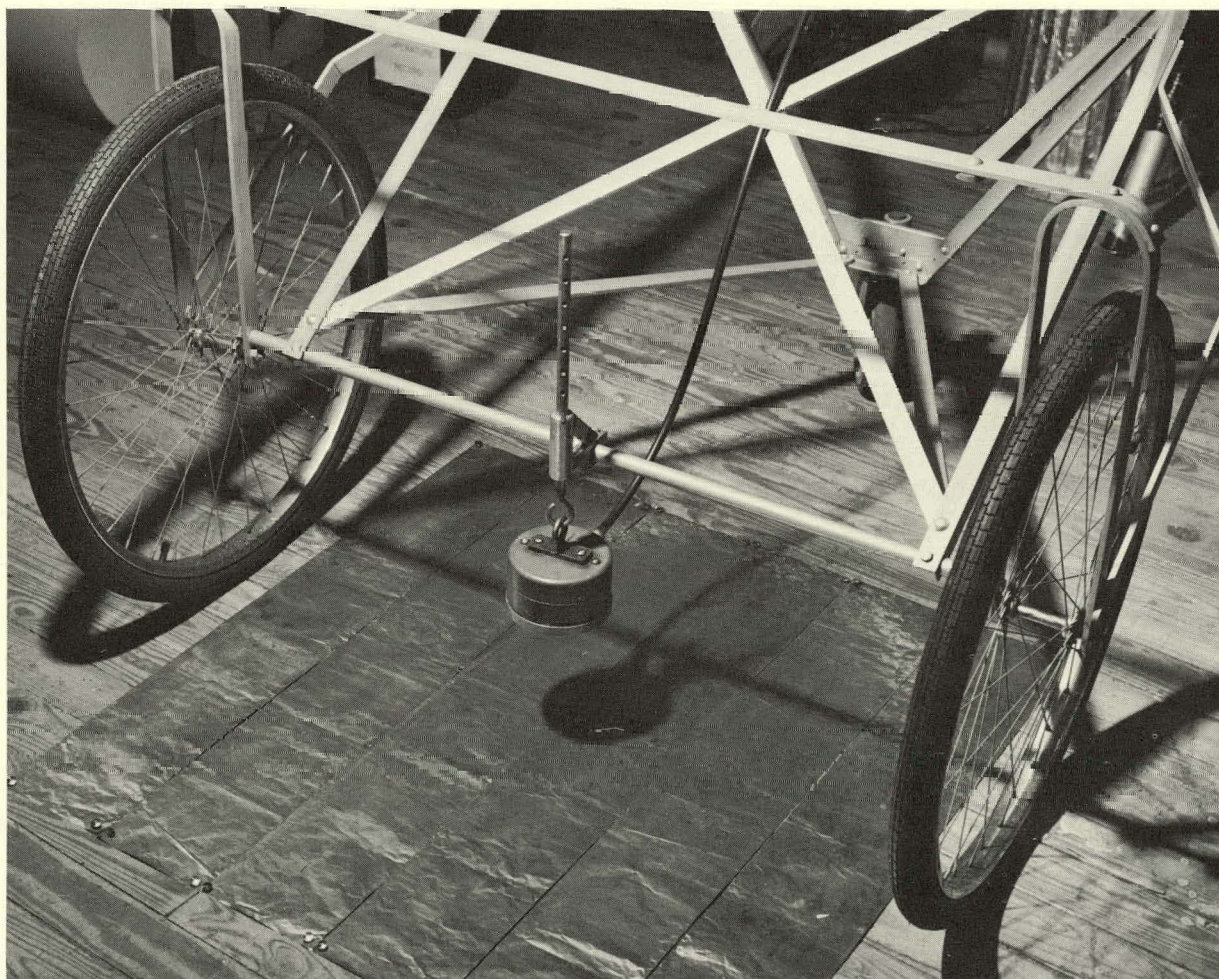
where

F = correction factor,
A = foil activity in dis/min/in.^{2}, and
n = net cart count in counts/min.

The constant includes area conversions and the dis/min to curie relationship.

The assumption made in this procedure is that the radiation in the field is distributed uniformly and without significant gradation within the area seen by the cart. Since >50% of the radiation detected by the cart is within 6 in. of a point below the center of the tube and 95% is within 18 in., and since autoradiographs show a uniform and rather dense distribution on the surface, this assumption is believed valid for field use.

A more serious objection may be made to the variable height of some particles in the field due to their positions on the grass rather than the soil surface itself. However, two pertinent factors, the attenuation by distance and the area seen by the detector, tend to counteract each other. Particles elevated on grass blades are closer to the tube and their radiation is subject to less attenuation than that from a point on the ground. On the other hand, fewer particles so elevated are within the volume seen by the tube. Normally, the grass is cut to a height of $\approx$3 in. before a test. This means that the vertical distance from some particles to the tube can range from 4.9 to 7.9 in. In one calibration test of this effect, lowering the tube 3 in. from its normal height increased the count by 22%. Under field conditions the change should be less. Furthermore, the distribution of particles with height probably does not vary greatly over the field in spite of some variations in density and composition of the vegetative cover.

SAFETY PROCEDURES

With the advice of the Health Physics Division, procedures for safe handling of Cu^{64} were established. Irradiated samples are stored and transported in a lead pig with a 2-in. wall thickness. The outside of the pig is surveyed for contamination and if necessary cleaned before leaving the isotope area of the reactor. Samples are removed from the pig and put into the dispensing solution by long-handled tools. The dispensing container is surrounded by ½ in. of lead and is approached closely only momentarily. Emission is controlled from a distant point.

