

Session 2

HEPA FILTER TEST METHODS

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CHAIRMAN: R.P. Pratt  
UK Atomic Energy Authority

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OPENING REMARKS OF SESSION CHAIRMAN:

Unfortunately, Mr. Jean Dupoux of France is unable to be with us due to illness and his paper will be read by Monsieur Rouyer. The papers at this session will cover both in situ testing and rig testing of filters using a wide variety of methods. I am sure that we are all going to know a bit more about the many methods that are available and, hopefully, a bit more about the pros and cons associated with each method.

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### IN SITU MEASUREMENT OF THE EFFICIENCY OF FILTRATION INSTALLATIONS IN THE NUCLEAR INDUSTRY BY THE SODA-FLUORÉSCÉIN (URANIN) AEROSOL METHOD - AFNOR STANDARD NFX 44.011

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#### Abstract

The in situ measurement of the efficiency of filtering installations in the French nuclear industry using the soda-fluorescein (uranin) aerosol method is presented.

A brief description and the interest of the method are discussed. Its use for the location of defects in filter elements and for in situ control of casing with tandem HEPA filter stages is described.

#### I. Introduction

The safety performance of a nuclear installation and the authorizations concerning gaseous waste discharge impose restrictions concerning discharges.

These imply that the purification systems must have the efficiencies defined in planned service conditions.

In order to obtain this result, it is very useful to control these systems at all stages of manufacture and assembly.

In France the controls are :

- establishment of a performance guarantee form for each type of filter element.
- conformity check of these elements.
- insitu testing of the filtering installations (1).

During all these controls, efficiency measurements are made. In France since many years only the method using a fluorescent soda-fluorescein aerosol has been used almost exclusively.

If the use of only one method has disadvantages, on the other hand, it is easier to compare the efficiencies of filter elements and filtering installations.

In practice the manufacturer guarantees that the decontamination factor for high efficiency filter elements at nominal air flow rate is greater than 5000 (penetration < 0,02 %).

For the majority of the filtering installations, decontamination factors between 1000 and 3000 are required by the safety performance for one stage of filtration.

These values are given considering the French AFNOR standard NFX 44.011.

Various reports (2), (3) have already described this method in detail, and its main features will be reviewed briefly here, so that its application may be discussed at length.

## II. Description of the method

### Principle

The test aerosol is produced by spraying of an aqueous soda-fluorescein solution with a rough formula  $C_{20}H_{10}O_5Na_2$ , also called uranin. After the elimination of large droplets by means of an inertia separator, the remaining liquid aerosol is evaporated by dilution with dry air. Aerosol samplings upstream and downstream of the test filter are collected on plane filters, from which the soda-fluorescein is then extracted by washing. The solutions obtained are finally titrated by measuring their fluorescence. The ratio of the amount of aerosol upstream of the test filter to the amount of aerosol downstream expresses the decontamination factor of the filter. Figure 1 shows a diagram of the test circuit, which may also be that of a filtering installation.

### Aerosol generation

Figure 2 illustrates an aerosol generator consisting of two sections : the spray system and the separator. The spray system consists of a shell (1) containing the soda-fluorescein solution, and a spray head (2) supplied with compressed air. The liquid aerosol produced enters the separator at (6).

The two-stage separator consists of shells (9), diaphragms (10) and nozzles (11). It is designed using the principle of the Hounam centripeter. The largest droplets are collected in settling vessels (12) or stopped by the filters (13). The fine aerosol liquid exits at (15). It is then dried almost instantaneously by simple mixing with dry air. The particle size distribution (Figure 3) corresponds to a standard logarithmic distribution with the following main characteristics :

- count median diameter                    0.08  $\mu\text{m}$
- mass median diameter                    0.15  $\mu\text{m}$
- geometric standard deviation 1.6

Generator operation

In normal service conditions, the spray system is filled with a 1% aqueous solution of soda-fluorescein, and the effective air supply pressure is 2 bars. The air flow rate is 30 liters per minute and the solution rate 40 cm<sup>3</sup> per hour. Evaporation of the droplets is achieved in less than half a second by dilution in a 1 : 6 ratio with air with less than 20% relative humidity (compressed air of industrial networks). Hence the total flow rate is 180 liters per minute, so that a 1 to 2 liter transit tube is adequate.

The amount of fine aerosol generated is about 30.10<sup>-3</sup> grams per hour. Self-contained operation between two solution fillings is five hours. Since the unit does not contain any moving parts, the only maintenance involved consists of periodic washing of the spray system and separator, and periodic replacement of the filter cartridges of the separator. Figure 4 presents a photograph of this portable generator, with the covers removed.

Aerosol sampling

Each aerosol sampling system includes a simple rectilinear probe with a filter holder, a compressed air ejector for air suction, and a gas meter for measuring the air volume sampled. The elbow probe with fine-edge nozzle for isokinetic sampling is not necessary, owing to the small size of the particles with negligible inertia in the test conditions. Fiberglass filter paper discs 4.7 cm in diameter are employed.

The upstream sampling rate generally ranges from 2 to 5 m<sup>3</sup> per hour TPN, corresponding to air velocities of 57 to 144 cm per second through the filters.

The downstream sampling rate is generally 12 m<sup>3</sup> TPN per hour.

Figure 5 shows upstream and downstream sampling devices with air discharge into the duct.

Processing of samples - Measurements

The soda-fluorescein collected on the sample filters is extracted by simple dipping in 100 cm<sup>3</sup> of ammonia-water upstream and 10 cm<sup>3</sup> of ammonia-water downstream. The solution obtained is titrated by means of a liquid fluorescence measurement instrument. This is a photometer whose light source is a xenon discharge lamp. The excitation and fluorescence wavelengths (490 nm and 520 nm respectively) of soda-fluorescein are selected by means of two optical interference filters.

The background of the washing solution for the type of sampling filter selected in terms of soda-fluorescein mass is about a few  $10^{-10}$  gram without centrifugation and less than  $10^{-10}$  gram with centrifugation. So the minimum weight of soda-fluorescein detectable is between a few  $10^{-10}$  gram and  $10^{-9}$  gram.

The instrument is calibrated with solutions obtained by successive dilutions of a mother liquor containing 10 grams per liter. The majority of fluorimeter equipped with 150 watt xenon lamp are suitable for this sensitive measurement (like TURNER Model 430 instrument).

Figure 6 shows equipment used for measurements : washing vessel automatic washing solution pourer, centrifugal instrument, analysis bath, fluorimeter.

### III. Interest of the method

For installations equipped with only one stage of filters as for the French electronuclear program, the method has been very well suited for in situ testing.

#### Main features of the aerosol

##### Fluorescent aerosol

The use of a fluorescent aerosol avoids any accidental increase of background on the level of the downstream sampling caused by the presence of dust in the filtering installation at this level.

An typical example is a downstream sampling located downstream of the fans through which undesired air of the room may enter.

It is indeed the only place to obtain representative sampling for compact filtering installations (PWR reactors) because the distance between the filters and the fans is very short.

The specific detection of this aerosol is of particular interest :

- for acceptance in situ of new installations situated in plants still dusty ;

- to know if the defects depend on the filters and their assembly or are side leakages like holes in the ducts, defects of the leaktightness of the access doors.

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### Size of the aerosol

Since the mass median diameter is equal to 0,15  $\mu\text{m}$ , the in situ efficiency measurement is made in the zone of maximum penetration of the filters.

The result includes also the total value of the leakages.

Since isokinetic sampling isn't necessary, identical straight probes can be used for filters testing or adsorbers testing as in the case of filtering installations of reactors (4). Simultaneous measurement of air flow rate is made using the ratio of the mass aerosol rate ( $\pm 10\%$ ) to the upstream concentration.

It is easy to compare the in situ efficiency with the efficiency of the filter elements since the same method is used in situ and for conformity checks (figure 6).

### Aerosol concentration

For example the aerosol concentration is equal to  $3 \times 10^{-6}$  gram per cubic meter for a 10.000 cubic meter per hour air flow rate circuit. The variation is inversely proportional to the air flow rate because of the constance of the mass aerosol rate generated.

On the other hand this concentration is generally too low to permit instantaneous location of defects with the ordinary use of a photometer.

### Other characteristics of the aerosol

The aerosol is nonradioactive, nontoxic and noncorrosive. It is slightly electrically charged (0,5 negative charge per particle) and is assumed to be solid for moisture air under 80% RH.

### Main characteristics of the method

#### Sensitivity

The method is sensitive because a penetrating aerosol ; a fluorescent titrating technique and a delayed measurement of the samplings are used.

Taking the case of the filtering installation of the auxiliary building 1200 MW PWR French power plants having an air flow rate of 200.000 cubic meters per hour (117 CFM).

The decontamination factor must be equal to or greater than 2000.

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The downstream sampling with an air flow rate of 12 cubic meter per hour must last 40 minutes to obtain a ratio of signal to background of 5.

The real in situ efficiency is not reduced by the sampling downstream of the filters, of the soda-fluorescein aerosol emitted in the room where the circuit is situated because the leakages of the generator are very low (leakages rate  $< 10^{-8}$  gram per hour).

### Selectivity

The classification in efficiency of high efficiency HEPA filters is correct (5).

### Reproductibility

A good reproductibility is obtained because of a rigorously constant particle size aerosol distribution. Indeed the generator which does not have any moving parts or electro-mechanical system is only supplied by air pressure regulated by a two-stage regulator. Moreover the particle size distribution of each generator is directly measured with a flame spectrophotometer during the calibration and periodically.

### Ease of use

It is obtained by the use :

- of a rugged and easily portable equipment
- of compressed air
- of distinct sampling systems upstream and downstream so there is no need to fear any pollution of the downstream sampling instrument by the aerosol which is present in a higher concentration upstream;
- of stainless steel generator and sampling holders which are easy decontaminated.

Generating and sampling may also be performed in the ducts under pressure and over pressure.

After they have been connected to the ducts, the generator and the sampling instruments don't need any survey by the personnel during the test.

Measurement with the fluorimeter

As a rule it presents the disadvantage of a delay because it is necessary to wash the sampling filters in ammonia-water and to measure water fluorescence. The practice shows that this disadvantage is minor.

Indeed since the fluorimeter is used on the test site, the efficiency of the installation can be determined in situ within 15 to 30 minutes after sampling is completed.

This measurement time can generally be neglected in comparison to the testing time. The testing time which is always more important than the sampling time depends as a matter of fact on local conditions such as conformity adjustment of the circuit, passages in a restricted zone, health physics inspection, etc.

IV. Location of defects in filter elements

The technique used to locate defects in filter elements depends on the design of the filtration installation and the possibility of access to the filter.

For filtering installations of reactors a typical detailed inspection is sufficient for the majority of cases to locate the defects.

In case of small filtering installation with a few number of filter elements, the replacement of these elements and the checking of the tightening are generally sufficient to remove the defects.

In the case of important filtering installation one may locate the filter casing or filter wall using efficiency measurement after sequential isolation of casings and walls. This means that a downstream representative sampling is made in practice. It is not always obtained.

In other cases a method like DOP aerosol method using higher aerosol concentration and a instant reading detector may be used.

We also point out a special use of the soda-fluorescein aerosol method for location of defects in walls of filter cells situated in PWR reactors.

These walls are housed in concrete bunkers closed by field doors. Hence access is possible to the downstream sides of these filter walls.

The technique consists of :

- isolating the filtering wall from the main air flow by closing the shutoff dampers of the bunker.
- generating aerosol into the prefiltration bunker using an outside fan with an air flow rate between a few hundred cubic meters by hour and 1000 cubic meters by hour.
- successively sampling air dowstreams each cell using a suction cap equipped with a probe connected to a TDA model 2D light diffusion photometer. In these condition the aerosol concentration is high enough to be measured using a photometer.

This technique is less easy to use compared to the DOP aerosol method for example.

On the other hand, each filtering cell is only submitted to a small mass of aerosol ( $5 \cdot 10^{-3}$  gram) and the workers are not exposed to the air flow rate.

#### V. In situ control of filtering installations with tandem HEPA filter stages

The sensitivity of the soda-fluorescein aerosol method is not sufficient to rapidly measure, the total decontamination factor of filtering installations equipped with tandem HEPA filter stages mounted in casings and generally situated in hot nuclear laboratories.

The new method using an intra cavity laser will permit this measurement (6).

So the measurement of the total decontamination factor corresponding to all stages is replaced by the measurement of the decontaminating factor of each filter stage.

The technique consists (Figure 8) of :

- isolating the casing from the ventilation by closing the shutoff dampers.
- connecting with air tight techniques upstream and downstream each stage of filters an external mobile loop equipped with a fan, tubes, an aerosol generator and sampling probes.

Generally the air flow rate used is the nominal air flow rate of the casing divided by 5.

The efficiency measurement is successively done on each stage. The casings may be equipped with special connectors as it is practiced in new French nuclear filtering installations.

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By using this technique we assure the production of a constant aerosol concentration upstream and downstream of each stage of filter since one can choose the form and the length of the connecting tubes.

It's necessary that equal air flow rate through each filter element is assured. This is obtained by using a special mixing baffle fixed inside each upstream connector of the casing.

So we assure that for the air flow rate used the decontamination factor of each stage is correct according to the aerosol size spectrum generated.

The real value of the decontamination factor of each stage at nominal air flow rate and of the total tandem stages must be calculated using laboratory results.

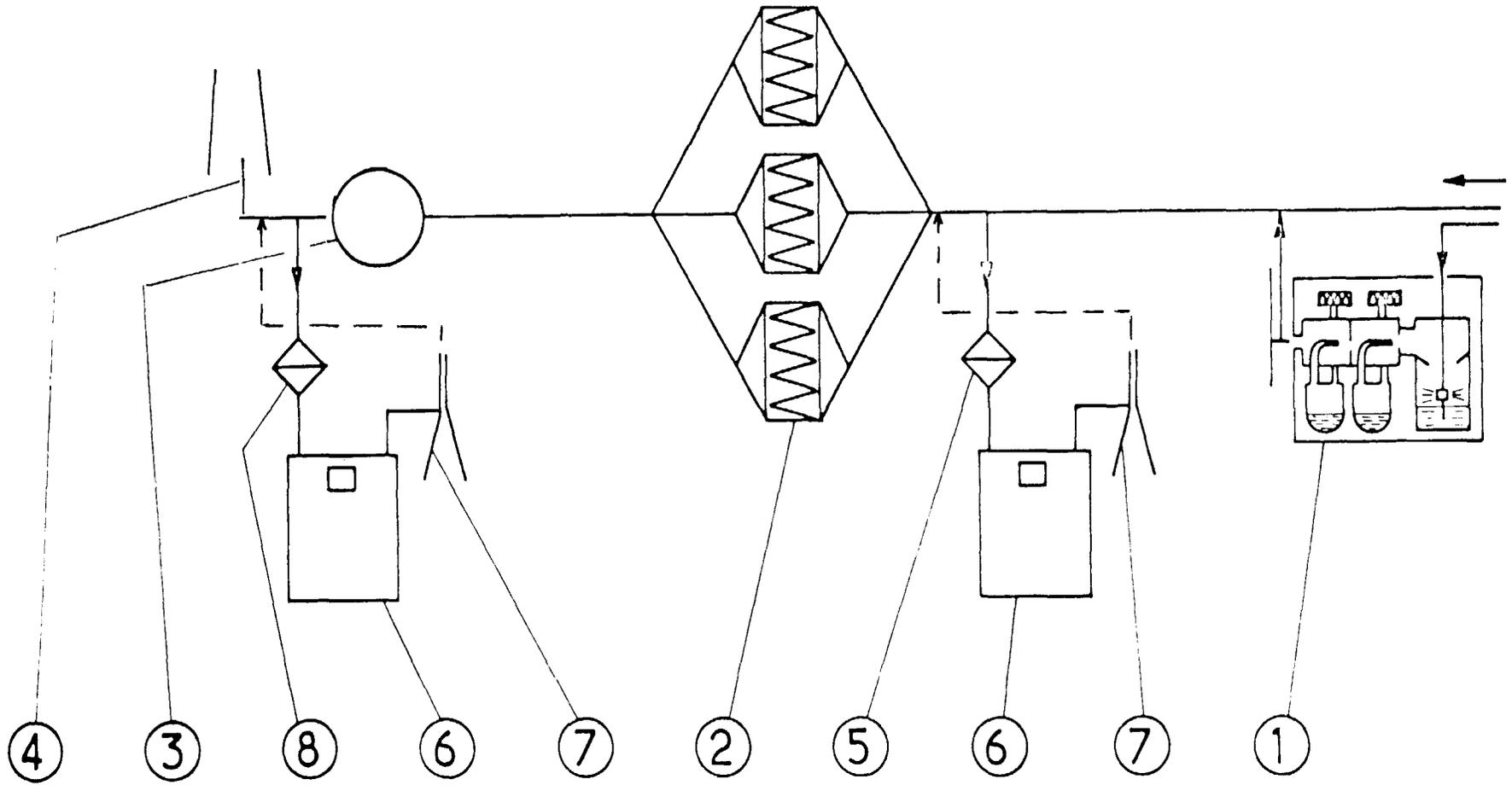
This technique would reduce, even for one filter stage casing, the constraint of ducts form and length. So installations would be more compact and less expansive.

Indeed French experiments using helium gas as a tracer show that a 10 length equivalent diameter of duct, sufficient to obtain a correct velocity distribution with turbulent air flow, does not generally allow the operating of a constant aerosol concentration when the generation of the tracer is made at a single point (7).

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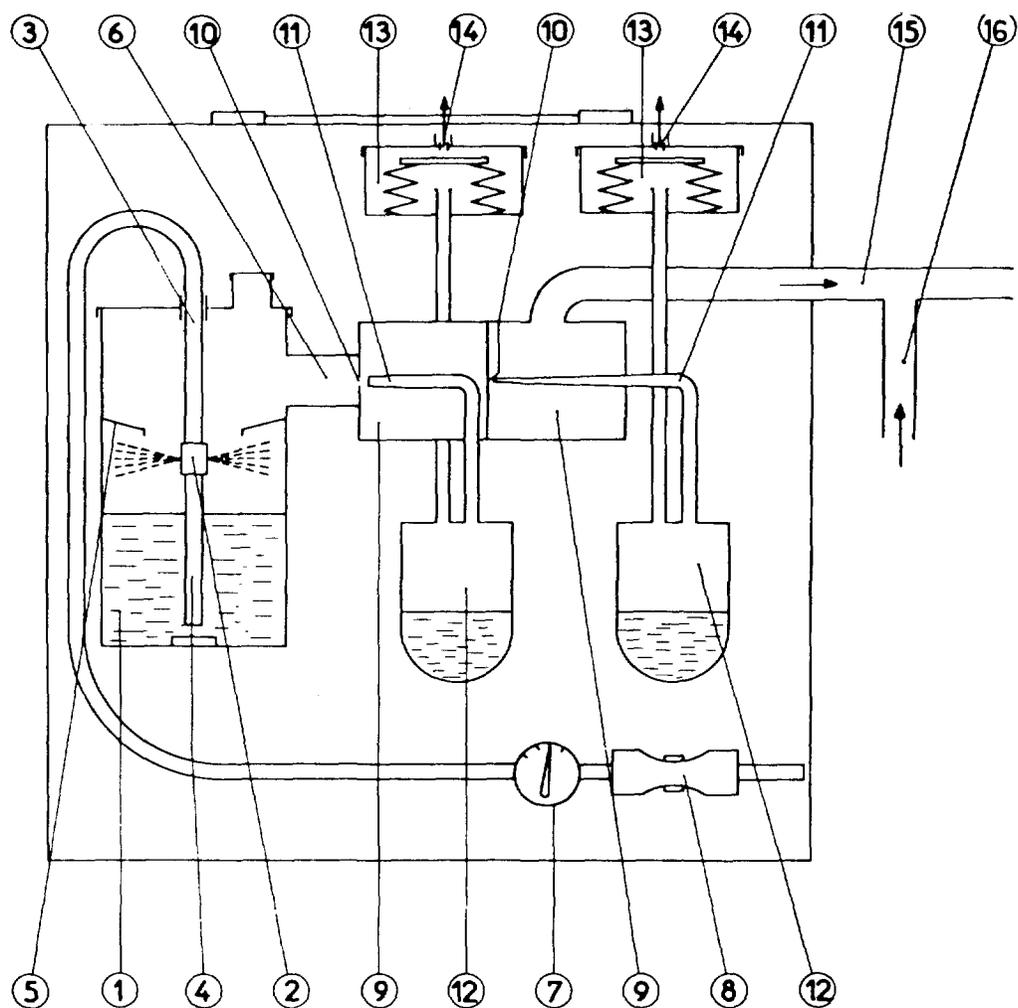
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Figure 1 Schematic diagram showing efficiency measurement of a filtration installation

- |                            |                               |
|----------------------------|-------------------------------|
| 1. generator               | 5. upstream sampling filter   |
| 2. filtration installation | 6. volume meter               |
| 3. blower                  | 7. air ejector                |
| 4. discharge               | 8. downstream sampling filter |



a. spray system

b. separator

- |    |                         |     |                           |
|----|-------------------------|-----|---------------------------|
| 1. | body                    | 9.  | body                      |
| 2. | spray nozzle            | 10. | inlet diaphragm           |
| 3. | sliding air intake tube | 11. | separation nozzle         |
| 4. | solution suction tube   | 12. | settling vessel           |
| 5. | drop arrestor           | 13. | filter                    |
| 6. | liquid aerosol outlet   | 14. | outlet diaphragm with tip |
| 7. | pressure gauge          | 15. | fine aerosol outlet       |
| 8. | needle valve            | 16. | dry dilution air          |

