REMOVAL OF HYDROGEN, CARBON MONOXIDE, AND METHANE FROM GAS COOLED REACTOR HELIUM COOLANTS

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ABSTRACT

It is necessary to maintain low contamination levels in Gas Cooled Reactor helium coolants and in-pile loops. The anticipated major non-radioactive, gaseous contaminents are H_2O, CO_2, H_2, CO, and CH_4. A two-step purification system for removal of these contaminants is proposed in which first all oxidizable gases are oxidized to H_2O and CO_2 and then the H_2O and CO_2 is removed from the helium by fixed bed co-sorption.

Although catalytic oxidation of the H_2, CO, and CH_4 by oxygen is possible, control problems make this method unacceptable when low contamination levels are desired. The alternate method of oxidation at low contamination levels is by fixed-beds of pelleted CuO. Kinetic tests on the reaction of H_2, CO, and CH_4 from a flowing stream of helium with fixed beds of CuO have been made in the temperature range of 400-600°C, pressure of 10-30 atm and mass flow rates of 0.085 to 0.255 g-moles/cm^2-min at contamination levels of 900-12,000 ppm by vol. It has been shown that the controlling reaction rate mechanisms for the reactions of H_2 and CO with CuO are mass transfer of the H_2 or CO from the bulk gas to the reaction site within the pellet while the CH_4-CuO reaction is dependent on CH_4 concentration in the bulk gas and available CuO surface area.

Finite difference type computer codes based on the determined reaction mechanisms were written and demonstrated which will describe the fixed-bed effluent concentration of H_2, CO, and CH_4 as a function of time, for any size bed in the range of experimental conditions tested.

Initial experimental results from kinetic tests on the co-sorption of H_2O and CO_2 by Molecular Sieves indicates that this process is feasible and the H_2O is sorbed preferentially to the CO_2.

INTRODUCTION

Contamination of coolant gases by chemical impurities is a major problem in gas cooled reactors and in-pile experimental loops. Recent gas cooled reactor concepts in the United States use helium as the primary coolant.(1, 2, 3)

The non-radioactive, gaseous impurities of most concern are H_2O, CO_2, H_2, CO and traces of hydrocarbons which can be represented by CH_4. These contaminants result from (1) out-gassing of the graphite reflector and moderator, and (2) inleakage of H_2O with resulting reaction with graphite, $H_2O + C \rightarrow H_2 + CO$. It is necessary to maintain these contaminants at low levels so that removal of graphite in the reactor core by chemical reaction is at a minimum, nuclear properties of the coolant are constant, and constant thermal properties of the coolant are maintained. In the Oak Ridge National Laboratory program of GCR Coolant Purification studies, processes for removal of the gaseous contaminants are being developed. Our ultimate goal is to develop design equations which will allow design of purification system components.

PURIFICATION FLOWSHEET

Production rates of the various gaseous contaminants are fairly low, therefore, removal of the contaminants can be achieved on a side stream from the main coolant stream. This side stream will probably be less than 1% of the total coolant flow. Use of such a side stream will also reduce the amount of useful heat lost from the coolant due to the necessity of operating part of the purification system at $< 100^oF$.

The proposed coolant purification flowsheet for non-radioactive, gaseous contaminants is a two-step process in which (1) all oxidizable gases are oxidized to H_2O and CO_2, and (2) the H_2O and Co_2 are removed by a fixed-bed sorption process (see Fig. 1). It is anticipated that the oxidation process step will be carried out at a temperature in excess of 400°C, while the sorption process will be at ambient temperature. Present GCR concepts are for system operating pressure of 20-60 atm, and it is desirable to operate the system at a minimum pressure drop.

Oxidation of H_2, CO, and CH_4

There are two possible process methods of oxidizing H_2, CO and CH_4: (1) catalytic oxidation by O_2, and (2) oxidation by inorganic oxides such as CuO.

Catalytic oxidation by oxygen in a fixed bed of solid catalyst appears to offer the best solution to this problem if allowable concentration levels are high ($>$ 10,000 ppm). Introduction of small amounts of the oxidant, oxygen, to the coolant would not add materially to the contamination problem. However, if contamination must be maintained at low levels ($<$ 1000 ppm) as would be the case for high temperature GCR,$^{(3)}$ the control problem of O_2 addition becomes very critical since small amounts of additional oxygen contamination to the coolant could not be tolerated.

For the cases of low levels of contamination by H_2, CO, and CH_4, fixed beds of CuO could be used to oxidize the contaminants to CO_2 and H_2O,

$$H_2 + CuO \rightarrow H_2O + Cu \qquad (1)$$

$$CO + CuO \rightarrow CO_2 + Cu \qquad (2)$$

$$CH_4 + 4CuO \rightarrow CO_2 + 2H_2O + 4Cu \qquad (3)$$

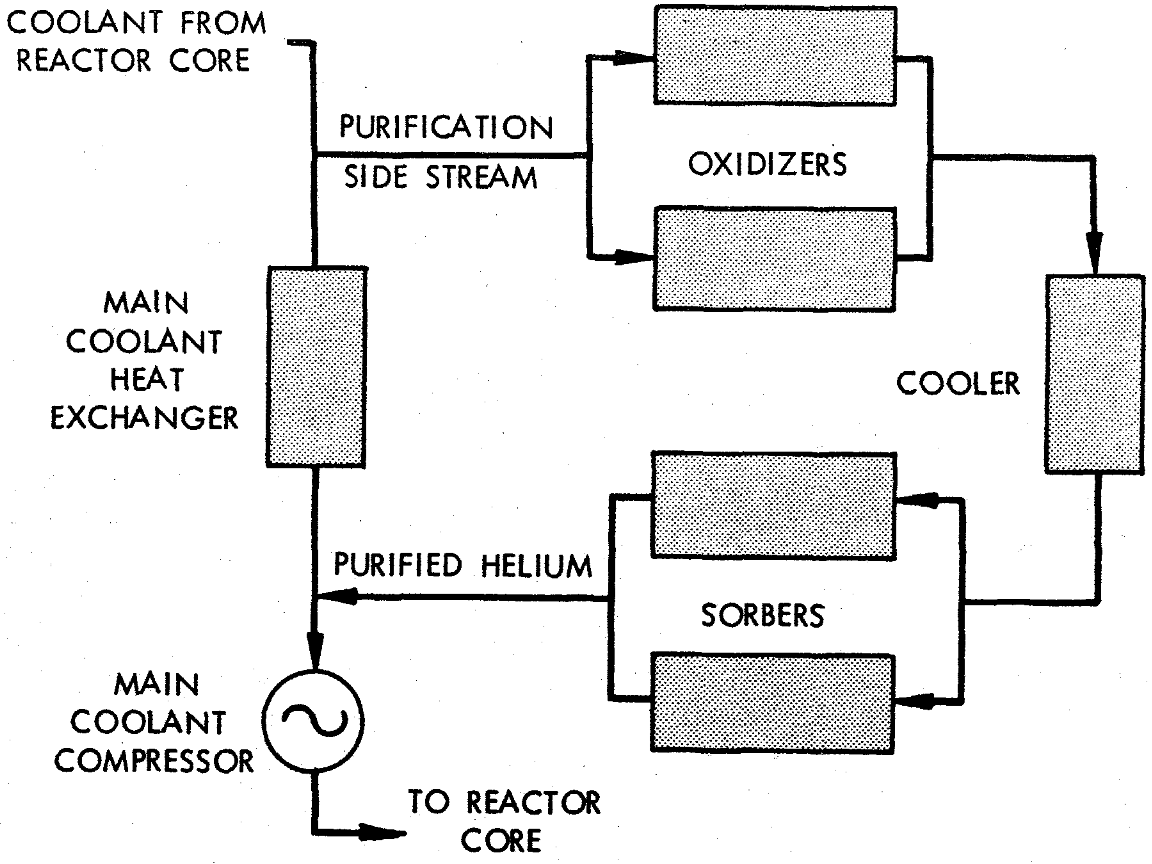

Fig. 1. Simplified flowsheet for a GCR helium purification system for the non-radioactive, gaseous contaminants, H_2, CO, CH_4, CO_2 and H_2O.

The resulting reduced beds of Cu could then be regenerated by CuO by use of air or O_2,

$$Cu + 1/2\ O_2 \rightarrow CuO \qquad (4)$$

The fixed bed-CuO oxidizer would not have the problem of oxygen contamination of the coolant (O_2 decomposition pressure is < 1 ppm at 600°C and 20 atm),(4) however, if continuous operation is desired dual oxidizers would be needed for cyclic operation of the system with alternate regeneration of the reduced bed.

Cosorption of CO_2 and H_2O

Removal of the CO_2 and H_2O from contaminated helium is most easily achieved by fixed beds of Molecular Sieves. This method of CO_2 and H_2O removal is necessary for low levels of contamination and it may be used in conjunction with a cooler-condenser if large amounts of H_2O are present. Since this process is also a fixed-bed process, there will also be a necessity for dual sorbers one of which will be active at a time, thus allowing time for regeneration of the second bed by dry air.

EXPERIMENTAL PROGRAM

Although a considerable amount of experimental work has been done on this entire subject by Oak Ridge National Laboratory, the program is not complete. In order to demonstrate the general techniques of experimentation and data analysis used to date, the work on the H_2-CuO reaction will be discussed in somewhat more detail. The other processes and reactions have been and are being investigated in a similar manner.

Our approach to the development of CuO fixed-bed oxidizers has been to determine the kinetic mechanisms of the reactions of interest in the operating ranges of interest. This is then the basis for derivation of the necessary design equation for system components.

Physical Properties and Operating Ranges

Since low pressure drops are necessary in the CuO oxidizer, it was decided to use pelleted CuO as the fixed bed material. The material used was CuO compacted into nominal 1/8-in. rt circular cylinders obtained from the Harshaw Chemical Company (see Fig. 2). This material is available in commercial quantities. The mass flow rates of interest were 0.25 g-moles/cm^2-min or less. The H_2 contamination level to be investigated was 1000 to 10,000 ppm (vol). Temperature range of interest was 400-600°C. The pressure range was 10-30 atm.

Rate Controlling Mechanism

It was found that when CuO pellets reacted with H_2 from a flowing stream of He, the partially reduced CuO pellet always had a well defined Cu-CuO interface within the pellet (see Fig. 3). This interface was so well defined that it suggested control of the

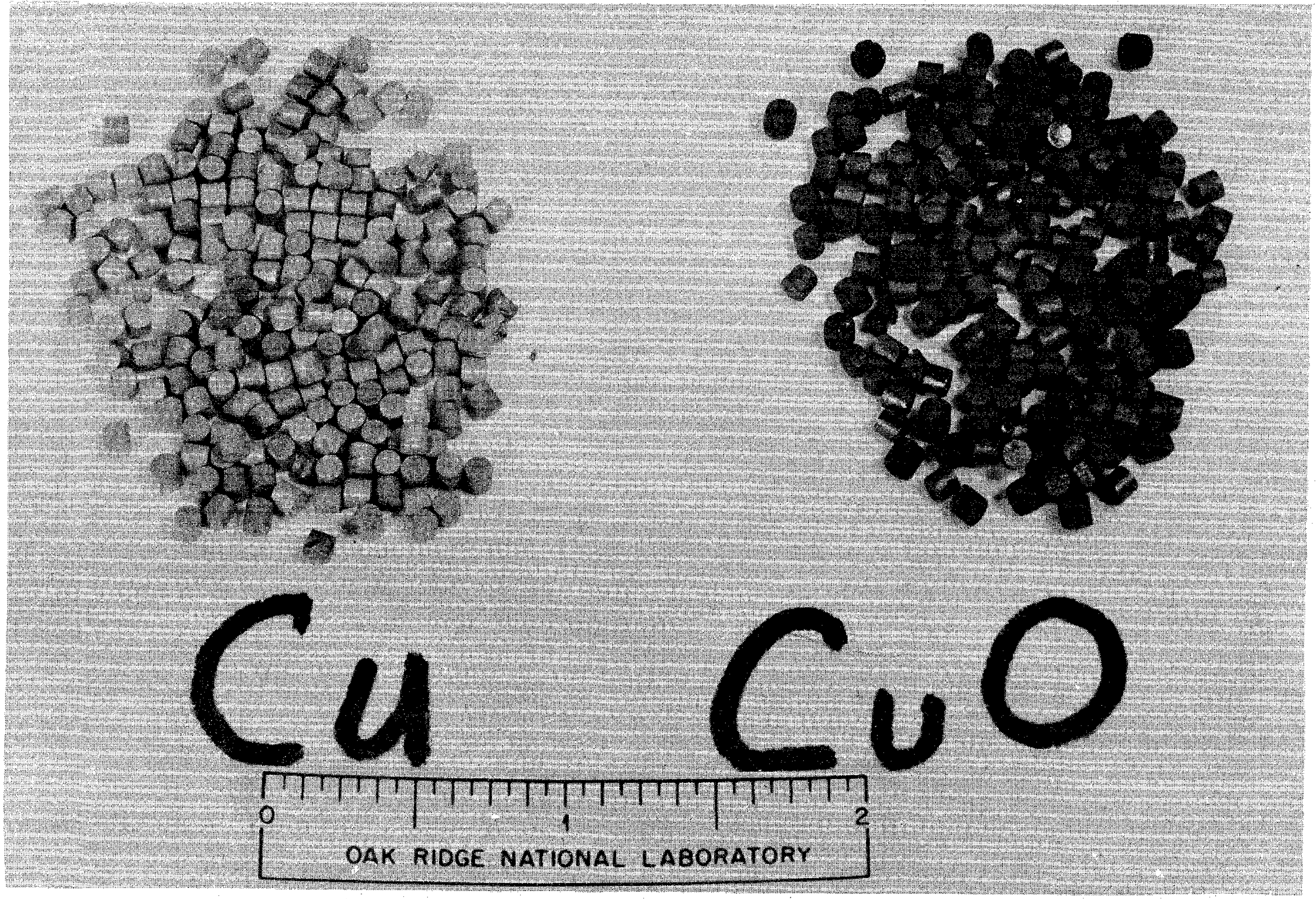

Fig. 2. Copper oxide pellets in oxidized and reduced state.

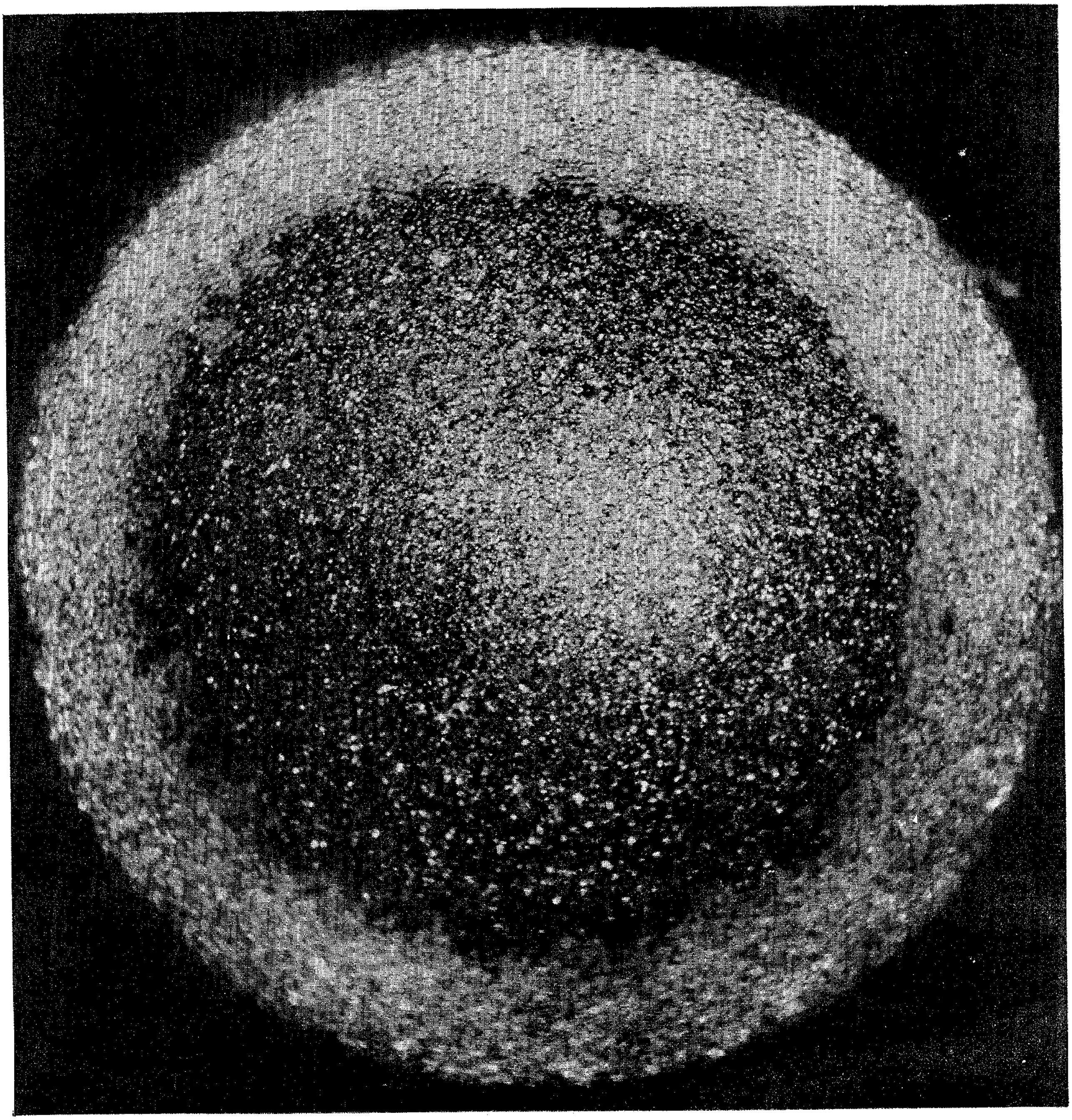

Fig. 3. Sectioned CuO pellet which has been partially reduced by hydrogen.

reaction must be due to mass transport of the H_2 from the bulk gas to the reaction site. That is, the reaction was very rapid and essentially irreversible. Thermodynamic considerations show that the H_2-CuO reaction is essentially irreversible.

The resulting mathematical model for such a system would be (see Fig. 4) a pellet, assumed to be a sphere, in which the H_2 diffused through an external stagnant gas film to the solid surface and then subsequently diffuses through the solid to the reaction interface. The reaction interface would be a spherical shell of radius r_o. Because of the irreversibility of the process, the H_2 concentration at the interface would go to zero and the H_2 concentration gradient within the sphere would be dependent on the internal diffusion assumed to be molecular diffusion through the pores of the solid. The external diffusion through the gas film would be assumed to be dependent on the driving force due to the concentration difference between the bulk gas and on the solid surface.