From the time the pig is opened, all personnel in the area are required to wear suitable protective clothing and film badges, supplemented in some cases by pocket dosimeters. After emission those entering the contaminated area wear complete protective clothing including coveralls, boots, rubber gloves, head piece, and respirator. Contaminated clothing is changed at a line established between the contaminated and work areas. Operations are normally monitored by a trained health physicist. A safety shower and eyewash are available in the field.

Maximum permissible air concentrations of Cu^{64} as recommended by the National Committee on Radiation Protection[6] are 7×10^{-6} μC/cc for a 40-hr week and 3×10^{-6} μC/cc for a 168-hr week. In only one run has the highest measured concen-

Figure 24. Meteorological instruments in the sampling grid. Thermocouples in the tubing openings near the right side are aspirated by the vacuum pump.
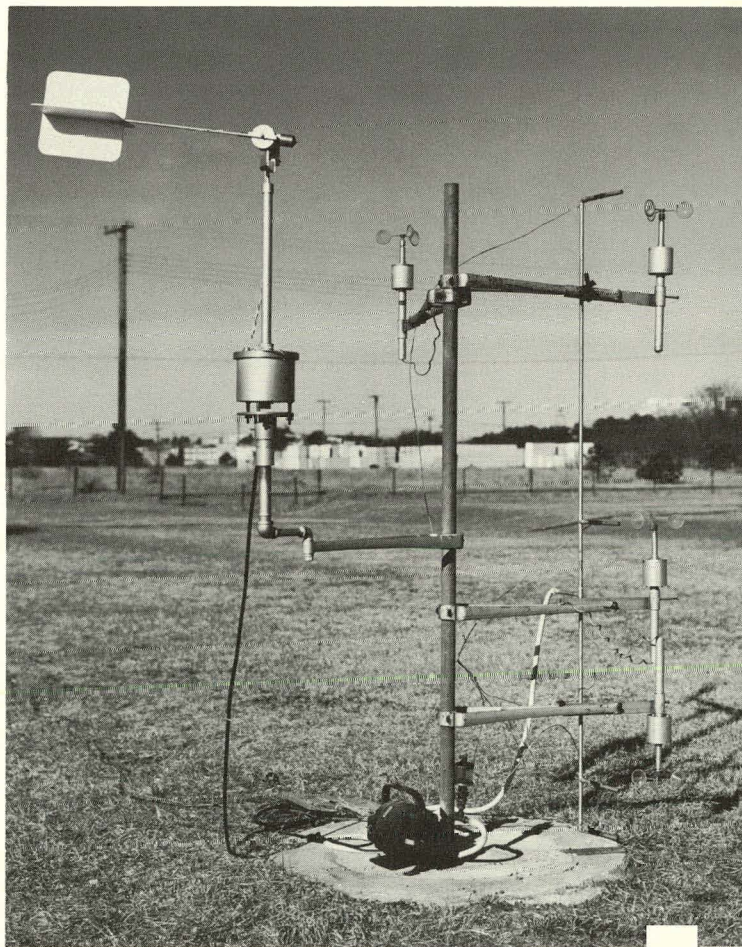
tration in the field exceeded the lower of these values, and then only for the duration of the emission at points directly in front of the nozzle. It seems certain that concentrations have never exceeded these limits except within 100 ft directly in front of the nozzle during emission. All personnel, however, are upwind at all times during the run, and no measurable exposures to airborne radiation have occurred. Termination of emission would be immediate if the wind shifted to bring the plume outside the sampling grid.

The chief concern in the conduct of the experiments has been not the safety of the personnel involved, but possible interference with sensitive counting operations being conducted elsewhere on site. For this reason runs are held only when the wind is blowing away from such areas, and every precaution is taken to avoid tracking particles by foot or vehicle outside the fenced area.

DATA

Data obtained from field experiments include the following:

1. Air concentrations of uranine at three heights.

2. Air concentrations of copper particles at three heights. These measurements are known to be low, particularly for the larger particles, because of the aerodynamics of the filter system.

3. Deposition per unit area measured by the mobile radiation counter.

4. Deposition per unit area measured by the sticky paper method.

The last two measurements are normally obtained at all 100 basic measuring positions and in a number of intermediate locations.

Meteorological measurements are taken in the sampling grid during each run (Figure 24). Total air passage is measured by sensitive anemometers with counter outputs mounted at 0.2, 1.0, and 2.0 m. A similar anemometer at 2 m gives a continuous trace of wind speed on an EA recorder. A Gelman-Gill bivane at 2 m gives recordings of horizontal and vertical angle. Aspirated thermocouples at ground level and at 0.2, 1.0, and 2.0 m above the ground give essentially continuous temperature traces on a Leeds and Northrup recorder. In addition, wind speed, wind direction, vertical angle, and temperature are measured at various levels of the 420-ft meteorology tower about one mile away. Other variables such as clouds, net radiation, air pressure, and relative humidity are

recorded, as well as the condition of the soil (wet, damp, or dry) and of the grass (green, dry, or mixed) and the grass height.

ACKNOWLEDGMENTS

Development of this sampling system and the associated procedures has required an extensive cooperative effort. The authors, while responsible for the over-all plan and many of the details, are most grateful for the assistance provided by many individuals and groups.

Within the Meteorology Group, particular mention should be made of the contributions of S.N. Rottenberg, who undertook development of the radioactive tracer technique and perfected many of the operational procedures, and L. Marcus, who completed a parallel development of the uranine system. F.E. Bartlett and R.M. Brown also contributed to the initial development of the fluorometric technique.

M. Slavin of the Chemistry Department and R. Dvorak of the Instrumentation Division were most helpful in the design and construction of the fluorometer. Various members of the Instrumentation and Health Physics Department assisted materially in the development of the several radiation detection devices and procedures necessary to the success of the program. H. Zulliger and R. Brennan were primarily responsible for the electronics of the mobile radiation counter.

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