Figure 2 Diagram of soda-fluorescein (uranin) aerosol generator

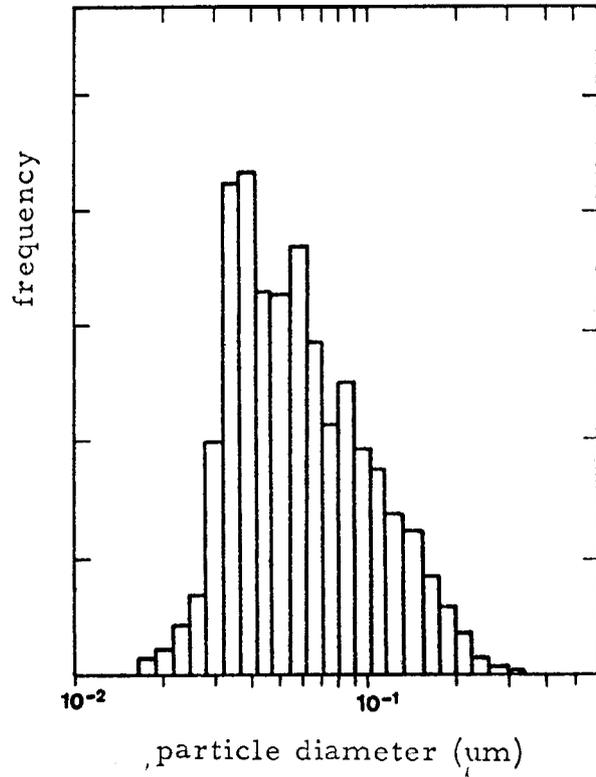


Figure 3

Size distribution of soda-fluorescein aerosol

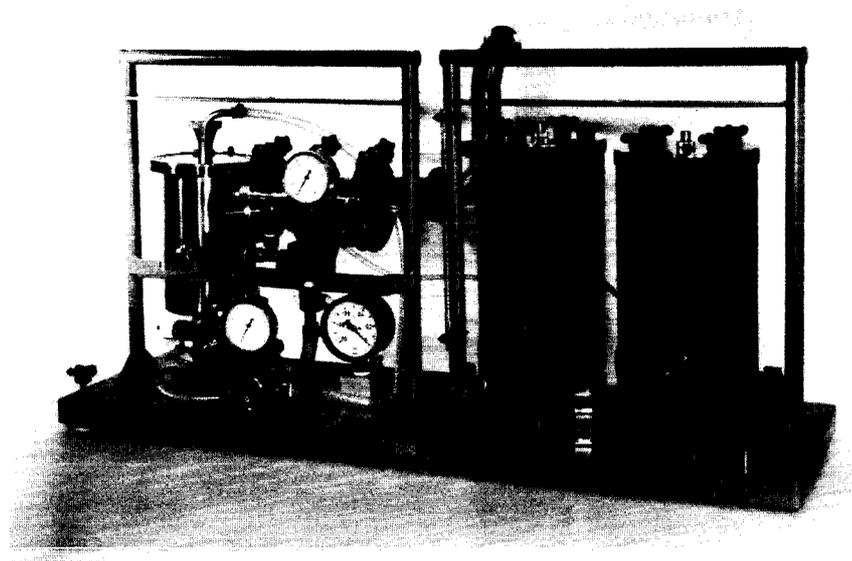


Figure 4

New model of soda-fluorescein aerosol generator

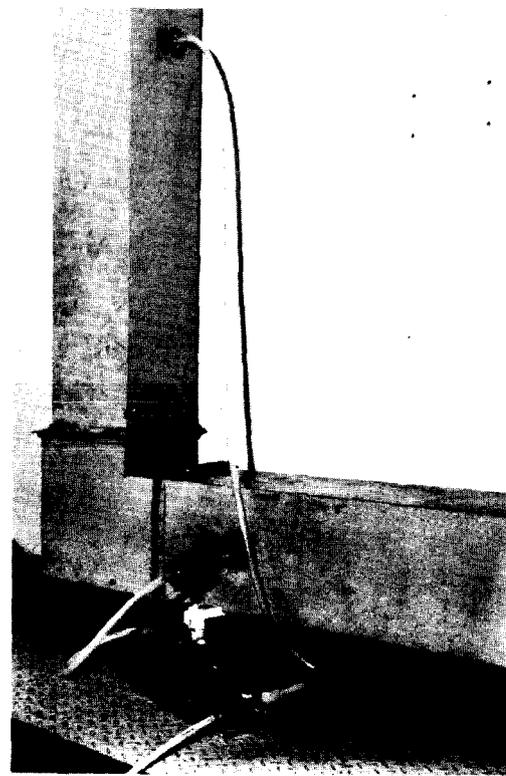
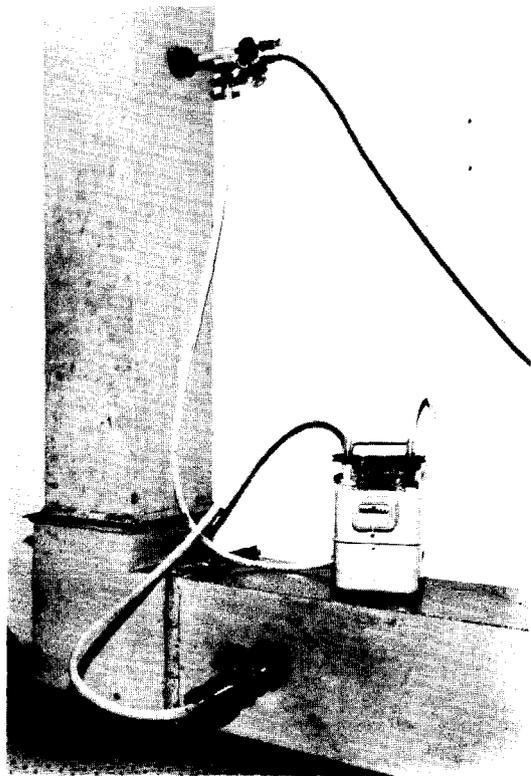


Figure 5

Upstream sampling of aerosol with air discharge through the air nozzle

Downstream sampling of aerosol with air discharge through the air nozzle

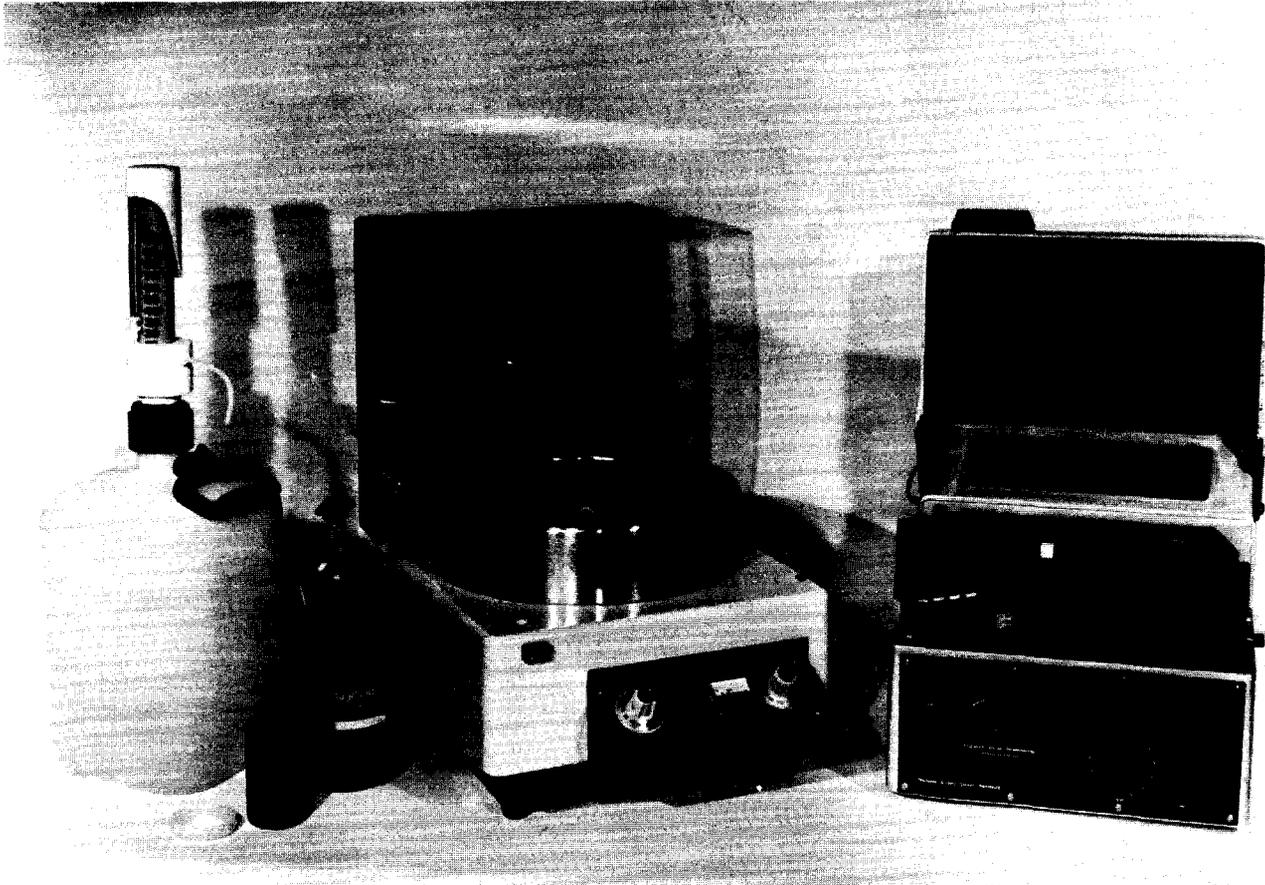


Figure 6 - Instruments used for the processing of samples and the measurements.

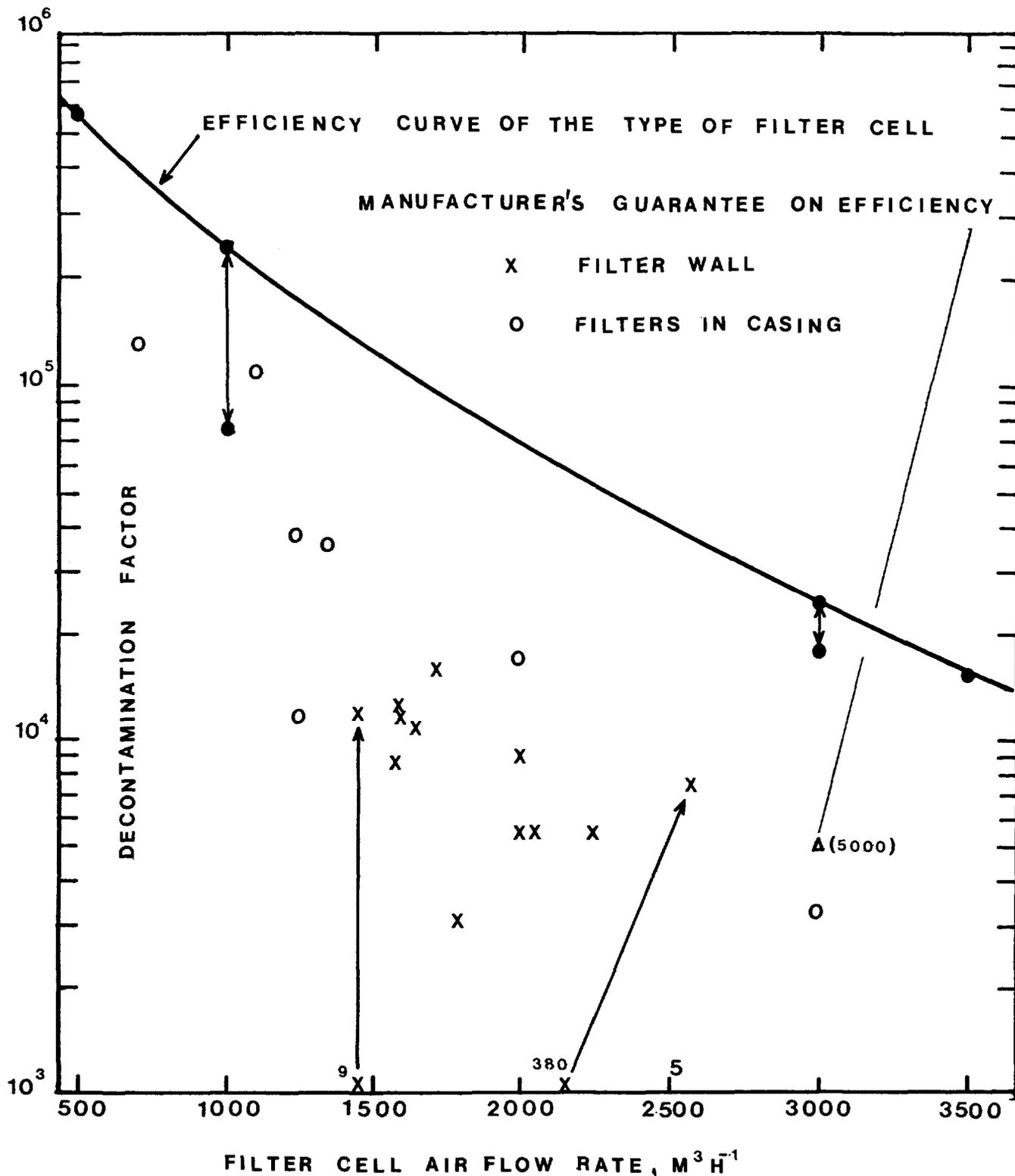


Figure 7 - Results of in situ measurement of the efficiency of filtration installations in a 2 x 900 MW PWR power plant.

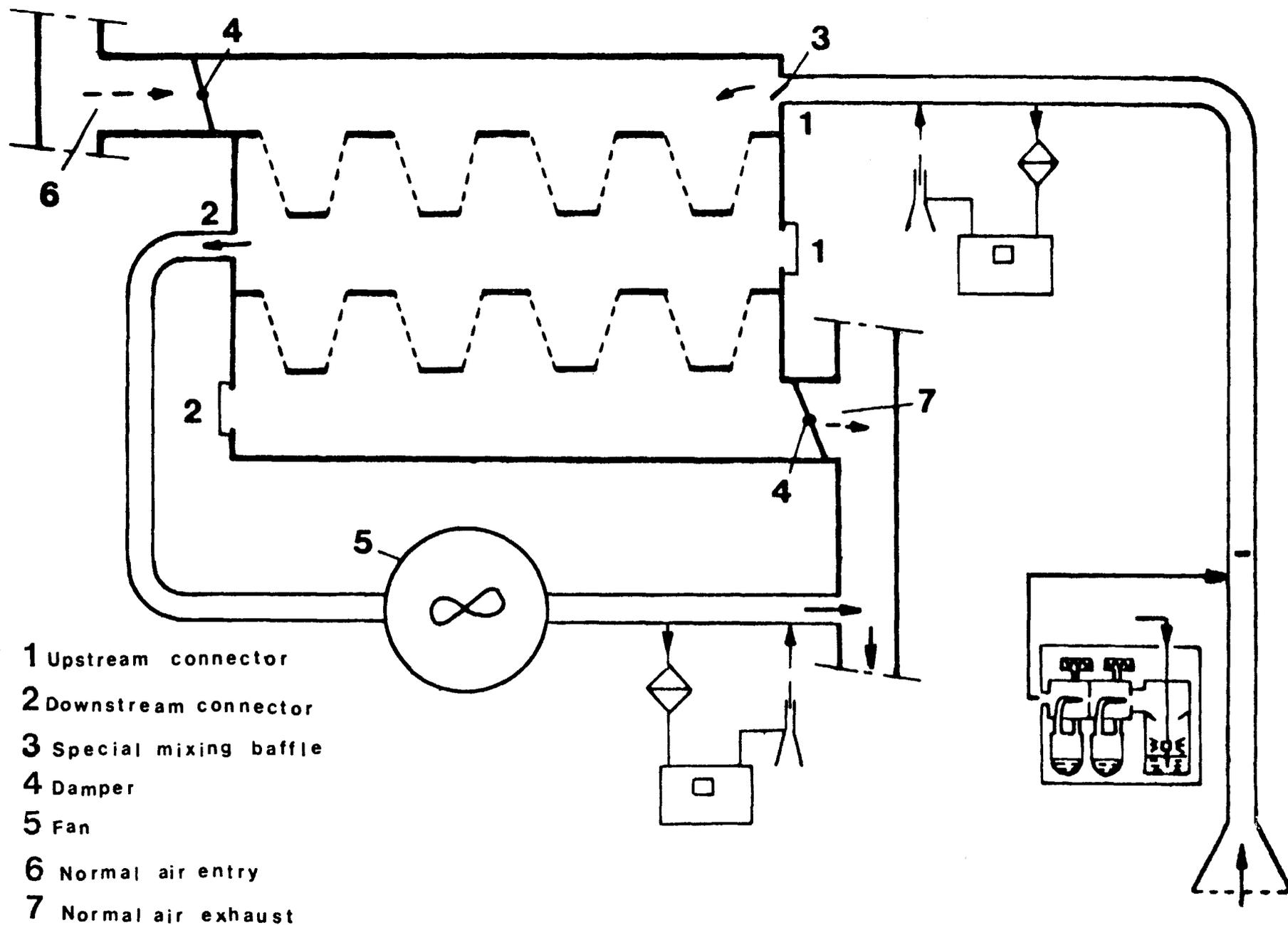


Figure 8 - In situ control of a casing equipped with 2 HEPA filter stages.

DISCUSSION

DORMAN: Have you any figures to compare the penetration due to uranine with atomized sodium chloride?

ROUYER: You can find comparative results for filter papers in the presentation of J. Dupoux and A. Briand: "Laboratory and plant measurements of the efficiency of filter elements and filter papers using the soda fluorescein uranine aerosol method - AFNOR-NFX 44011" at the Seminar of the Absolute Filtration of Aerosols in Nuclear Industry (Aix en Provence - 22-25 Nov. 1976) edited by the European Communities. I know that, more recently, Mr. Dupoux has obtained comparative experimental results for filtering cells manufactured in Great Britain.

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## A COMPARATIVE STUDY OF IN-SITU FILTER TEST METHODS\*

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### Abstract

Available methods of testing high efficiency particulate aerosol (HEPA) filters in-situ have been reviewed. In order to understand the relationship between the results produced by different methods a selection has been compared. Various pieces of equipment for generating and detecting aerosols have been tested and their suitability assessed.

Condensation-nuclei, DOP (di-octyl phthalate) and sodium-flame in-situ filter test methods have been studied, using the '5000 cfm' (9000 m<sup>3</sup>/h) filter test rig at Harwell and in the field. Both the sodium-flame and DOP methods measure the penetration through leaks and filter material. However the measured penetration through filtered leaks depends on the aerosol size distribution and the detection method. Condensation-nuclei test methods can only be used to measure unfiltered leaks since condensation nuclei have a very low penetration through filtered leaks.

A combination of methods would enable filtered and unfiltered leaks to be measured. A condensation-nucleus counter using n-butyl alcohol as the working fluid has the advantage of being able to detect any particle up to 1 µm in diameter, including DOP, and so could be used for this purpose.

A single-particle counter has not been satisfactory because of interference from particles leaking into systems under extract, particularly downstream of filters, and because the concentration of the input aerosol has to be severely limited.

The sodium-flame method requires a skilled operator and may cause safety and corrosion problems.

The DOP method using a total light scattering detector has so far been the most satisfactory. It is fairly easy to use, measures reasonably low values of penetration and gives rapid results. DOP has had no adverse effect on HEPA filters over a long series of tests.

### 1. Introduction

The aim of this work is to compare in-situ filter-test methods throughout the U.K. Many methods are currently in use and although each has its particular merits it is desirable to introduce some uniformity in the types of methods used and to understand the relationship between the results produced by the various methods.

There is a need (i) to ensure that the performance of a given filter installation is within that specified for the plant and (ii) to determine when filters require changing. The first requirement is met by in-situ testing of filters when first installed and after each subsequent filter change or major plant modification. For this purpose the test method should preferably give rapid results. The second requirement ideally involves continuous testing but in practice the frequency of testing is determined by the likelihood and the consequences of

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\* Work performed under contract from British Nuclear Fuels Ltd., Windscale, U.K.

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filter failure.

After a brief review of available methods this paper describes and evaluates selected test methods and instruments in the laboratory, in limited field trials and using the new '5000 cfm' filter test rig at AERE, Harwell.

### 2. Review of Test Methods

When a filter system is routinely challenged with radioactive aerosols then it may be possible to measure the real system efficiency with this aerosol. However, in general, particularly with filters installed to prevent significant releases under accident conditions and filters in standby systems, a test aerosol is required. Ideally this aerosol would have the same characteristics for filtration as the radioactive aerosol to be removed by the filters. Since these characteristics are generally not known it is usual to choose a test aerosol of particle size close to the maximum-penetrating particle size for the filter i.e. in the range 0.1 to 0.5  $\mu\text{m}$ . This will give a lower limit to the protection factor for the system.

For a polydisperse aerosol the particle size distribution downstream of the filter may be different from that upstream due to partial filtration, in which case different methods of detection will give different values for the protection factor. Since the radioactivity present will generally be proportional to the mass of an aerosol, then a mass measurement is preferred. However, since small particles are relatively more hazardous to the population, a measurement based on number concentration may be more relevant.

Any in-situ filter test system must be capable of measuring protection factors  $>10,000$  (for a single stage of filtration) and should be reasonably portable and easy to use. The test aerosol should of course be stable and reproducible and have no adverse effects on the filters. Methods of aerosol generation and detection considered are given in Table I with appropriate cross references.

Various methods are unsuitable for in-situ testing at large flow rates because the equipment is too unwieldy (e.g. monodisperse hot DOP, Colliston atomisers, Dautrebande atomisers). Paraffin oil was considered to be similar to DOP. Methods requiring sampling and subsequent analysis (e.g. sodium-24, sodium fluoresceinate) are restrictive since results are not available to allow immediate checking and retesting. There is also the possibility of cross-contamination of the samples, particularly from downstream of the filter system. Radioactive methods require appropriate handling and shielding. Availability of equipment also restricted the methods used.

The test aerosols chosen for comparison were condensation nuclei, DOP (di-octyl phthalate), produced by pneumatic and thermo-pneumatic generators, and sodium chloride, produced by the Porton flame generator. These are detected respectively by Pollak counters, total light scattering devices and sodium-flame photometers. An individual particle counter has also been tested.

### 3. Condensation-Nuclei Test Methods

#### 3.1 Detection of Condensation Nuclei

Most detectors of condensation nuclei are based on the design of L.W. Pollak (see ref.1) in which air is humidified and then expanded in a ceramic-lined tube. This produces supersaturation and subsequent condensation on nuclei with diameters

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in the range 0.0001–0.1  $\mu\text{m}$ . Concentrations from about 260,000 particles per  $\text{cm}^3$  down to about 3 per  $\text{cm}^3$  are determined from the reduction in light transmission. Several versions differing only in their auxiliary components (pumps, valves, display, etc.) are in use of which four were chosen for study; a single-tube laboratory model, a semi-automatic single-tube commercial instrument (Polkinghorne Industries Ltd,<sup>(2)</sup> Workington, U.K.), a twin-tube automatic model developed by AEE, Winfrith, U.K. and an Environment One Counter (Environment One Corporation, New York). In preliminary work results with the Environment One were not reproducible and our suspicions<sup>(3)</sup> that the humidity was not being maintained were confirmed by Cooper and Langer<sup>(3)</sup>. This instrument was not used further.

One instrument (Condensation Nucleus Counter, TSI Inc., St. Paul, Minnesota, Model 3020) uses n-butyl alcohol and the diffusion cloud-chamber principle to produce condensation on nuclei and uses light scattering for detection. At low concentrations individual particles are detected while higher concentrations are determined from the total light scattered. The measurable range is from  $10^{-2}$ – $10^7$  particles/ $\text{cm}^3$ .

### 3.2 Sources of Condensation Nuclei

There is normally a fairly high concentration of condensation nuclei present in any environment and this has been used for filter testing. However the inlet concentration can vary wildly over short time periods leading to inconsistent results. Sources of nuclei (e.g. in-leaks in negative-pressure systems or fans) between the upstream and downstream sampling points, particularly downstream of the filter, will lead to erroneously high penetration values. Also when testing high-efficiency installations the inlet concentration must be high enough to give sufficient dynamic range for the detector. The ambient aerosol is not therefore recommended as a source of condensation nuclei for filter testing.

A common method of generating condensation nuclei is to burn a 3% solution of ammonium sulphide in methylated spirits. One methylated spirits burner is sufficient to produce the highest measurable concentration for a Pollak counter in a flow rate of 8500  $\text{m}^3/\text{h}$ . The particles have a count median diameter of 0.02  $\mu\text{m}$  with a geometric standard deviation (GSD) of 2.2<sup>(4)</sup>. Alternative methods are to burn tributyl phosphate in methylated spirits or to use propane gas burners.

## 4. DOP Test Methods

DOP has been chosen as a suitable aerosol because it is relatively easy to produce in large quantities of an appropriate size, it has a low vapour pressure, it is stable and it has a low toxicity. It is used extensively in the USA<sup>(5)</sup>.

### 4.1 Detection of DOP

DOP aerosol is detected by light-scattering devices. These are not specific to DOP but will be affected by any particles greater than about 0.1  $\mu\text{m}$  in diameter. Three instruments have been tested, the TDA-2C (Air Techniques Inc., Baltimore) and its successor the TDA-2DN, which are total light-scattering detectors, and a single-particle light-scattering detector (Royco Instruments Inc., Menlo Park, California, Model 225). The TSI condensation nucleus counter can also be used for DOP measurements.

