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USE OF DEPOSITION COLLECTOR PARTICLE-SIZE DATA IN THE INVESTIGATION OF ENVIRONMENTAL EXCHANGE MECHANISMS OF PARTICULATE POLLUTANTS

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DISCLAIMER

INTRODUCTION

Presently, one of the greatest environmental controversies is the importance of plutonium-in-soil measurements to the assessment of environmental impact and population dose. The consensus at a recent "Workshop on Environmental Surveillance Methodology" was that with the installation of more and more elaborate filtration systems in plutonium handling facilities, expected incremental fresh deposition can be expected to be indistinguishable in the present plutonium-in-soil inventory. Surveillance of recent depositions and re-entrainment, therefore, will have to be evaluated by some means other than soil sampling, i.e. deposition collectors.

Particulate deposition collector data have been generated by enviromental surveillance programs since the onset of concern over radioactive fallout. However, these devices have seldom in recent years been recognized for their unique collection and data characteristics and exploited to the maximum extent. Concentration and particle-size distribution analysis techniques are presently being used to extend the application of deposition collector data to the assessment of population dose and the investigation of environmental exchange mechanisms of particulate pollutants.

PARTICLE SIZING APPARATUS

The particle-sizing for this study was accomplished through the utilization of a new concept in the sizing of radioactive or non-radioactive airborne particulates.[1,2,3] The glow-discharge perturbation method economically generates particle-size data which can be directly accumulated and spectrometrically analyzed by a multichannel analyzer.

The device shown schematically in Figure 1 makes use of a constricted d.c. glow-discharge maintained between two electrodes separated by a disc containing an orifice. If a particle is allowed to pass through the orifice, a momentary interruption, proportional to the particle size, occurs in both the discharge current and voltage. These perturbations can be analyzed to determine the particle sizes and numbers.

The glow discharge tube consists of two glass sections, joined to each other by a standard taper ground-glass joint. Overall dimensions of the tube are 3.8 cm diameter by 9 cm length. The lower section houses a 1.9 cm diameter by 0.64 cm thick molybdenum cathode and acts as a supprort for the 2.5 cm disc which contains the sizing orifice. The upper section contains a tungsten ring-electrode and the particle injection stem. The anode-cathode separation distance is 2.5 cm. Particles leave the injection stem 0.4 cm above the orifice.

Air is admitted to the system through the inlet impactor. The impactor is composed of the first stages of an Anderson Cascade impactor and serves as a roughing device to eliminate particles with diameters larger than the orifice. The air stream is then split; one part going through the gas discharge tube and the other going through a filter.

Air is drawn through the gas discharge tube to the maximum capacity of the vacuum system and the orifice, with particulates in this stream being sized.

The filter arm of the system serves to provide: (a) the flowrate necessary to make the cascade impactor functional, (b) the variability

of flowrate necessary to make the system applicable to particle sizing and sampling situations which require near isokinetic sampling, and (b) a sample of the airborne particulates which can be analyzed chemically or physically to provide information on the composition of the particles being sized. Both arms of the vacuum system are monitored for pressure and flowrate.

Figure 1 includes the schematic of the elctronic circuit used with the glow-discharge apparatus. Negative high voltage is applied to the cathode of the discharge tube and the anode is grounded. When a particle passes through the orifice in the discharge tube, it momentarily reduces the current flowing in the tube. A voltage pulse proportional to current is coupled from the cathode circuit through a voltage dividing network and to the input of a linear amplifier/pulse shaper. The linear amplifier insures that the pulses reaching the multichannel analyzer are proportional to the pulses leaving the cathode circuit of the discharge tube, and at the correct polarity. A pulse shaper circuit is needed to widen the very narrow pulses generated by the discharge tube to one acceptable by the multichannel analyzer (MCA).