This mathematical model of the rate controlling mechanisms of H_2 in a flowing stream of helium reacting with a fixed bed of CuO pellets can be expressed as three differential equations:

Bed Material Balance:

$$\left(\frac{\partial C}{\partial t}\right)_z + v\left(\frac{\partial C}{\partial z}\right)_t = -\frac{1}{\beta}\left(\frac{\partial n}{\partial t}\right)_z \tag{5}$$

Reaction Rate:

$$\left(\frac{\partial n}{\partial t}\right)_z = k_g aC \left[1 - \frac{k_g a}{\tau}\left(\frac{R - r_o}{4\pi\alpha r_o R + k_g a/\tau \quad (r - r_o)}\right)\right] \tag{6}$$

Position of Reaction Interface:

$$r_o(1 - r_o/R)\, dr_o = -\frac{\alpha D}{b}\left[\frac{k_g aC}{\tau}\left(\frac{1}{\frac{4\pi\alpha D r_o R}{R - r_o} + \frac{k_g a}{\tau}}\right)\right] dt \tag{7}$$

where,

C = gas-phase concentration of hydrogen

t = run time

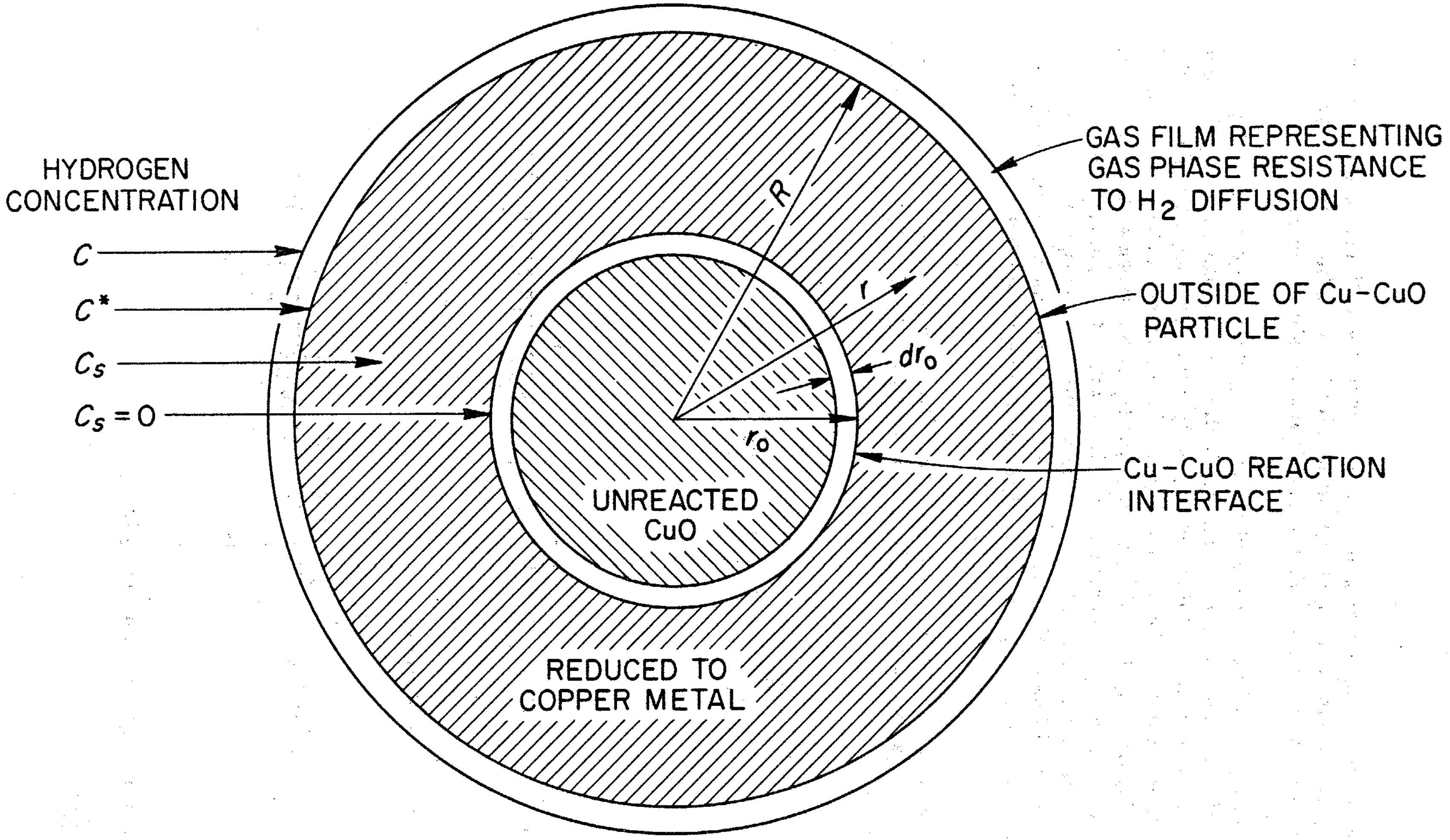

Fig. 4. Section of the spherical model used to describe the case of the rapid, irreversible reaction of H_2 and CuO with the reaction rate controlled by gas phase and solid phase diffusion of H_2.

z = bed height measured from bottom of bed

V = interstatial gas velocity

β = bed porosity

n = bed molar density of CuO

k_g = film mass transfer constant

a = exterior surface area of CuO pellets

τ = number of CuO pellets per unit volume of bed

α = effective porosity in CuO pellets

R = radius of pellet

r_o = radius of Cu-CuO interface in pellet

b = molar density of CuO in pellet

D = molecular diffusivity of hydrogen

These equations can be solved simultaneously by a finite difference technique on a digital computer.

Internal Diffusion

In order to determine if the assumed mechanism of internal mass transfer was correct, molecular diffusion through pores, a series of experiments were made in which differential beds of CuO pellets were reacted with H_2 in high velocity streams of He in the pressure range of 10-30 atm, temperature range of 400-600°C and H_2 concentration range of 0.1 to 1.0%. If external diffusion does not contribute to the reaction rate control, an expression for molecular diffusion can be derived for the differential beds of CuO:

$$D = \frac{b}{\alpha C t}\left(\frac{R^2}{6} + \frac{r_o^3}{3R} - \frac{r_o^2}{2}\right) \qquad (8)$$

In the differential bed tests, the monolayer of pellets were sectioned after each test and the resulting position of the Cu-CuO interfaces were determined and used in equation (8) to obtain the molecular diffusivity. It was found that the temperature and pressure dependence for the molecular diffusivity were correct in the range of operating conditions tested. However, the effective porosity of the pellets was found to be about 1/5 of the total porosity of the pellets (~40%). This effect has been noted by several other workers.

External Diffusion

When resistance to mass transfer of the contaminant from the bulk gas to the CuO pellet surface was assumed to be the conventional film diffusion, it was found that the mass transfer factor, J_D, for this transfer in the range of Modified Reynold's Numbers tested (~4-15) could be expressed as a function of Modified Reynold's Number:

$$J_D = 0.24\ (Re')^{-0.69}$$

where,

J_D = mass transfer factor

Re' = Modified Reynold's Number

This expression is similar to others found in the literature for various solid-fluid mass and heat transfer systems.

Deep Bed Tests

The data on internal and external mass transfer and the three differential equations describing the mathematical model can be utilized to predict the H_2 concentration at any point in the deep bed. Finite difference solution of the three equations, (5), (6) and (7), was made by an IBM 7090 digital computer. A series of deep bed tests were made in which a flowing stream of pre-heated He contaminated with H_2 was continuously introduced to a bed of CuO pellets and the effluent from the bed was periodically analyzed for H_2 content by a gas adsorption chromatograph (see Fig. 5). These resulting H_2 effluent concentration history curves were than used as the basis for comparison of experimental and predicted results (see Fig. 6). It was found that agreement was adequate for the assumed model and the three differential equations from the model can be used as design equations.

Other Work

Work has also been completed on the CO and CH_4 oxidation by CuO and the reaction mechanisms for these reactions have been determined. The CO-CuO reaction appears to have the same mechanism as the H_2-CuO reaction while the CH_4-CuO reaction is dependent on CH_4 concentration in the bulk gas and available CuO surface area. The co-sorption of H_2O and CO_2 by type 5-A Molecular Sieves is currently being investigated in a manner similar to that outlined for the H_2-CuO reaction. Initial experimental results indicate that this process is feasible and the H_2O is sorbed preferentially to CO_2.

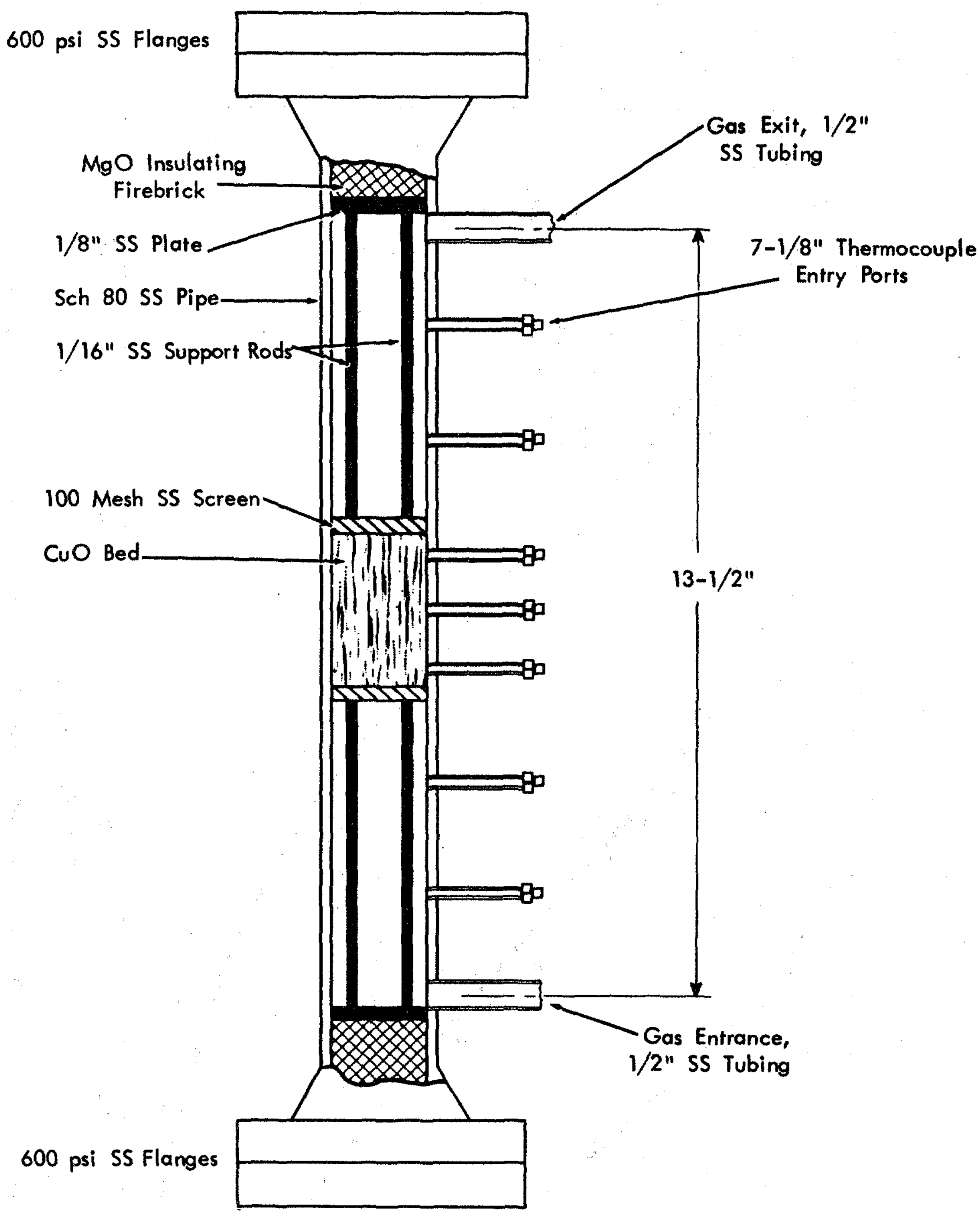

Fig. 5. 2-in. oxidizer used in deep bed tests for the H_2-CuO reaction.

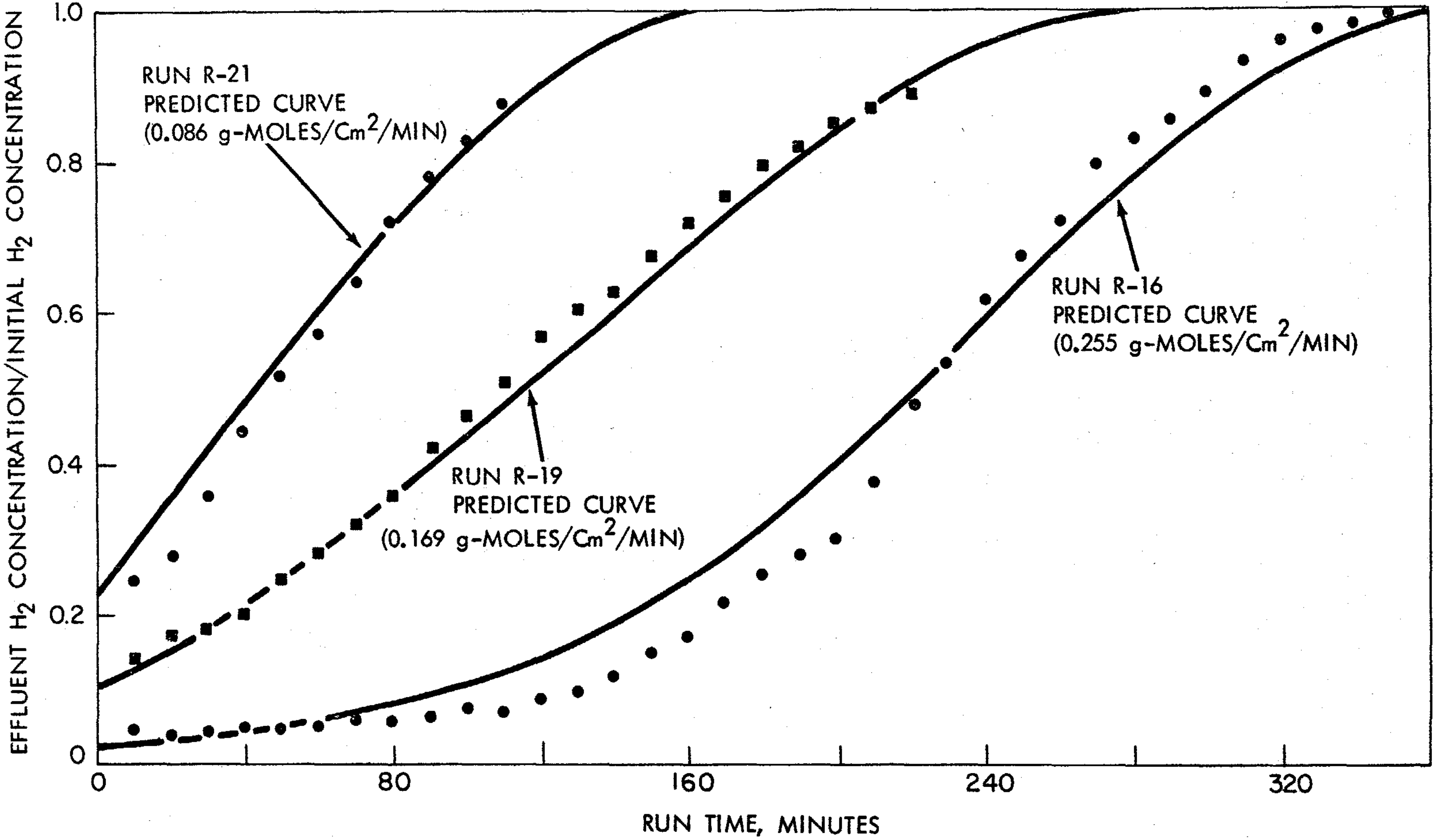

Fig. 6. Comparison between predicted and experimental H_2 effluent concentration history curves for the H_2-CuO reaction showing dependence of mass flow rate.

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2. Anderson, F. A., Proposed Helium Purification System for the Experimental Gas-Cooled Reactor (EGCR), ORNL-2819, October 2, 1959.

3. Scott, C.D., Finney, B.C., and Suddath, J.C., The Helium Purification System for the Proposed Pebble Bed Reactor Experiment, ORNL-CF-60-10-31, October 25, 1960.

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DISCUSSION

RILEY: Have you done any tests on regenerating these beds?

CHESTER (ORNL): Yes, with oxygen. There is a very slight loss activity after many regenerations, and you have to be careful not to run the temperature up too high, but all you do is put oxygen to them and they go right back.

RILEY: Isn't it a fact that these temperatures are rather low for satisfactory oxidation of methane?

CHESTER (ORNL): 400 to 600° Centigrade?

RILEY: I thought you said Fahrenheit at the beginning.

CHESTER (ORNL): I am sorry. I meant Centigrade.

FIRST: Could you discuss the difference between your theoretical porosity and your measured porosity on these pellets?

CHESTER (ORNL): Well, the porosity in the pellets can be measured by a number of methods, one of which is the takeup of some solvent such as toluene and what we measured in the experiments was the product of the porosity and the diffusivity of the gas in question. These appeared lumped together.

We found that this number, if we used the textbook value for the diffusivity of hydrogen at the temperatures in there and the porosity measured by, say, toluene absorption, disagreed with our experimental measurements, the number we had to use to make the curves fit the data.

We performed many experiments and found that the temperature and pressure behavior of the diffusivity agreed with what was in the texts, and from this the assumption was made that the textbook value for the diffusivity of hydrogen was all right.

Therefore, the assumption was made that the disagreement between the outside calculations of the product of diffusivity times porosity and the observed value of this product, in the experiments, in the actual beds, must be due entirely to a disagreement in porosity. The effective porosity, as measured by, say, toluene absorption, is not the same as that observed.

As to the basis of this model, by assuming that the porosity is simply one-fifth of the measured porosity, we can have design equations that work and that we think pretty well explain what is going on.

FIRST: You don't have any other porosities on which to make a similar calculation? You worked only with this one type of pellet, is that correct?

CHESTER (ORNL): In this particular reaction, yes.

METEOROLOGY AND AIR CLEANING

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ABSTRACT

Brief descriptions are given of several new meteorological techniques for measuring atmospheric transport and dilution that have been developed in the past five years. Among these are the use of atmospheric tracer techniques, direct application of wind measurements to diffusion, and the determination of trajectories over distances in the excess of ten miles.

In 1955 at the 4th AEC Air Cleaning Seminar at Argonne National Laboratory I had the privilege of discussing this same topic. The intervening six years has brought about many changes due primarily to the growth of the nuclear industry. Reactors are more sophisticated, fuel processing techniques and waste disposal methods are more varied and the technology of the nuclear industry has, in general, become more complex. At the same time there has been a marked growth of the nuclear industry. Figure 1 shows how widespread reactor sites have become.

Concurrent with the growth and spread of the industry has been the improved understanding of the environmental and health problems of waste disposal. Attention has shifted from gross external gamma irradiation to consideration of the inhaled and ingested radiation exposures from specific radionuclides. Our accident experience to date, while fortunately scanty, has directed additional attention to the deposit of radioactivity on vegetation and on the ground.

To keep pace with these changes, it has been necessary for the meteorologists to do two things simultaneously; first to continue studies which improve our ability to estimate the dilution of radionuclides, and second to invent new techniques for estimating the major features of atmospheric dispersion at a wide variety of sites with a minimum of equipment and manpower. The remainder of this discussion will describe some of the work directed to these goals.

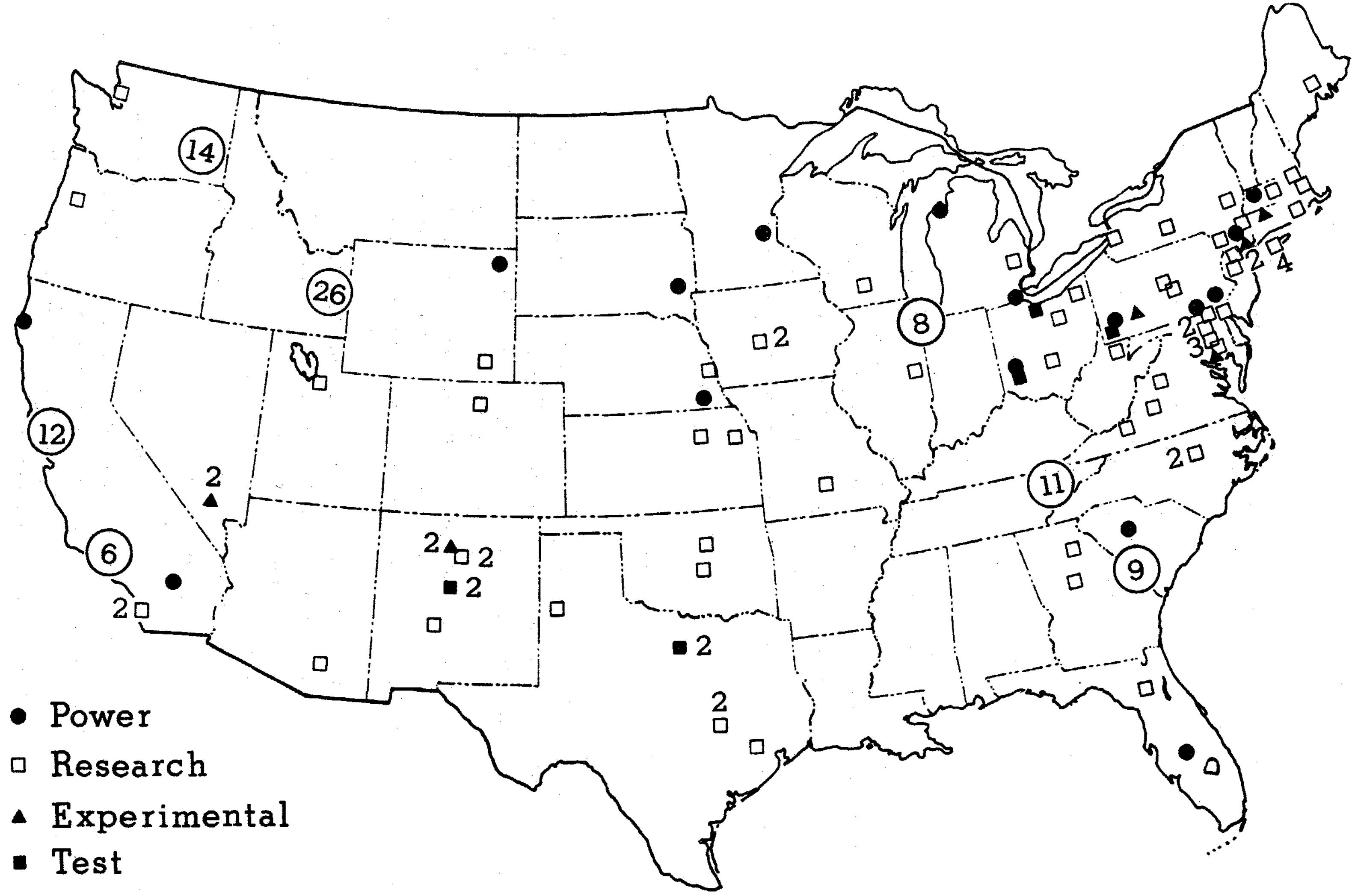


Figure 1

There has been an impressive increase in field experimentation to measure diffusion using tracers. The Air Force supported program has resulted in Project Prairie Grass (1), in Project Greenglow (2), and in field experiments using fission products as tracers (3), (4), Greenglow and the FRT-II are especially interesting in the attention given to diffusion in stable conditions over distances greater than 10 miles. Considerable benefit is also expected from work sponsored by the Chemical Warfare Service such as the Windsoc tracer experiments over several tens of miles in central Texas. Other current experiments near seacoasts and in irregular terrain will provide further useful information. In addition to these planned tracer releases the dispersion of radionuclides from various AEC activities have been utilized in meteorological studies. The ANP operations at NRTS and the Rover and Pluto Projects at the Nevada Test Site are experiments where there have been sufficient weather instrumentation to permit comparative studies of meteorological parameters and the spread of radioactivity.