### 4.2 Generation of DOP

DOP aerosol for in-situ filter testing is normally produced by a pneumatic or thermo-pneumatic generator. An example of each has been used in this study. The

aerosol outputs and size distributions have been determined in the laboratory and details are given in Table II. The pneumatic DOP generator (Air Techniques Incorporated, Model TDA-4A) uses compressed air at a gauge pressure of about 140 kPa, which is passed through from 1 to 8 Laskin nozzles immersed in a tank of DOP<sup>(6,7)</sup>. The thermo-pneumatic DOP generator is made by C.F. Taylor (Metal Workers) Ltd., Wokingham, U.K. DOP is entrained in a stream of carbon dioxide, vapourised by a heater and then condensed. A small cylinder within the equipment contains sufficient carbon dioxide for 40 min at a gauge pressure of 280 kPa.

### 5. Sodium-Flame Test Methods

This method is more specific than the other methods tested in that it detects only sodium, which is generated as a sodium chloride aerosol. It was developed at CDE, Porton<sup>(8)</sup>. The name (sodium flame) is derived from the method of detection (viz. a flame photometer).

#### 5.1 Detection of Sodium Chloride Aerosol

A portable flame photometer is used for in-situ filter testing<sup>(9)</sup>. A sample of air containing salt particles is drawn by a fan into a hydrogen flame and the sodium light emission is measured. There is a continuous analogue output so that concentration variations across a duct can be measured easily. Sodium chloride concentrations in the range  $0.1 \mu\text{g}/\text{m}^3$  to  $13 \text{ mg}/\text{m}^3$  can be measured giving a minimum detectable filter penetration of 0.001%.

#### 5.2 Generation of Sodium Chloride Aerosol

The method used in the British Standard Filter Test Rig (using Colliston atomisers to spray up a solution of NaCl which is then dried) is not suitable for in-situ filter testing since it is not portable and has insufficient output. The generator used<sup>(10)</sup> consists of an oxy-propane gas torch with an annular nozzle through which a salt stick (12.5 mm diameter) is fed into the flame at a controlled rate. The aerosol has a mass median diameter of  $0.3 \mu\text{m}$  and is produced at a rate of 0.2 to 5.0 g/min. Higher rates may be obtained.

### 6. The '5000 cfm' Filter-Test Rig

#### 6.1 Description

The rig was designed as a scaled-up version of the 1000 cfm ( $1700 \text{ m}^3/\text{h}$ ) British Standard Sodium Flame Filter Test Rig to provide filter-test facilities at flow rates closer to those commonly found in building extract systems. A scale diagram of the rig is shown in Fig.1.

The rig consists of PVC ducts with a diameter of 610 mm leading to and from a filter housing. The housing holds  $4 \times 1700 \text{ m}^3/\text{h}$  HEPA filters in a conventional frame. The design allows other filter mountings to be fitted if required. The rig can be operated under extract or under pressure using the appropriate fan. A baffle at the downstream end of the system controls the airflow. Air entering the system can be prefiltered.

The test-aerosol injection point is positioned about 20 duct diameters upstream of the filters under test. Sampling points are provided about 6 duct diameters upstream and 20 duct diameters downstream. At each position there are two sampling points at right angles so that concentration or velocity profiles can be obtained across two perpendicular duct diameters. A Stairmand disc about 15 duct diameters upstream of the filters ensures good mixing of the test aerosol while

another disc about 10 duct diameters downstream mixes the penetrating aerosol.

## 6.2 Velocity and Concentration Profiles at Sampling Positions

These were measured across two perpendicular duct diameters at both sampling positions.

Velocity profiles are shown in Fig. 2. The average velocity was about 8.6 m/s corresponding to a flow rate of 9000 m<sup>3</sup>/h. The velocity tended to increase across the duct due to bends which direct the air flow outwards. The maximum deviation from the average was about 20%.

Concentration profiles were measured using the DOP aerosol from a TDA-4A generator with a TDA-2DN light scattering photometer. At the upstream sampling point the variation was 6% without and 2% with the Stairmand disc. Downstream the aerosol was injected at the side of the duct to simulate an edge leak in a filter which is probably the most severe test of mixing. At the sampling position the concentration variation was 20% without and only 4% with the mixing disc. The Stairmand discs therefore provide useful mixing, particularly downstream of the filter, and were used during all the experimental work. If aerosols of large particles (>5 μm diameter) were used in the rig, larger variations in concentrations would be expected due to bends. However all aerosols used up to the present have had mass median diameters well below 1 μm.

## 6.3 Experimental Arrangement

A bypass was installed in the filter housing consisting of a 20 mm diameter pipe and valve. The pipe penetrated 300 mm into the duct upstream and downstream of the filter mounting. Air flows of up to 6.8 m<sup>3</sup>/h (4 cfm) were obtained corresponding to leak rates of up to 0.08%. The rig was normally operated under extract at 9000 m<sup>3</sup>/h (5300 cfm).

Before mounting on the rig, filters were individually tested on the British Standard Filter Test Rig to obtain matched sets of four. Two sets of matched filters have been used, HEPA filters each with about 0.009% penetration on the standard rig and a set of lower efficiency filters with a similar construction to HEPA filters but with a 2% penetration. The Royco measurements used a further set of filters, not tested on the standard rig, but giving a penetration of 0.04% measured with DOP (TDA-4A generator and TDA-2DN detector).

## 7. Experiments on Filter Test Rig

### 7.1 Comparison of Test Methods with HEPA Filters

Comparisons were made at various times depending on the availability of equipment and results are given in Table III. In general, good agreement was obtained and the penetrations provided by the bypass leaks were adequately measured by all methods.

For the Pollak counters, at least three readings have to be taken for each condition to ensure that the readings have stabilised and to give an average value. This is because downstream concentrations have uncertainties associated with the precision of measurement of light transmission and the subtraction of the background concentration. Typical standard deviations on given concentrations have been obtained (Table IV) and penetration values derived from them for an upstream concentration of 200,000 particles/cm<sup>3</sup> and negligible background. Lack of precision at penetrations <0.01% could be a significant factor in the discrepancies between

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condensation-nuclei tests and sodium-flame tests found by Davis and Clifton<sup>(2)</sup> and reported by Fraser<sup>(1)</sup>.

With both the Polkinghorne and the automatic Pollak counter it was often found that reliable measurements of the background concentration downstream of the filters could only be made after measurements had been made with the injected aerosol. Initial measurements of the background concentration downstream are often higher than with the injected aerosol. It may be that the tube requires a very long time to settle down. No hysteresis effect could be demonstrated using the Polkinghorne counter (low/high/low concentration measurements).

The DOP test results (with both aerosols) indicated an increase in penetration during the tests (0.005 to 0.008% for pneumatic DOP and 0.008 to 0.012% for thermo-pneumatic DOP). This increase was not shown by the condensation-nuclei tests but at this level of penetration the errors associated with the Pollak method are large. The increase was not due to a decrease in filter efficiency as a repeat test on the British Standard Test Rig showed no change. It was probably due to increased leakage round the filters which had been loosened for some tests.

The DOP and sodium-flame tests each took about 5 min while the condensation-nuclei tests took about 20 min with the automatic counter and about 30 min with the single-tube versions since several readings were required.

### 7.2 Comparison of Test Methods with Low Efficiency Filters

Filters with an average penetration of 2% were tested using condensation-nuclei and DOP methods (Table V). The opportunity was taken to try out a TSI Condensation Nucleus Counter which was being demonstrated.

The condensation-nuclei methods showed very low penetrations compared with DOP methods, but yet still measured the by-pass leak. Results with the DOP methods varied considerably depending on the method but were all similar to results obtained for the filters on the Standard Sodium Flame Filter Test Rig. The bypass leak was not detectable at the measured penetrations.

This indicates that penetration through low-efficiency filters (and presumably through filtered leaks in high-efficiency filters) is highly dependent on the particle size. Condensation nuclei are stopped very efficiently while DOP from the Taylor generator is more penetrating than from the TDA-4A as measured by a light scattering device. The TSI Condensation Nucleus Counter responds to the number of particles and not to the light scattered and therefore the difference between the two types of detectors is not surprising since the size distribution of the aerosol will have changed in passing through the filter. When used with condensation nuclei, the TSI counter gave similar results to the Pollak counters as expected.

### 7.3 Royco Counter Measurements

The high sensitivity of the Royco Counter allows much lower concentrations of input aerosol to be used than for other methods. Indeed, because of problems of dead time at high count rates, the upstream concentration must be limited to less than 600,000 counts/min. Both the pneumatic and thermo-pneumatic DOP generators produce over 90% of particles with diameters  $<0.3 \mu\text{m}$ . These are not individually detected but in large quantities give a measurable signal due to pile-up resulting in a non-linear response with concentration. This further limits the usable upstream concentration. Thus very low concentrations of test aerosol are obtained downstream. This makes the method very sensitive to in-leakage of particles and to

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variations in the upstream concentration of 'natural' aerosol. Measurements were taken with the rig under extract (with and without inlet filters) and under positive pressure (Table VI).

With no inlet filter, the DOP concentration could not be made much greater than the natural concentration due to count-rate problems. With an inlet filter, but still under extract, the background downstream was unstable and for large particles could be much larger than that upstream. This led to negative values of the DOP concentration downstream in some cases. This instability was probably due to in-leaks of environmental particles since much more stable results were obtained with the rig under positive pressure. Tapping the ducting downstream of the filter could also cause significant bursts of particles due to resuspension. Reducing the sampling rate of the counter from 0.17 to 0.017 m<sup>3</sup>/h (0.1 to 0.01 cfm), with a corresponding increase in the input concentration, would reduce these effects. However it was found that at the 0.01 cfm setting the sampling rate was drastically affected by the pressure in the duct and so this was not used.

The results obtained with the rig under positive pressure are comparable with the DOP test measurements on these filters (0.04%, see Section 6.3) The effect of background is negligible. However statistics on the outlet concentration are poor and could only be improved by increasing inlet concentration (with consequent danger of pile-up and dead-time problems) or by considerably increasing the sampling time. An alternative would be to desensitise the Royco counter by a factor ~100 but it would then have a similar sensitivity to some total light scattering detectors and the advantage of low input concentrations would be lost.

### 8. Other Experiments

#### 8.1 Field and Rig Comparisons

During routine field tests using the DOP method at AERE, Harwell and independently using the condensation nucleus method at AEE, Winfrith a batch of HEPA filters was detected which was below specification (penetration >0.05%). The opportunity was taken at AERE to test some of these filters on the British Standard and the '5000 cfm' test rigs (Table VII). Some filters from another manufacturer were also measured. The limited results show good agreement in general.

#### 8.2 Field Tests

DOP and condensation-nuclei test methods were compared for several sets of filters in an operating plant. A Taylor generator and TDA-2DN detector were used for the DOP tests while Polkinghorne counters with natural aerosol and aerosol produced by ammonium sulphide in methylated spirits or by a petrol driven 4-stroke motor were used for the condensation-nuclei tests. Extracts from the results (Table VIII) highlight a number of points.

In one case (Filter 1), using injected aerosols, DOP showed the higher penetration. From the test-rig experiments this would suggest a low-efficiency filter since DOP is more penetrating than condensation nuclei.

Condensation nuclei testing, using the natural aerosol only, gave higher values of penetration than the DOP method in another case (Filter 2), whereas when injected aerosol was used similar results were obtained. The natural aerosol probably gave a high value due to in-leaks downstream of the filter.

The efficiency of mixing of the test aerosol before the upstream sampling

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point should be checked, particularly where two or more ducts lead to the same set of filters and the test aerosol is only injected into one duct. Injection points should be sufficiently far upstream and downstream sampling points should be sufficiently far from the filters to ensure reasonable mixing. However 'dead' volumes between the injection point and the filters (e.g. injecting the aerosol into a cell) should be minimised so that equilibrium in the system can be reached in a reasonable time.

### 8.3 Effect of DOP Loading

Initial and final measurements on the British Standard rig showed that there was no significant change in sodium chloride penetration due to the 36 DOP tests over a period of 3 months. Each HEPA filter would then be loaded with about 60 g of DOP. This loading had no effect on the pressure drop across the filters. In the laboratory a glove box filter has been loaded with 5 g of DOP (equivalent to 600 g on a 1700 m<sup>3</sup>/h filter) with no detectable effect after storage for three months.

Experiments in the U.S.A. on charcoal filters<sup>(12)</sup> have shown no effects at concentrations several orders of magnitude greater than those normally expected.

### 9. Comments on Performance and Use

The results indicate no significant differences for HEPA filters in good condition between the condensation-nuclei, DOP and portable sodium-flame test methods both in the experiments on the '5000 cfm' rig and in the limited field trials. Results also agree with penetrations measured on the British Standard Sodium Flame Filter Test Rig.

However results with low-efficiency filters show that condensation-nuclei test methods do not detect filtered leaks although they detect direct leaks (e.g. gasket leaks). This has also been observed by Powell and Wilkinson<sup>(13)</sup>. The penetration through filtered leaks also depends on the particle size distribution and on the detection method and may vary by a factor of over 2. This effect was also discussed by Dorman<sup>(14)</sup> and depends on the variation in filter efficiency with particle size. Differences between methods is probably not important for comparative measurements.

Any of the methods are therefore suitable, in principle, for leak testing of filters in-situ so that a choice depends on such factors as the minimum measurable penetration, ease of operation, speed of operation, portability and suitability for a given plant.

Condensation-nuclei test methods are unsuitable for in-situ testing of filter efficiency. However a combination of a condensation-nuclei method with another method would enable the effects of leaks and changes in filter efficiency to be separated. A detector such as the TSI Condensation Nucleus Counter could be used for both methods but further tests are required.

### 9.1 Condensation Nuclei

Generation of condensation nuclei is a simple matter. Two methods have been used in this work (burning ammonium sulphide or tributyl phosphate in methylated spirits) but other methods are probably as suitable (e.g. burning propane gas).

Unless the background concentration is negligible its effect must be removed by taking measurements with and without the generated aerosol. The generated aerosol must have a concentration considerably higher than that of the ambient

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aerosol to minimise the effect of fluctuations in background and to improve sensitivity. Adventitious sources of condensation nuclei downstream of the filters (e.g. from in-leaks or fans) must also be minimised.

The dynamic range of Pollak counters is about 50,000:1 which results in a minimum measurable penetration of 0.002%. However poor statistics preclude reliable measurements below 0.01%.

To obtain reliable results with Pollak counters several samples must be taken upstream and downstream. Even with a recycling counter, such as the Winfrith design it takes about 10 min to obtain 3 or 4 samples from each position. To measure background and aerosol requires 15-20 min. The time is doubled for single-tube counters. It is therefore not possible to determine concentration variations across a duct in a reasonable time.

The counters are generally very heavy (>50 kg for the Winfrith and Polkinghorne models) and are not easily transported to in-situ test locations. The ceramic lining of the tube needs wetting every 100-200 measurements after which the optical alignment has to be checked on all models. An experienced operator is generally required.

There is no simple way of checking the calibration of a Pollak counter. There seems to be no standard other than visual counting of droplets<sup>(2)</sup>. The laboratory model used in this work gave consistently high values of concentration compared with other laboratory models of identical construction and with the Polkinghorne counter. The difference was not uniform over the scale but amounted to a difference in light transmission of 3-5% at mid scale corresponding to a concentration difference of 30%. A similar difference was obtained for filter-penetration measurements.

A condensation nucleus counter such as the TSI Model 3020 could have three main advantages over other types; a) virtually instantaneous readings are produced giving rapid results and enabling variations in concentration across ducts to be determined; b) the dynamic range is increased using the single-particle mode to 10<sup>5</sup>:1 although in practice this would be limited by the background effect; and c) the total light-scattering calibration can be checked against the single-particle mode where their ranges overlap. It could also be used with other sub-micron aerosols for filter-efficiency determinations.

An improved version of the Environment One counter with an effective humidifier and suitable for sampling from ducts under extract would also provide rapid results.

### 9.2 DOP

Both of the DOP generators tested are adequate for flows about 10,000 m<sup>3</sup>/h and can be used up to 100,000 m<sup>3</sup>/h (see Table IX). The thermo-pneumatic generator is more convenient to use in practice since, unlike the pneumatic type, it does not require a large supply of compressed air. It can also be used for up to 20 min after disconnection from a mains electrical supply. Both generators are reasonably portable.

The detectors are also robust and fairly portable and can measure penetrations down to 0.001% under reasonable conditions. Their response is fast so that checks of the uniformity of aerosol concentration can rapidly be made. The position of leaks in or around filters can also be determined by scanning immediately downstream of the filters.

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The sensitivity depends on the aerosol. For a fixed gain setting (5 turns) on the TDA-2DN the concentration to give 100% reading<sup>3</sup> was 330 mg/m<sup>3</sup> for DOP from the TDA-4A generator (MMD 0.5  $\mu\text{m}$ , GSD 2.8) and 130 mg/m<sup>3</sup> for the Taylor generator (MMD 0.3  $\mu\text{m}$ , GSD 1.5).

At the DOP concentrations used the natural levels of aerosol in systems give insignificant readings. High loadings of DOP have had no measurable effect on the characteristics of HEPA filters. Other work has shown that testing with DOP does not affect charcoal filters<sup>(12)</sup>.

DOP also has the advantage that it is used in American Standard tests<sup>(5)</sup>.

### 9.3 Single-Particle Counters

The results obtained with the Royco counter are probably typical of other single-particle counters. They are designed to detect low concentrations but cannot distinguish between extraneous particles and the test aerosol. They are therefore unsuitable for in-situ testing of plant filters where the system is under extract and subject to in-leakage. This problem has also occurred with multiple filter testing<sup>(15)</sup>. Single-particle counters which only detect specific aerosols (e.g. fluorescent particles) could overcome this problem<sup>(16)</sup>.

Where there is little extraneous aerosol the minimum measurable penetration is limited by the maximum measurable concentration upstream and the sampling time downstream. Particles of diameter  $<0.3 \mu\text{m}$ , although not recorded, cause pile-up and dead-time effects at high concentrations. This equipment could be used with relatively coarse aerosols (e.g. 1-5  $\mu\text{m}$ ) to reduce such problems. Where in-leakage is not a problem it might be useful for long-term sampling.

### 9.4 Portable Sodium Flame

Generation of the aerosol uses a salt stick in an oxy-propane flame which could present some safety problems. The sodium chloride aerosol could also cause corrosion. Current output is sufficient for a 17,000 m<sup>3</sup>/h flow rate and higher outputs are possible.

The minimum penetration detectable is  $\sim 0.001\%$ . The analogue output provides a rapid measurement of penetration and measurements of variations in concentration can be made easily. The detector requires a cylinder of hydrogen which may also pose safety problems.

Although the generator and detector are not large, when gas cylinders are included the equipment is cumbersome to transport. The operator requires to be highly trained.

## 10. Conclusions

Condensation-nuclei test methods can be used to measure the leakage of filter systems in-situ. They are not suitable for measuring changes in filter efficiency. The methods tested have been slow and the equipment unwieldy. Counters based on the diffusion cloud chamber principle, such as the TSI Condensation Nucleus Counter, could be useful but further tests are required.

DOP and sodium-flame test methods measure the total penetration through leaks and filter material. However the measured penetration through filtered leaks will depend on the aerosol size distribution and the detection method. These methods have the advantage of speed over the condensation-nuclei methods tested and the

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ability to measure lower levels of penetration.

The sodium-flame method requires a skilled operator and poses safety problems due to the use of an oxy-propane flame and cylinders of oxygen, propane and hydrogen. Sodium chloride may also cause corrosion.

Single-particle counters are not satisfactory due to interference from in-leakage in systems under extract and due to severe limitations on input concentration.

The DOP method has so far been the most satisfactory. It is fairly easy to use, measures reasonably low values of penetration and gives rapid results. DOP has had no effect on HEPA filters over a long series of tests nor does it affect charcoal filters.

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Table I Possible Aerosol Generation and Detection Methods

No.	Generation Method	Detection Method
G1	DOP, pneumatic	D1,2
G2	" , thermo-pneumatic	D1,2
G3	" , monodisperse,hot	D1,2
G4	Condensation nuclei, natural	D3,4
G5	" " , generated	D3,4
G6	Sodium chloride, Colliston atomiser	D1,5,6
G7	" " , Dautrebande atomiser	D1,5,6
G8	" " , Porton flame	D1,5
G9	Sodium fluoresceinate (Uranine)	D7
G10	Paraffin oil (German DIN 24184)	D1,2

	Detection Method	Generation Method
D1	Individual particle light scattering detector	G1-3,6-10
D2	Total light scattering detector	G1-3,10
D3	Condensation nuclei counters, Pollak type	G4,5
D4	" " " , diffusion type	G4,5
D5	Sodium-flame photometer	G6-8
D6	Sodium-24, filter paper sampling and counting	G6,7
D7	Filter paper sampling and fluorimeter	G9

Table II Properties of DOP Aerosols

Generator	Gauge Pressure kPa	Aerosol Output (g/min)	Mass Median Diameter(a) (µm)	GSD	Count Median Diameter (µm)
TDA-4A(b)	69	-	0.84	2.9	0.03
"	138(c)	0.34(d)	0.64	2.9	0.02
"	207(e)	0.46	-	-	-
Taylor	138(f)	1.6 (g)	-	-	-
"	276	2.8	0.28	1.7	0.12

Notes

(a) The mass distribution was obtained using an Andersen sampler<sup>(17)</sup>. The count median diameter was calculated.

(b) The supplied glass-fibre outlet filter was removed. This did not significantly change the particle size distribution. With the filter in place the output for 1 jet at 138 kPa(gauge) was reduced to 0.29 g/min.

(c) Air consumption 70 l/min at 138 kPa(gauge).

(d) Output per jet, The output increased in proportion to the number of jets. Lieberman and Haslop<sup>(6)</sup> have reported an output of 2.25 g/min for 6 jets (cf 1.8 g/min)

(e) Increasing the pressure over that recommended (138 kPa(gauge)) does not significantly increase the aerosol output.

(f) At 138 kPa(gauge) the generator tends to splutter.

(g) The output may be slightly underestimated due to difficulty in collecting all the aerosol.

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Table III In-situ Tests of HEPA Filters on the 5000 cfm Test Rig (a)

Expt. No.	Filter Bypass (m <sup>3</sup> /h)	Penetration (%)				
		DOP Generator TDA-4A	DOP Generator Taylor	Condensation NH <sub>4</sub> S	Nuclei(b) TBP	NaCl Generator
1	None	0.004 (c)	0.008	0.007	0.006	
		-0.006	-0.008	-0.008		
	6.8(d)	0.10	0.11	0.09	0.095	
		-0.11	-0.13			
	Large(e)	0.4	0.4	0.29		
2	None	0.007	0.011	0.007	0.006	0.006
		-0.008				
	3.4	0.058	0.055	0.048		0.058
	6.8	0.11	0.11	0.087	0.014	0.09
3	None	0.008	0.012	0.0075(P)		
				-0.018		
	3.4	0.055	0.06	0.04 (P)		
	6.8	0.11	0.12	0.07		
				-0.13 (P)		
Filters removed for re-test on British Standard Rig						
4	None	0.008	0.015	0.008 (P)		
				0.065 (L)		
	3.4	0.05		0.047 (P)		
	6.8			0.085 (P)		
				0.11 (L)		
Mean (f)	None	0.007	0.010	0.009	0.010	0.006
		(0.002)	(0.002)	(0.004)	(0.004)	
	3.4	0.057	0.058	0.053		0.058
				(0.015)		
	6.8	0.10	0.12	0.10	0.13	0.09
	(0.007)	(0.008)	(0.035)	(0.05)		

**Notes**

(a) Four new filters were used. Each gave a penetration of 0.009% on the British Standard Rig before installation. After 36 DOP tests (60 g loading per filter) the individual penetrations on the standard rig were 0.009, 0.010, 0.007 and 0.012%.