Using the MCA in the pulse height analyzer mode then creates a one-to-one correspondence between incoming pulse height and increasing channel number. Thus the output of the MCA yields the pulse height spectra of the particles passing through the discharge tube during a given time period. For a typical room air particle-size spectrum, the integrated system background contributes only three percent to the gross particle-size distribution.

DEPOSITION-PRECIPITATION COLLECTOR

A combination precipitation/particulate deposition collection device has been designed to maximize the utility of the generated data.[4,5] The collector which is used at ground level consists of a funnel of 25 cm diameter (483 cm^2) as indicated in Figure 2. Clamped into the top of the funnel is a 1/2 inch-thick disk of rigid fiber air conditioner dust filtering material. Attached to the stem beneath the funnel is a plastic bag which collects the precipitation if any occurs. The device is inserted into a stainless steel funnel stand at the sampling location.

Since particle-size distribution data are being extracted from these devices, it was found useful to provide a protective lip around the fiber disk. In this case a funnel with a 10 cm inherent lip was selected. Witout the lip, especially at ground level, particle-size analysis is inhibited by the presence of massive particles and other debris.

Selection of the collection medium also proved to be very important. Since the particles are necessarily re-entrained by the vacuuming action of the particle-sizing device, the possibility of using sticky paper was eliminated. Significant resuspension of collected particulate material seemed to occur with the use of relatively fine pore filter paper types. Air conditioner fiber dust filtering materials were observed to give the coarse surface necessary to hold the particles until removed for analysis and also proved to be acceptable for transmission of precipitation to the plastic bag.

PARTICLE-SIZE ANALYSIS

One of the basic uses of particle-size spectroscopy is qualitative identification of particle-size distributions in environmental samples.

E. S. Gladney, et al[6] have shown that identifiable contaminats can be expected to be associated with distinctive particle-size distributions. As has been contended previously[3] Gladney states that the size distributions are so distinctive that particle-size data can be used to identify sources of elements when emissions from various types of sources have been characterized by the same methods.

If only one single distribution peak existed in the spectrum, with no larger particle-size distributions contributing any counts to this peak area, no component resolution problems exist. However, this is very seldom the case, an allowance must be made for overlapping distributions. The Compton Continuous subtraction method[7] has been adapted to the spectrum stripping of composite particle-size distributions of environmental origin. Application of this technique involves locating distribution peaks on the relative frequency vs particle-size plots and drawing a line along their base. (This technique insures that when the inserted line is subtracted from the relative frequency coordinates of the peak coordinates at the same particle size, the resulting particle-size distribution peak will have an upper bound.) Starting at the peak representing the largest particles, this subtraction is done and the subtraction peak is examined.

Similar to the Davies procedure[8] the assumption is made that even the most complex particle-size spectra are finite sums of simple log-normal distributions. The examination of the subtraction peak therefore involves the plotting of this distribution on log probability paper and the determination of its linearly on this plot; linearly denoting the presence of a single distribution nonlinearly denoting a mixture. If a

mixture still exists the procedure is iterated on this subgroup of the original spectrum.

Following the resolution of the first major peak of the composite distribution, the next lower particle-size peak of the composite spectrum is analyzed. The technique as applied to a fictitious composite (d) of (a) Hanford background[9], (b) Washington airborne particulates[10] and (c) simulated fallout[11] is illustrated in Figure 3. Line (e) is the subtraction line. There is no precise "right" way to draw this line. It is largely a matter of practice and judgment.

Identification of a resuspension distribution from deposition collector particle-size data is illustrated in Figure 4, with results being in general agreement with earlier work.[9,12]

ATMOSPHERIC CONCENTRATION

It has been demonstrated[13,14] that large numbers of nuclide/medium combinations exhibit long-normal distributed environmental concentrations when these are plotted for a location over time or for one time over many locations. In fact it has been said that when the applicable distribution is unknown, a log-normal one is the first alternative to try.[13] Figure 5 shows a typical background environmental concentration distribution for a single time for 48 locations. Note that when plotted on log-probability paper a straight line results. For many sets of contaminant concentrations in environmental media, it has been found that slopes are near 2.