These extensive experiments can and have provided data which permit an empirical selection of coefficients to adapt the Sutton type of diffusion equation. At the same time efforts have been numerous to identify weather measurements which would be more directly related to dispersion. Most of us have felt, intuitively that the turbulence represented by the swinging of a wind vane should be related to diffusion. A quantitative method of relating the wind fluctuations to dispersion has been developed (5), (6) and shows considerable promise. This approach, which uses the Gaussian diffusion formulation shown in Figure 2, relates wind vane fluctuation statistics to the lateral (σ_y) and vertical (σ_z) spread of a plume of diffusing material. The theory has been extended into a very practical method of estimating concentrations from accidents by Pasquill (7) and described in detail in the U. S. literature by Gifford (8), (9), (10) in Nuclear Safety. Gifford's three articles have recently been issued as a combined reprint by the Oak Ridge Technical Information Extension Office.

This approach, while refreshing and based on physical measurements more readily obtainable, is not a panacea, and its limitations are nowhere better recognized than by the meteorologists who conceived it. Further testing will be required and one of the major activities of the Weather Bureau Research Station at the NRTS is an extensive series of tracer experiments to compare wind statistics and dispersion.

The first in this series of tests has been described by Islitzer (11) and Figures 3 and 4 are from this work. Figure 3 illustrates the calculation of the point of maximum concentration using vertical wind fluctuation statistics and Figure 4 shows a comparison of the observed and calculated concentrations using wind measurements as prediction parameters. Figure 5 is included to illustrate the particular wind installation used to support these tracer tests.

The tracer technique used in this work has intrinsic interest and is also described by Islitzer. Building on the initial suggestion by Robinson (12) a successful uranine dye technique has been developed which is relatively easy to use in the field. The dye which is atomized from a water solution becomes a very small

$$\chi = \frac{Q}{2\pi\,\sigma_y\,\sigma_z\,\bar{u}}\ \mathrm{EXP}\left\{-1/2\left(\frac{y^2}{\sigma_y^2}+\frac{z^2}{\sigma_z^2}\right)\right\}$$

χ = CONCENTRATION............units/m^3

Q = SOURCE..............................units/sec

y = CROSSWIND DISTANCE....... meters

z = VERTICAL DISTANCE......... meters

$\bar{u}$ = MEAN WIND SPEED .... m/sec

σ_y = STANDARD DEVIATION OF PLUME WIDTH

σ_z = STANDARD DEVIATION OF PLUME HEIGHT

RELATION TO SUTTON

$$\sigma_y = 1/2\ C_y{}^2\ x^{2-n}$$

$$\sigma_z = 1/2\ C_z{}^2\ x^{2-n}$$

Figure 2

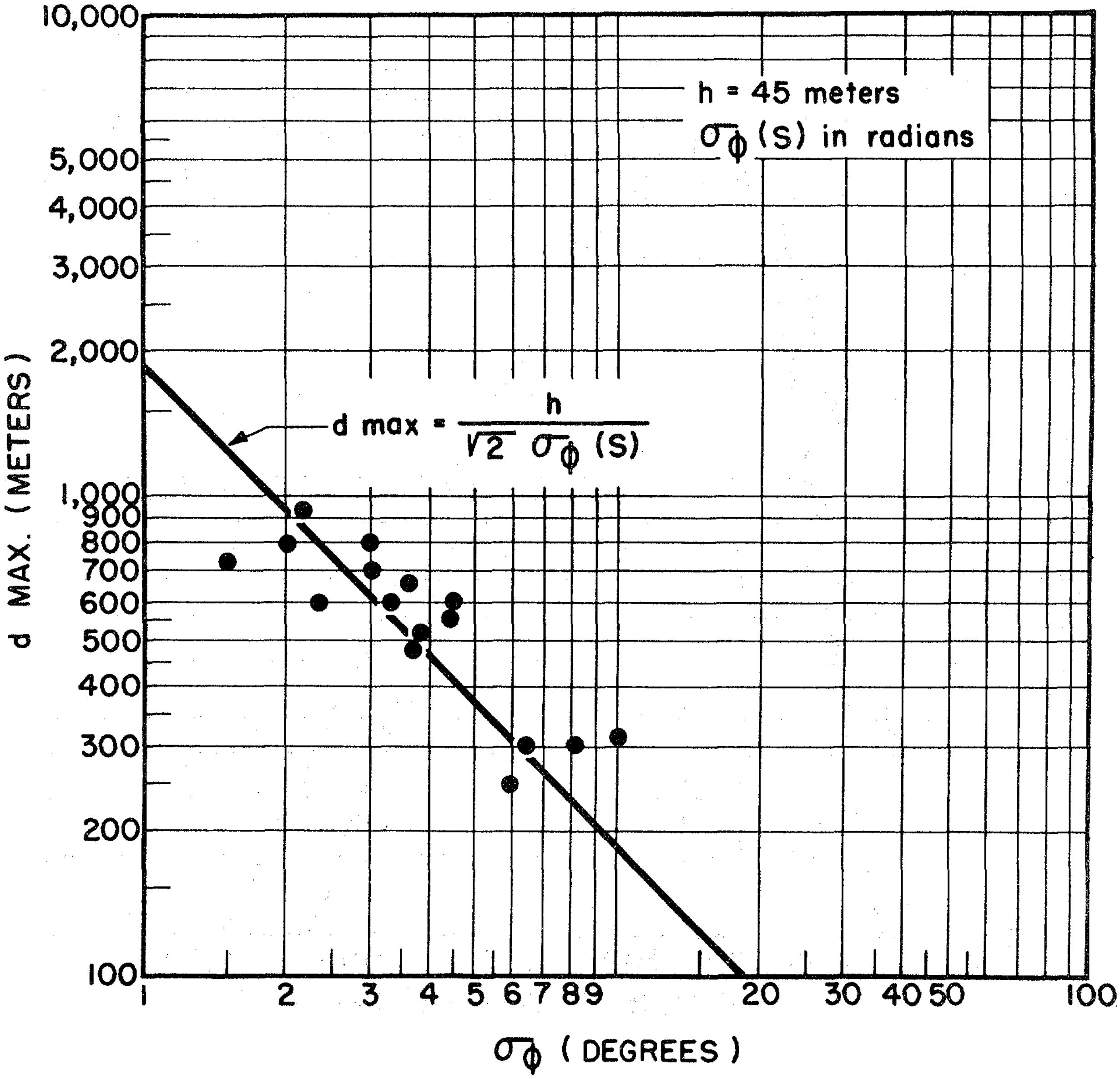

h = 45 meters
σ_ϕ (S) in radians
d max = $\frac{h}{\sqrt{2}\,\sigma_\phi(S)}$
d MAX. (METERS)
10,000
5,000
4,000
3,000
2,000
1,000
900
800
700
600
500
400
300
200
100
1
2
3
4
5
6
7
8
9
20
30
40
50
100
σ_ϕ (DEGREES)

Figure 3

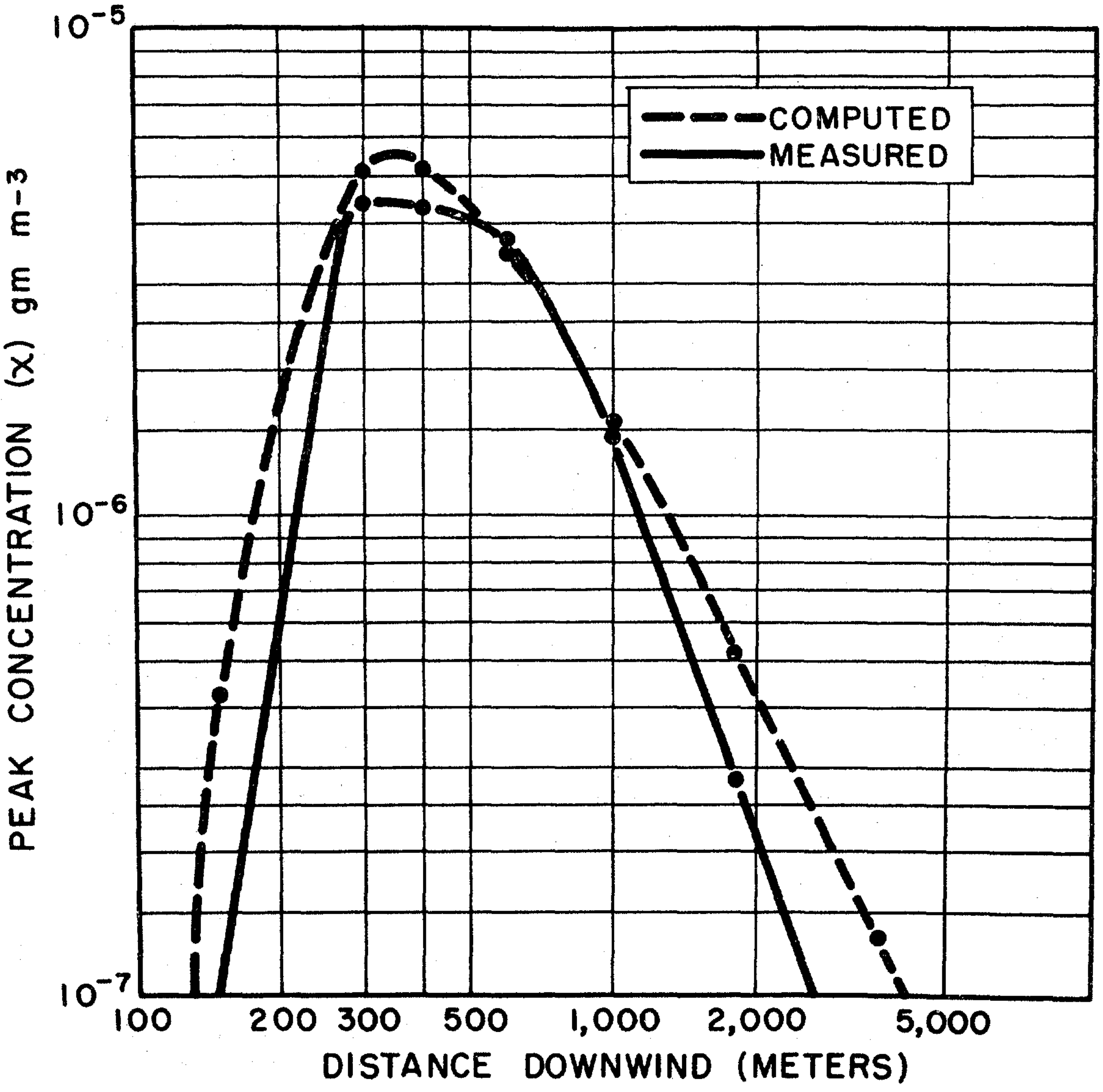

10⁻⁵
10⁻⁶
10⁻⁷
PEAK CONCENTRATION (χ) gm m-3
100
200
300
500
1,000
2,000
5,000
DISTANCE DOWNWIND (METERS)
COMPUTED
MEASURED

Figure 4

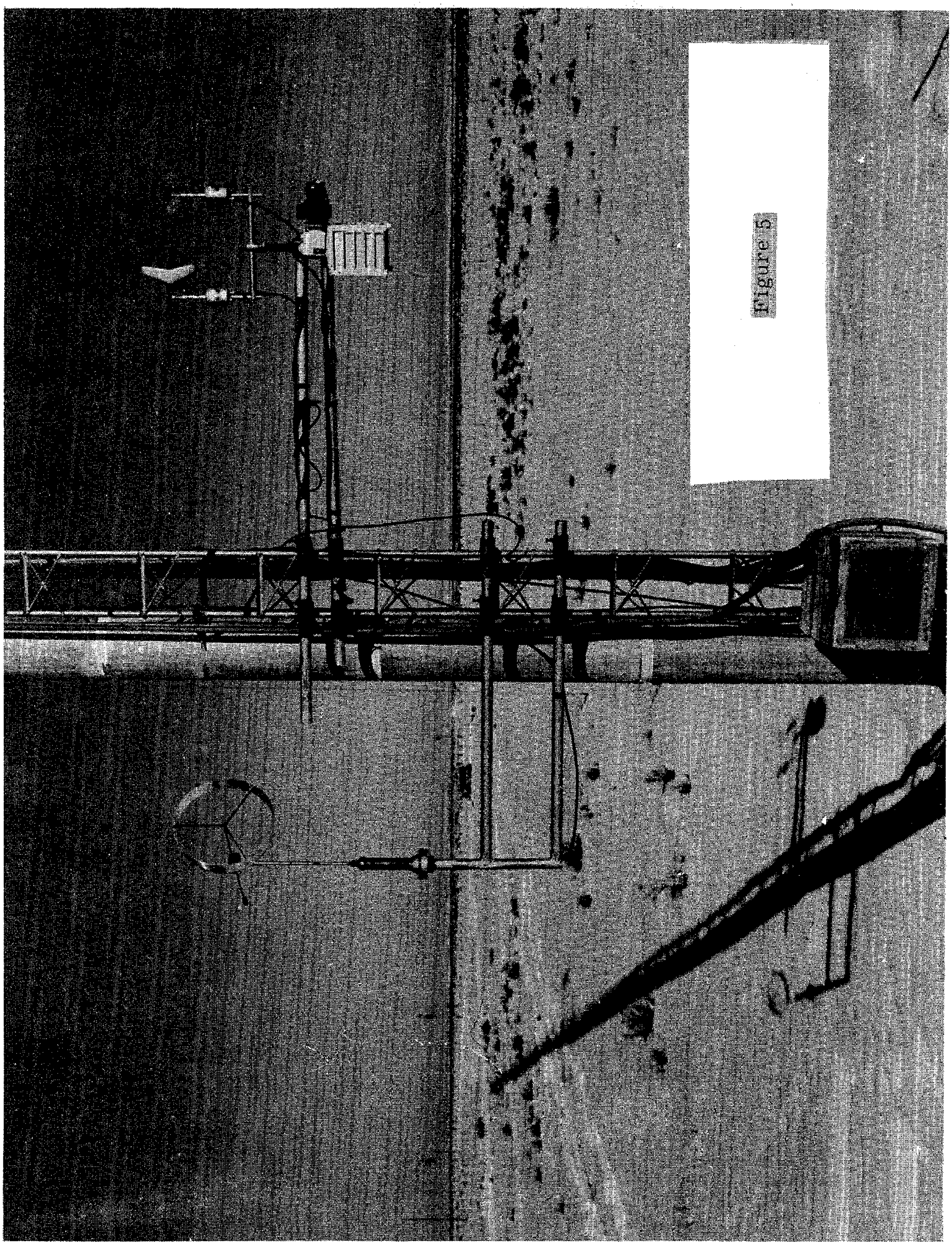


Figure 5

particulate in the micron size range. Quantitative tracer measurements involve only the washing of the filter paper collectors and measuring the fluorescence of the resulting solution. The technique is quite sensitive (10^{-9} to 10^{-10} gm/m^3) and rapid since two men can completely process 100 filters in a single eight hour day.

While an extensive program such as this is necessary to test meteorological diffusion theories and which may be useful and necessary at locations where there is a significant airborne waste problem, generally it is neither possible nor desirable to have such an elaborate program. The use of visible tracers in determining atmospheric transport has been common for many years, and it has the advantage that one does not have to chase about at random because there is no way of determining whether or not you are within the cloud of airborne material. Recently Gifford (13) has formulated a mathematical basis for quantitative determination of diffusion parameters based only on observations of the geometry of smoke plumes. Culkowski (14) has developed photographic techniques for recording smoke plumes which permits a relatively easy application of this technique. Figures 6, 7, and 8 illustrate what can be done with time exposure photography to determine dispersion over different time intervals. Figure 6 shows the smoke from a standard Chemical Corps smoke pot. The photograph was taken at an exposure time of 1/100 of a second. Figure 7 shows smoke from the same source; the exposure time was 10 seconds. Here we see how the violent gyrations of the plume are beginning to average out. Figure 8 shows the same smoke plume, but the exposure time is now 8 minutes. Here we see that the visible portion of the plume begins to resemble the classic elliptical shape. Comparisons of the diffusion coefficients determined from smoke releases accompanying quantitative tracer tests showed an almost one-to-one comparison. Using pictures such as these and the mathematical techniques of Gifford, it is possible to determine quantitative values of diffusion parameters for individual sites. Obviously this can be done for different times of the day, for different wind directions, and for different seasons of the year.

Once one has related wind statistics to quantitative diffusion parameters determined from smoke or other tracers and has continuing records of the wind, it is possible to quantitatively compute the spatial distribution of concentration over various periods of time. A numerical technique for doing this has been developed at Oak Ridge (15) and an example of this technique applied at the National Reactor Testing Station (16) is shown in Figure 9, where isopleths for two different sources have been computed over a one year period.

The studies of overlapping diffusion patterns from multiple sources has, to date, been of real interest to only a few locations. A glance at Figure 1 however shows that the time is not too far in the future when quantitative knowledge of the total impact on the environment of a nuclear "complex" will be necessary. However when more than two or three sources are involved the calculation of cumulative concentrations over a large geographical area becomes extremely complex and laborious. At this point hand calculations must give way to the computer. This has been shown to be entirely feasible. In connection with studies of the multiple source urban air pollution program a diffusion model appropriate to computer solution has been developed (17). This model will handle more than