The rig flow rate was 9000 m<sup>3</sup>/h and the pressure drop across the filters was 320 Pa (1.3" water gauge) throughout the experiments.

(b) Each result for the condensation nucleus counters is the mean of several readings (see Table IV). The Winfrith condensation nucleus counter was used except where indicated by a (P) (Polkinghorne) or (L) (Laboratory model).

(c) Range of values where more than one measurement was made.

(d) The 6.8 m<sup>3</sup>/h by-pass is equivalent to a penetration of 0.07% the 3.4 m<sup>3</sup>/h by-pass to 0.035%.

(e) Filter mounts loosened.

(f) Mean (standard deviation) for expts. 1-3 only.

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Table IV Measurement Uncertainties for Condensation Nucleus Counters  
(Pollack type)

(a) Typical uncertainties on concentration measurements

Mean light transmission(%)		Concentration(particles/cm <sup>3</sup> )	
Range	Std. Devn.	Range	Std. Devn.
96-100	0.2	0-119	6
92-96	0.5	119-292	15-25
40-50	0.8	11,000-20,000	500-900
6.5-15	1.0	124,000-250,000	10,000-30,000

(b) Typical uncertainties on penetration measurements.

Downstream concentration (particles/cm <sup>3</sup> )	10	20	200
Std. devn. on concentration	9	9	26
Penetration (%)	0.005	0.01	0.1
Relative std. devn. on penetration (%)	90	46	16

Notes

- (a) Upstream concentration 200,000 particles/cm<sup>3</sup> with negligible background.
- (b) Downstream background concentration in the range 0-100 particles/cm<sup>3</sup>.

Table V In-situ Tests of Low Efficiency Filters on the 5000 cfm Test Rig (a)

Expt. No.	Filter Bypass (m <sup>3</sup> /h)	Penetration (%)		
		DOP Generator TDA-4A	Taylor	Condensation Nuclei (b)
1	None	1.6	3.4-3.7	0.03-0.06(P) 0.01-0.06(L)
	3.4	1.6	-	0.15(P) 0.10(L)
2	None	1.8	4.5	0.06(P)
	3.4			0.10(P)
3	None	2.0		
		4.1(T)		0.03(T)
	3.4	4.4(T)		0.07(T) 0.10(P)

Notes

(a) Four nominally 95% filters were chosen which had individual penetration of 2.4, 2.1, 1.9 and 1.9% on the British Standard Rig.

The rig flow rate was 9000 m<sup>3</sup>/h and the pressure drop across the filters was 125 Pa (0.5" water gauge).

(b) Ammonium sulphide.

(P),(L),(T) are respectively the Polkinghorne, laboratory model and TSI condensation nucleus counters. For the Polkinghorne and laboratory counters each result is the mean of several readings (see Table IV).

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Table VI Royco Counter Measurements (a)

Measurement	Particle-Size Range (um)				
	0.3-0.5	0.5-0.7	0.7-1.6	1.6-3.0	>3.0
<b>NO INPUT FILTER ON RIG</b>					
Background					
Inlet	30610	9230	3240	1094	68
Outlet	85	49	63	11	0
DOP+Background					
Inlet	52268	29741	26249	6547	92
Outlet	118	56	42	79	17
Net DOP					
Inlet	21658	20511	23009	5453	24
Outlet	33	7	-21	68	17
Penetration (%)	0.15	0.03	-	1.2	71
	Overall Penetration 0.17%				
<b>INPUT FILTER ON RIG</b>					
Background					
Inlet	2800	1421	1092	249	2
Outlet	256	129	151	62	37
DOP+Background					
Inlet	82493	45913	35127	9568	35
Outlet	424	160	100	32	8
Net DOP					
Inlet	79693	44492	34035	9319	33
Outlet	168	31	-51	-30	-29
Penetration (%)	0.21	0.07	-	-	-
<b>RIG UNDER POSITIVE PRESSURE</b>					
Background					
Inlet	60	45	22	3	0
Outlet	0	0	0	0	0
DOP+Background					
Inlet	25335	26698	29196	7627	111
Outlet	3	6	3	1	0
Penetration (%) (b)	0.01	0.02	0.01	0.01	0.0
	Overall Penetration 0.015%				

Notes

(a) Counts in 1 min at sampling rate of 0.17 m<sup>3</sup>/h (0.1 cfm).  
 The filters had a penetration of 0.04% to 'cold' DOP measured with a TDA-2DN detector.

(b) Background negligible, not subtracted.

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Table VII Field and Test-Rig Comparisons of Selected Filters

Filter	Test	Rig	Penetration (%)	Comments
1. Man. 1 (HEPA)	DOP(hot)	Field test	0.13-0.15	Filter in safe change mount Above filter after removal
	NaCl	BS (AERE) (a)	0.15	
2. Man. 1 (HEPA)	NaCl	BS (AERE)	0.031	Replacement for above After installation
	DOP(hot)	Field test	0.01	
3. Man. 1 (HEPA) (Set of 4)	DOP(hot)	Field test	0.3	After removal Filters mounted together
			0.3 max 1.2	
			0.2 max 0.5	
			0.2 max 0.4	
	NaCl	BS (AERE)	0.4-0.55	
	DOP(cold)	5000 cfm rig	0.35	
	DOP(hot)	5000 cfm rig	0.4	
4. Man. 2	NaCl	BS (Man. 2)	0.008	
	NaCl	BS (AERE)	0.012	
	DOP(cold)	BS (AERE)	0.01	
5. Man. 2	NaCl	BS (Man. 2)	0.062	Disagreement possibly due to travel. Man. 2 and AERE BS rigs normally agree
	NaCl	BS (AERE)	0.12	
	DOP(cold)	BS (AERE)	0.14	

Notes

(a) Man. 1 and Man. 2 refer to two manufacturers.

(b) BS (AERE) and BS (Man. 2) refer to the British Standard Sodium Flame Filter Test Rigs at AERE and at the premises of manufacturer 2 respectively.

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Table VIII Results of Field Tests

Filter	Flow rate (m <sup>3</sup> /h)	Test (a)	Concentration		Penetration	Comments
			Upstream	Downstream		
1	3400	DOP	100%	0.10%	0.1%	
		C.N.	36,800/cm <sup>3</sup>	<<16/cm <sup>3</sup>	<<0.04%	Natural aerosol
			200,000/cm <sup>3</sup> (20,000)(b)	22/cm <sup>3</sup> (10)	0.011% (0.005)	Injected aerosol
2	42,500	DOP	20%	0.15%	0.8%	See note (c)
		C.N.	17,900/cm <sup>3</sup> (800)	737/cm <sup>3</sup> (190)	4.1% (1.0)	Natural aerosol
			61,500/cm <sup>3</sup> (9,500)	1340/cm <sup>3</sup> (160)	1.4% (0.7)	Injected aerosol
			DOP	20%	0.04%	0.2%

Notes

- (a) DOP - Taylor generator and TDA-2DN detector.  
C.N. - Condensation nuclei and Polkinghorne counters.
- (b) Standard deviation in parentheses.
- (c) The test point was only a few duct diameters downstream for this and the C.N. tests.
- (d) The aerosol was injected into a large cell leading to the duct so that the aerosol concentration built up slowly with a half time calculated as 14 min. The result quoted is the mean of several sets of upstream and downstream readings during which time the concentration varied by a factor of two.
- (e) The downstream test point was moved about 15m further downstream thus ensuring good mixing.

Table IX Minimum penetration limits using TDA-2DN detector (gain 6 turns)

Generator	Aerosol Output (g/min)	Filter Flow (m <sup>3</sup> /h)		
		1700	17,000	85,000
		Minimum Penetration(%)		
TDA-2C, 3 jets	1.0	0.002	0.02	0.1
TDA-2C, 6 jets	2.0	0.001	0.01	0.05
TDA-2C, 8 jets	2.7	0.001	0.007	0.04
Taylor ('Hot')	2.8	0.001	0.004	0.02

Notes

- (a) The upstream reading can not always be set to 100% and lower values are then used.
- (b) A further factor of 5 may be obtained by increasing the gain but at the expense of instrument stability.
- (c) The TDA-2C is about 10 times less sensitive than above limits.
- (d) Extra sensitivity may be obtained by increasing the aerosol input rate e.g. by using extra generators.

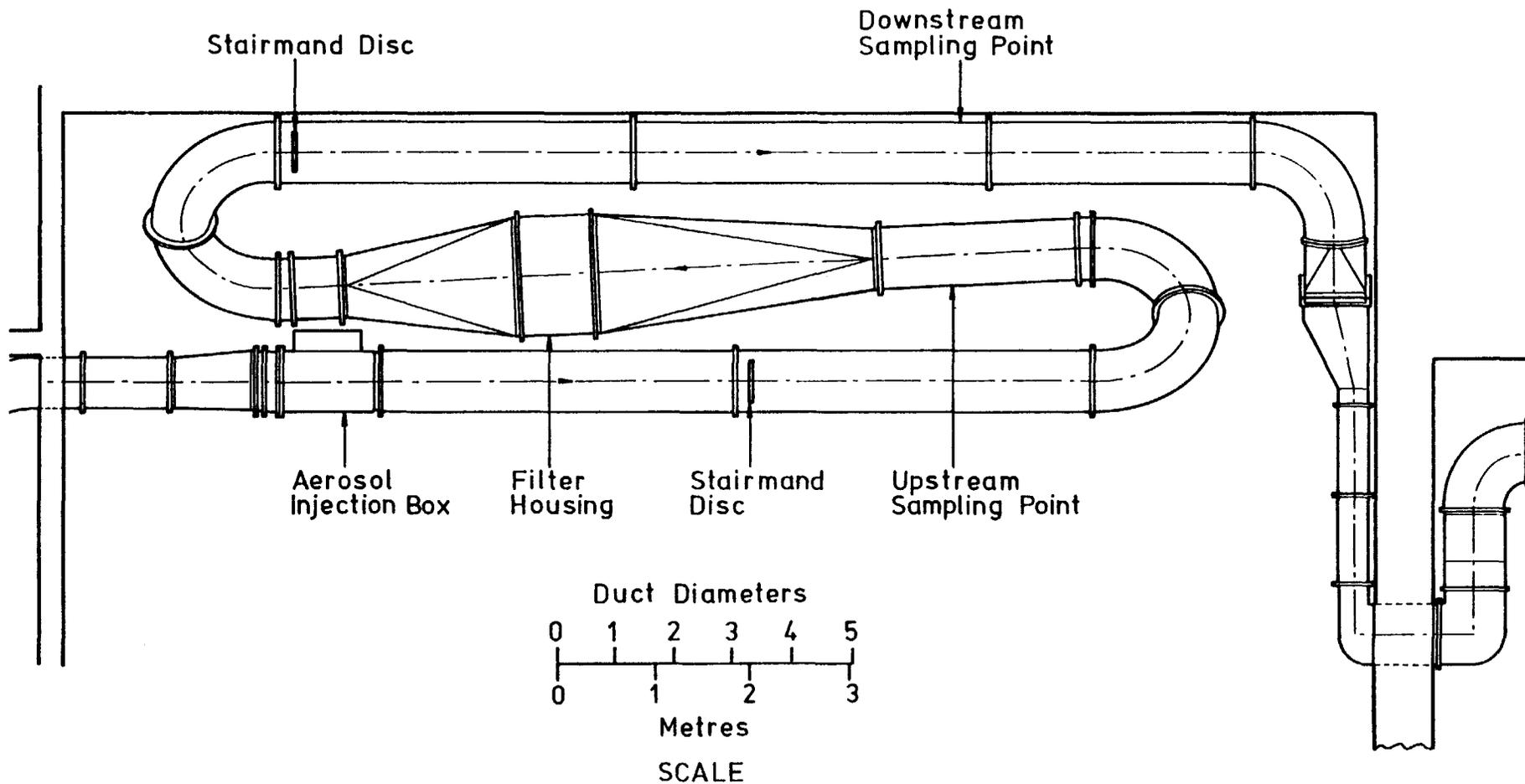


FIG.1. DIAGRAM OF '5000 CFM' FILTER TEST RIG

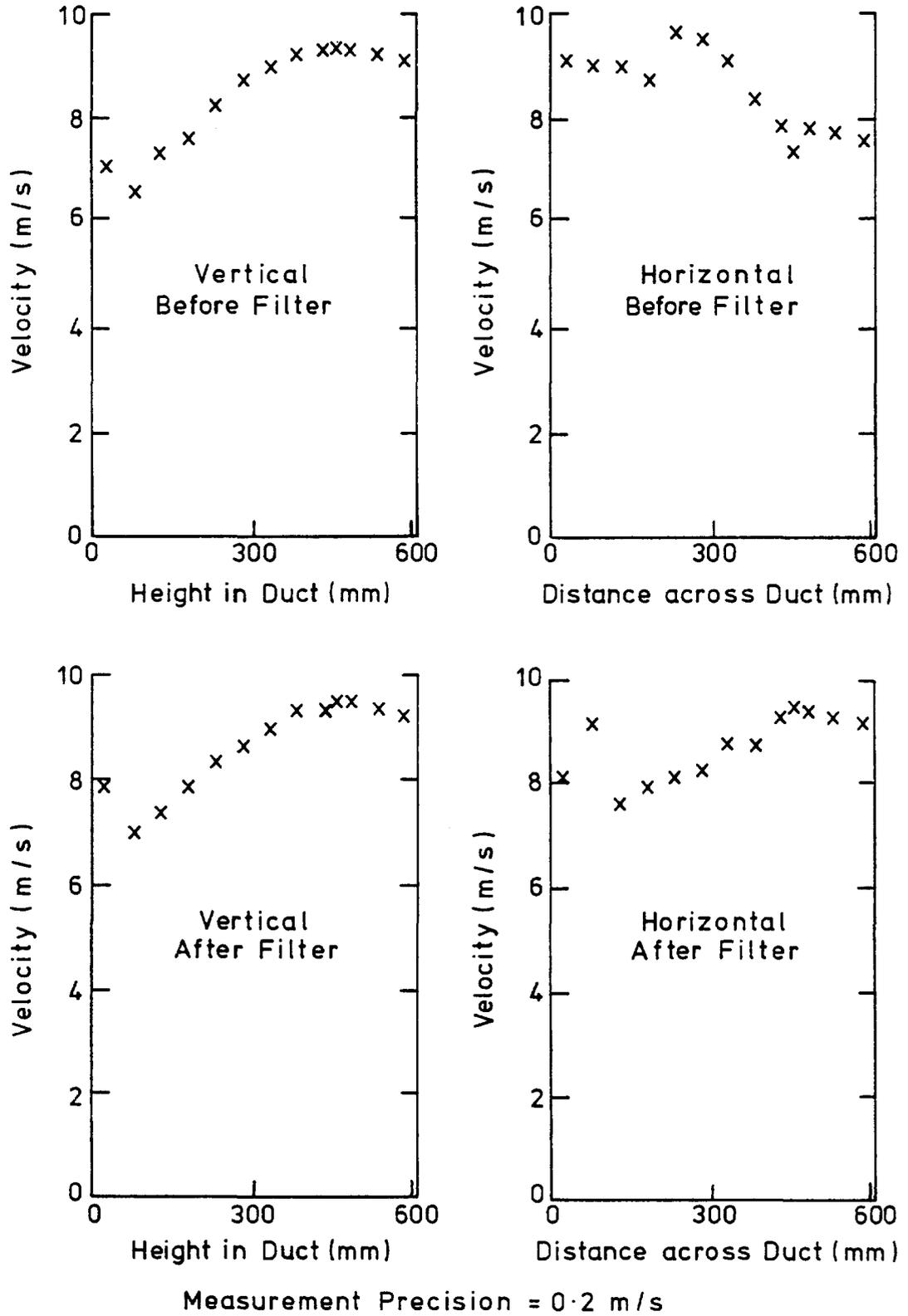


FIG.2. VELOCITY PROFILES AT SAMPLING POSITIONS

DISCUSSION

STEINBERG: What was the concentration you were operating at? You mentioned penetration, but you did not mention the challenge DOP concentration when you were comparing sodium chloride, DOP, and condensation nuclei count.

MARSHALL: About 100 micrograms per liter.

STEINBERG: Was that measured by a gravimetric method?

MARSHALL: Yes.

A COMPARISON OF METHODS FOR PARTICULATE TESTING OF HEPA FILTERS

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Abstract

In 1976 a suggestion was made at a European Economic Community Seminar in Aix-en-Provence that there should be an attempt to correlate results of the various methods of testing HEPA filters. From previous results with the Q127 DOP equipment there seemed to be a reasonable correlation with the sodium chloride test employed in the United Kingdom when sheets of 100cm<sup>2</sup> were tested, while a few tests with HEPA filters also showed fair agreement with figures from the British Standard 3928 method. Little, however, was known of comparisons with other methods when intact HEPA filters were tested. An investigation, begun under an EEC contract in 1979, included as its object both rig and on-site assessments at various European Establishments.

Transport of large filters leads to damage of unknown magnitude, whilst seating into test ducts may not always be perfect; on the other hand, testing the same filter many times may lead to erroneous comparisons due to clogging. In consequence in the work, which has continued in 1980, a large number of sheets of 100cm<sup>2</sup> area, held in a leak-proof jig, has also been employed. Some 140 sheets of each of five grades of filter paper were obtained. Tests on randomly chosen sheets indicated consistent penetration figures (variation within any grade less than 15%) so that there were good grounds for assuming that the filters were suitable for comparison purposes. Ten different methods of rig and on-site tests have been investigated, with results reported in this paper. As expected, there are wide differences in absolute penetrations, in velocity/penetration curves and in sensitivity. Figures for penetration of the paper sheets are still awaited from ten other test houses and a detailed analysis will not be attempted until these have been received.

I. Introduction

The need for this study arose from the multiplicity of tests used in the assessment of the particulate efficiency of filters employed in the atomic energy industry, in testing of small sheets of the medium, in the manufacturer's initial test of the whole filter, and later, when installed. This last may be divided into (a) first installation and (b) routine tests at predetermined intervals.

It is obvious that manufacturers and users should be able to

compare the products which they make, buy and use, with products from other firms. Plainly, too, the system should be of as high an efficiency as practicable, leading to the conclusion that in the rig test the aerosol should contain at least a large fraction of highly penetrating particles. Among other desiderata in a rig test are sensitivity, speed of assessment, stability of equipment and that the aerosol should not be a health hazard. Ideally the aerosol for on-site tests should also consist of particles in the most penetrating sizes, but particle size is generally not so important, as measurements are mainly concerned with searching for holes and leaks due to poor sealing which are less size selective than an intact medium.

## II Preliminary Tests

In a rig test, mixing of the aerosol should be such that the concentration variation across the upstream filter face is less than 10% about the mean, while downstream it must be sufficiently good for a centrally situated sampling point to give a representative sample (within  $\pm 10\%$ ), even when there are edge leaks or pinholes on the periphery of the filter. Dorman and Yeates (1) found that adequate mixing of submicrometre aerosols was provided upstream by a  $D/2$  baffle,  $10D$  from the filter. Downstream mixing was attained in a duct of diameter  $d$  with an orifice plate  $3d$  from the filter with a central sampling point a further  $8d$  downstream, followed by a straight length of  $5d$ . Anisokinetic sampling within  $x^{1/4}$  to  $4x$  duct velocity was permissible. As mixing does not always seem to have received much attention - for example, one nuclear publication merely states that 'downstream probe positions should be at least 10 diameters from the filter unless artificial turbulence inducers are used, in which case 5 diameters are sufficient' - it was decided to carry out tests with HEPA filters containing holes of known size. Clearly testing with aerosols of various size distributions, the object of this paper, loses much of its value if sampling is faulty.

Experiments were carried out on the BS3928 rig. Briefly, with four evenly spaced atomizers upstream of the filter there was a concentration variation of 2:1 across the duct without a mixing baffle. When the upstream concentration was corrected and a hole of 0.4cm diameter made in a corner of the HEPA filter (of negligible penetration when intact) a central sampling point  $10d$  downstream indicated 0.0065% penetration. When the aerosol was properly mixed the central sampling point gave a penetration of 0.033%, an increase of  $x5$ . On the other hand, with a central hole the unmixed reading was 0.145%, falling with correct mixing to 0.04%. The foregoing shows that  $10d$  length of straight ducting is not sufficient for good mixing. As a matter of interest the penetrations were very close to theoretical calculations.

Further, it is essential that filters in rig tests should be totally enclosed in a housing in order to test the integrity of the filter case. There are still test houses where the filter is

simply clamped between two flanges with the case exposed to ambient air.

These faults - holes, incorrect sampling and fixing of filters - all militate against accurate comparison of HEPA filter test methods, both in rig and on-site work. In order to eliminate these factors it was decided that, in addition to  $1700\text{m}^3/\text{h}$  assessments, measurements would also be made of penetration through small areas of filter paper held in a leak-proof jig. The aerosol was taken via a central sampling pipe upstream of the HEPA filter, passed through the filter sheet at measured flow rates and thence to the detector.

Five different grades of glass fibre paper, 140 sheets of each grade were kindly given by an English manufacturer. The homogeneity of the paper was established by testing six or seven randomly selected sheets of each grade on the sodium flame apparatus at face velocities of 2 to  $10\text{cm/s}$ . The assessments were made on the British Standard 3928 rig with Collison atomizers, on the same rig but with Dautrebande atomizers and also on the British Standard 4400 respirator test rig. New sheets were employed for each test. The range of penetrations for the five grades was from about 0.001 to 20%. Measured penetrations of any grade varied by at most  $\pm 15\%$  about the mean for the grade (ie  $0.001 \pm 0.00015$ ). From these tests it was concluded that the papers were suitable for reliable comparisons to be made with different aerosol distributions. A large number of sheets and a filter holder of  $100\text{cm}^2$  were therefore taken to various Establishments with the intention, when time permitted, of testing up to three fresh sheets of each grade in the velocity range of 2 to  $10\text{cm/s}$ . In the event it was not always possible to carry out complete tests, nor was it always possible to test during a visit due to, for example, equipment breakdown or laboratory reorganization. In such cases promises were made that the work would be done as soon as possible and results forwarded by post. In addition some samples were mailed to test houses in Europe and the USA.

#### Types of rig on which tests have been made

1. Paraffin oil, pressure atomized, particle counter detector. Aerosol stated to be mainly  $0.3-0.55\mu\text{m}$  diameter.
2. Similar to (1) with DOP aerosol.
3. TDA 4A generator and TDA 2D or 2DN detector.
4. TDA 5A generator and TDA 2D detector.
5. Atmospheric nuclei, augmented by oily flame.
6. Atomized paraffin oil, Sartorius photometer VM102. DIN24184.
7. Sodium chloride Collison atomizers, Harwell type flame detector BS3928 rig.

8. As in (7) but with Porton hydrogen flame detector. Standard BS 3928 rig.
9. Dautrebande atomizers, otherwise as (8).
10. British Standard 4400 respirator test rig.
11. DOP Q127 respirator test rig.
12. DOP Q107, tap test.
13. Uranine aerosol. French Norme Afnor NFX44011.

It is also hoped to test with radioactively tagged aerosol as standard in West Germany.

### III Results to Date

Results are shown in Tables 1 to 4. The test houses are designated by numbers.