The presence or absence of plant or re-entrainment contributed contamination to this medium/nuclide combination can be detected by a change in the slopes of these curves. Figure 6a indicates the concentration distribution resulting from a re-entrainment contribution. The composite distribution, the sum of background and other contributions, as it was actually found to exist in the environment is shown in Figure 6b. Because of the additive nature of environmental concentration distributions, the single components can be resolved directly from the log-normal plots for relatively simple composite spectra. Complex shapes have been observed and experience is still being developed to lend a physical interpretation to each of these.

SYSTEM APPLICATIONS

The potential contributions of this deposition/particle-sizing approach to investigations of particulate pollutant behavior in the environment appear to be many. Applicability of this approach to some difficult problems in environmental data interpretation and utilization are worth mention. Noteworthy among these is the partition of responsibility for population radiation exposure among local contributors of airborne particulates to shared environments where differentiation by isotopic spectrographic means is difficult or impossible.

As a result of the trend toward the development of multipurpose, multi-owner nuclear industrial parks, it seems likely that situations will arise in which the determination of regulatory compliance will be undeterminable by isotopic analysis of environmental samples. It is

possible to develop a Responsibility Factor (RF) for use in determining compliance based upon data generated by particle-size spectroscopy using the gas discharge perturbation method. Assume that all particle sizes are of equal concern. The two quantities necessary to establishing a RF are: (a) that part of the "total particle-size distribution integral" that is the effluent contribution, and (b) the "total particle-size distribution integral" itself. The RF is simply defined as the quotient of these two quantities, the part over the whole. Possible values would be in the range $0 \leq RF \leq 1$. In the radioactive particulate case, the calculated RF would be multiplied by the radiological results of radiological data to establish the contribution of a particular facility to the reported numbers.

CONCLUSIONS

The construction of environmental contamination pathway models has demonstrated the acute need for re-entrainment data[4] in the assessment of population doses. Experience is showing that the deposition collector particle-size technique has the capability to provide information on both concentration and particle-size distributions in the environment. The plutonium deposition and re-entrainment question is only one of the many areas in which this technique is applicable.