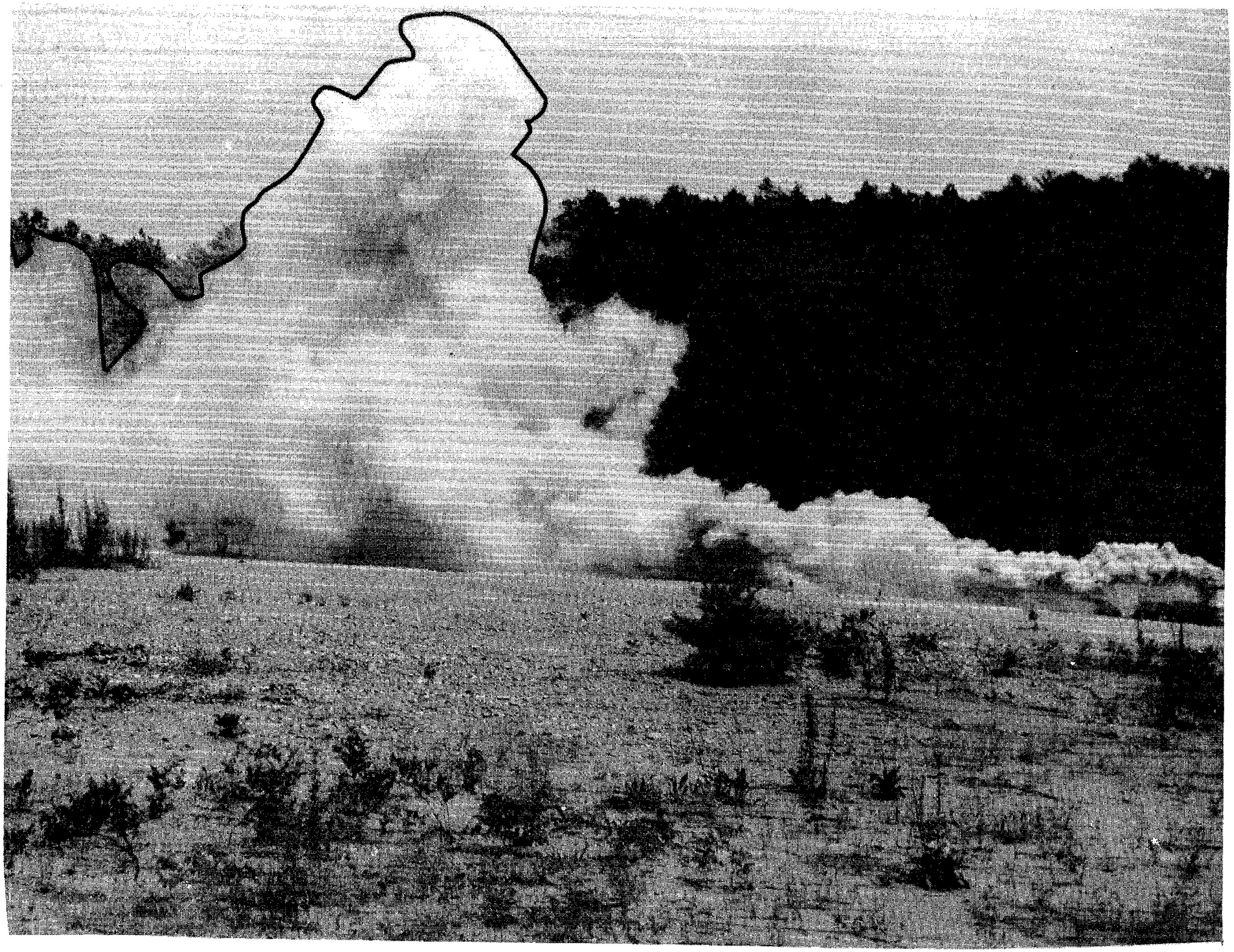


Figure 6

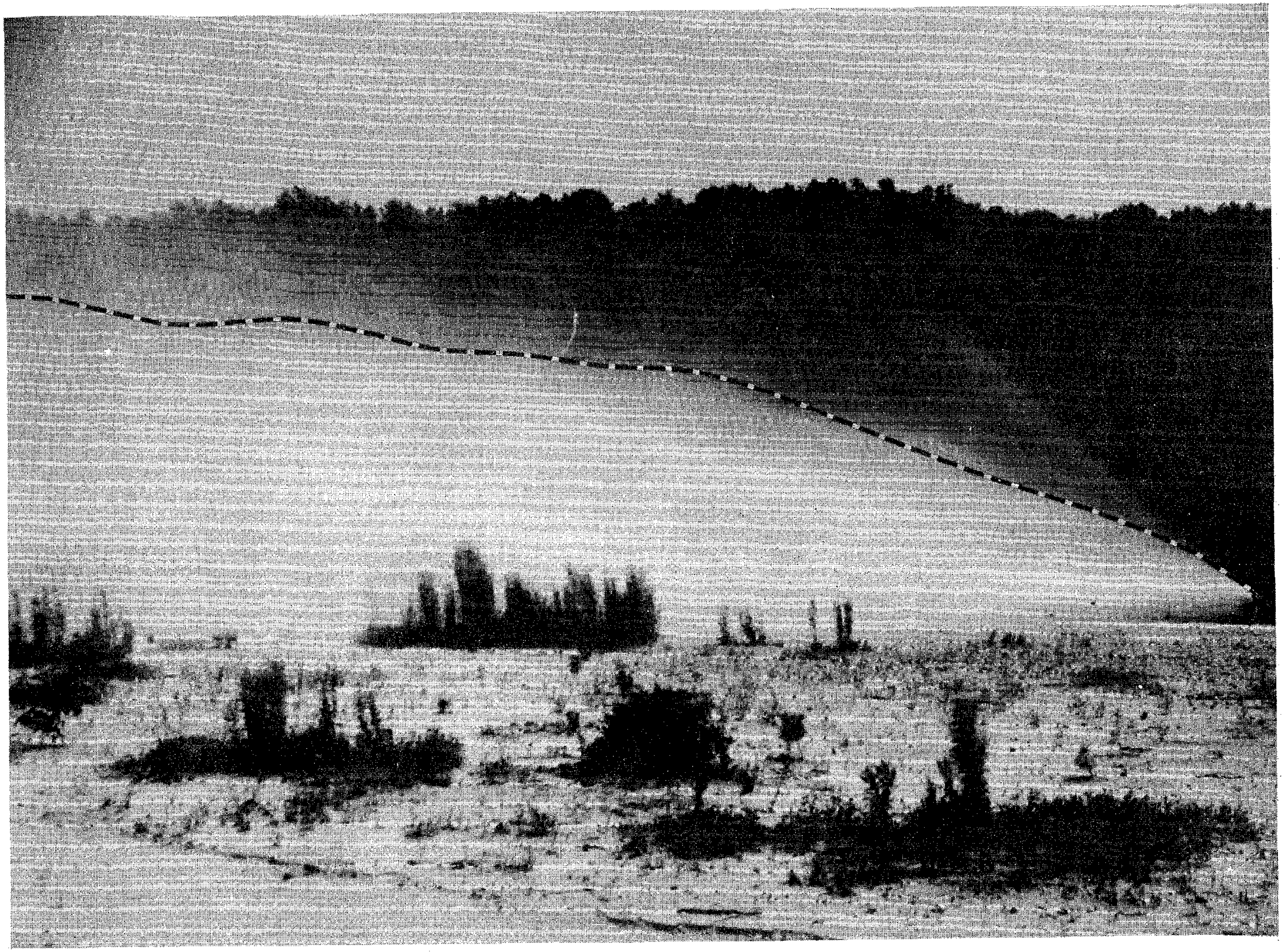


Figure 7

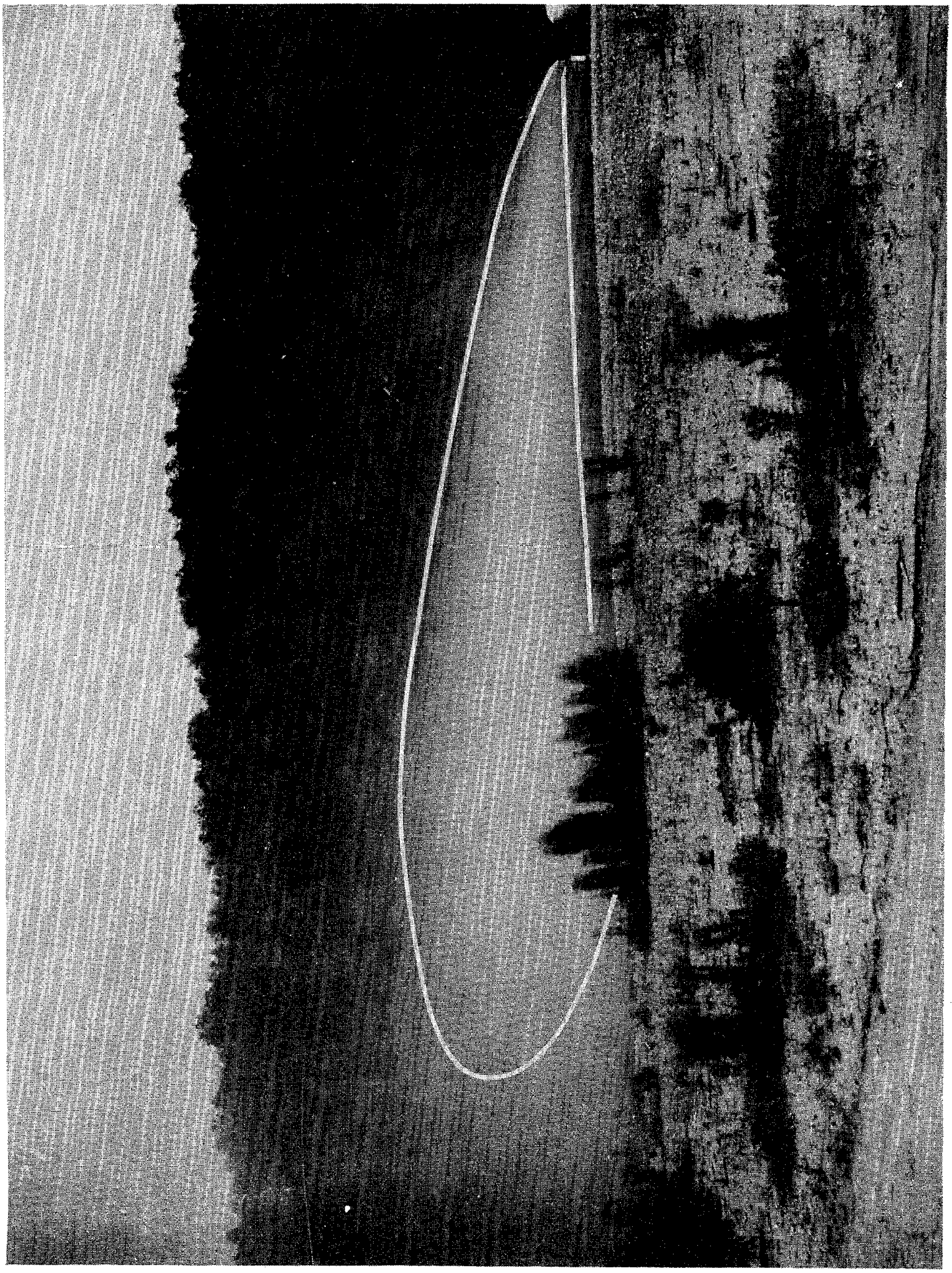


Figure 8

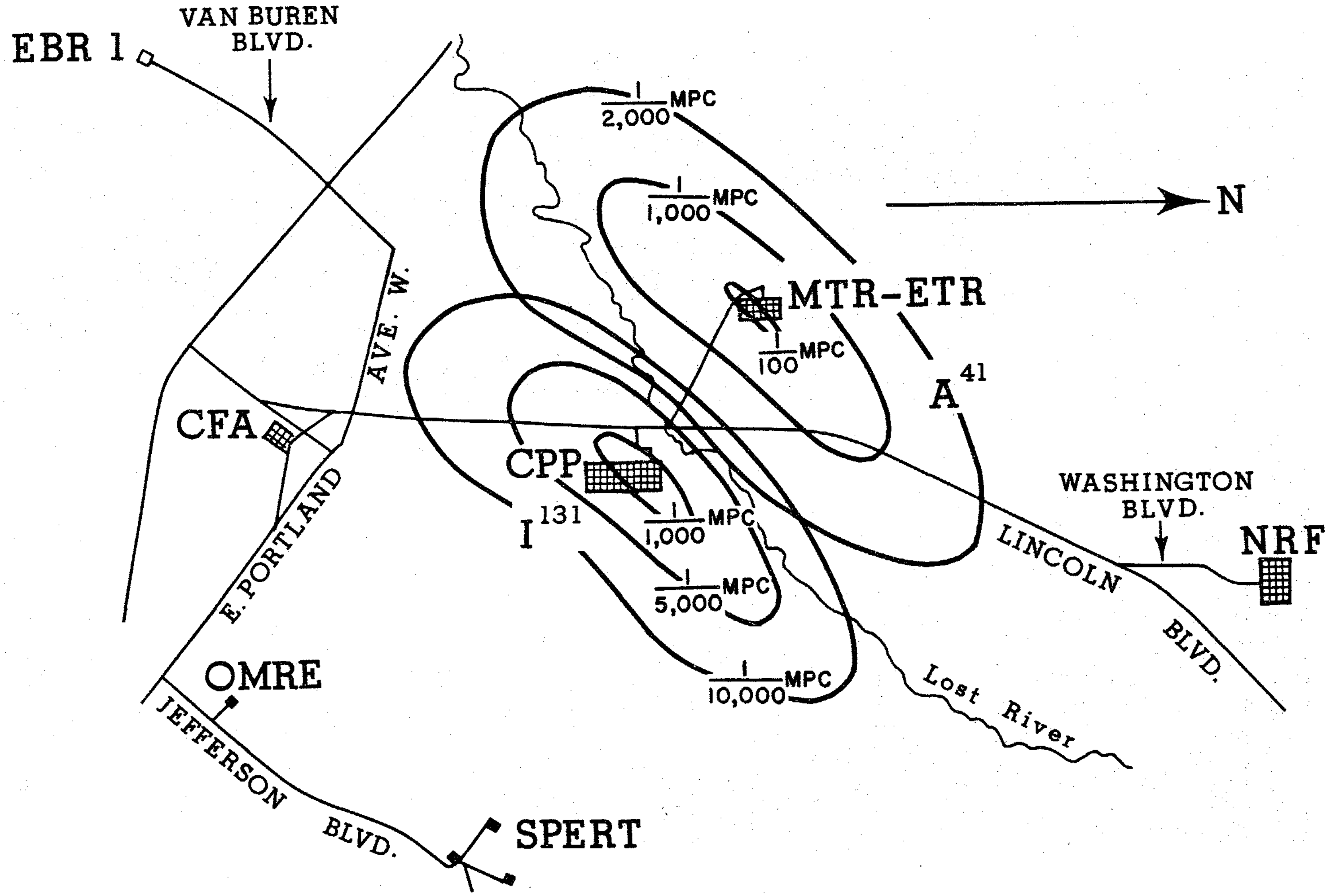

VAN BUREN BLVD.
EBR 1
1/2,000 MPC
1/1,000 MPC
N
MTR-ETR
1/100 MPC
A41
AVE. W.
CFA
CPP
I131
1/1,000 MPC
1/5,000 MPC
1/10,000 MPC
E. PORTLAND
WASHINGTON BLVD.
NRF
LINCOLN BLVD.
Lost River
OMRE
JEFFERSON BLVD.
SPERT

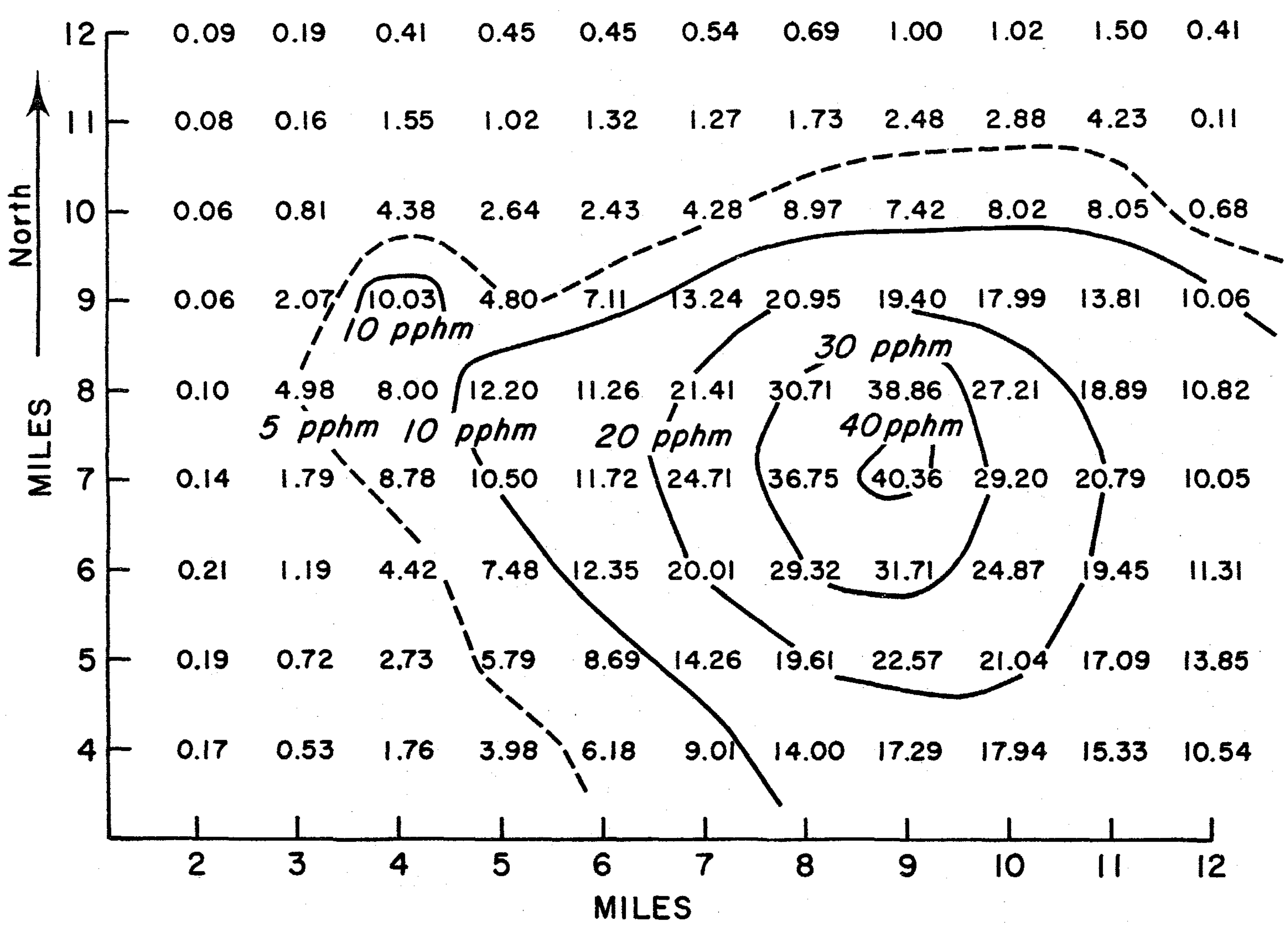
Figure 9

200 separate source locations and compute their cumulative effect at more than 100 locations. Figure 10 is the machine output of this model. Relatively short period air concentrations can be computed and summed to obtain daily, monthly, or annual averages.

As a part of the input to evaluation of the large area, long term environmental levels of radioactivity is some knowledge of the spatial variation of atmospheric transport and dilution. This information is also useful for comparative evaluation of nuclear site locations. A first approach to establishing a diffusion climatology of the United States has been made by Hosler (18) who has summarized all the low level inversion data from radiosonde observations in the United States. He has shown that these data provide good estimates of the seasonal and annual frequency of slow atmospheric dilution.

An entirely different problem is posed when attention is shifted from average concentration to the problem posed by specific experiments and to the need of determining precisely where a cloud of radioactivity may go. Over distances in excess of a few miles the angle subtended by a "target", even if this "target" is a city, is relatively small and it becomes at least as important to know whether the material will pass over the "target" as it is to know what the atmospheric dilution will be. For such problems particularly for distances between 10 and 100 miles experiments have proved the feasibility of using small constant volume balloons as tracers (19), (20), (21). The balloons currently in use are shown in Figure 11. A set of trajectories obtained at the Nevada Test Site by this technique is shown in Figure (12). Experiments of this type have shown the complexity of the paths followed by the air. Figure 13 shows the path of a balloon off the Southern California coast and shows that, almost 8 hours after release the balloon had made a loop and almost crossed its initial release point.

Many other projects are underway, too numerous in type and objective to cover here but special mention should be made of the direct comparison of wind tunnel models and atmospheric tests by Argonne National Laboratory, of the studies of dispersion differences over land and water by the University of Michigan and the Weather Bureau, and of the increased use of analogue computers for diffusion modelling by the University of Michigan and by Oak Ridge.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MILES
12 0.09 0.19 0.41 0.45 0.45 0.54 0.69 1.00 1.02 1.50 0.41
11 0.08 0.16 1.55 1.02 1.32 1.27 1.73 2.48 2.88 4.23 0.11
10 0.06 0.81 4.38 2.64 2.43 4.28 8.97 7.42 8.02 8.05 0.68
9 0.06 2.07 10.03 4.80 7.11 13.24 20.95 19.40 17.99 13.81 10.06
8 0.10 4.98 8.00 12.20 11.26 21.41 30.71 38.86 27.21 18.89 10.82
7 0.14 1.79 8.78 10.50 11.72 24.71 36.75 40.36 29.20 20.79 10.05
6 0.21 1.19 4.42 7.48 12.35 20.01 29.32 31.71 24.87 19.45 11.31
5 0.19 0.72 2.73 5.79 8.69 14.26 19.61 22.57 21.04 17.09 13.85
4 0.17 0.53 1.76 3.98 6.18 9.01 14.00 17.29 17.94 15.33 10.54
2 3 4 5 6 7 8 9 10 11 12
10 pphm
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Figure 10