#### Comments on results:

##### Table 1. HEPA filter tests.

- i. Dautrebande atomizers produce a finer aerosol than that from the standard Collisons, giving about a factor of x2 augmentation of penetration.
- ii. The Harwell type detector generally indicates a rather higher penetration than the hydrogen flame detector. As the two equipments were originally calibrated to give the same figures this could be due to a change in calibration or to seating leaks in ducts.
- iii. The DOP equipment designed for on-site tests was not sensitive enough for testing filters of the highest efficiency.
- iv. Uranine penetrations of filters (a) and (d) are far in excess of those found by other test methods. Such high penetration was not found for filter (g), nor was it noted by Dupoux and Briand (2) who gave a uranine/NaCl ratio of about  $1\frac{1}{2}$ .
- v. Aitken nuclei results do not correlate well with either NaCl or cold DOP.
- vi. Nil penetration was recorded at Establishment 8. The reason for this is not clear as penetrations were measurable on the best quality sheets of 100cm<sup>2</sup>.
- vii. At Establishment 4 the maximum flow attainable was 1000m<sup>3</sup>/h. With a paraffin oil aerosol and a particle counter for the 0.3-0.55µm range penetration of filter (b) was 0.002%.

Table 2. NaCl Jig Tests

Grades 1 and 2 show good agreement with both Porton detector (hydrogen flame) and Harwell detector employing Collison atomizers. The figures for Grade 3 are less good but still acceptable. Although not noted in the Table the penetrations measured by the BS4400 respirator rig were always within 10% of the BS3928 (hydrogen flame) rig.

As with HEPA tests the Dautrebande atomizers indicate up to twice the penetrations given by Collison atomizers.

Table 3. (a) Paraffin Oil Aerosol with light scattering detector and (b) Paraffin and DOP Aerosols with particle counting

Paraffin oil penetrations at 2cm/s are in fair agreement with NaCl Collison atomized aerosol. With increase of velocity paraffin penetrations fall well below NaCl. (Establishment 8).

At Establishment 4 and 5 although tests have considerable similarity agreement is poor.

Table 4. TDA Jig Tests

Agreement is good between all three Establishments at velocities in excess of 4cm/s, although Establishments 6 and 10 were not able to detect any penetration through the two papers of the highest quality. At 2cm/s Establishment 7 found lower penetration than that found with sodium chloride for the Grade 2 paper but much higher for Grade 1.

### Untabulated Results

Comparative results from six HEPA filter tests (four dating from the 1960's) between Q107 DOP and Collison atomized NaCl have shown a ratio of DOP/NaCl near equality. Four have given a 1:1 ratio within the limits of  $\pm 10\%$  while in the other two the ratio has been 1:4.

Q127 tests at Establishment 1 on the five grades of paper indicate that DOP penetrates more easily than NaCl at 2cm/s with NaCl penetrating more easily at velocities above about 6cm/s. Another test house, however, has found DOP to penetrate less well than NaCl even at 2cm/s. Further tests are expected to be made in the USA during September of this year (1980).

### IV General Comments

There is a wide scatter of results from different methods and, indeed, between the same methods when testing papers of the same nominal efficiency. A full analysis must wait receipt of the test figures from other Establishments. The cold and hot DOP equipment

is popular for on-site tests and is simple to use. With more stringent requirements it appears that an improvement of at least a factor of x10 in sensitivity is desirable.

For rig testing the sodium chloride method employing Dautrebande atomizers may be the best. Personal experience with the monodisperse HEPA DOP rig does not permit any comments on its reproducibility or convenience.

Whatever aerosol and detector are used it is essential, both for rig and on-site work, that adequate mixing is achieved. For this it is necessary that attention is given to the design in new installations, a matter that has surely been neglected in the past.

#### Acknowledgements

The work has been made possible through the cooperation of numerous people in Europe and the USA who will be named in the final report and to whom my thanks are due. This paper is published with the permission of the Health and Safety Directorate of the European Economic Community.

#### References

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2. Dupoux, J., and Briand, A., Proc. Seminar on High Efficiency Aerosol Filtration in the Nuclear Industry, p249; (pub. EEC Luxembourg, 1977).

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TABLE I 1700m<sup>3</sup>/h HEPA filter tests

Etab No	Type of test	% Penetration						
		Filter Designation						
		a	b	c	d	e	f	g
1	NaCl BS3928 Collison Atomizers Hydrogen Flame	0.0011	0.0075	1.2	0.016	1.3	0.0001	0.0015
1	NaCl BS3928 Dautrebande Atomizers Hydrogen Flame	0.0022	0.0144	3.3		2.4		0.0025
9	NaCl Alternative BS3928 Collison Atomizers Harwell Detector	0.002			0.016	1.9		0.0048
17	" " " "					1.8	0.0002	
6	Cold DOP TDA4A TDA2D		NIL (limit 0.01)	1.7		1.2		NIL (limit 0.002)
7	Cold DOP TDA4A TDA2DN	0.003 ±0.003			0.006 ±0.002	1.7		NIL (limit 0.001)
10	" " " "	NIL (limit 0.001)			0.004	0.96		NIL (limit 0.002)
8	Paraffin oil Sartorius Photometer VM102	NIL			NIL			NIL
7	Uranine	0.325			1.25	1.0		0.0016
7	Aitken nuclei flame augmented	0.01 (limit)			0.11 ±0.02	2.1		0.002

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TABLE 2  $100\text{cm}^2$  sheets tested with NaCl aerosol

Etab No	Type of test	vel cm/s	% Penetration				
			Filter Grade				
			1	2	3	4	5
1	NaCl BS3928 Collison Atomizers Hydrogen Flame	2	0.0008	0.005	0.2	2.6	7.4
		4	0.0044	0.017	0.4	4.4	11.8
		6	0.011	0.037	0.62	5.7	14.2
		8	0.019	0.058	0.85	6.8	15.8
		10	0.028	0.082	1.1	7.5	17.0
9	NaCl Alternative BS3928 Collison Atomizers Harwell Detector	2	0.0007 0.0009 0.0009	0.0035	0.35		
		6	0.014 0.011 0.012	0.032	0.9		
1	NaCl BS3928 Dautrebande Atomizers Hydrogen Flame	2	0.0014	0.0065	0.28	3.7	10
		4	0.0072	0.013	0.6	6.5	15.2
		6	0.0154	0.046	0.95	8.7	18.3
		8	0.025	0.075	1.32	10.3	20.9
		10	0.038	0.116	1.73	11.2	22.7

TABLE 3  $100\text{cm}^2$  sheets tested with liquid aerosols, particle count and light scattering detectors

Etab No	Type of test	velv cm/s	% Penetration					
			Filter Grade					
			1	2	3	4	5	
8	Paraffin oil VM102 Sartorius Photometer	2	0.0015	0.0021	0.17	3.7	9.4	
			0.0014	0.0021	0.16	3.7	9.6	
			0.0015	0.0017	0.19	3.4	12.1	
	90% of particles less than $1\mu\text{m}$ Stokes diameter	5	0.003	0.0042	0.29	5.0	14.6	
			0.0029	0.0045	0.29	5.0	14.1	
			0.0026	0.0041	0.30	4.7	15.1	
		10	0.0039	0.0092	0.41	6.0	16.9	
			0.0039	0.0084	0.39	5.9	15.5	
			0.0039	0.0056	0.40	6.0	17.5	
	4	Paraffin oil Particle counter in $0.3-0.5\mu\text{m}$ range Aerosol stated largely $0.3-0.55\mu\text{m}$ diameter	2		0.004	0.8		
			4		0.0065	1.21		
			6		0.0085	1.39		
8			0.006	0.0085) 0.01 )	1.49) 1.35)	~2.5	>6	
5	As (4) above with DOP aerosol Penetration in $0.3-0.5\mu\text{m}$ diameter range	2	NIL	0.01 $\pm 0.02$	0.17 $\pm 0.08$	3.4 $\pm 0.2$		
		10	NIL	0.17 $\pm 0.17$	0.2 $\pm 0.03$	4.9 $\pm 0.4$		

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TABLE 4 100cm<sup>2</sup> sheets tested with TDA equipment

Etab No	Type of test	velv cm/s	% Penetration				
			Filter Grade				
			1	2	3	4	5
6	TDA2D TDA4A	2	NIL	NIL	0.12	3.7	11
		~3.6	NIL	NIL	0.24	3.8	11
7	TDA4A TDA2DN	2	0.007 0.028		0.03 ±0.004	0.9 ±0.05	3.2 ±0.1
		~4	0.05		0.17 ±0.02	2.8 ±0.01	9.7 ±0.1
		~6			0.27 ±0.01		14 ±0.2
		~10		0.003 ±0.001	0.33 ±0.02		14 ±0.4
10	TDA4A TDA2DN	5			0.3 0.34	3.7	11
6	TDA5A TDA2D	2	NIL	NIL	0.13	3.3	10
		~3.6	NIL	NIL	0.15	4.3	10.8

DISCUSSION

JOHNSON: On slide 3, there was a variety of significant figures listed for the results of some of your testing. How accurate can we really claim our photometers to be, and what is a reasonable accuracy for the method? You had, I think, anywhere from two to four significant figures on that viewgraph.

DORMAN: You are probably referring to sodium chloride. I think that between different establishments using the same equipment that is well looked after, there should not be more than 15% difference. In other words, if somebody gets 0.001% and somebody else gets 0.00115%, this is the sort of difference you might expect. It is possible with sodium chloride, though, when the equipment is made sensitive by methods I won't go into now, to test down to something like 0.00005%. Now, at that level, I would say 0.00005 could be 0.00003 or 0.00007. A variation of, say, 30% about a mean is likely. But it is possible, certainly, in the range of 0.001% to be within 15% with equipment that is well calibrated.

JOHNSON: Do you happen to know similar numbers for the DOP system?

DORMAN: I'm no expert with DOP. I've used DOP equipment, a Q127, but I've never used the Q107. I think you will be much better advised to direct your question to others here, who have far, far more experience than I.

Comparison of HEPA Filter Test Methods  
in Corrosive Environments

L. P. Murphy, S. J. Fernandez and B. G. Motes

Exxon Nuclear Idaho Company  
Idaho Chemical Programs - Operations Office  
Idaho National Engineering Laboratory

ABSTRACT

The in-plant testing of process off-gas HEPA filters is an important quality control activity of the ALARA (As Low As Reasonably Achievable) policy at nuclear facilities. Imprecise and irreproducible data were recorded during DOP testing at the Atmospheric Protection System (APS) of the Idaho Chemical Processing Plant (ICPP). The tests at the APS are performed in an environment that has high humidity, high temperatures and has  $\text{NO}_x$  present.

An evaluation of three HEPA filter test methods in corrosive environments was conducted: the dioctyl phthalate (DOP) method (US Standard Method ANSI N-101.1-1979) the sodium chloride method (British Standard 3928:1969), and the soda-fluorescein or uranine method (French Standard AFNOR STD NFX 44-011). The effects of high humidity, temperature and oxides of nitrogen ( $\text{NO}_x$ ) on each method was examined. The effects of each variable and any interaction between variables on the test methods were examined. Recommendations for changes in the standard methods to reduce erratic on-line results are presented.

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\* Work performed under USDOE contract DE-AC07-791DO1675

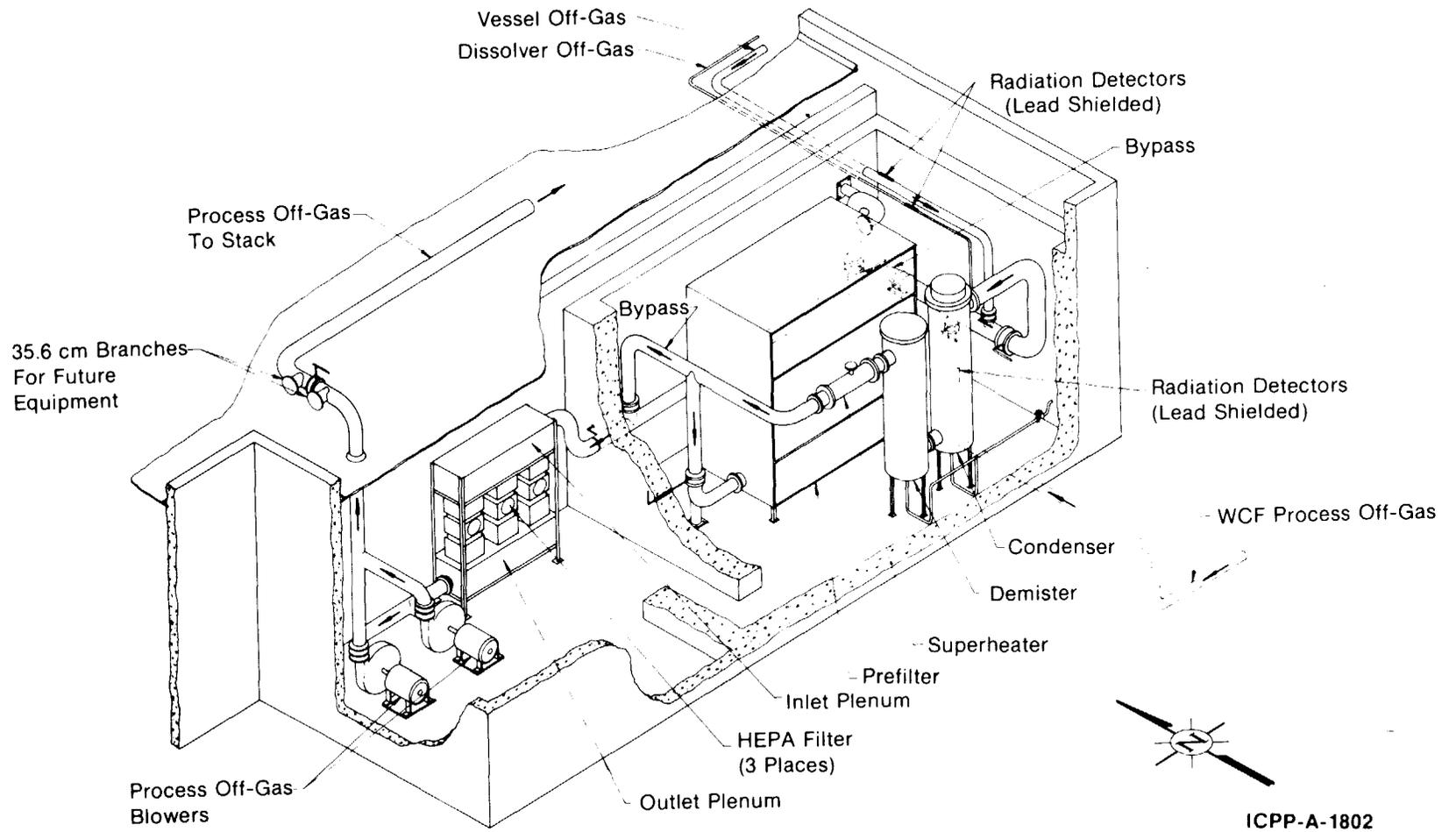
## I. INTRODUCTION

The Idaho Chemical Processing Plant (ICPP), located at the Idaho National Engineering Laboratory, is operated by Exxon Nuclear Idaho Company for the Department of Energy to recover uranium from spent fuel elements. Aqueous wastes from the recovery process are solidified in a fluidized bed waste calcining facility.

The Waste Calcining Facility (WCF) produces an off-gas stream that contains high concentrations of nitrogen oxides ( $\text{NO}_x$ ), and has high temperatures and high relative humidities. The final process off-gas clean up system before emission to the environment is the Atmospheric Protection System (APS). The APS consists of a condenser, demistor, superheater, prefilter, and finally three HEPA banks. An isometric view of the APS process off-gas area is shown in Figure 1.

To ensure that the HEPA filters are functioning properly, it is necessary to ascertain whether these filters have sustained damage during transportation to or installation in the APS and that the assembly of the filter elements is satisfactory. Thus, testing these HEPA filter banks in place in the corrosive environment is required.

To provide the most appropriate filter test method, the American DOP, the British NaCl and the French uranine filter test methods were evaluated in the laboratory to determine the combined effects of temperature, humidity, and  $\text{NO}_x$ . The most appropriate method as determined by this laboratory evaluation was demonstrated in-plant.



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Figure 1. Isometric View of APS Process Off-Gas Area

## II. DESCRIPTION OF THE TEST METHODS

The development of HEPA filter test methods has been conducted in several countries. Some, such as the American DOP method use liquid droplets as a challenge aerosol, while others such as the British NaCl and French uranine methods use solid particles. The Nuclear Energy Agency of the Organization for Economic Cooperation and Development, recommended <sup>(1)</sup> at a meeting for the exchange of information on gaseous wastes, that test conditions for HEPA filters be defined with respect to high temperatures, humidity and radiation conditions. The participants at this meeting considered the DOP, uranine, and sodium chloride methods to be most important and recommended that these methods be investigated as to suitability for in-situ tests in all process stream conditions. Brief descriptions of these methods are given in the following sections. More detailed descriptions are presented elsewhere. <sup>(2,3, 4)</sup>

A. Diocetyl Phthalate (DOP)

This method was originally developed in the United States and employs either a monodisperse or polydisperse liquid aerosol of DOP. The aerosol can be produced by either a thermal or pneumatic type generators.

An aerosol at a concentration of  $100 \text{ mg/M}^3$  from either type of generator is introduced upstream of the filter to be tested. To determine the filter efficiency, the air is sampled before and after the filter and the efficiency is determined by comparing particle concentrations upstream and downstream of the filter. These particle concentrations are measured with a forward light scattering photometer. The DOP method is useful when there are many tests to be performed and/or no interferences are present since the penetration measurements are practically instantaneous.

B. Sodium Chloride (Sodium Flame)

In the sodium flame test, performed according to British Standard 3928, an aqueous solution of sodium chloride is atomized. The moisture evaporates, leaving a dry cloud of solid cubic salt particles. These particles range from 0.01 to 1.7

$\mu\text{m}$ , with a mass median diameter of  $0.6 \mu\text{m}$ . To measure filter efficiency, samples of the air before and after the filter are directed to a flame photometer which measures the intensity of the characteristic yellow sodium emission line produced in a hydrogen flame. The intensity of this emission line is directly proportional to the mass of sodium chloride present.

This method, like the DOP method, measures penetration in real-time.

For our studies, the single particle laser aerosol spectrometer<sup>(5)</sup> was substituted for the flame photometer, because the sampling conditions at the APS were not practical for the flame photometer.

### C. Soda-Fluorescein (Uranine)

In France, a fluorescent aerosol is used to evaluate HEPA filters. The aerosol is produced by atomizing an aqueous solution of uranine. After the elimination of large droplets with an inertial separator, the remaining liquid aerosol is evaporated by dilution with dry air. A solid aerosol results with an average particle size corresponding to the size of maximum penetration ( $0.08 \mu\text{m}$ ) for HEPA filters at the face velocity encountered at the APS.<sup>(6)</sup>

The test aerosol is injected upstream of the filter being tested and samples are collected before and after the HEPA on 47 mm membrane filters. The uranine is extracted from the sample filters with ammonium hydroxide and the extracts measured fluorimetrically (496 nm excitation, 517 nm emission). This fluorimetric analysis requires about 45 minutes.

## III. LABORATORY EVALUATION

1. Description of Laboratory TestsA. Experimental Apparatus

Because of the highly radioactive environment of the WCF and APS off-gas, an experimental system was constructed to simulate the off-gas conditions. The studies were conducted with 20 cm x 20 cm x 15 cm (8 inch x 8 inch x 5 7/8 inch) HEPA filters in tandem. Measurements of aerosol concentrations were taken upstream of both filters, between the filters, and after the final filter.

This test assembly is shown in Figure 2. The DOP, NaCl and uranine methods were adapted to the apparatus with appropriate sampling modifications. The system included an aerosol generator, injection and sampling points for water vapor,  $\text{NO}_x$  and the test aerosol. Also included was a reaction bomb for partial conversion of NO to  $\text{NO}_2$ . This reaction provided a final mixture of 50/50 NO/ $\text{NO}_2$ .

The gas flow through the system was measured by linear mass flow transducers and the flow was maintained at 25 scfm. Humidity and  $\text{NO}_x$  concentrations were measured with a hygrometer<sup>a</sup> and chemiluminescent<sup>b</sup> analyzer, respectively. The temperature was monitored with thermocouples and controlled with heat tapes.

B. Experimental Design

The experimental design used in this study was a two-level factorial design containing three variables (temperature, humidity, and  $\text{NO}_x$ ). Factorial designs are, in general composed by selecting a fixed number of levels of each variables, and performing all the possible unique tests. The greatest strength of a factorial design is that the controlled variables can be varied simultaneously, thereby providing information about interaction of variables.

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a Series 1200 AP Optical Dew Point Hygrometer manufactured by General Eastern Corp.

b Model 10, NO- $\text{NO}_2$ - $\text{NO}_x$  Chemiluminescent Analyzer Manufactured by Thermo Electron Corp.

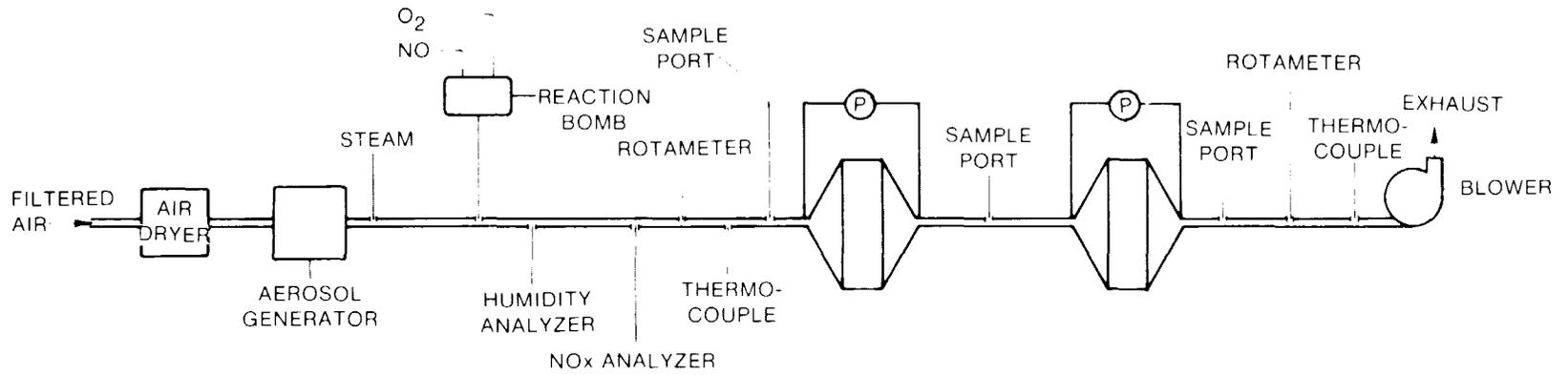


Figure 2 HEPA Filter Test System

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The  $2^3$  factorial design, which is useful for ease of analysis and for its predictive capability for responses that do not have strong curvature in the region of interest, requires that two levels be chosen and all eight ( $2^3 = 8$ ) combinations tested. The two levels chosen for this study were: for temperature, 66 and 93°C; for humidity, 50 and 95%; and for  $\text{NO}_x$  concentration, 500 and 10,000 ppm.

## 2. Results and Discussion of Laboratory Evaluation

### A. Diethyl Phthalate (DOP)

The DOP studies were conducted by measuring penetration with the conventional forward light-scattering photometer and the laser single particle aerosol spectrometer. The method used followed as closely as possible that detailed in ANSI N-101.1-1979. When testing streams with high dew points (above 60°C) a drying tube was used to lower the sample dewpoint without removing the aerosol. This dryer is based on selective permeation of water vapor as described by Fernandez, *et al.*<sup>(7)</sup> The effects of humidity,  $\text{NO}_x$ , and temperature on the standard DOP in-place test method were examined. The results of these examinations were reported at a previous Air Cleaning Conference<sup>(8)</sup>, however they are discussed briefly in the following sections.