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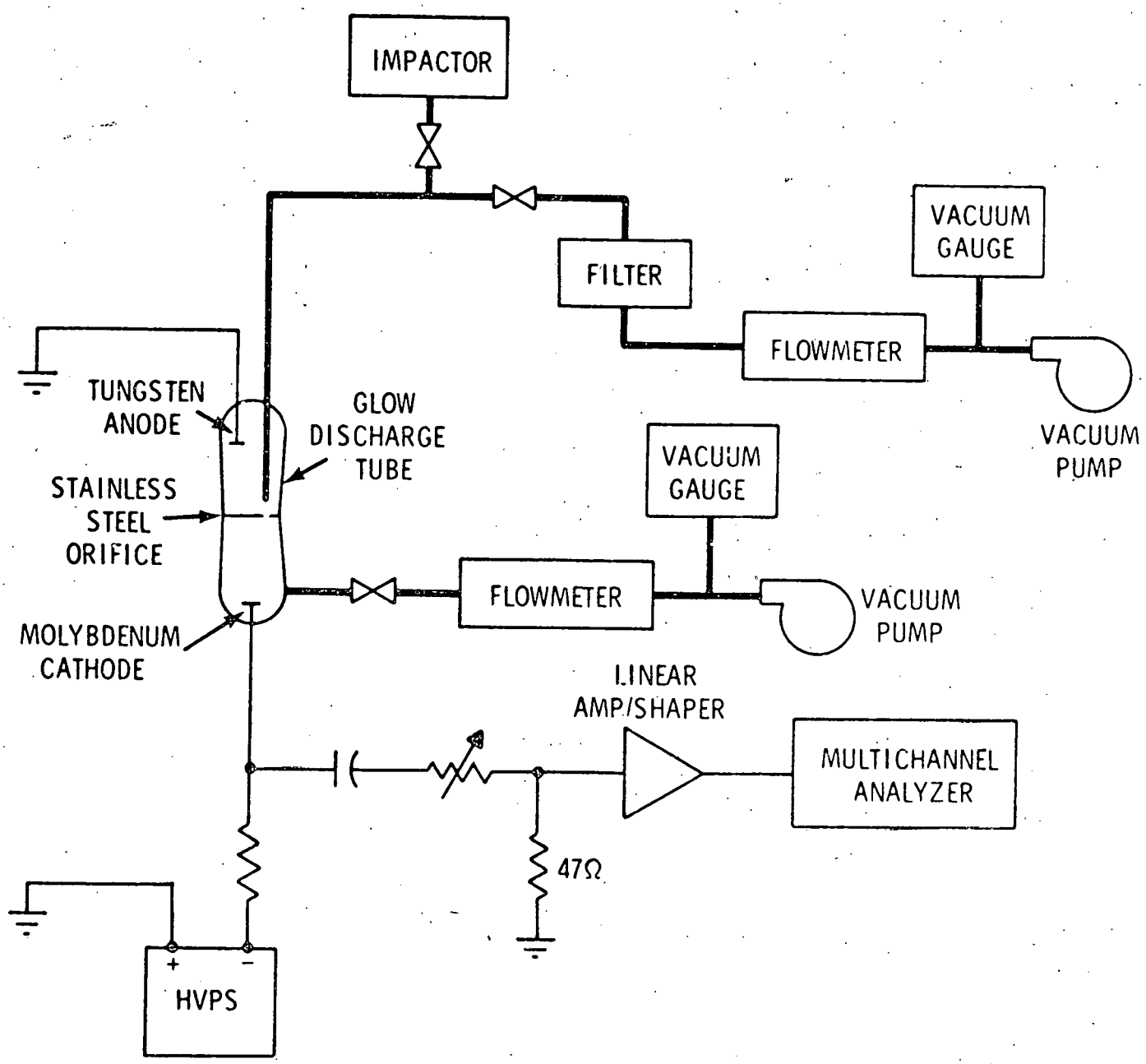
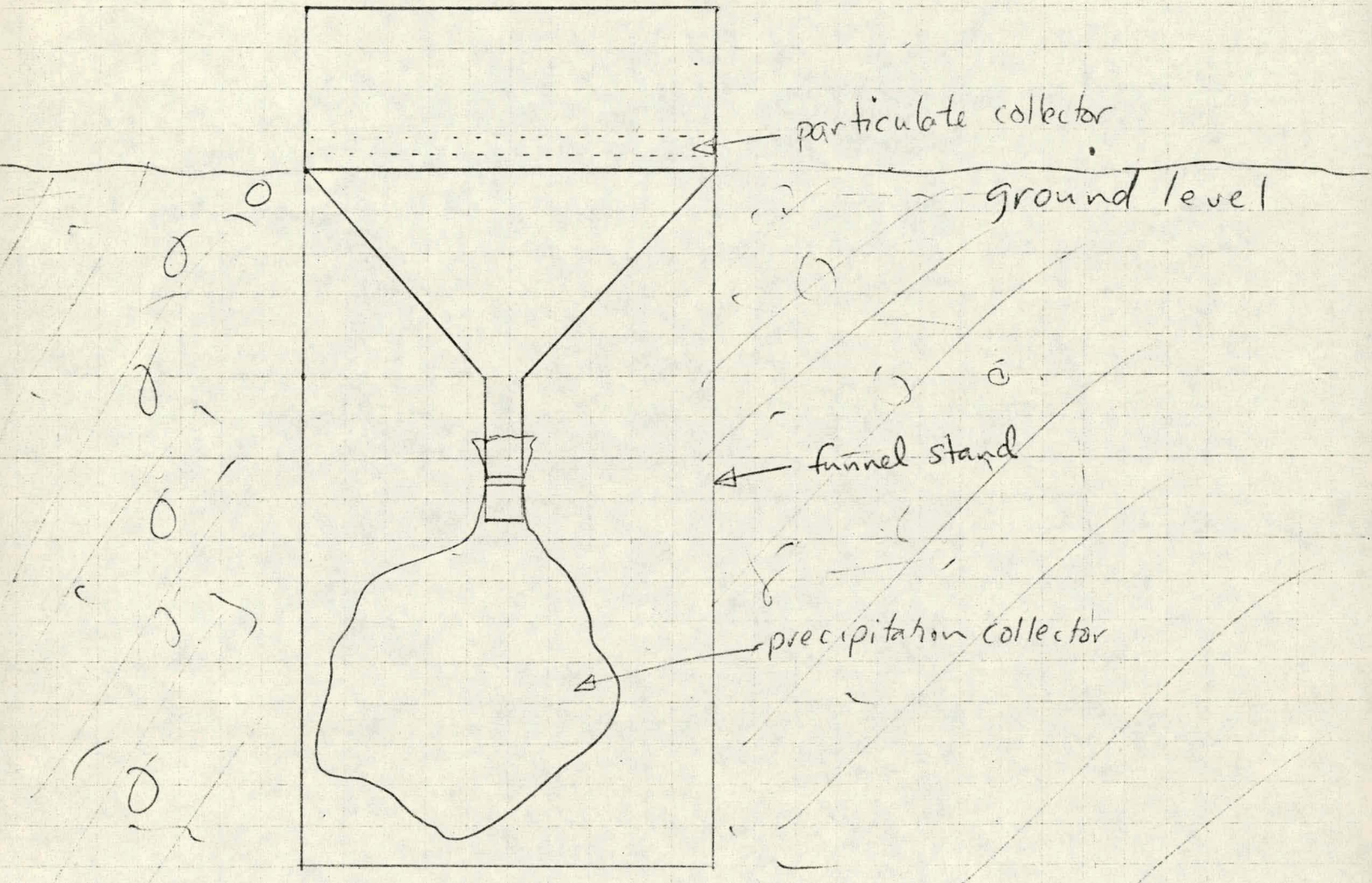