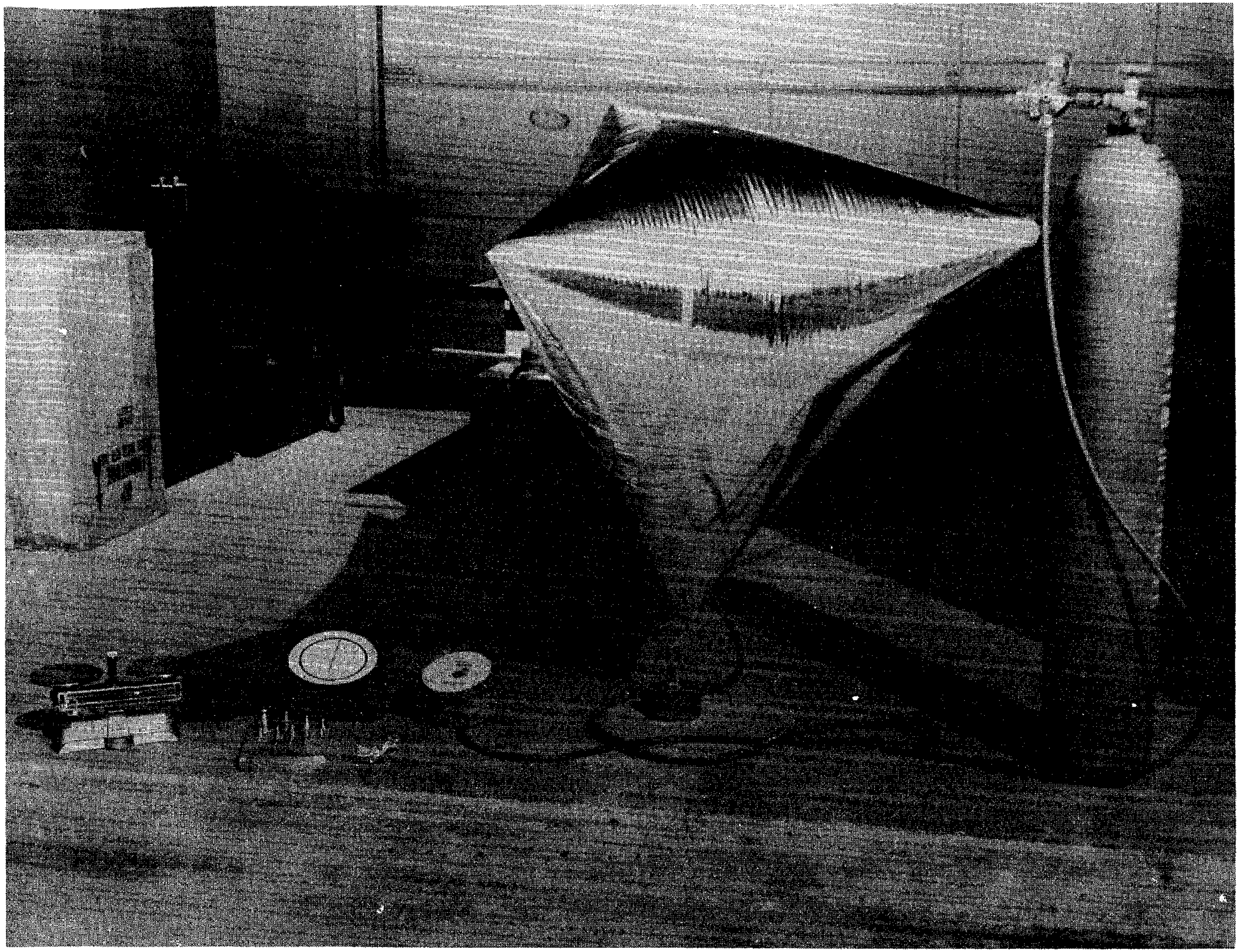
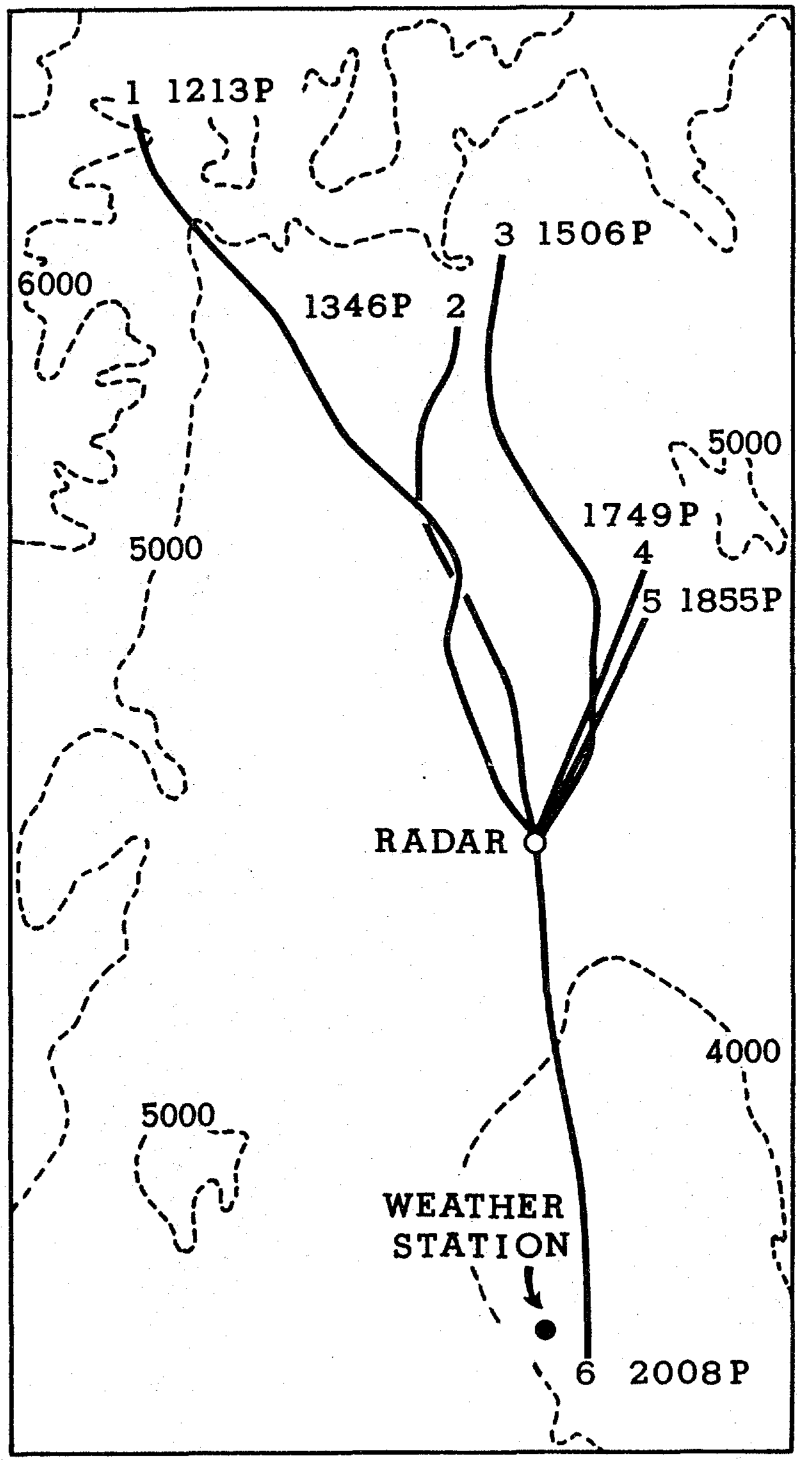


Figure 11


1 1213P
3 1506P
6000
1346P 2
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1749P
4
5 1855P
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RADAR
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WEATHER STATION
6 2008P

Figure 12

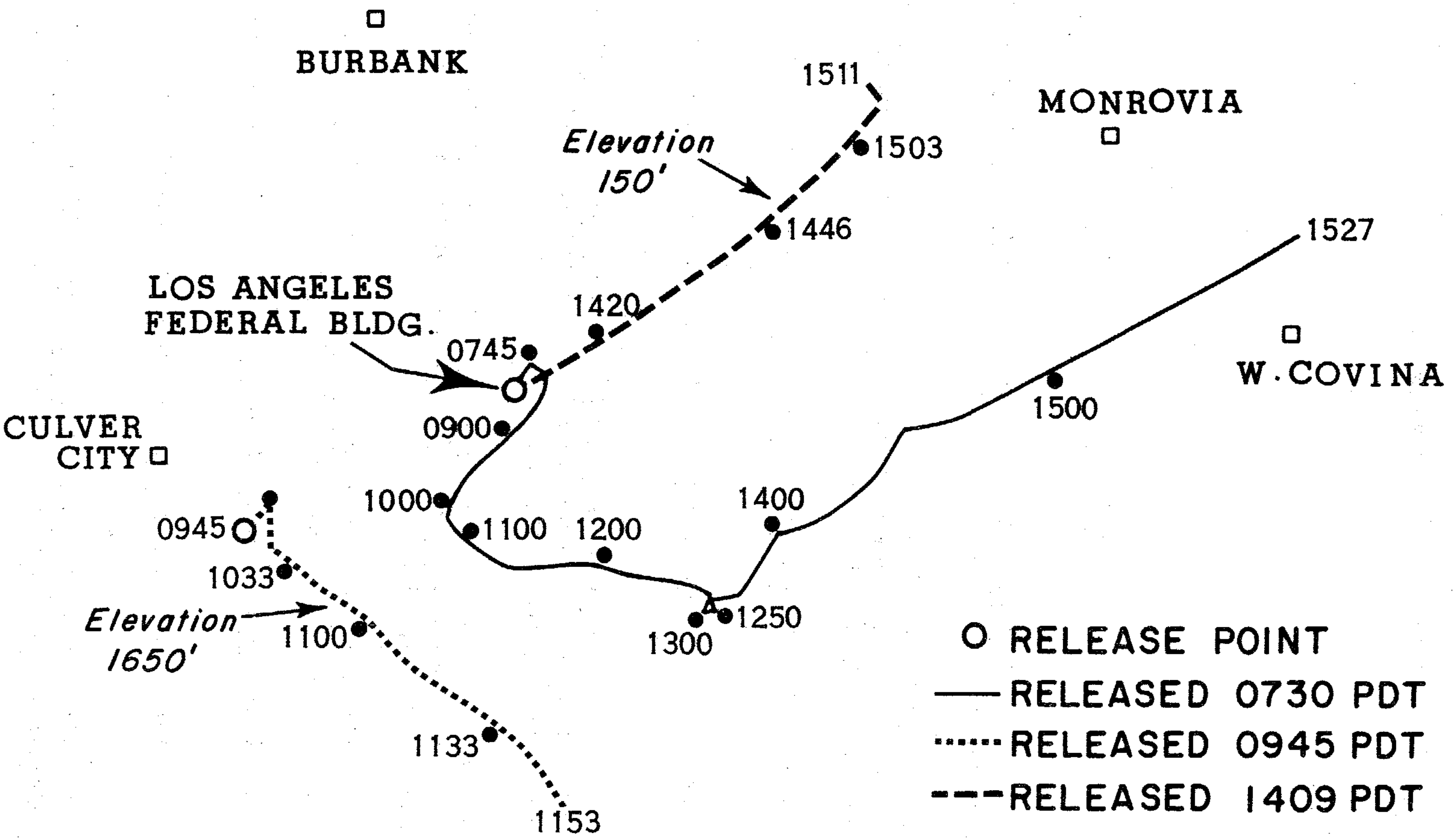
BURBANK
1511
MONROVIA
Elevation 150'
1503
1446
1527
LOS ANGELES FEDERAL BLDG.
1420
0745
W. COVINA
1500
CULVER CITY
0900
1000
1400
0945
1100
1200
1033
Elevation 1650'
1100
1300
1250
1133
1153
○ RELEASE POINT
RELEASED 0730 PDT
RELEASED 0945 PDT
RELEASED 1409 PDT

Figure 13

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COMBINED DEPOSITION AND DIFFUSION STUDIES

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ABSTRACT

The Brookhaven Meteorology Group is undertaking a series of combined diffusion-deposition field studies to provide detailed information on the behavior of small particles in the atmosphere, and to permit evaluation of this behavior in terms of meteorological parameters. In this work, a dual particulate emission consisting of uranine dye and radioactive copper particles is used in conjunction with a variety of sampling techniques. This paper is an interim progress report, describing the background of the studies, the equipment and procedures and the nature of the resulting data. Plans for the forthcoming analysis are reviewed.

OBJECTIVES

During their participation in the study of the safety of large power reactors in 1957 (1), several members of the Meteorology Group became convinced that a full, detailed study of the deposition of small particulates was needed. The literature reviewed in the course of the safety study failed to reveal even one series of experiments containing the data necessary for evaluation of any of the theoretical models of the process. It was also clear that most of the data were by-products of other experiments, and hence were often of a lower order of reliability than one would wish. The work described in this paper is a direct outgrowth of this review, and has been planned to provide as complete information as possible on the deposition processes over a limited area.

Specifically, these experiments include the measurement of (a) the deposition of particles directly on the ground, (b) deposition on prepared collection devices, (c) the low-level airborne distribution of the deposition tracer, (d) a similar airborne distribution of very fine aerosol particles substantially unaffected by deposition and (e) the meteorological parameters influencing the foregoing.

IMPLEMENTATION

Considerable time and effort has been devoted to the development of procedures and procurement of the equipment required and, while many of the techniques are not new, extensive refinement and testing has been necessary.

Of first importance was the selection of suitable particulates for the work. It was decided quite early that the primary interest would be in particles ranging from 0.1 to 20 microns in diameter, since there is very little deposition of smaller particles and the larger ones are affected principally by gravitational settling. Of the many possibilities, it was decided that a radioactive isotope of moderately short half-life would prove the most effective deposition tracer. Radioactivity would not only provide a straightforward mean of measuring the deposition directly on the ground surface and vegetation, but the experiments could be repeated at the same site without interference from previous work.

Initially, Lanthanum in fused clay spheres was selected, since the strong β and γ radiation of La^{140} and the 40-hour half-life were virtually ideal. Unfortunately, although a technique was developed for creating a suitable range of spherical particles, it proved impossible to eliminate long-lived radioactive constituents of the clay to the degree necessary. Repeated experiments would have resulted in a gradual increase in the field background which in time would have rendered it useless.

The deposition tracer finally selected consists of solid spheres of extremely pure copper, in which Cu^{64}, having a 13-hour half-life, is the detectable isotope. The copper is obtained commercially in a polydisperse powder form, after which it is separated into 3 size groups by water elutriation. They have mean mass diameters of 2.67, 4.60 and 9.62 microns.

While the individual groups do not contain the truly monodisperse particles one would like, they are quite suitable for most of the experiments planned. Fig. 1 shows a typical size distribution and Fig. 2 is a photograph of the Group III particles after final elutriation. The copper spheres are of course sized in their stable form, and are irradiated several hours before the run. Residual radioactivity from impurities is insignificant from any practical viewpoint.

These size groups will be recognized as relatively large for some of the experiments which might be of interest. To solve this problem, copper in colloidal suspension will be used in place of the elutriated spheres when studies of the finer particles are undertaken.

While the copper serves as a deposition tracer, fluorescent uranine dye has been chosen as the diffusion aerosol. This dye, first used as an atmospheric tracer by Robinson (2), is very easy to handle, and when ejected in solution in a water spray, forms particles of significantly less than 1 micron diameter. Furthermore, it is detectable at the level of approximately 5×10^{-10} gm/ml of water. The detection is accomplished on a fluorometer especially designed for the purpose by the Brookhaven Instrumentation Division.

Both the copper and the uranine are dispersed from identical though separate, Chicago atomizers. The uranine of course is in solution, while the copper is suspended in the water by mechanical agitation, and each provides the desired dry particulate after evaporation of the drops. In order to provide complete

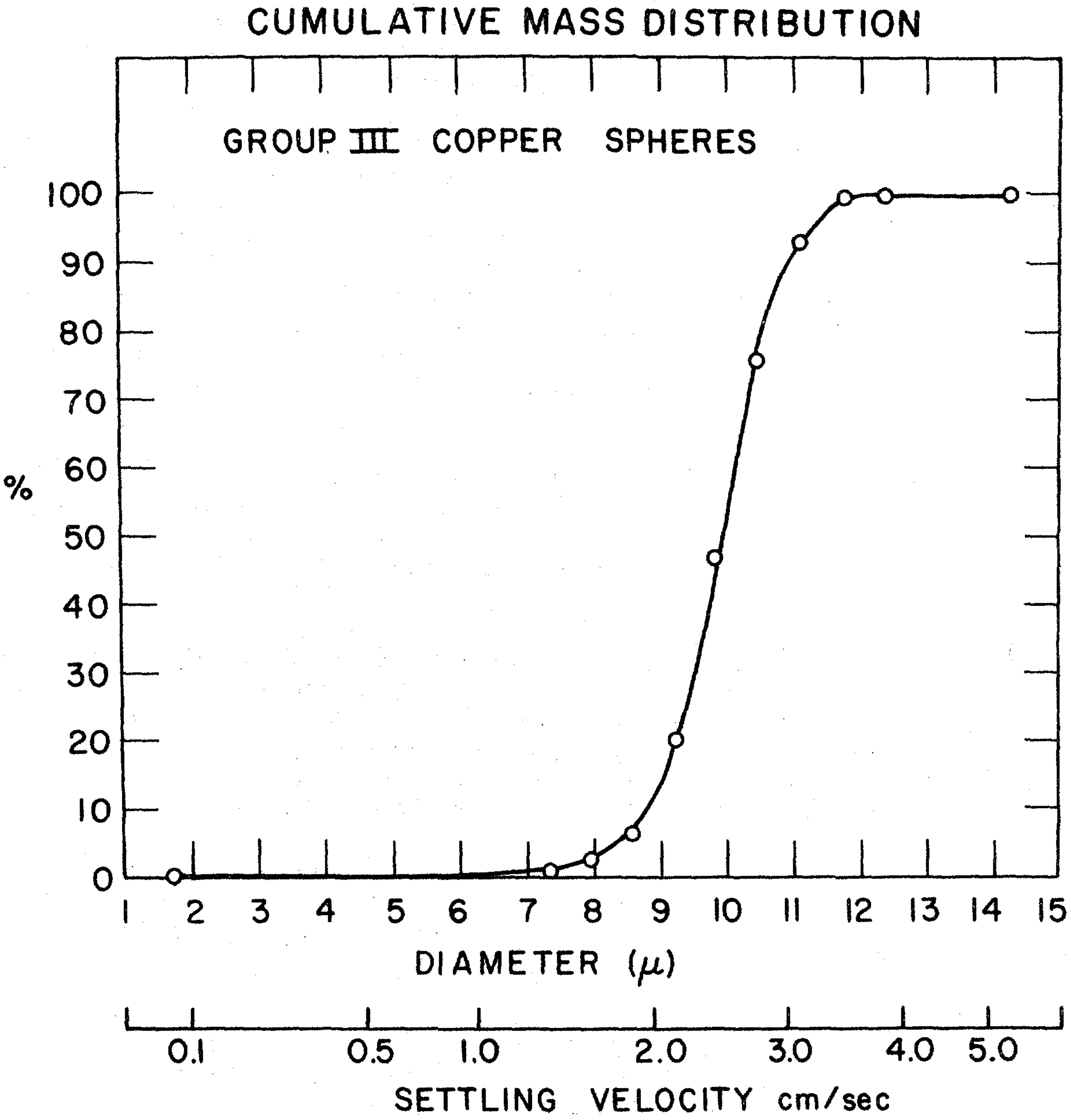

CUMULATIVE MASS DISTRIBUTION
GROUP III COPPER SPHERES
%
100
90
80
70
60
50
40
30
20
10
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
DIAMETER (μ)
0.1
0.5
1.0
2.0
3.0
4.0
5.0
SETTLING VELOCITY cm/sec

Figure 1

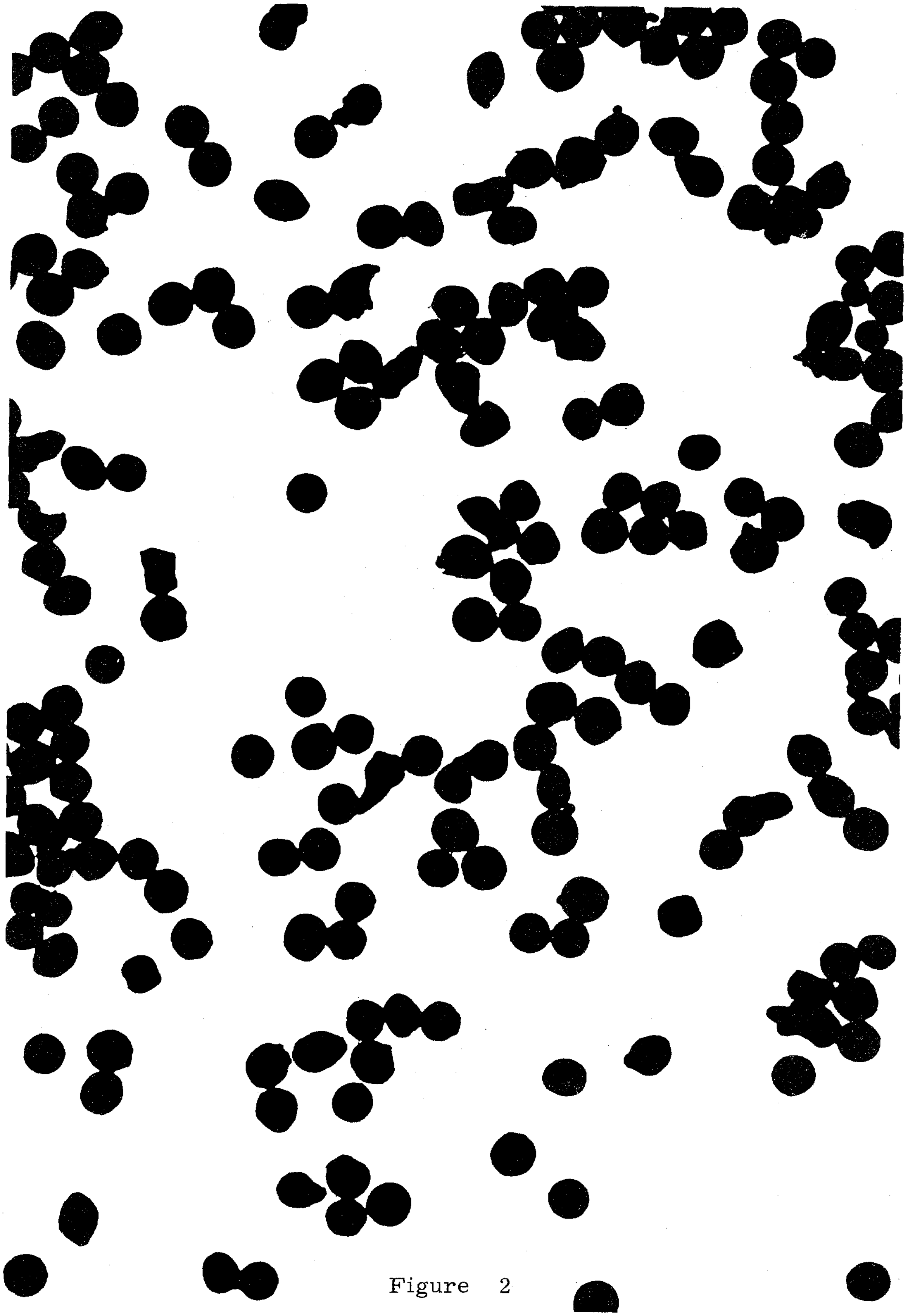


Figure 2

flexibility, the dual source assembly can be moved to any desired location and its height above ground is adjustable.

The sampling area is located in the eastern portion of the Laboratory site, and consists of a grass-covered field 100 meters on a side, equipped with sampling stakes at 10-meter intervals. The 100 stakes are served by a pair of underground vacuum systems, whose exhausts can be diverted downwind of the field to prevent fluorescent contamination of the filters. Currently, an individual stake supports three Millipore filter samplers at 0.1, 1.0 and 2.0 meters above ground. Each is equipped with its own mercury manometer and control valve for air flow adjustment. These samplers of course collect both the uranine dye and the radioactive copper spheres, although with different efficiencies. The small uranine particles are sampled properly, but the catch of the copper tracer is a function of particle size and wind speed. At present, we know of no perfect solution to the latter problem.

Deposition is currently measured by three different means and will probably be supplemented by other techniques as the work progresses. The standard sticky-paper squares, used extensively in fallout evaluation, are placed at the base of each stake and at a number of additional locations close to the source. Following each run, these papers are ashed and counted. Additional deposition plates are used for radioautographs, the current purpose of which is to establish that the particles are not agglomerating. At a later date, radioautographs may be used at all locations to differentiate between two different-sized particle groups emitted simultaneously. Fig. 3 shows a typical radioautograph of Group III particulate deposition.