#### a. Light-Scattering Photometer

Preliminary tests were performed to establish baseline behavior for the photometer by measuring DOP concentrations in room air. The tests were performed at a flow of 25 scfm and temperatures of 66 and 93°C. Each test lasted two minutes and 100 mg/M<sup>3</sup> of DOP challenged the filters.

The results of these measurements indicated a background penetration of 0.002% ± .001% (0.2 parts per ten thousand (ppt)).

After the baseline measurements were completed, scoping tests with DOP and conditions ranging from 16% to 50% relative humidity (RH) and from 500 to 10,000 ppm  $\text{NO}_x$  were performed. The apparent penetrations for all of the tests at 50% relative humidity varied rapidly from 0% to 40% penetration. When measurements were attempted with room air and 500 to 10,000 ppm of  $\text{NO}_x$ , a negative reading was obtained.

Although the reason for this behavior is far from obvious, an explanation consistent with the data is that at high humidities the water vapor condensed within the scattering chamber causing unrealistically high penetration values. The negative response during the dry NO<sub>x</sub> runs was attributed to absorption of the light by the brown-colored NO<sub>2</sub>. During the high humidity, high NO<sub>x</sub> tests, the NO<sub>x</sub> was scrubbed from the scattering chamber by the condensation, yielding the same response as the high humidity, no NO<sub>x</sub> case.

The problem of condensation occurring in the photometer was solved by installing a drying tube to remove water vapor without removing the aerosol. The NO<sub>x</sub> interference was eliminated by making the photometer stray light adjustment while sampling the stream containing the NO<sub>x</sub>.

Once these modifications were made, experiments were carried out following the experimental design shown below. The eight test design was duplicated, and the average penetration measured was  $1.9 \pm 0.9$  pptt.

Figure 3

2<sup>3</sup> Experimental Design

Relative Humidity %	NO <sub>x</sub> (PPM)	Temperature °C
50	500	66
95	500	66
50	10,000	66
95	10,000	66
50	500	93
95	500	93
50	10,000	93
95	10,000	93

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The design was performed in random order. Each test point was duplicated.

b. Laser Aerosol Spectrometer

Tests similar to the ones performed with the photometer were performed with the laser aerosol spectrometer. These tests established baseline behavior for the laser by measuring DOP concentrations in dry air.

The results of these tests indicated background penetration of  $.02 \pm .03$  pptt for the laser spectrometer in a well filtered gas stream.

The baseline measurements were followed with tests designed to ascertain if there were effects due to humidity or  $\text{NO}_x$ . The results of these measurements indicated that the laser showed no humidity or  $\text{NO}_x$  effect, yielding the same penetration as the baseline measurements.

The HEPA filter efficiencies in a corrosive environment using the laser were obtained. The average penetration was  $1.7 \pm 1.6$  pptt.

In summary the statistical analysis of the two-level design at the 95% confidence interval gave the following results. Using the photometer, an effect of  $+1.5 \pm 0.2$  pptt was observed due to water vapor. All other effects were insignificant at the 95% confidence interval. This water vapor effect may be due to residual condensation in the scattering chamber.

Using the laser spectrometer, a small temperature effect of  $0.30 \pm 0.09$  pptt was observed. The effects of all other variables were insignificant at the 95% confidence interval. The cause of this effect is unknown but may be due to the fact that the vapor pressure of DOP is sufficiently high at temperatures above  $80^\circ\text{C}$  to cause a wicking effect through the filter. In any event, this temperature effect is much less than the observed 1.7 pptt variability in the overall average.

By comparison, the laser spectrometer is superior to the photometer since it is three times more sensitive than the photometer and has no water vapor or  $\text{NO}_x$  interference.

The variability of the DOP method, i.e., 1.5 pptt, is unacceptable. For example, many acceptable filters are in the 99.97 to 99.95% efficiency region, the added 1.5 pptt penetration bias would cause a good filter to fail the DOP test.

## B. Sodium Chloride

British Standard 3928: 1969 calls for a solid challenge aerosol to be generated by atomization of an aqueous solution of common salt from large collision - type atomizers. Subsequently, measurements are made upstream and downstream of the filters with a flame photometer. In this study, the standard procedure was modified by substituting the single particle laser aerosol spectrometer for the flame photometer. The flame photometer was thought to be impractical from safety considerations and the laser allows particle size determinations in addition to concentration.

In these experiments, the challenge aerosol was produced by atomizing a 5% solution of NaCl. The particles were polydisperse and 95% were in the range of 0.15 to 1.0  $\mu\text{m}$ . The entire range was from 0.10 to 1.72  $\mu\text{m}$ .

As with the DOP tests, when experiments based on a  $2^3$  factorial design were performed, the variable that caused the greatest interference was water vapor. No results were obtained at 95% relative humidity and 95°C due to condensation inside the sample line and spectrometer. Heat tracing the sample lines was not possible due to the laser's temperature limitations. Attempts to remove the water vapor with the drying tube mentioned earlier were successful, but the plateout of aerosol inside the dryer was as high as 16%. This plateout was significantly greater than with DOP, probably due to differences in the particle size distributions.

Under normal operating conditions, the relative humidity at the APS rarely falls below 60%. Consequently, any satisfactory system must be suitable for use in humid gas streams. Upon completion of the two-level experimental design, the water vapor effect was determined to be 5.5 pptt (see Table I). Since the maximum penetration allowed is 5.0 pptt, high humidity conditions will cause a "fail" test no matter the actual efficiency of the filter. One possible explanation was suggested by Dorman<sup>(9)</sup> who observed that in tests with humidities above 50%, the NaCl aerosol takes on moisture providing a mechanism for particles to pass through the filter.

TABLE I

Measured Filter Efficiencies in Corrosive Environments (NaCl)

<u>Test Conditions</u>			<u>Penetration</u>
<u>Relative Humidity %</u>	<u>NO<sub>x</sub> (PPM)</u>	<u>Temperature °C</u>	<u>Parts/10,000 (pptt)</u>
15	500	25	0.6
55	500	25	5.2
15	5,250	25	0.7
55	5,250	25	6.3
15	500	60	0.5
55	500	60	4.4
15	5,250	60	0.6
55	5,250	60	7.5

An analysis of the data listed above based on a two-level design and the 95% confidence interval indicate the standard deviations for the measurements was  $\pm 0.4$  pptt. The minimum significant factor effect was  $\pm 0.5$  pptt. If the computed factor effect is twice the absolute value of the minimum significant factor then we conclude that there is an effect.

The effects of NO<sub>x</sub> and temperature were insignificant within the 95% confidence limits as were the interactions. These findings are consistent with the DOP experiments that showed that the laser was unaffected by NO<sub>x</sub> atmospheres. In addition, the NaCl test method has been recommended for temperatures as high as 400°C.<sup>(10)</sup>

In conclusion, the NaCl test method is not recommended for humid atmospheres due to the hygroscopic nature of the test aerosol.

#### C. Soda-Fluorescein (uranine)

The French soda-fluorescein or uranine method was the final method evaluated. The standard method for the testing of high efficiency filters in France (AFNOR Standard NFX 44011) was studied under high humidities, high NO<sub>x</sub> concentrations and high temperatures. In the standard test, aerosol samplings are collected upstream and downstream of the test filter on 47 mm glass fiber filters. The sample flowrate was 2 to 5 m<sup>3</sup>/hr. The uranine was extracted with 5%

ammonium hydroxide solution. The fluorescence of the solution was then measured at the characteristic uranine wavelengths (496 nm excitation and 517 nm emission). The ratio of the uranine mass concentration downstream of HEPA filter to the quantity of aerosol found upstream is the measured penetration.

To conduct our laboratory evaluation, a uranine generator, filtration pots, and sampling devices were purchased from Societe Setra, Ormesson, France. A Varian Model SF-330 Spectrofluorometer was used to measure fluorescence.

Preliminary tests were conducted at ambient conditions to check the performance of the equipment, the reproducibility of the generator, and the reliability of the sampling and measurement techniques. The amount of aerosol generated was about  $18 \pm 2.5$  mg/hr. Once the initial adjustments were made the generator functioned without attention for three hours with the only requirement being a 100 psi air supply.

The particle size of the aerosol was measured with a diffusion battery.<sup>(11)</sup> All particles were smaller than  $0.2 \mu\text{m}$  with a geometric mean diameter of  $0.08 \mu\text{m}$ .

Penetration measurements were performed on the same test apparatus as the DOP and NaCl tests.

The average penetration of the uranine method was  $3.4 \pm 1.0$  pptt with a range of 2-5 pptt.

The results of the measurements using a  $2^3$  factorial design indicated that there was no main effect for relative humidities up to 80%, temperatures up to  $90^\circ\text{C}$  and  $\text{NO}_x$  concentrations up to 10,000 ppm. Also, no detectable interaction between temperature, relative humidity and  $\text{NO}_x$  was observed. However, when condensation was detected on the sample filter the uranium was leached from the filter. This was eliminated by heating the sample filter significantly above the sample dew point.

This laboratory evaluation revealed the French uranine method was satisfactory for use in humid, high temperature, corrosive atmospheres and that it had the following advantages over the DOP and NaCl filter test methods:

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- 1) The uranine aerosol maintains its integrity in temperatures up to 120°C and relative humidities as high as 80%.
- 2) The uranine aerosol is a solid with an average particle size of 0.08 μm. This makes the uranine more representative of the particles that penetrate APS HEPA filters.
- 3) The fluorimetric assay eliminates the interferences from in-leakage of non-fluorescent ambient aerosols.

### IV. IN-PLANT DEMONSTRATION

The French uranine method was selected for in-plant demonstration tests. This demonstration was performed in conjunction with another task for extending the uranine method to relative humidities greater than 80%. The uranine and DOP methods were used simultaneously to determine the efficiencies of the HEPA filters at the APS.

The APS process off-gas system consists of a condenser, demister, superheater, glass fiber prefilter bank, and three HEPA filters (see Figure 4). The total flow of the combined process off-gases is limited to 4500 cfm and this flow is split equally between the three HEPAs.

A comparison of DOP and uranine was made under WCF conditions. Table II shows the result of this comparison.

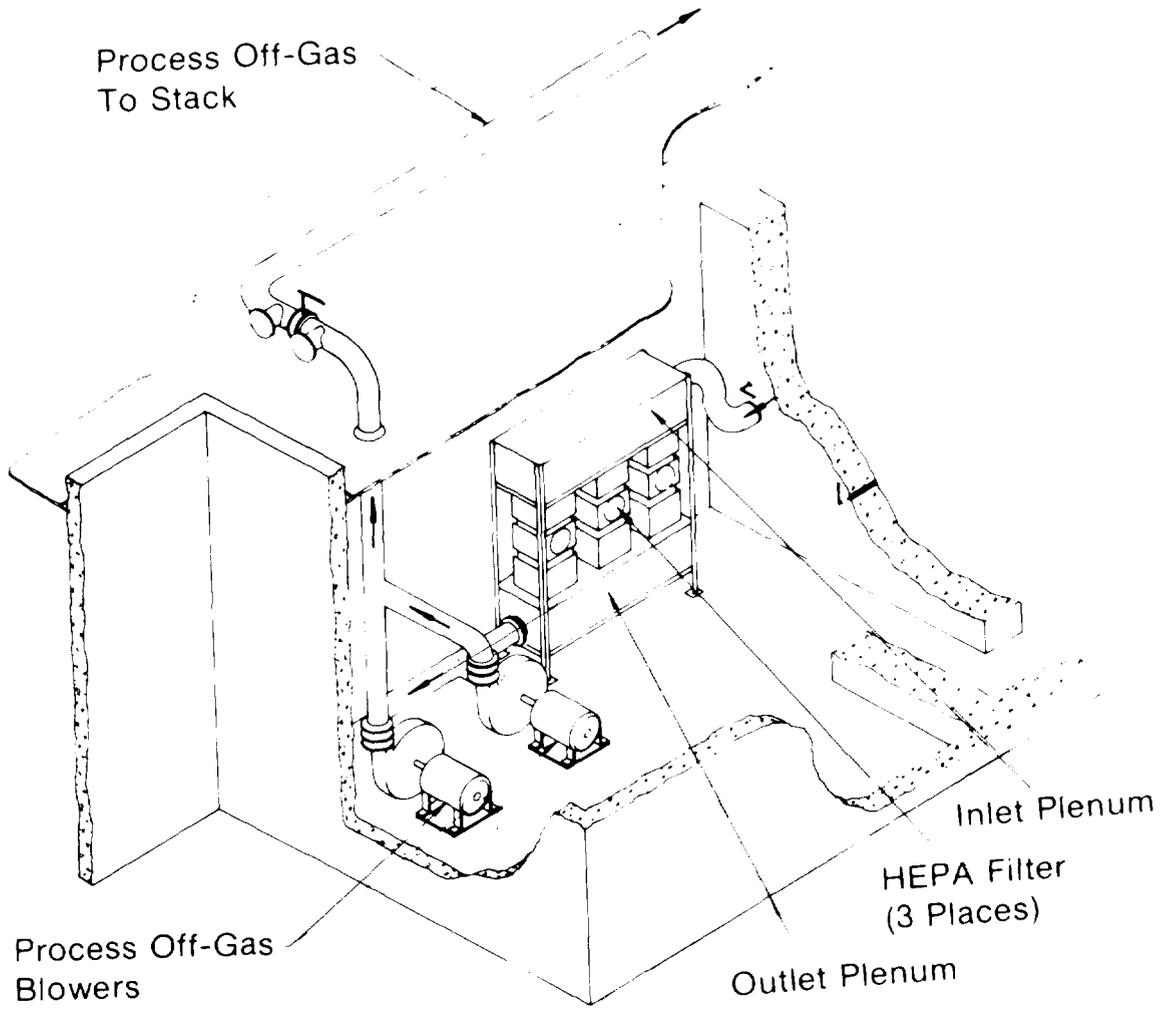
TABLE II

In-Plant Measurement W/DOP and Uranine

Filter #	NO <sub>x</sub> Present	Measured Efficiencies	
		DOP	Uranine
OGF-100	YES	—	85%

Measurement with DOP and the photometer were not possible while the calciner was operating. The measurement of that filter was made after the calciner shut down (no NO<sub>x</sub>) and it failed the DOP test.

More tests were performed during the WCF shut down. The results are shown in Table III. In these measurements fluorescein and eosin-y were used to attempt to extend the uranine method to relative humidities above 80%.



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**Figure 4. APS Process Off-Gas Filters**

TABLE III

## In-Plant Measurements W/DOP and Other Fluorescent Aerosols\*

<u>Filter #</u>	<u>Measured Efficiencies</u>		
	<u>DOP</u>	<u>Fluorescein</u>	<u>Eosin-Y</u>
OGF-100	a99.96	99.8 <sup>b</sup>	-----
-101	a99.94		97.4 <sup>b</sup>
-102	99.98		99.93 <sup>b</sup>

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\*Note: WCF was shutdown

a These filters failed the DOP test

b The difference in the efficiencies between DOP and the fluorescent aerosols is thought to be due to the smaller diameter of these aerosols.

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In summary, because imprecise and irreproducible data were recorded during past DOP testing <sup>(12)</sup> at the Atmospheric Protection System during WCF operations, we collected data using the uranine aerosol at this point. But, the WCF has been down for an extended period and the data from in-plant demonstrations has been limited. However, brief operating experience has shown that measurements are possible with uranine at the APS in NO<sub>x</sub> containing streams.

V. CONCLUSIONS AND RECOMMENDATIONS

In conclusion, the French uranine method has the following advantages over the NaCl and DOP filter test methods:

- 1) The uranine aerosol is a solid aerosol, rather than a liquid like the DOP aerosol. This makes the uranine aerosol more representative of the solid particles in the WCF off-gas stream.
- 2) The fluorimetry assay used in the uranine method eliminates interferences from in-leakage of non-fluorescent ambient aerosols. The DOP and NaCl methods are subject to these interferences.
- 3) Without modifications the uranine method can be used in relative humidities as high as 80% (with modifications the humidity limits can be extended) and temperatures as high as 120°C. The DOP method is only recommended for ambient conditions; while the NaCl method can withstand temperatures as high as 400°C, it is not recommended for relative humidities above 50%.

The advantages listed above underscore the results of the 2<sup>3</sup> factorial design. The design indicated that there were no main effects or interactions on the French uranine method from relative humidities up to 80%, temperature up to 90°C, and NO<sub>x</sub> concentrations up to 10,000 ppm. Based on this satisfactory performance, the uranine method was selected to determine the particle retention efficiency of the Atmosphere Protection System HEPA filter banks.

The results under the in-plant demonstration are limited due to operation of the waste calciner. We recommend further measurements at the APS filters to study the effect of NO<sub>x</sub> and uranine particle size on the French uranine method.

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DISCUSSION

ETTINGER: I wonder if you monitored the size of the several aerosols, i.e., sodium chloride, DOP, uranine, and eosin, because of the great importance of size on filter performance? This would be important, first, because there is a natural variation when generating aerosols of presumably constant sizes and, second, aerosol size variations at high humidity. I also think you would have difficulty generating a small uranine aerosol because of the hygroscopic nature of uranine. I wonder if you measured the size of any or all of these aerosols on a continuing basis as a function of the other variables you've used, e.g.,  $\text{NO}_x$ , temperature, and humidity.

MURPHY: We did not do that for DOP and sodium chloride. We have an independent study going on now at the INEL where monitoring particle size in relation to the effects of these same conditions on the sizes of the uranine aerosol is the purpose of the study. This work was referenced in the paper.

SIGLI: I have an answer to your question about relative humidity. Up to 80% relative humidity, you cannot measure variation in the size of the aerosol. At more than 80% relative humidity, you have absorption of moisture on the aerosol so that the size is not as constant as at lower relative humidity. In reply to your question, you can assume that you have no variation in the aerosol size up to 80%.

PRATT: You mentioned in your paper that you used an aqueous solution to generate the sodium chloride aerosol. I am directing my question as much to Mr. Dorman as to Mr. Murphy. I wonder if you have any comments on whether one would expect a different result using a salt stick generator.

DORMAN: Probably. I think that whatever the method of generation and size of the salt aerosol, it is going to take up moisture. One point I would take issue with, though, is the statement that sodium chloride cannot be used above 50% relative humidity. It is perfectly all right up to 65% and it can be used up to 75%, but above 65% it does begin to grow slowly, and this will affect the results for filter penetration. That is to say, penetration at 75% humidity will be less than at 65% because of the particle size change. To repeat, I think that however you generate your salt particles, they are going to take up moisture sooner or later.

IN-PLACE TESTING OF TANDEM HEPA FILTER  
STAGES USING FLUORESCENT AEROSOLS\*

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Abstract

Fluorescent test aerosols were used in field testing of large multiple-stage HEPA filter systems. The technique excluded interference from non-fluorescent background particles known to leak into the plenum or ducting between the filters and the downstream sampling probe. This technique solved the problem of measuring extremely low concentrations of the test aerosol in the presence of background aerosol.

The upstream fluorescent test aerosol was diluted with clean air and drawn into a single particle aerosol spectrometer capable of counting, sizing, and detecting fluorescence of each particle. The particle sizing function was performed on light scattered by the particle passing through the beam of a helium-cadmium laser. Concurrently the fluorescence excited by the laser illumination was detected at a longer wavelength. Since spectrometer response in the fluorescent mode was <2 percent of naturally occurring aerosols, background aerosols were insignificant as an interference to the downstream concentration measurement. Decontamination factors (DF) on the order of  $10^8$  were measured in the field studies on >9.4 m<sup>3</sup>/s (20,000 cfm) systems. Additional generator capacity and acceptably lower test aerosol to background aerosol concentration ratios could be used to extend this capability to measure DF greater than  $10^8$ .

Dye-tagged DOP aerosols were generated either by gas-thermal or sonic nozzle generators. Experiments with the gas-thermal generator showed only 20 percent of fluorescence from the dye was degraded by the vaporization process. A single sonic nozzle was shown to aerosolize 0.7-1.0 L/hr of dye-tagged DOP aerosol in the proper size range for HEPA filter testing. A multiple sonic nozzle generator is a practical consideration to provide greater capacity.

I. Introduction

This project was directed toward development of new methods of in-place testing of air cleaning systems containing multiple stages of high efficiency particulate air (HEPA) filters. Earlier work produced a highly sensitive method for testing two stages of HEPA filters with a capacity of up to 14.2 m<sup>3</sup>/s (30,000 cfm) using a single-particle, particle-size spectrometer<sup>(1)</sup> in combination with

\*Work supported by the Office of Nuclear Waste Management and performed at the Los Alamos Scientific Laboratory operated under the auspices of the US Department of Energy, Contract No. W-7405-ENG-36.

an aerosol diluter.(2,3) Usefulness of this method in testing some systems was restricted by extraneous aerosol leaking into the downstream structure, causing an excessive background count. Development of an aerosol spectrometer which could detect a tagged or self-identifying test aerosol was undertaken. Coordinating its development directly with an instrument manufacturer has made the fluorescent particle spectrometer immediately available.

Testing an air cleaning system as a unit was considered advantageous over stage-by-stage tests using the less-sensitive photometer method of ANSI/ASME Standard N510(4) for the following reasons: future air cleaning systems could be simplified and made more compact; existing systems could be tested without need for auxiliary ducts and valves to inject challenge aerosol between the first and second stages; testing could proceed without interruption of system operations; and a conservative decontamination factor (DF) of the system could be determined without the possibility of overestimation when individual stage DF's are multiplied together. This overestimation was attributed in earlier testing(3) to challenging the second stage with an aerosol containing larger particle sizes normally removed by the first stage.

The additional sensitivity which made the laser spectrometer method applicable to high DF measurement was gained from the complete lack of instrument noise in its digital counting electronics and from its ability to integrate particle count over any required time period. Its particle sizing capability allowed determination of system DF in terms of the ratio of upstream to downstream particle count or volume (mass) in any particle size interval, rather than the ratio of integrated values of instantaneous light scattering signals from an aerosol which undergoes major size change during filtration. Improved accuracy is inherent in the method, although the need for aerosol dilution adds a source of error which offsets this gain somewhat. A disadvantage of the spectrometer method was the more complex, more expensive, and less rugged nature of the spectrometer equipment; however, extensive field testing showed this to be a manageable problem.

## II. Experimental Apparatus and Techniques

Typical system arrangement for testing a two-stage filter system is shown in Fig. 1. Upstream and downstream sampling locations should be selected in conformance with aerosol mixing and sampling recommendations in ANSI/ASME N510.(4) The spectrometer and dilution systems are described in detail as their use was applied to several phases of the experimental program. The photometer is considered optional for continuously monitoring dilution ratio.