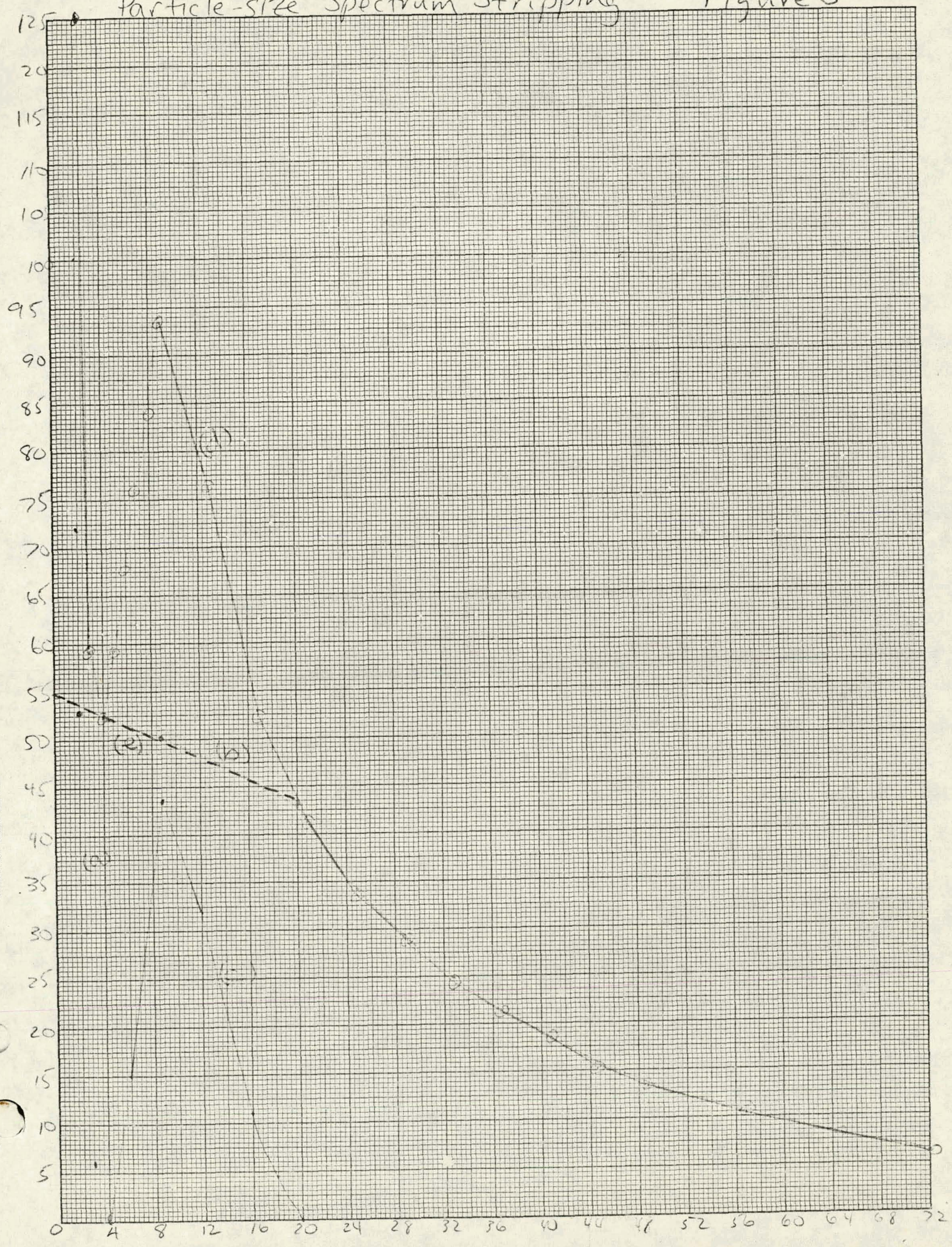
Particle Sizing

FIGURE 1.1 Apparatus Schematic Diagram

Deposition Collector Schematic Figure 2



Particle-size Spectrum Stripping Figure 3



Re-entrainment Particle-Size Spectrum: Stripping

Figure 4