The sticky papers represent a catch on a surface which is not identical to that of the natural ground and vegetation surfaces, and one of the aims of the study is to determine the relation between true deposition and that indicated by various collection devices. To provide direct evidence of true deposition, a cart carrying a complete radiation detection system has been constructed and this is used to survey the grid immediately after the termination of a run. The number and location of the sampling points includes all the stake locations and others at the discretion of the operator.

The meteorological instrumentation currently in use at the site consists of three very light cup anemometers designed and built by the Meteorology Group, a Gelman-Gill bivane, a sensitive, standard vane with a Styrofoam fin and a thermocouple temperature-difference system. These appear to be perfectly adequate for the present, but they will probably have to be supplemented at a later date, when work with very fine particles begins.

PROCEDURES

All of the initial testing is being done with wind directions ranging from NW to N to NE to obviate any possibility of disturbing sensitive counting operations at other locations within the Laboratory. Experience has already shown that this is

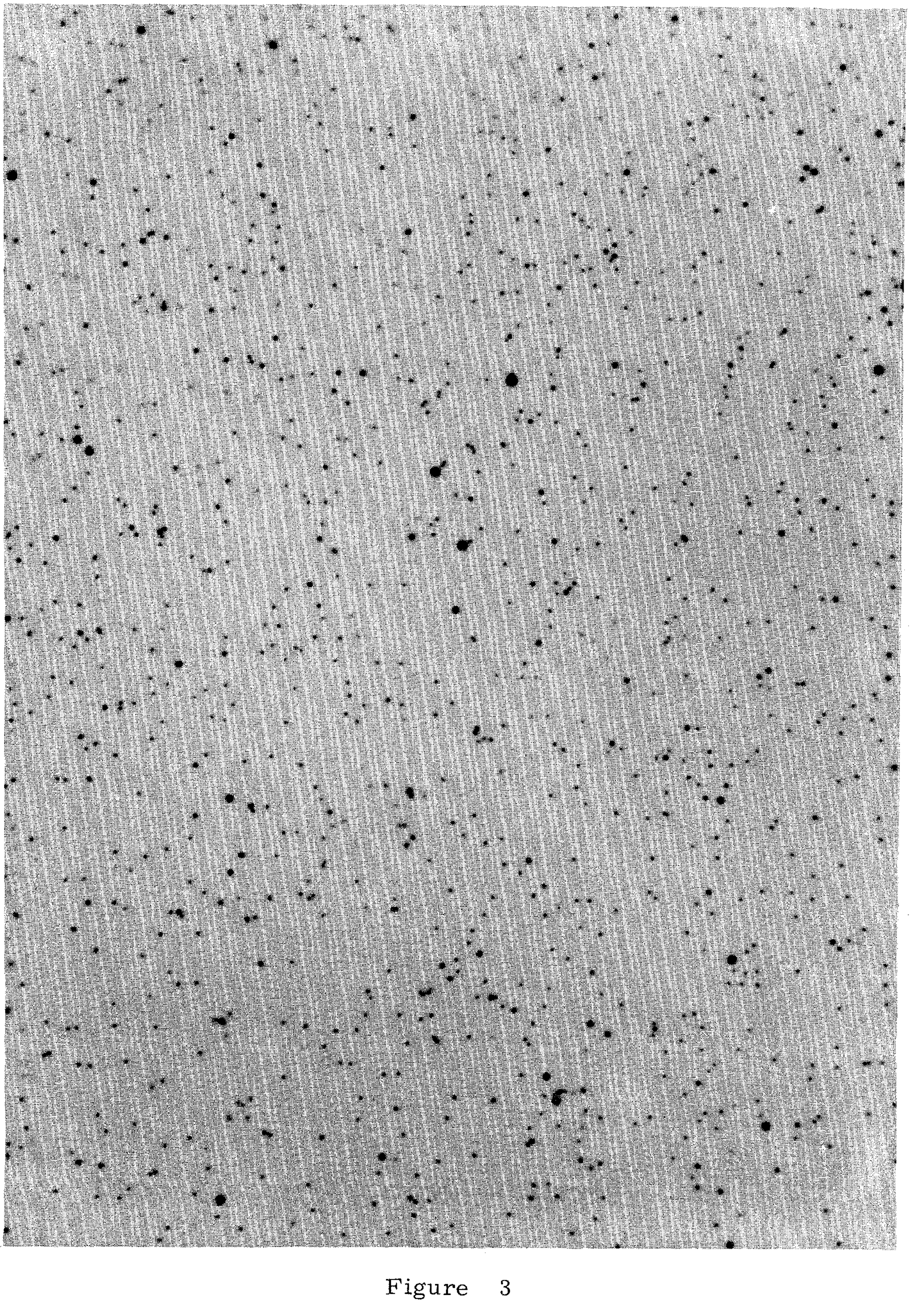


Figure 3

most unlikely, since the radioactivity is barely detectable by sophisticated means at the field boundary. However, it is desirable to establish the contamination potentialities quite carefully at first. The two wind vanes and the exhaust trails from the vacuum pumps are used as visual monitors during the course of each run. If the wind direction shifts to an undesirable quadrant for any length of time, the operation is terminated.

30 minutes is the normal operational period for each run, and the actual length is dependent upon the exhaustion of the copper-water slurry. Emissions totalling 0.5 to 1.5 curies produce readily-measured concentrations and deposition. Immediately following the run, all sticky paper samples and filters are collected and processed for counting, and the cart survey is begun. Virtually the entire Group staff participates in the operation at this stage, and the counting of the radioactivity is continued without interruption until all samples are assayed. In the course of the counting, the filters are not disturbed physically, and the uranine measurements are completed when the radioactivity dies away.

The 13-hour half-life of the copper is sufficiently long to permit a certain amount of re-evaluation following the initial survey. If, for example, one desires a bit more definition of certain areas or if there is question of the validity of some of the initial measurements, recounts can be made up to two days later at the hottest points. This afterthought procedure is of course dependent upon the continuation of favorable weather. The short half-life allows repetition of the experiments in about the same time period required to complete processing and preparation of the equipment for renewed operation (approximately 10 days). While this appears to limit the rate at which experiments can be performed, it does not constitute a serious defect since full evaluation of a given run could probably consume a month and a half at the very least.

RESULTS

Since it is anticipated that it will take most of the current winter to analyze the data accumulated recently, it is premature to attempt any firm conclusions at this time. However, it is interesting to review the data from a particular run to see what sort of information is becoming available. The August 11 test is probably as appropriate as any; at least it is realistic in that the distributions are not Gaussian, and certain portions of the analysis will not apply. The operation was conducted during a rather typical summer afternoon, with a mean lapse rate of -4.9° C over the 2-meter height interval. The mean wind speed at the 2-meter position was very light (2.6 m/sec) and the direction distribution was curious to say the least. Approximately 1.5 curies of copper was distributed in 26 minutes, accompanied by 1.2 grams of uranine. The source height was 1.75 meters and the location 5 meters west of the NW corner of the grid. The particles were of the Group III, having a mean mass diameter of 9.62 microns.

Perhaps the most interesting feature of the data is the comparison between the sticky paper and the cart surveys. One would expect relatively good agreement between them if gravitational settling were the dominant process, and this would be most likely with the large particles and light winds. In this case, there is little doubt that the two techniques have shown substantially identical results. Fig. 4 shows the two patterns, and even without formal correllation, it is apparent that even the small details are well-matched in both plots. It is also interesting to see the advantage of subsequent evaluation with the cart, for the fine detail in the vicinity of the maximum was obtained after the initial survey had been completed.

A second interesting plot is that of the vertical cross sections of the uranine and copper concentrations along the centerline of the plume. In this particular instance of course, the centerline is poorly defined, but Fig. 5 still illustrates the point. Both distributions are highly concentrated near the height of the source during the first few meters, but thereafter, as the uranine distribution becomes more uniform with height, the copper concentrations decrease much more rapidly, particularly near the surface. Such plots in runs with more normal wind distributions will allow direct evaluation of deposition velocities.

It is clear that the comparisons of the copper concentrations just above the ground and the deposition immediately beneath will constitute the most directly practical information available from the tests, but of equal importance is the variety of additional evaluations which can be made. For example, one of the most quoted references in deposition problems is that of Chamberlain (3). He has assumed that deposition for particles of this general size range can be approximated by applying a deposition velocity directly to the Sutton diffusion equation, and correcting for the depletion of the material. In these experiments, the uranine concentrations will permit direct evaluation of the Sutton diffusion parameters, and the effective deposition velocities can be computed and compared with settling velocities. These presumably will show good agreement with Stokes settling for the larger sizes, but may not be simple functions for the smaller sizes.

Similarly, the treatment of Rounds (4) and Godson (5) suggests a relationship between the distance at which all particles would settle if there were no diffusion, and that at which the maximum crosswind integrated dosage actually occurs. This is a function of the wind profile, the settling velocity of the particles and a virtual diffusion velocity. The latter is a parameter of considerable interest to the meteorological fraternity, and all the required data are available to assess this approach.

For analyses such as these, the field is suitable as it now stands, except for the addition of somewhat more detail near the region of the maximum, and this is easily provided. Later experimentation is expected to include studies in the forested area directly south of the present grid, where significant differences in the results may be anticipated. Washout of the particulates by rain may also be included, although this will be undertaken only after considerable preliminary work with dry deposition.

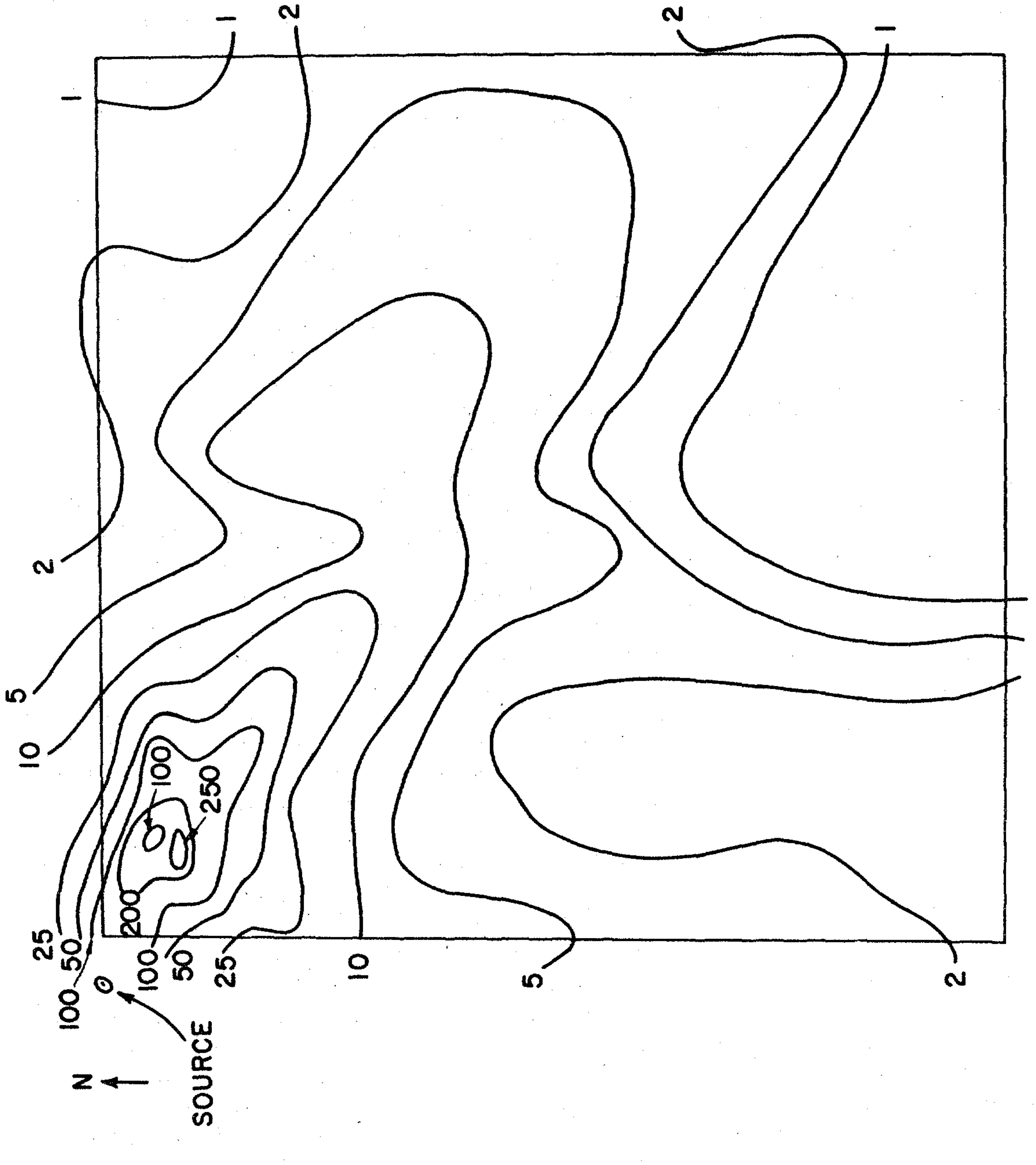


Figure 4A

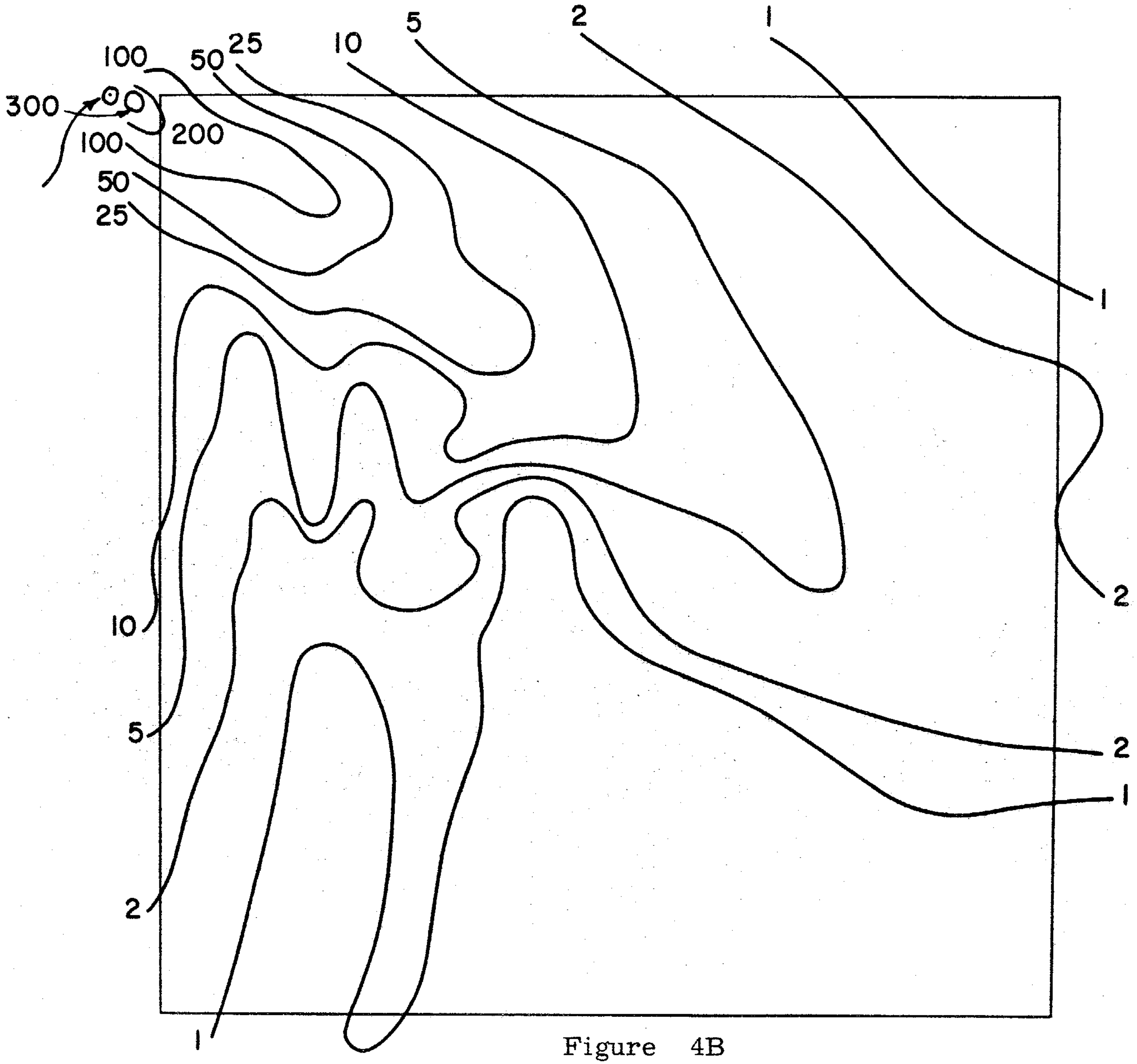
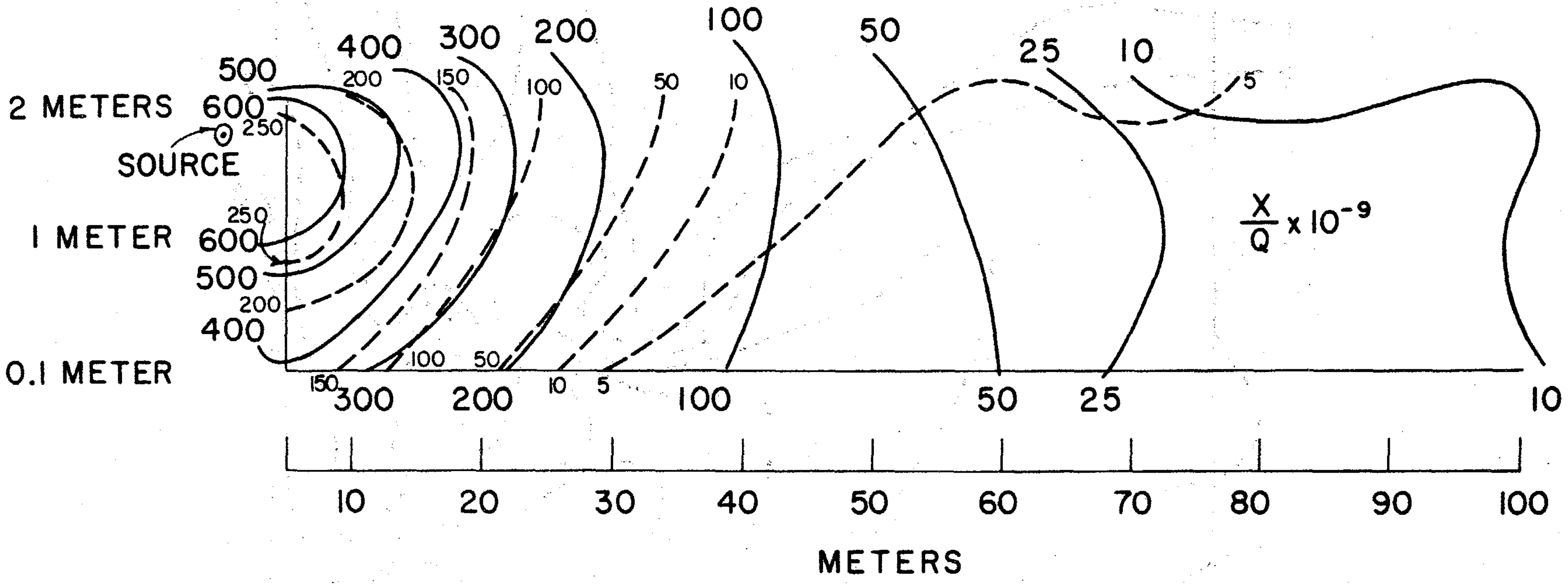


Figure 4B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$\frac{X}{Q} \times 10^{-9}$
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METERS

Figure 5

ACKNOWLEDGEMENTS

This paper consists of a broad survey of a major phase of Group activity and it should be apparent that the author is by no means the only individual involved. Details of the facility, instrumentation and analysis will be forthcoming under appropriate authorship. However, it is fitting, at this point, to recognize certain indispensable contributions. Gilbert Raynor, Seymour Rottenberg and Leonard Marcus have completed the design and implementation of the grid facility and established the operational and initial evaluation procedures; a long and arduous task. Robert Brown, Ansel Tuthill and Michael Butler have been responsible for the meteorological instrumentation, and Irving Singer and Ben Davidson (New York University) have been most helpful in planning the analysis. The Group has also received invaluable assistance from others in the Instrumentation and Health Physics Department. The Instrumentation Division, Calibration Laboratory and the Health Physics Division have all cooperated fully in developing equipment and establishing safe operating procedures.

Figures

1. Cumulative Mass Distribution, Group III Copper Spheres. The graph emphasizes the narrow particle size distribution achieved by careful water elutriation. The standard deviation is 14% of the mean diameter, as is true of the 2.7 and 4.6 micron particles also.

2. Group III Copper Spheres. A typical group of the 9.6 micron particles is shown after elutriation. While a few odd sizes and shapes are apparent, the vast majority are nearly spherical and of uniform size.

3. Radioautograph of Particles Collected in the Field. The film was exposed to the surface of a sticky paper collector which had been located in the deposition field. Most of the spots are quite uniform, but further examination is needed to determine whether the few large spots are true agglomerations or simply several particles very close together.