### Description of Fluorescent Particle Spectrometer

The laser fluorescent particle size spectrometer, PMS Model ASAS-XF (Particle Measurement Systems, Inc., Boulder, CO.), was designed to identify tagged (fluorescent) particles over a size range of 0.125 to 3.1  $\mu\text{m}$  diameter. Two selectable ranges covered this total range, with each range having fifteen linear size intervals. The light source for the sizing of particles was a 10 mW

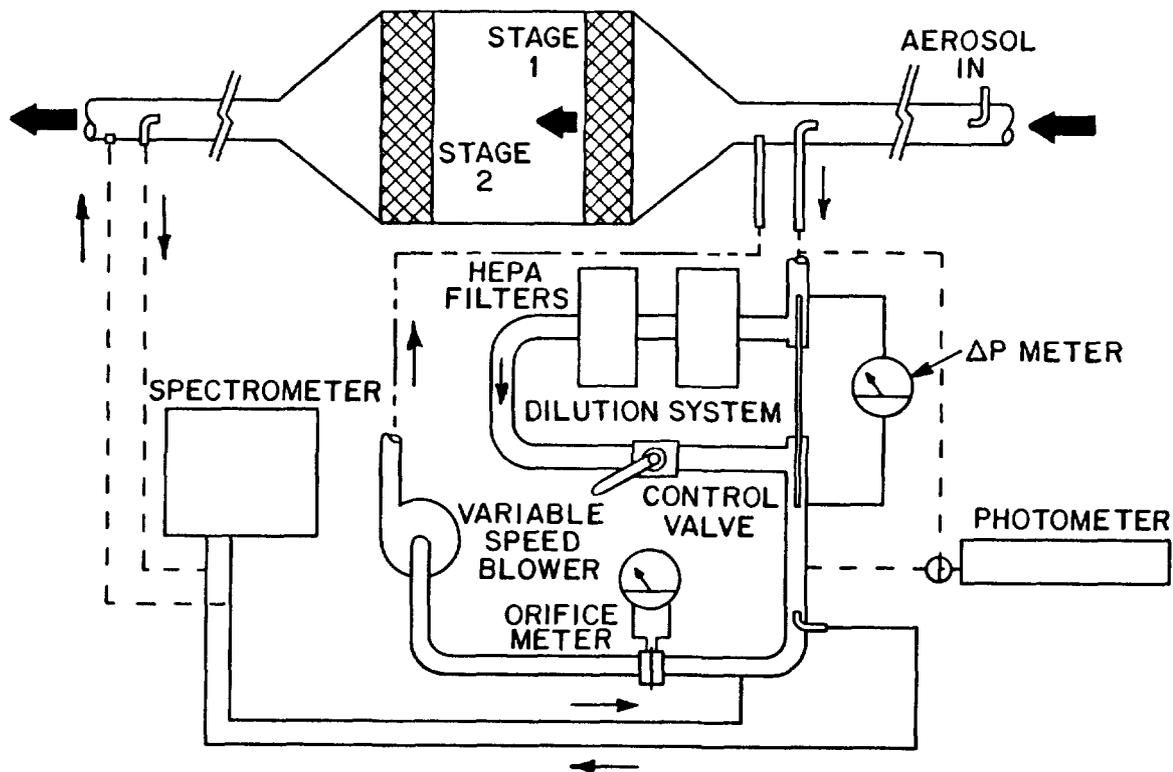


Figure 1.  
HEPA filter test system connection.

HeCd laser operating at a wavelength of 442 nm (blue light) while the fluorescent detection wavelength was greater than 475 nm. This separation between laser (excitation) and fluorescent (emission) wavelengths allowed separate detection of the scattered light for sizing a particle and the emitted light for fluorescent identification.

The optical system is shown schematically in Fig. 2. Light scattered from a particle passing through the active volume of the spectrometer was collected by a parabolic mirror over the angle  $35^\circ$  to  $120^\circ$  and directed to a photomultiplier via mirror and beamsplitter. Intensity of the light pulse was proportional to the physical diameter of the particle, allowing pulse amplitude to be used as a measure of particle diameter. The output of the preamplifier was applied to a pulse height analyzer composed of sixteen voltage comparators and latches. As a voltage pulse was received and sized, it was counted in an appropriate channel of a 16 channel accumulating memory. Coincident with sizing, light emitted from a fluorescent particle at the longer wavelength was producing a pulse from the fluorescent photomultiplier which enabled a count to be registered when the mode selector was set for fluorescent counting. Either fluorescent (FL) or all particle (ALL) mode could be selected by positioning a single switch. At the end of each sampling interval, the final count values were printed on a paper tape record along with time-of-day and total elapsed time. A built-in oscilloscope

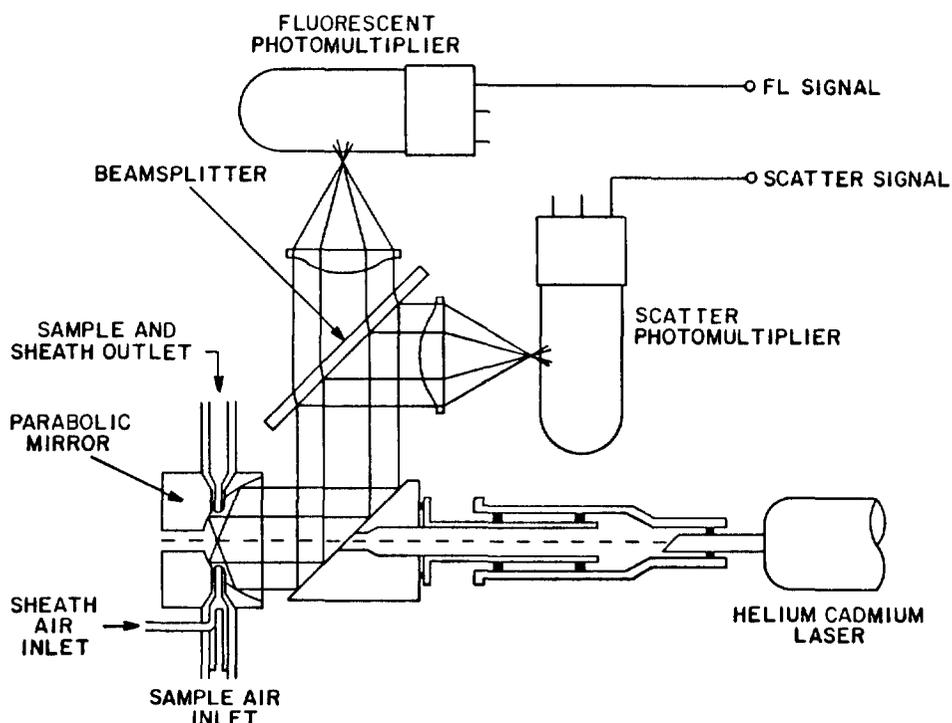


Figure 2.  
ASAS-XF optical system diagram  
(courtesy of Particle Measurements Systems).

displayed a histogram of particle size distribution which could be frozen at any time to allow detailed analysis of a distribution. Counting rate limitations were observed to avoid entering a non-linear region. This occurred at counting rates above  $10^4/s$  where more than one particle was consistently in the active volume at one time. Sampling flowrate was  $3.0 \text{ cm}^3/s$  (maximum). Clean sheath air was used to surround the hydrodynamically focused, particle-laden airstream as it entered the laser beam. Laminar flow conditions and isokinetic merging of the two airstreams restricted turbulence and mixing within the sample inlet. Sampling flow and sheath flow rotameters were calibrated using a precision bubble meter and were found to be within  $\pm 4$  percent over their ranges.

Calibration of the ASAS-XF spectrometer was performed by the manufacturer using twenty monodisperse aerosols of polystyrene latex spheres (Dow Chemical Corp., Midland, MI). A possible difference in response of the spectrometer due to difference in refractive index of the DOP aerosol used in field testing (1.50) and the polystyrene spheres used in calibration (1.58) was investigated by the manufacturer. His Mie scattering calculations indicate DOP particles would be sized with approximately  $+0.02 \text{ }\mu\text{m}$  error near the  $0.10 \text{ }\mu\text{m}$  calibration point and  $+0.2 \text{ }\mu\text{m}$  at the  $1 \text{ }\mu\text{m}$  calibration point; i.e., a  $1 \text{ }\mu\text{m}$  DOP particle would be registered one channel above a  $1 \text{ }\mu\text{m}$  polystyrene particle. Since the DOP test aerosol was almost totally composed of droplets smaller than  $1 \text{ }\mu\text{m}$ , this source of error was considered to be insignificant.

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The ASAS-XF laser spectrometer was contained in three packages; one for the laser optical system (18.6 kg, 41 lbs), a second for the electronics enclosure (25.5 kg, 56 lbs), and the third for the laser power supply (5 kg, 11 lb). These packages were cart-mounted for use in the field as shown in Fig. 3 and were easily transported. Operational problems were not serious, involving occasional misalignment of the parabolic mirror aperture. Readjustment of this aperture required 5-10 min. Cleaning of optics was rarely required.

### Sample Dilution

The limitations on count rate specified for the fluorescent particle spectrometer prohibited direct upstream sampling of the challenge concentration required to test large two-stage HEPA systems. Dilution of the sample on the order of 500 to 700 was required. A suitable dilution system was developed with adjustable dilution over the range 500 to 1000. This system, shown schematically in Fig. 1 and also in Fig. 3, provided a diluted sample for the spectrometer from a sidestream duct which responded to pressure fluctuations in the main duct. Connecting the suction and exhaust of the dilution system to the main duct avoided fluctuations in sampling flow noted in earlier dilution systems. Nominal dilution system flow rate was  $0.0024 \text{ m}^3/\text{s}$  (5 cfm), induced by an  $0.038 \text{ m}^3/\text{s}$  (80 cfm) variable-speed, centrifugal blower. This blower was oversized to accommodate the heavily negative static pressure in some HEPA filter systems [up to  $-38 \text{ cm}$  (15 inches) wg]. The major portion of the dilution stream was directed through two HEPA filters ( $0.012 \text{ m}^3/\text{s}$ , 25 cfm capacity) and was recombined with the small unfiltered sample stream passed through a small diameter tube ( $0.162 \text{ cm}$  diam, 33 cm long). Differential pressure measurement across a standard orifice meter provided flow indication. Dilution ratio was selected by setting differential pressure across the small diameter tube by positioning the ball valve. The dilution ratio measurement was calibrated by determining the ratio of light scattering intensity upstream and downstream of the small diameter tube. This calibration indicated an expected error (one standard deviation) of approximately  $\pm 7$  percent at dilution ratios 700 and above, and  $\pm 2$  percent at dilution ratios of 500 or less. This was compatible with the expected error of the DF measurement across two HEPA filter stages. Particle size characteristics ( $\text{cmd}$  and  $\sigma_g$ ) upstream of the diluter were reduced less than 5 percent by passage through the diluter. Actual particle losses in the tube were accounted for in the calibration procedure.

### Fluorescent Dye Selection

Features of interest in selection of a dye tag were solubility of the dye in DOP, fluorescent intensity at ambient temperature, adequate separation between excitation and emission wavelengths, and retention of fluorescence during aerosol generation from a high-capacity, gas-thermal generator. Potomac yellow No. 838 (Dayglo Color Corporation, Cleveland, OH) was selected over many other dyes after its tag was readily detected in small droplets ( $0.15 \text{ }\mu\text{m}$  diam) by the laser fluorescent particle spectrometer. It had a 440 nm excitation wavelength, well within the band width of the HeCd laser of the fluorescent particle spectrometer, and an emission wavelength of 490 nm, adequately separated from the excitation wavelength for

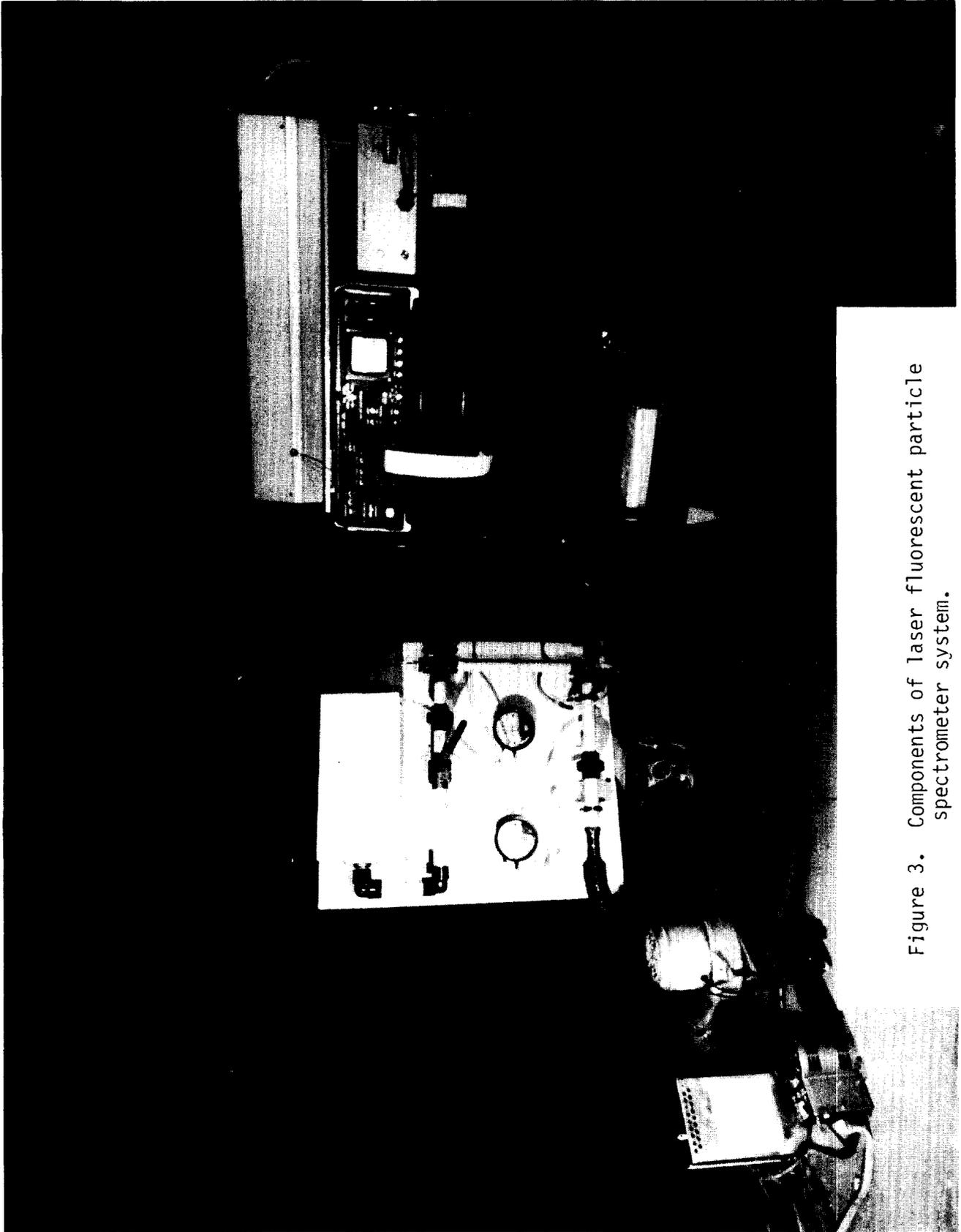


Figure 3. Components of laser fluorescent particle spectrometer system.

discrimination by the spectrometer. Solubility of Potomac yellow dye in DOP was determined to be 2.0 g/L. This amount of dye was best dissolved by heating the DOP to 90°C for approximately one hour.

#### Fluorescent Aerosol Background

Naturally occurring or spurious fluorescent particles were known to be present in the atmosphere and in the ventilation systems of buildings. The effect of these particles on the fluorescent particle test method was evaluated by sampling the background aerosol in open building areas, in ventilation ducts both upstream and downstream of HEPA filter systems in buildings housing chemical and metallurgical research, and at several locations outdoors. The laser fluorescent particle spectrometer was used in both FL (fluorescent) and ALL (all particle) modes to provide the FL/ALL particle ratio. The highest FL/ALL value was 0.016 encountered in open building areas. The lowest value was 0.007, measured outdoors at 1.6 m above ground level. At the sampling points downstream of two HEPA filter stages, only once in 20 counting periods of 10 min each was a fluorescent particle registered by the spectrometer. These data confirmed that the fluorescent particle background downstream was negligible.

#### Aerosol Generation

Dye-tagged aerosols suitable for testing HEPA filters were generated using two methods: (1) gas-thermal generation in which bulk solution containing the DOP and the dye are vaporized and recombined in condensed droplets of the proper size and (2) atomization generation by acoustic or sonic breakup of droplets into smaller droplets. A sonic nozzle generator was developed to produce large quantities of dye-tagged particles without need for vaporization. A sonic nozzle [Sonicore No. 035 (Sonic Development Corporation, Upper Saddle River, NJ)] was mounted in a generator assembly as shown in Fig. 4. This nozzle directed a jet of compressed air containing large droplets of DOP into an acoustic cavity<sup>(5)</sup> where a sonic standing wave broke up the droplets and directed the fog outward toward the wall of the horizontal cylinder. Larger drops impinged on the cylinder wall, requiring collection of DOP in a drain bottle for recycling. A small fan provided airflow to remove the aerosol from the generator.

Fig. 5 shows performance characteristics of the sonic nozzle generator at an optimized primary air pressure of  $4.1 \times 10^5$  Pa (60 psig). The feedrate increased with fluid pressure to a maximum of approximately 17 ml/min at  $1.1 \times 10^5$  Pa (16 psig) and above. Losses of large droplets impinging on the generator walls and recovered in the drain bottle reached a peak of 41 percent at  $1.1 \times 10^5$  Pa (16 psig) and decreased at higher pressures. Optimum settings provided particle concentration of  $2.6 \times 10^5$  particles per  $\text{cm}^3$ , adequate for testing 0.47  $\text{m}^3/\text{s}$  (1000 cfm) systems. Larger systems would require several nozzles mounted in the same generator. Particle size characteristics over this fluid pressure range averaged  $0.27 \pm 0.04$   $\mu\text{m}$  [ $\pm 1$  std dev on 46 values of count median diameter (cmd)]. Mean geometric standard deviation of the DOP aerosol over the same pressure range was  $1.55 \pm 0.06$  ( $\pm 1$  std dev). A single

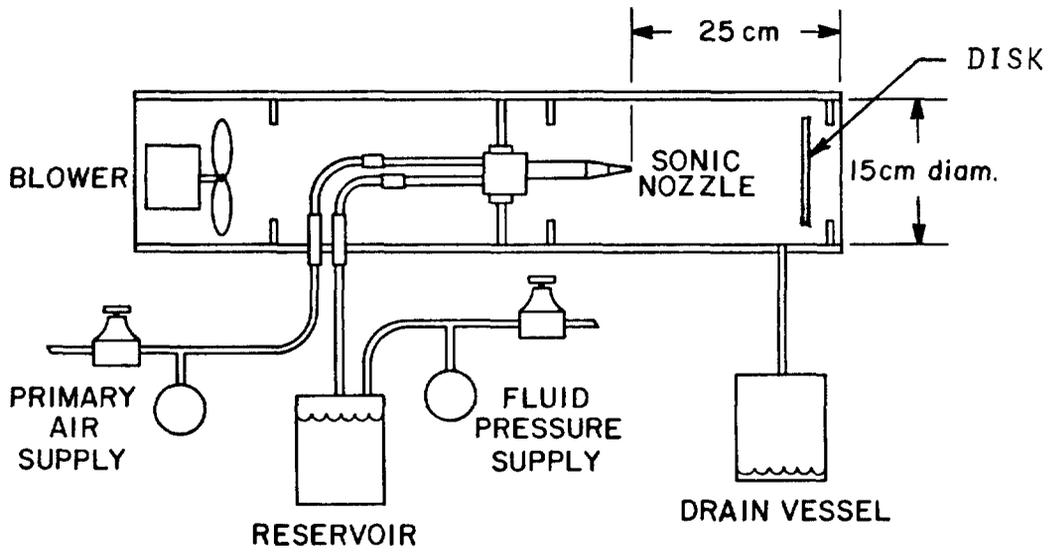


Figure 4.  
Sonic nozzle aerosol generator.

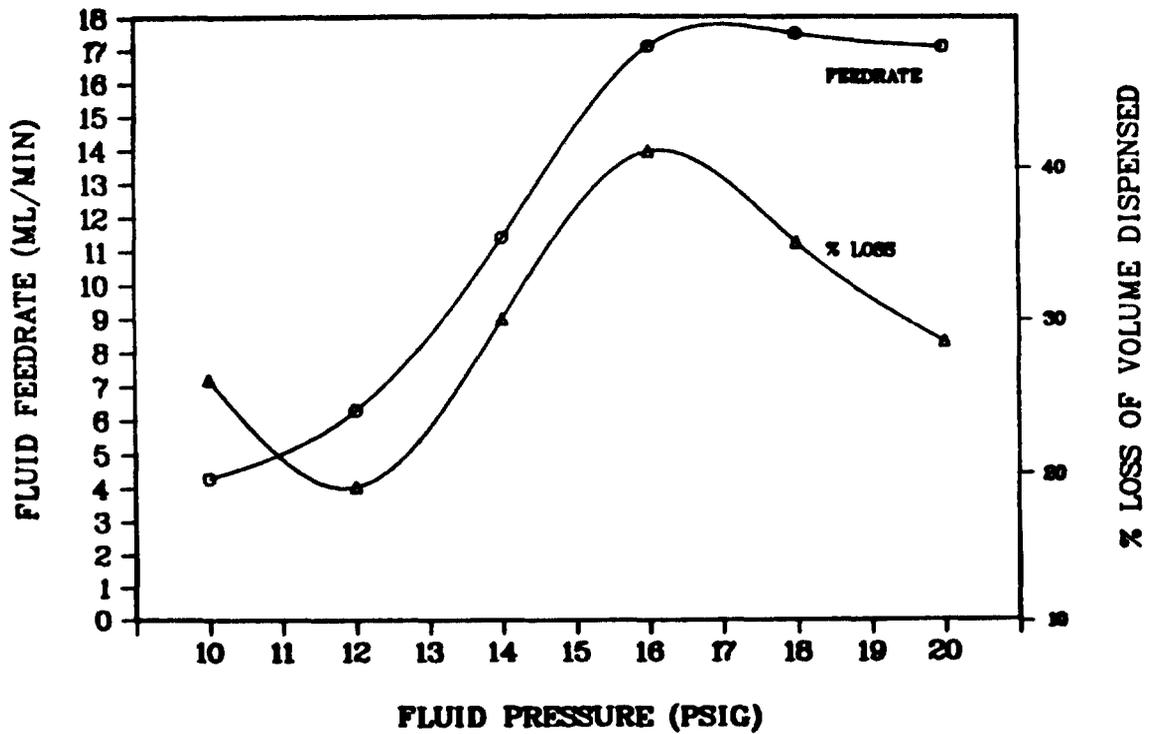


Figure 5.  
Performance characteristics of the sonic nozzle generator.

sonic nozzle generator required only  $9.4 \times 10^{-4} \text{ m}^3/\text{s}$  (2 cfm) compressed air and a 115 Vac supply for its operation and provided a convenient, high-output source of liquid droplet aerosol.

Higher feedrates and smaller particle size distributions were obtained with two versions of a gas-thermal generator in which a mixture of  $\text{CO}_2$  gas and the dye-DOP solution was forced under pressure into a vaporization chamber operating at approximately  $315^\circ\text{C}$ . As the  $\text{CO}_2$  - DOP vapor mixture exits the generator through a jet, it cools and condenses on available condensation nuclei in room air, forming a dense cloud. The smaller of these generators, the Cloud-maker (Model 11-48, Testing Machines, Inc., Amityville, NY) provided a feedrate of 10 ml/min with none of the wall losses noted with the sonic nozzle generator. Average FL/ALL particle ratio for this generator was  $0.78 \pm 0.10$ , as some particles were not counted as fluorescent. Size characteristics of the aerosol were the same whether sized in the FL or ALL particle mode of the spectrometer, indicating that loss of fluorescence was not size dependent. Size characteristics of the DOP aerosol from the small gas-thermal generator were  $0.20 \pm 0.01 \text{ } \mu\text{m}$  ( $\pm 1$  std dev) cmd and  $1.24 \pm 0.04$  ( $\pm 1$  std dev)  $\sigma_g$ , indicating the gas-thermal generator produces a smaller and narrower size distribution than the sonic nozzle generator. Number concentrations as high as  $5 \times 10^6$  particles/cm<sup>3</sup> were generated consistently.