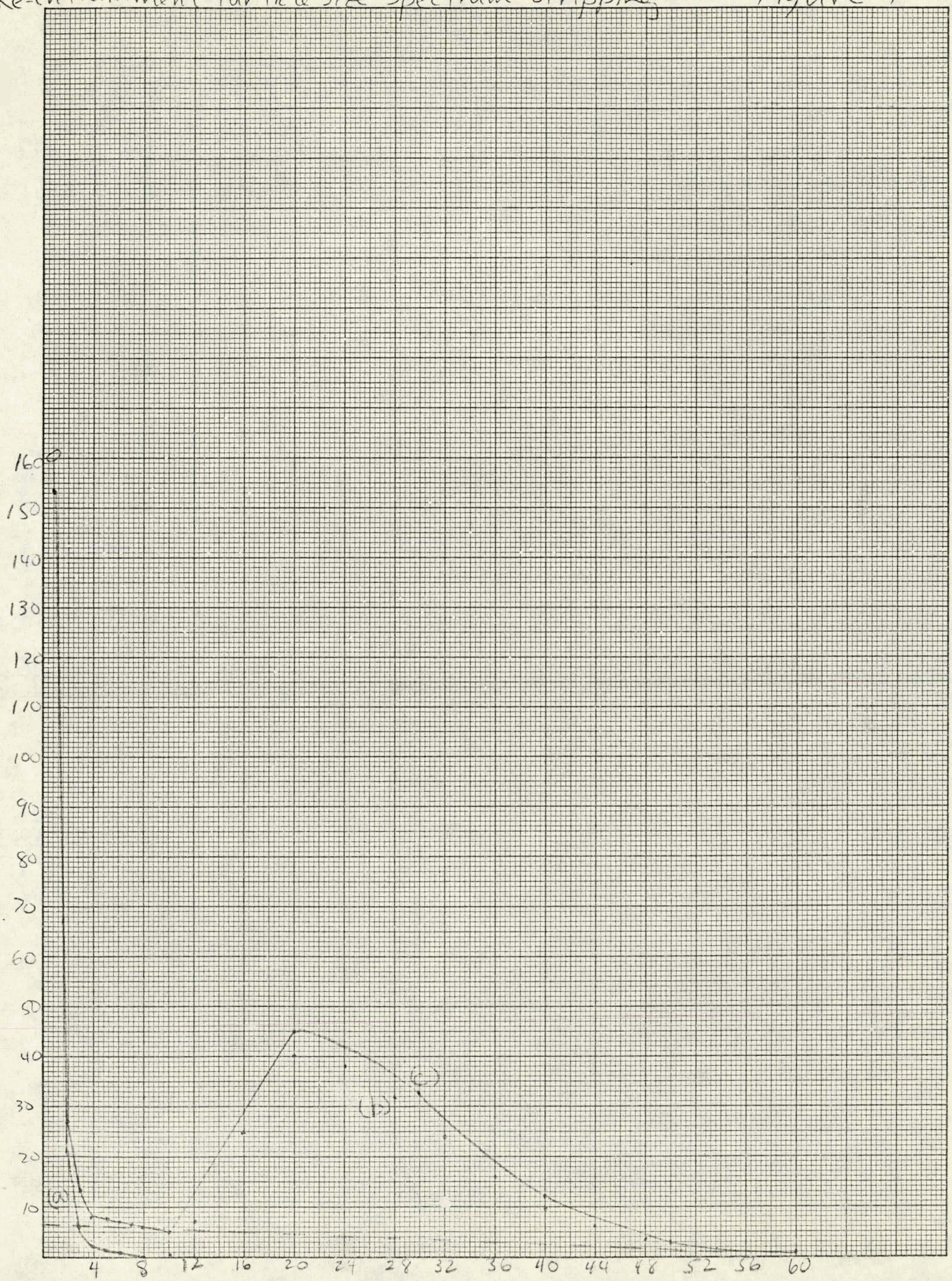


Figure 5

Contaminant

LOGNORMAL DISTRIBUTION OF ENVIRONMENTAL CONCENTRATION Background

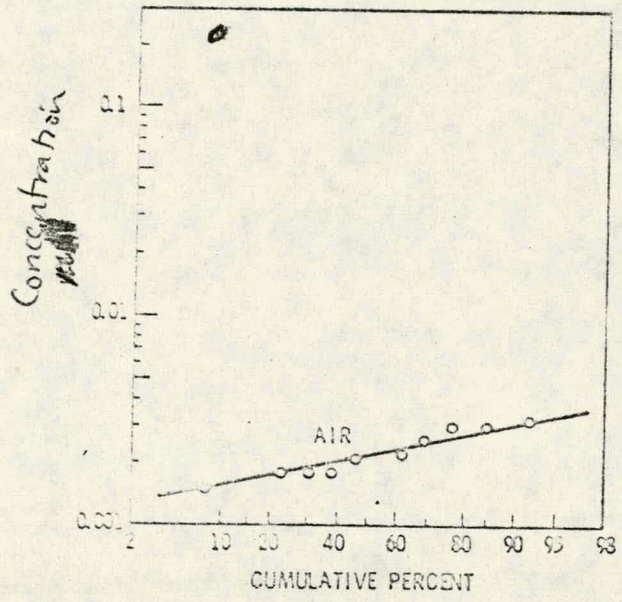


Figure 6

Lognormal Distribution of Environmental Contaminant Concentration
Plant Contribution