4. Deposition Patterns, Run No. 124. Deposition measured by two techniques is compared. Figure 4A shows the pattern obtained by means of the cart measurements and 4B those obtained from the sticky papers. Data in both cases are in curies/m^2 x 10^{-6}. Although not identical, the cart results and sticky paper results are quite similar. Note the superior definition of the cart data in the vicinity of the maximum.

5. Centerline Concentrations, Uranine and Copper, Run No. 124. In this graph, the uranine concentrations are presented as solid lines and the copper concentrations as dashed lines. Note that the uranine concentration becomes fairly uniform with height with increasing distance, while the copper drops off sharply. Also, the absolute values of the copper concentrations decrease much more rapidly than those of the uranine.

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ANALOGUE DEVICES FOR DISPERSION ESTIMATES

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ABSTRACT

The introduction, in 1959, of an electronic circuit to measure, directly, the standard deviation of meteorological parameters has provided the needed impetus for devising rapid and low cost methods of estimating atmospheric dispersion conditions. This paper discusses the Jones and Pasquill circuit, a few of its variations, its present use and possible future applications. In addition, a few examples of other analogue techniques are presented.

INTRODUCTORY REMARKS

At the Sixth AEC Air Cleaning Conference in 1959, I presented a paper describing a method which could be used to determine concentration and deposition estimates over periods of months and years. The procedure was straightforward and required only standard wind rose data. Concentration estimates of short duration, however, were, and still are, based almost entirely on Sutton parameters, although a second empirical model by Pasquill (1) and Meade (2) was beginning to gain adherents.

Coincidentally, a paper by Jones and Pasquill describing a simple, inexpensive electronic system for estimating the standard deviation of wind direction (for time periods of one hour or less) was appearing in the Journal of the RMS in July 1959 (3). Our experiments with this device, a similar device by Brock and Provine (4), and their potential application to pollution control are the subjects of my talk today.

Figure 1 is an imaginary wind direction trace; the ordinate representing direction, the abscissa time, T. This figure helps define the terms "Sampling Time"S, and "Averaging Time" τ. Parenthetically, I should say that these terms are not universal and may be called differently by various authors. Unfortunately, the terms are even interchanged, so it behooves the reader to proceed with caution whenever the literature mentions "Sampling" or "Averaging" times. The nomenclature here is that which is followed by Jones and Pasquill in their circuit description.

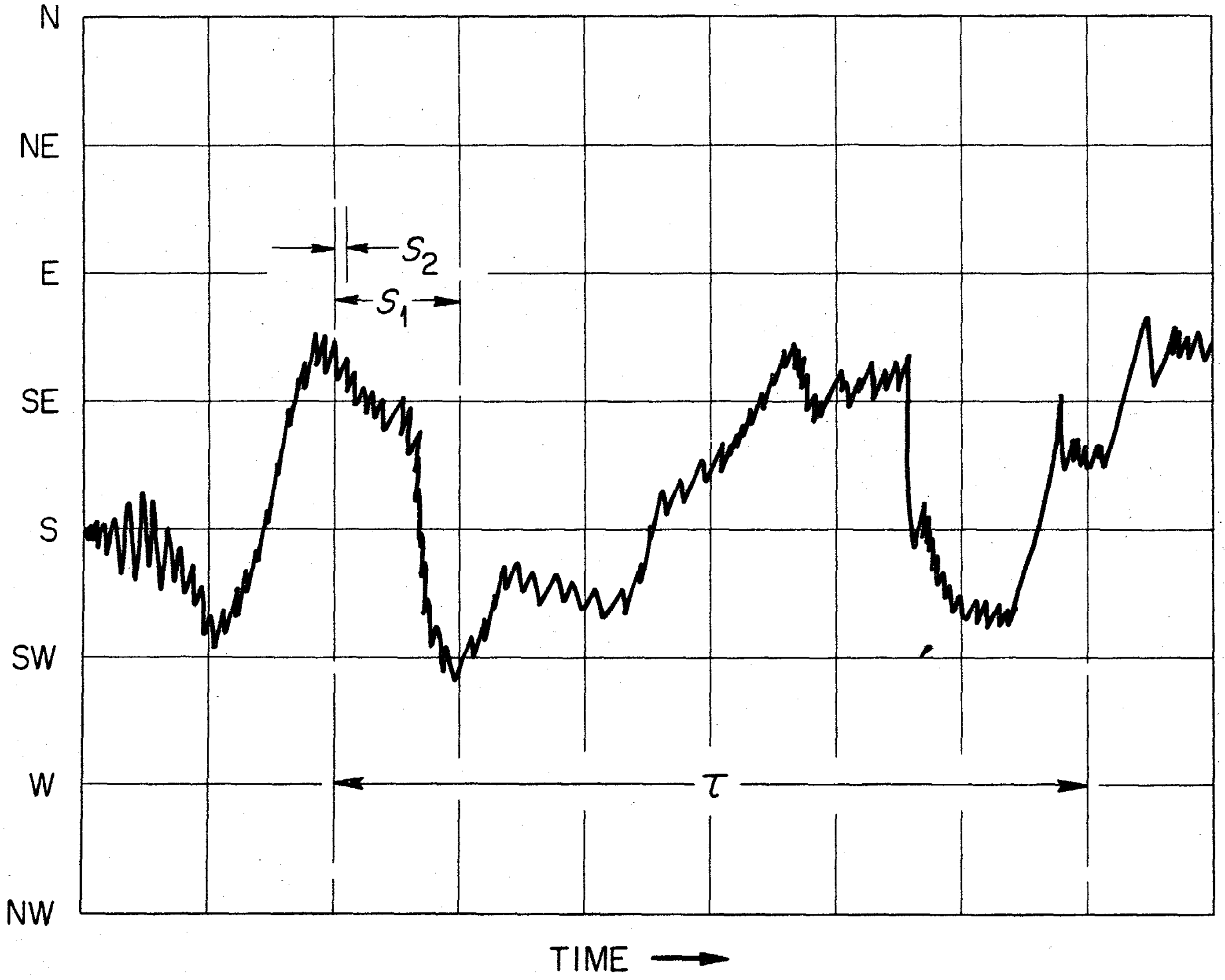


Figure 1 Relation of Sampling Time "S" to Average Time τ.

The sampling time "S" is the time interval between samples, i.e., one second, minute, day, etc. The value of S may be either the instantaneous value at the interval point or the average value over the interval, depending on the nature of the problem and the preference of the researcher. We can see, however, that a long interval, as represented by S_1 is insensitive to high frequency oscillations whereas a shorter period S_2 will enable higher frequencies to be detected. The moral, of course, is to select as short an interval as is consistent with the problem at hand. We'll come back to this principle in a little while.

The second parameter we are concerned with is the averaging time " τ ". A simpler term would be "period of interest." This period of interest might be the time an effluent takes to reach a nearby town or facility-or it may be a period of weeks or years, depending on the nature of the problem. For example, estimates of effluent behavior about an air intake or crew of men near a stack requires a τ of a few seconds. Industrial damaged to foliage, surfaces, etc., may be several miles distant and the period of interest might range from several hours for an accidental release, to many years from continuous releases.

Figure 2 illustrates the effects of sampling and averaging times of the frequency response of the experiment. Instrument men and High-Fidelity hobbyists will recognize this form of presentation. The ordinate is the percentage "power" (in this case wind vane movement) permitted to pass, while the abscissa is the frequency in cycles/second with which we are concerned.

The curve on the right labelled "one second samples" represents the response permitted by sampling once each second. Actually, this curve is smoothed a great deal. On the high and low ends are a series of damped oscillations which need not concern us here. It is interesting to note, however, there are works in the literature which display a function of these lobes on the low end and their meaning often seems to have escaped the authors.

From figure 2 it is obvious that one second samples are almost two orders of magnitude too coarse to fully utilize the sensitivity of wind vanes we have today. Yet digital analysis of small 20 minute run at the one second interval means punching and analyzing 1200 cards. Clearly, the millions of cards required for a run of one entire day is impractical.

Here is where the analogue technique shows its clear superiority. With an analogue averaging circuit, "S" is limited only by the response of the sensor. Since sampling is continuous all the sensitivity of the sensor is utilized, and a continuous record over days and weeks may be kept.

Having dealt with "S" we now turn to τ . Since τ is determined by the problem at hand, we cannot be specific in our remarks. At Oak Ridge we have a unit which measures the energy components from 0-5 seconds, 0-50 seconds and 0-500 seconds, the significance of these three intervals will be explained later.

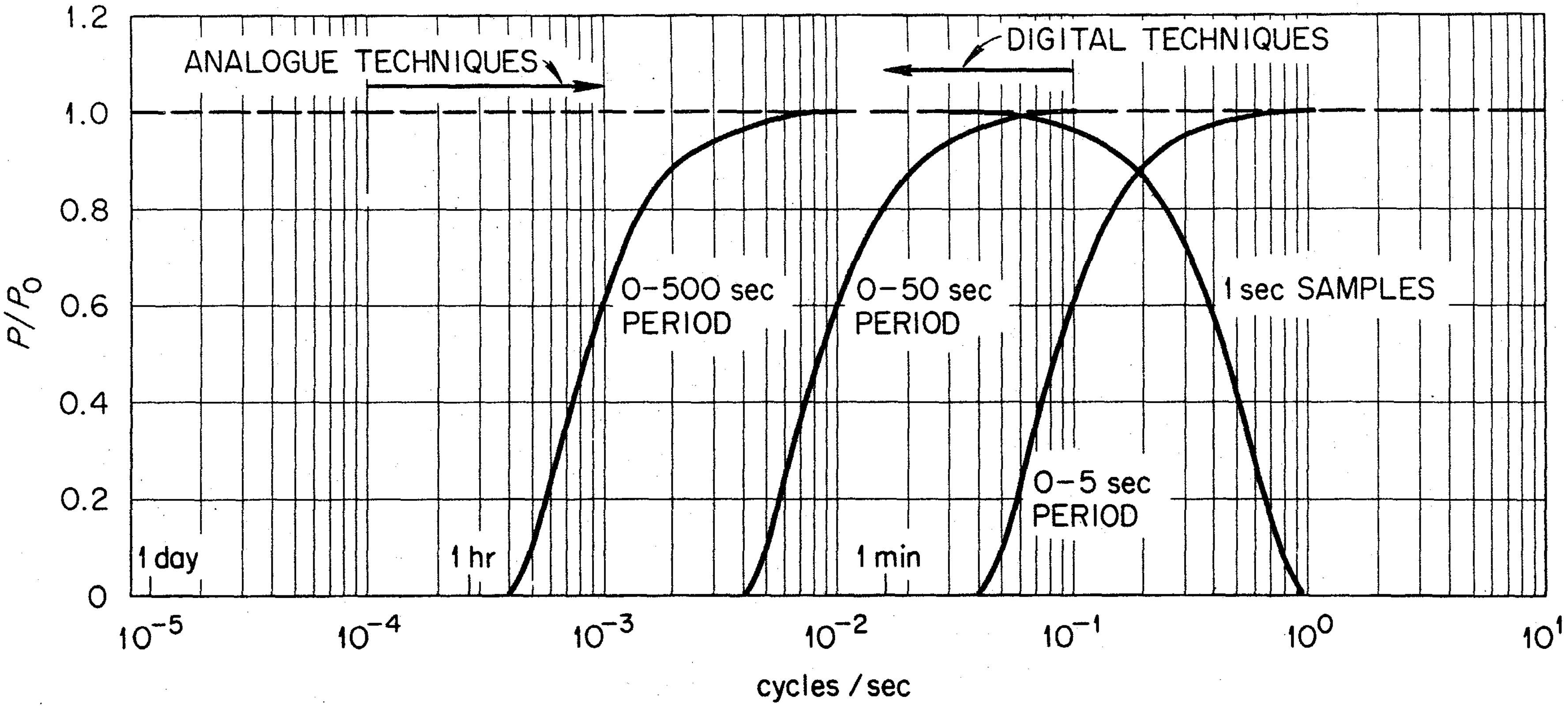


Figure 2 Frequency Response vs Analysis Techniques.

Finally, Figure 2 shows two arrows labelled "Analogue Technique" and "Digital Techniques." These represent, more or less, the practical limitation of each technique as they are now employed. The Digital methods may be used down to about 10^{-1} cycles/second although at considerable expense. They are clearly superior, however, in determining long (10^{-4} cycles/second or fewer) period wave behavior. Analogue techniques, on the other hand, have a definite superiority below averaging times of one hour.

I must add here, rather parenthetically, that Humphrey and Wong (5) have developed a low cost mechanical analogue system for extended period sampling. It can run indefinitely and gathers data by counting the amplitude of, and number of, wind vane fluctuations. At present its resolution is about 5°, but it seems apparent that this resolution could be improved to about 0.5° with standard commercial components. It would be extremely useful in gathering data on sampling times of from one hour to several years. It promises to obviate the need for punch cards, tape, etc. Our discussion here, however, deals with short range, high resolution devices such as the Jones and Pasquill circuit.

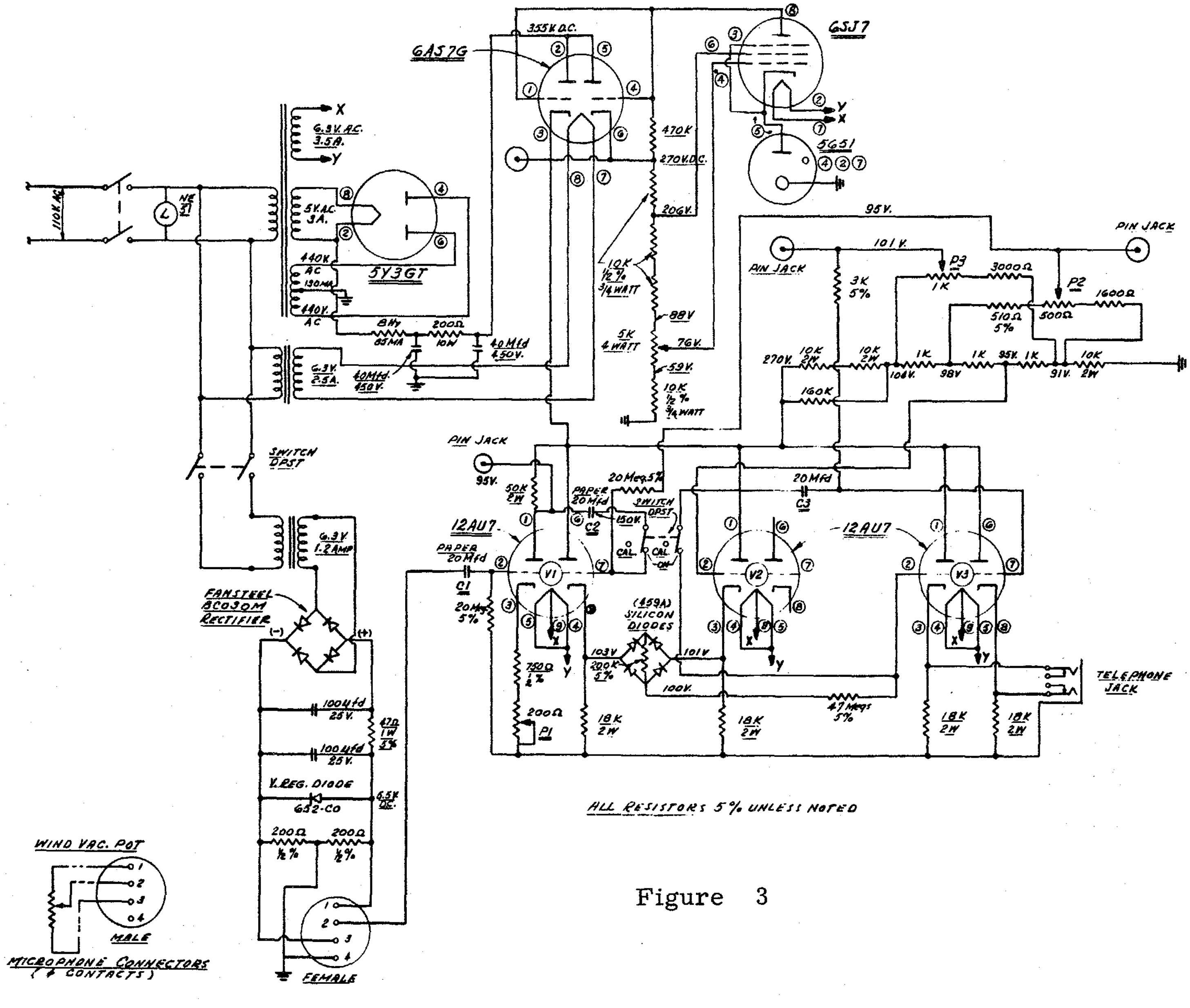
Without going into great detail about the Jones and Pasquill circuit, I would like to point out that the response curves shown in Figure 2 can be approximated by a two stage Resistance-Capacitance circuit, in short, a wave analyzer. The circuit is quite simple, using inexpensive materials throughout. Since "inexpensive" is a relative term, I might add that total parts for the three-channel model we built at Oak Ridge cost about $250, exclusive of labor.

Let us now turn to practical applications. The significance of the first circuit you will see hinges on the fact that the response of the wind direction sensor is a direct function of the dispersive property of the atmosphere. Thus this device will show, immediately, when good or poor dispersion exists, by indicating the standard deviation of wind direction fluctuations.

Figure 3 is our single channel version of the Jones-Pasquill circuit, showing the power supply, filter and output circuit. This particular schematic is for a standard deviation sampling time of 720 seconds. Subsequent versions of this model are essentially identical except for a multiplicity of channels and, consequently, a heavier power supply.

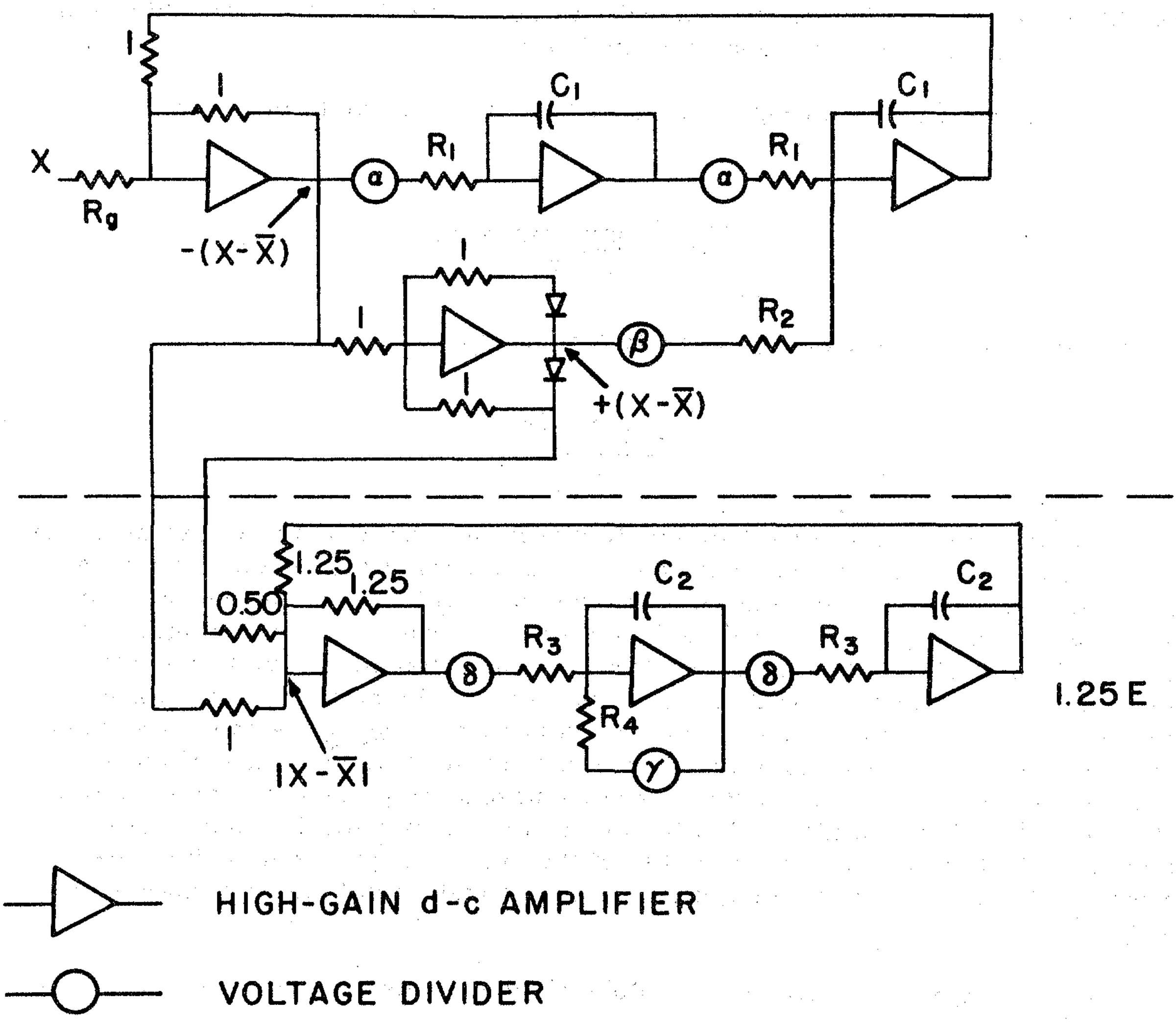
Briefly, the signal enters from the wind vane plug, is filtered thru capacitor C_1 and 20 million ohms, amplified by the tube V_1, filtered again by capacitor C_2 and 20 million ohms which shapes the response curve to approximate those in Fig. 2. Since this is an a.c. signal, we rectify it through a bridge circuit of four diodes, smooth it through another RC stage and read the result across the cathode of V_3.

For those of you familiar with, or have access to, a general purpose analogue computer, Figure 4 may be of interest. This is a design by Brock and Provine of the University of Michigan. The two stage differentiator (wave shaping) circuit, the rectifier, and the integrating (smoothing) circuits are quite apparent.



ALL RESISTORS 5% UNLESS NOTED

Figure 3



Brock and Provine's Computing Circuit to Measure Standard Deviation. All Resistors in megohms, All Capacitors in microfarads. (From Brock and Provine 03632 U. Michigan.)

Figure 4

The variations of these circuits are obviously quite numerous. One design we are toying with at Oak Ridge is outlined in Figure 5. This is a device for warning of low dispersion conditions. It is not a rigorously correct circuit, but it would serve as a useful tool to supervisory personnel in industries which must keep their effluent concentrations at a low level.

The centerline concentration of an effluent from a point source is

$$\chi_{C_L} = \frac{Q}{\bar{u}\sigma_\theta\sigma_\phi 2\pi f(x)}$$

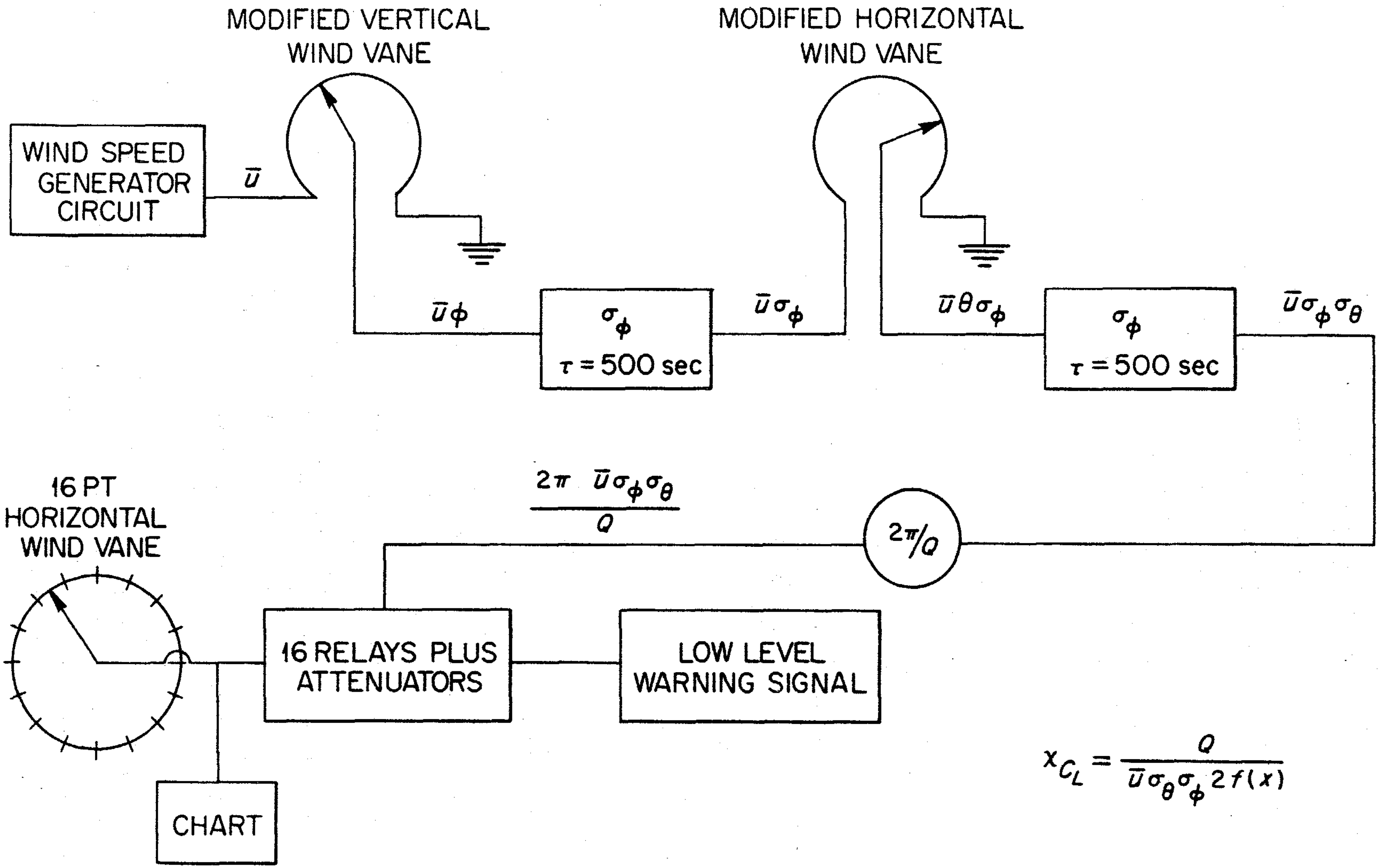
By electrical means we can approximate the inverse,

$$\frac{\bar{u}\sigma_\theta\sigma_\phi 2\pi f(x)}{Q}$$

The power for this circuit is supplied from an anemometer whose output is proportional to the wind speed averaged over a few minutes. If the wind speed were to drop below the anemometer's threshold, a voltage representing a constant wind speed (e.g. one mile/hour) would be generated. This voltage, proportional to $\bar{u}$ would supply the power for a vertical standard deviation circuit whose long time constant would, in turn, be fed into the horizontal standard deviation circuit. This voltage, which approximates $\bar{u}\sigma_\theta\sigma_\phi$ is then attenuated by a potentiometer whose value is proportional to the emission rate of the effluent in question. The final stage is a conventional 16 point wind vane, chart, and relay network which attenuates the signal inversely with the most critical distance in each direction. If the value of the output dropped below the predetermine "safe" level a warning signal appears and simultaneously marks the chart.

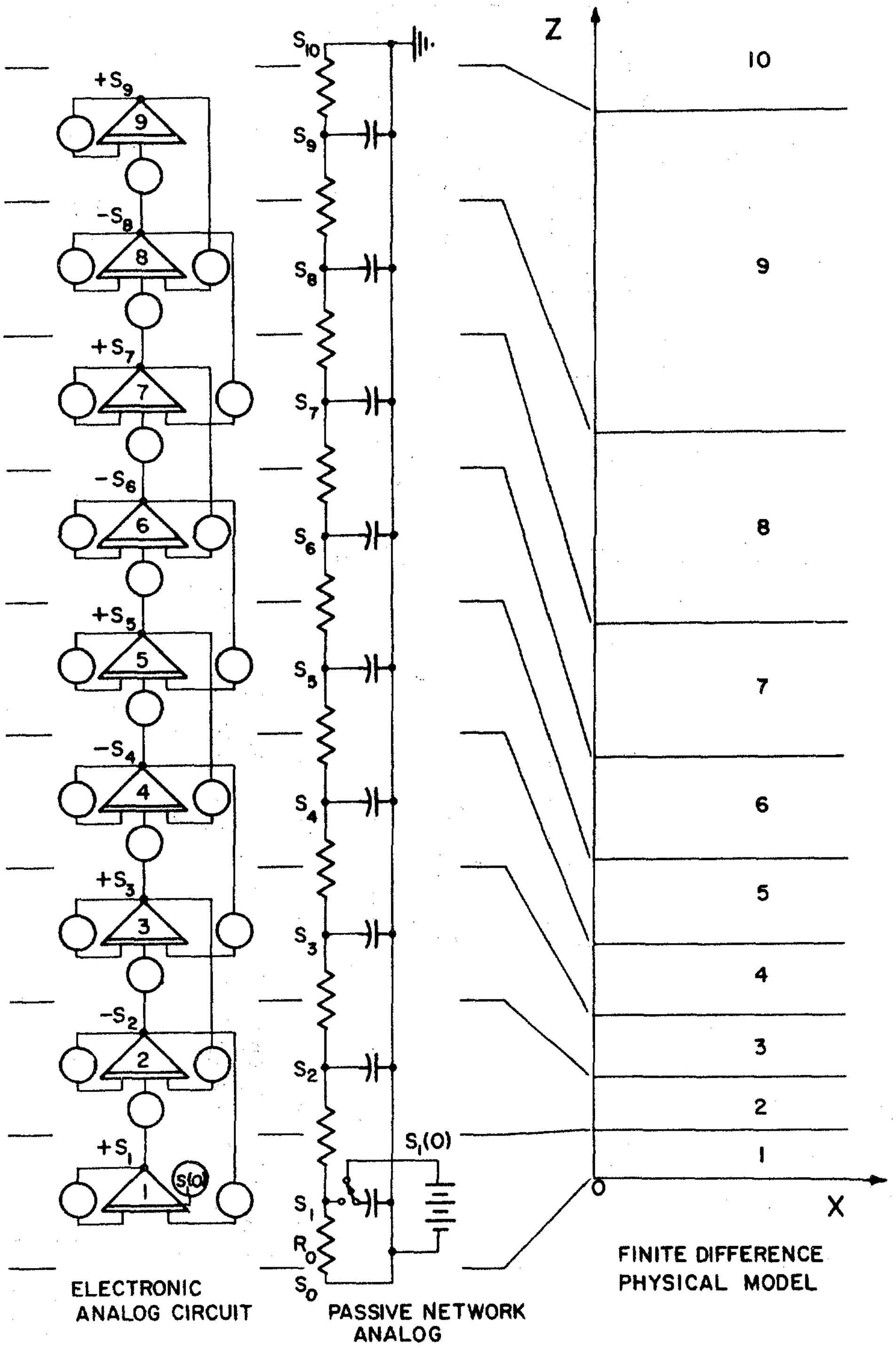
Now Figure 5 is somewhat of a "Rube Goldberg" device, but it could serve quite well in many situations.

Before we leave the subject of analogue design, I'd like to point out an interesting circuit by Brock for estimating concentration and deposition. Figure 6 shows Brock's circuit (6) on the left which uses standard analogue components and Karplus and Allder's (7) passive network on the right. The interesting point here is the approach that can be used. By using non-dimensional parameters these two circuits relate the physical capacity of the atmosphere to retain an effluent to the electrical capacity of a resistance-capacitance network. By appeal to the non-dimensional arguments and manipulation of the circuit components, deposition, absorption, concentration and other atmospheric and surface to atmosphere processes can be simulated. This particular model is for a continuous crosswind line source although dimensionally it is equivalent to an integrated crosswind point source. Of course, this model and others similar to it can be modified to simulate multiple sources, sources of various heights, etc. There is virtually no limit on the variety of problems we can subject to this approach.



Low Dispersion Level Warning Device

Figure 5



Electronic Analog, Passive Network Analog, and Physical, Finite Difference Model of the Atmosphere. Note that Each Group of Components Represents a Layer of the Atmosphere. (From F.V. Brock 03632-2-T, U. Michigan.)

Figure 6

Up to this point, the emphasis has been on what we can do with these devices. Perhaps this is the proper place to illustrate what has been done. Pasquill has worked extensively with his circuit to measure the spectrum of wind behavior. Brock and Hewson have applied their analogue circuits to theoretical models of the atmosphere. At Oak Ridge, however, we have been operating on a more basic level. We have attempted to gather continuous data of the wind behavior over an extended period of time, (i. e. one month). We selected the vertical wind distribution as an initial study since it lends itself easily to analysis yet little is really known about it.

Figure 7 represents the average standard deviation, by hour, of the vertical wind fluctuations for the month of August 1961, as obtained from a bivane in Oak Ridge, Tennessee. Three curves are shown here, the dark area from the baseline to the first curve represents the energy measured from the lowest limits of the wind vane response to a 5 second averaging time, the light area to the next curve represents energy from 5 to 50 seconds and the final dark area, the energy from 50 to 500 seconds.

The reason for choosing 3 sampling times may be explained by an analogy. To water a lawn with a garden hose, one sets the nozzle at an optimum spray angle and simultaneously uses an oscillating motion to cover those areas not included in the direct spray of the nozzle. Similarly, an effluent from a stack will disperse at some initial angle, but will appear to undulate downwind. This was described by Gifford (8) in his paper on undulating plumes. The concentration of effluent, accordingly, varies both in time and space. If we average the signal over differing periods of time, we obtain a comprehensive picture of wind statistics, enabling us to describe this concentration in both its point-time and space-time behavior.

Figure 7, indicates that the greatest vertical mixing in the summer months occurs from two hours to four hours after sunrise. Shortly after noon the lower layers are somewhat less unstable and we have approximately 10% less mixing in the vertical than at the peak hour. Vertical mixing decreases throughout the afternoon until sunset when it becomes fairly constant.

From the raw data (not shown) it follows that there are times when vertical mixing stops almost entirely. It is also evident, again from the raw data, that night-time conditions vary substantially more than day-time conditions. A complete analysis of the information Figure 7 represents would provide enough material for several papers, but the important fact I'd like to stress here is that it is possible to gather wind fluctuation data on the same continuous, routine basis as temperature and rainfall data. These data can be fed directly into computers for immediate use or stored for reference purposes. By applying the analogue techniques to our sensors, the problem of data reduction has been held to an absolute minimum. This means that we can have, continuously available, a measure of the atmosphere's dispersion ability, applicable at various preselected distances from a source, and with the capability of accounting for the effect of varying sampling times. The peculiarities of the effluent elements themselves, (adhesion, impaction, change of state, combination, etc.) should therefore become the limiting factors in concentration estimates.

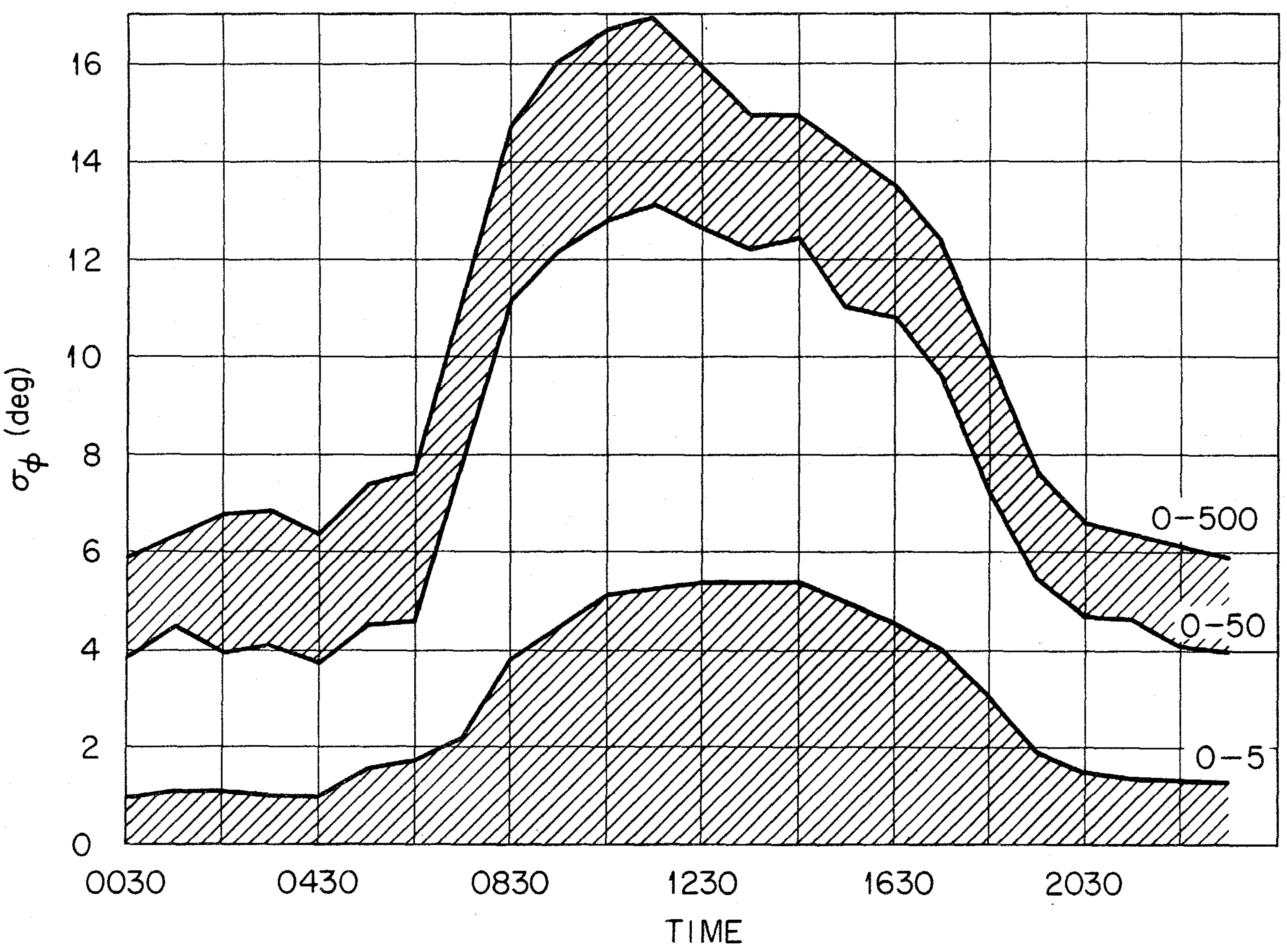


Figure 7 σ_ϕ vs Hours (July 28 – September 1).

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DISCUSSION

DENNIS: I think, in view of the complications involved in weather analyses that we are very fortunate in having had two beautiful, sunny days and only one cloudy day here in Brookhaven. I would like to set a maximum time period of ten minutes for any questions that the audience would like to direct to Mr. Pack, Mr. Smith or Mr. Culkowski.

ANTON: This question is for Mr. Smith: Will you be getting into the hot spot problems? I know you pick up hot and cold spots in your field, but does your setup provide a tool to go into the hot spot problem?

SMITH: I presume you mean what occurs in the larger scale fallout? I don't think we will have a hot spot problem in our work. We will have variations that won't fit the patterns one would like them to, but every indication so far is that we are not going to have the same type of thing that happens in the large scale fallout, where a single large particle may give a very high reading, and the rest would be considerably lower.

It is really hard to say what is going to come out of this series of tests. I find myself uncertain about that at the present time.

FISH: There is some indication in the literature that in certain parts of the country the dispersal of fallout back from the ground by wind and by other agents might be important. I wonder if you have any plans to examine this factor here at Brookhaven.

SMITH: We already know from the studies we have made that this is an area in which very little redispersion does occur. We have considerable evidence of various types to show that. The Hanford area is far more appropriate for it and they are already working on this problem, so that we feel that we are ill-equipped in every respect to do it here. I think we will ignore it, largely. It certainly will not affect our own tests, except in a very high wind.

May I make a comment on both Mr. Pack's and Mr. Culkowski's presentations without detracting in the least from their value? I think it is important for everyone to recognize that using the standard deviation of the wind fluctuations is of maximum value over short time scales, distances, and simple terrain. Much has been done in England and in this country (for example, at Prairie Grass, Davidson's work at Peekskill and Cramer's analysis) to show that effluent does travel for a considerable distance in straight lines from the source and consequently the standard deviation of the wind directions both vertical and horizontal, reflects what is happening in the atmosphere. However, as the scale of the problem increases, it is obvious that a single wind vane can no longer represent the flow completely. The problem then becomes a matter of large scale diffusion, trajectories and things of that nature. The important thing is to recognize that this application is extremely helpful in stack problems and much less helpful at longer range.

PACK: I agree with that. For that reason we are taking several hours of digitalized one second bivane data to look into the statistical characteristics of how it changes over long averaging times, which you can't do with the analogue techniques. At some place the dispersion ceases to be as the inverse power squared of the distance and becomes something between the inverse square and the first power. This is why we extended our tracer test to two miles, so that we would have more information on that.

Certainly as far as terrain influences are concerned, we must know more about these, and we have a program going now to measure wind fluctuation in the Blue Ridge Mountains in as irregular a terrain as we can find at our test and development center, Sterling, Virginia, which is rolling farmland and over the water at Chesapeak Bay. We want to compare these data and see just what happens as a function of terrain. The work under way now and the results are beginning to show some significant differences.

STERN: I could agree that the Chicago atomizer would be ideal for short range dispersion of the copper particulate. Have you any plans for the long-range application of this technique?

SMITH: No, I have none at all. The entire effort has been designed to take care of a local, low level, highly concentrated study. I have no large scale plans, certainly not here.

WEHMANN: Mr. Smith, how are you going to handle the problem that cropped up on September 1st with the fallout, as far as your program is concerned?

SMITH: We have been rather careful to look over the area to make sure that fallout is not bothering us and, at present, we have been unable to detect any significant increase in the local background or significant variations in the background of that particular field.

Since the fallout began in earnest again, we have made rather certain that we surveyed the field prior to the operation.

UNKNOWN: You have no single channelizer, so you are measuring the gross activity?

SMITH: Gross activity above the background of the field itself, which we have determined to be at a low level and of fairly constant value. Remember, we are not observing directly at the surface, and we are talking about several orders of magnitude difference at the present time.

DENNIS: We are going to hold off discussion until we complete our agenda this morning. I am going to announce now that we do have a foreign representative, Mr. John Riley, from Risley, England, who is a member of their reactor group. His concern has been principally high temperature reactor effluent cleaning. We are pleased to have him here. He has volunteered, without any advance warning, and with no opportunity to prepare a formal manuscript, to present a brief discussion on containment problems in their reactors.