A high-capacity, gas-thermal (HCGT) generator was developed for use in field testing large HEPA filter systems. One of several configurations of HCGT generator is shown in Fig. 6. Some difficulty in obtaining high, steady numbers of fluorescent particles was encountered at DOP feedrates above 60 ml/min, although the generator produced satisfactory DOP aerosol at higher feedrates for non-fluorescent particle testing. Fluorescence quenching was attributed to gradual cooling of the vaporization chamber below the temperature at which the dye would vaporize. The solid dye particles left after evaporation of the DOP carrier would then only occasionally recombine with a freshly condensed DOP droplet to give it a dye tag. Further modifications of the HCGT generator are expected to render it more stable at higher feedrates. Particle size characteristics of the HCGT generator were similar to the smaller generator characteristics, yielding  $0.19 \pm 0.02 \text{ } \mu\text{m}$  ( $\pm 1$  std dev) cmd, and  $1.22 \pm 0.04$  ( $\pm 1$  std dev)  $\sigma_g$ . The FL/ALL ratio for the HCGT generator was  $0.77 \pm 0.08$  ( $\pm 1$  std dev). As discussed later, this fluctuation in FL/ALL required several checks during field testing to assure consistent results.

#### Decontamination Factor and Uncertainty Calculations

Performance of each two-stage filter system was calculated in terms of decontamination factor DF by the equation:

$$DF = \frac{DC_D}{t_D} \left( \frac{C_{ds}}{t_{ds}} - \frac{B_{ds}}{t_B} \right)^{-1} \quad (1)$$

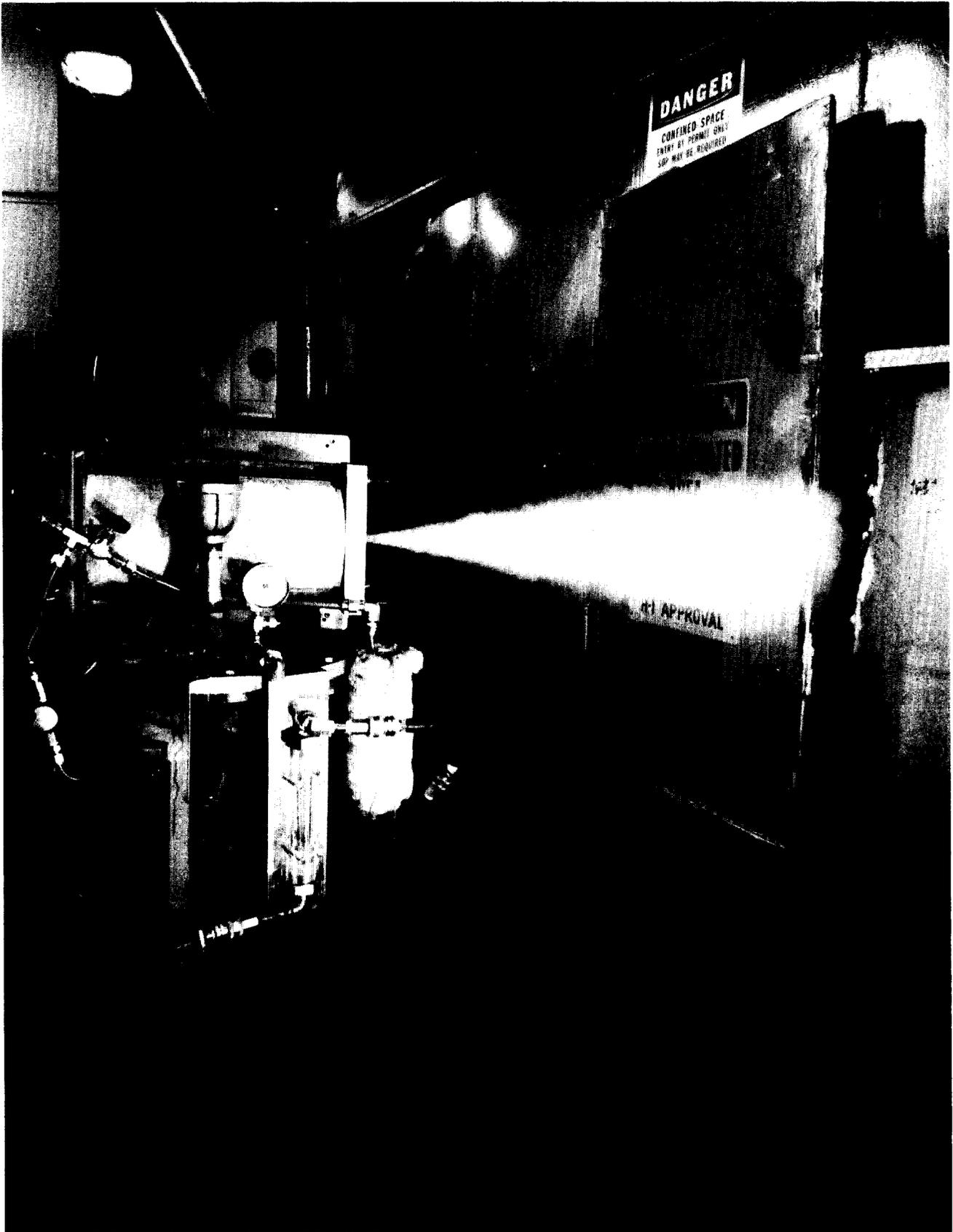


Figure 6. High-capacity, gas-thermal aerosol generator.

where  $D$  = dilution ratio,  
 $C_D$  = total count in the diluter (upstream sample),  
 $t_D$  = upstream sampling time,  
 $C_{ds}$  = downstream total count,  
 $t_{ds}$  = downstream sampling time,  
 $B_{ds}$  = downstream background count,  
 $t_B$  = downstream background sampling time.

Stage 1 decontamination factor by spectrometer was calculated by the following equation:

$$DF_1 = \frac{D C_D}{t_D} \left( \frac{t_1}{C_1} \right) \quad (2)$$

where  $C_1$  = total count downstream of Stage 1.

$t_1$  = sampling time downstream of Stage 1.

Stage 2 decontamination factor was calculated by the following equation:

$$DF_2 = \frac{C_1}{t_1} \left( \frac{C_{ds}}{t_{ds}} - \frac{B_{ds}}{t_B} \right)^{-1} \quad (3)$$

where the terms were as defined earlier.

The fractional standard deviation ( $\Delta DF/DF$ ) in the decontamination factor measurement across multiple stages was estimated by assuming that the random counting process of the spectrometer was a Poisson process, in which the standard deviation is approximated by the square root of the count number.<sup>(6)</sup> Therefore, the uncertainties based on one standard deviation of downstream count  $C_{ds}$  and downstream background  $B_{ds}$  were expressed as  $\Delta C_{ds} = \sqrt{C_{ds}}$  and  $\Delta B_{ds} = \sqrt{B_{ds}}$ . By assuming equal counting times to obtain  $C_{ds}$  and  $B_{ds}$  and no error in measuring the counting times,  $\sqrt{C_{ds}}$  and  $\sqrt{B_{ds}}$  could be combined as the root mean square of the sum as follows:

$$\frac{\Delta (C_{ds} - B_{ds})}{C_{ds} - B_{ds}} = \frac{(C_{ds} + B_{ds})^{1/2}}{C_{ds} - B_{ds}} \quad (4)$$

By combining this calculated uncertainty with the uncertainty terms for the dilution ratio  $D$  and upstream count  $C_D$ , and assuming these are all random and independent, the uncertainty in  $DF$  of multiple stages became:

$$\frac{\Delta DF}{DF} = \left[ \left( \frac{\Delta D}{D} \right)^2 + \frac{1}{C_D} + \frac{C_{ds} + B_{ds}}{(C_{ds} - B_{ds})^2} \right]^{1/2} \quad (5)$$

The fractional standard deviation in  $D$  was based on one standard deviation of the experimental results of the diluter calibration and subsequent checks. A conservative value for  $\Delta D/D$  was assumed to be

0.10 in this case. The term  $C_D$  was always large ( $>10^4$ ), permitting the  $1/C_D$  term to be neglected.

The effect of background count on the uncertainty of the DF measurement was calculated by Equation (5) to determine what conditions would favor the use of fluorescent particles. Figure 7 shows calculated  $\Delta DF/DF$  as a function of particle count above background ( $C_{ds} - B_{ds}$ ). The error is lowest for the fluorescent particle case ( $B_{ds} = 0$ ) and shows that errors can be maintained below  $\pm 0.50$  by counting downstream until 4 or more counts are collected. In non-fluorescent testing, a background of only 20 counts in a given time interval would require 15 counts above background (approximately 35 total) to maintain the error below  $\pm 0.50$ . These extra counts in all particle mode require counting times 3-4 times that in the fluorescent particle mode.

III. Experimental Results and Discussion

Laboratory Scale HEPA Filter Testing

Following development and calibration of the aerosol dilution system and checkout of the laser fluorescent particle spectrometer, a series of 20 tests of a two-stage,  $0.39 \text{ m}^3/\text{s}$  (820 cfm) HEPA filter system was performed preliminary to the field testing program. Two aerosol generators were used: the small gas-thermal generator for eight tests and the prototype sonic nozzle generator for 12 tests. Decontamination factors (DF) of the two-stage system averaged  $2.5 \times 10^7$  with a standard deviation of  $1.1 \times 10^7$  for 20

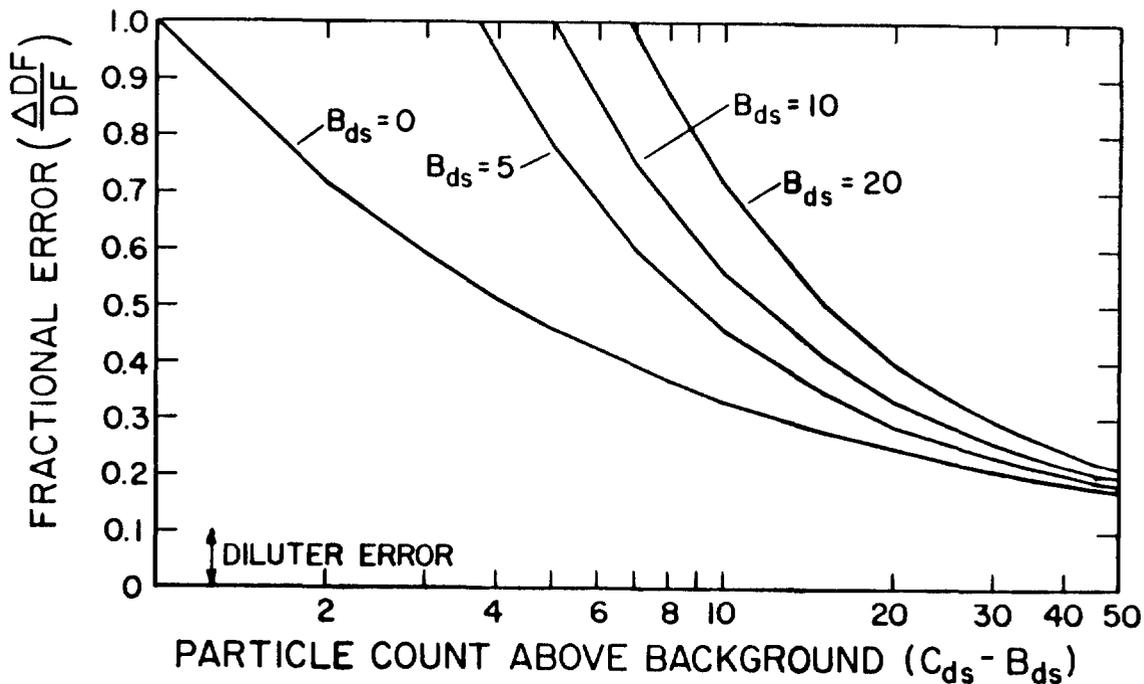


Figure 7. Error estimates based on background counts.

tests. The fractional standard deviation for these 20 tests (0.44) was larger than expected from test data of a single system. However, by selecting data taken when only the small gas-thermal generator was used at a single dilution ratio (700), the fractional standard deviation was only 0.15 for 5 tests.

The DF values above were based on totals of counts in all channels of the spectrometer. Since very few particles in the downstream sample appeared above Ch. 4 ( $>0.30 \mu\text{m}$ ), DF of particles in a narrower size range (Ch. 1,2,3,4) was calculated to determine whether this would provide a significantly different and more conservative result. Where the sonic nozzle aerosol generator was used, DF was reduced by approximately 28 percent; using the small gas-thermal generator, the reduction was only 14 percent since fewer larger particles were generated in the gas-thermal process.

### Field Testing of Large Two-Stage Systems

Decontamination factors of two large two-stage HEPA filter systems at the Chemistry and Metallurgy Research Building, LASL, were measured in FL and ALL modes of the ASAS-XF aerosol spectrometer and by the standard ANSI/ASME N510 photometer method. System FE-14, a 5 x 12 filter system rated at  $28.3 \text{ m}^3/\text{s}$  (60,000 cfm) was operating at  $22.9 \text{ m}^3/\text{s}$  (48,500 cfm) with a nominal pressure drop across Stage 1 of 3.2 cm (1.25 inches) w.g. at the time of testing. System FE-33 was operating at a reduced capacity of  $10.4 \text{ m}^3/\text{s}$  (22,000 cfm) at the time of testing. Pressure drop across its first stage was 1.0 cm (0.4 inches) w.g.

Results of multiple tests of these systems are shown in Figs. 8 and 9. The mean and standard deviation of FE-14 DF were  $0.90 \pm 0.26 \times 10^8$  for three FL tests;  $0.58 \pm 0.26 \times 10^8$  for three ALL tests. The single photometer test yielded  $\text{DF} = 0.27 \times 10^8$  for this system calculated by multiplying the individual DF's of Stage 1 and Stage 2. The mean and standard deviation of FE-33 tests were  $1.6 \pm 1.0 \times 10^8$  for four FL tests;  $2.0 \pm 0.7 \times 10^8$  for four ALL tests. The photometer test of FE-33 yielded a calculated DF of  $5 \times 10^8$ . DF measured by photometer was expected to exceed the spectrometer DF, particularly at Stage 2, due to reduced sensitivity of the photometer to the smaller mean particle size downstream of a filter stage.<sup>(3)</sup> This was not evident in these test results but the photometer results were too few to allow any generalizations to be made. However, the photometer data have been included in Figs. 8 and 9 to show the methods agreed reasonably well (mean values agreed within a factor of three) and all test results exceeded the DOE DF guideline of  $4 \times 10^6$  (99.95 percent efficiency on each of two stages).

Estimates of uncertainty in the FE-14 and FE-33 experimental results were calculated by Equation (5). Downstream FL counts ranged from 6 to 22, accounting for some of the variation in  $\Delta\text{DF}/\text{DF}$  values. The fractional standard deviations ranged from 0.20 to 0.46 for the ALL particle tests and from 0.24 to 0.42 for the FL tests. These experimental uncertainties showed reasonable consistency with the theoretical fractional standard deviations discussed earlier. In general, uncertainties of  $\pm 0.50$  should be expected when measuring DF on the order of  $10^8$ , whether by the non-fluorescent or fluorescent spectrometer method.

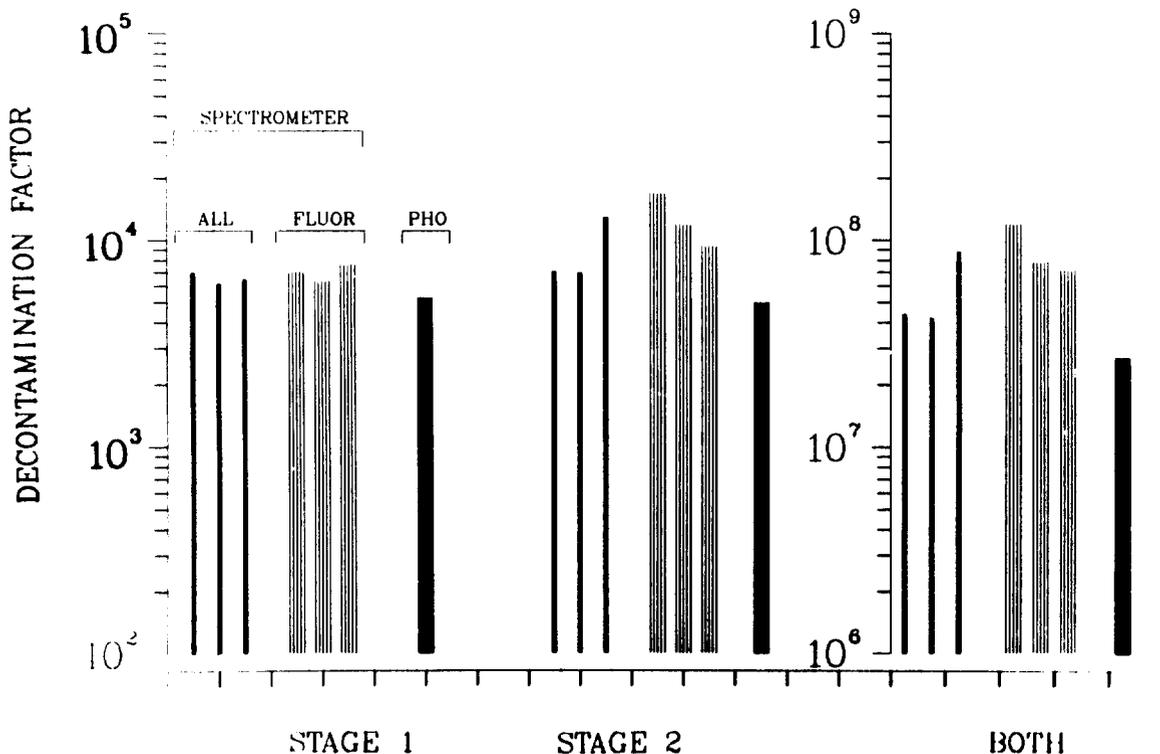


Figure 8. Performance of FE-14, a large two-stage HEPA filter system.

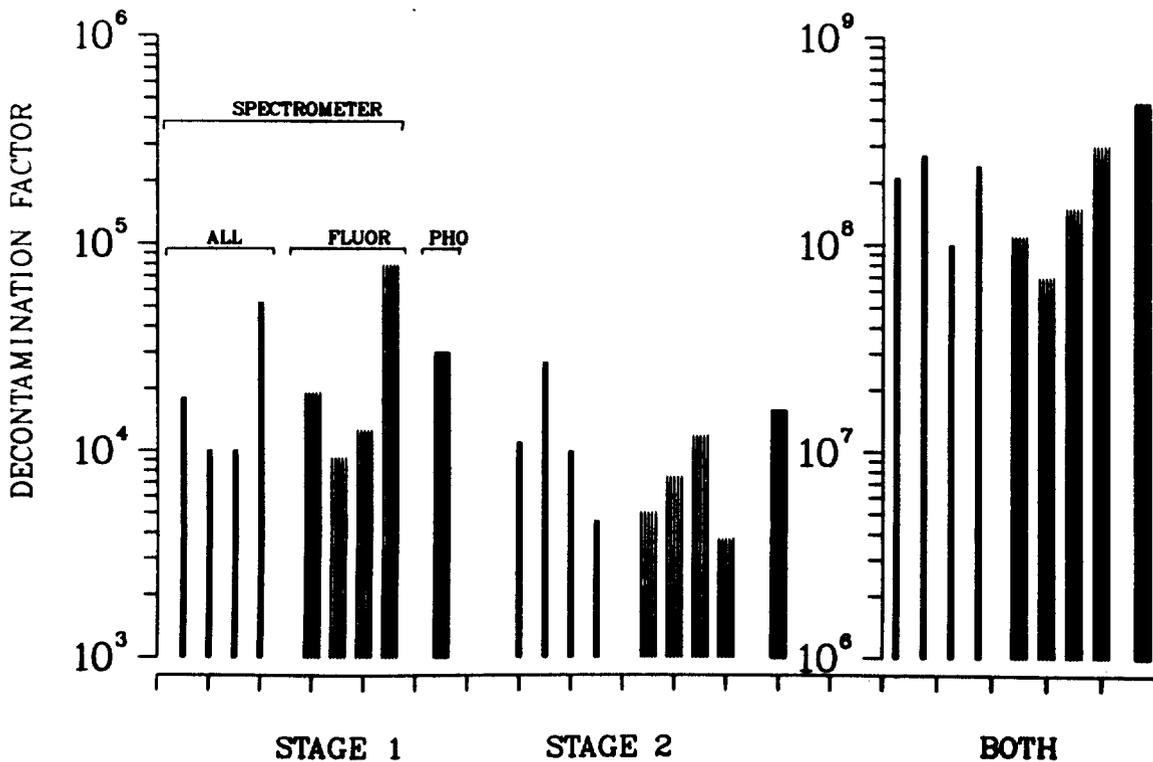


Figure 9. Performance of FE-33, a large two-stage HEPA filter system.

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These  $\Delta DF/DF$  values indicated no clear-cut advantage was gained by using fluorescent particles to reduce uncertainty in testing these particular systems, nor were sampling times significantly shortened. However, these systems are considered to contain a typically low background non-fluorescent aerosol concentrations. Future testing of most large two-stage systems is expected to benefit from fluorescent particle use and from improved stability of the HCGT generator.

### Sampling Line Losses

Particle loss in the sampling tube attached to the spectrometer was estimated by sampling a polydisperse DOP aerosol which had undergone filtration through one stage of HEPA filters. All large particles ( $>0.4 \mu\text{m}$ ) were removed by this filtration and the aerosol was rendered similar in particle size characteristics to the aerosol sampled downstream of two HEPA filters. Numbers of particles sized in the first three channels of the spectrometer ( $0.125 - 0.250 \mu\text{m}$ ) were compared by sampling from a chamber using a 33 cm (13 inches) sampling tube and a longer (142 cm, 56 inches) sampling tube. Both tubes were 0.32 cm i.d. plastic tubing. The longer tube was identical in length and configuration to the tube used in the field testing. The sampling tube was connected to a 122 cm, 0.48 cm i.d. sampling probe. Numbers of particles collected with the short tube exceeded the number collected with the longer tube by 2 to 5 percent. This indicated that errors in the particle counting were not large. The sum of losses in the probe and sampling tube was expected to be in the range of 5 to 10 percent; however, a correction factor was not applied to count results since upstream and downstream errors were expected to approximately offset each other.

### Filter Loading

Filter loading as a potential problem in two-stage HEPA filter testing by the fluorescent particle method was monitored during the field tests in terms of differential pressure across the first stage. As a general observation, it was noted that the  $28.3 \text{ m}^3/\text{s}$  (60,000 cfm) systems displayed very little  $\Delta P$  increase during as much as 2 hours generation time if the original  $\Delta P$ 's were low. For example, System FE-33 Stage 1  $\Delta P$  was 1.0 cm (0.40 inches) w.g. at the beginning of testing and 1.12 cm at the end of 100 min; it returned to 1.0 cm within 48 hours of the end of aerosol generation. As an illustration of a system starting with a somewhat higher loading, FE-14 Stage 1  $\Delta P$  started at 2.8 cm wg and increased to 3.2 cm after 175 min of testing. This  $\Delta P$  also returned to its original value; while the actual time required was not noted it was less than five days. Although the dye tag must have imposed a permanent loading on the filters, it was not measurable. Loading of a filter system by this test method does not appear to be a significant problem.

### Decontamination Factor As A Function Of Particle Size

The particle counts within each size interval provided by the spectrometer were used to calculate DF of Stage 1 as a function of particle size. The scatter in the data above Ch. 4 prevented correlation with earlier efforts to describe the size of minimum DF(2); however, all DF data from the field study indicate

continuously diminishing DF over the operating range of the spectrometer, from which the size of minimum DF could be inferred to be below  $0.15 \mu\text{m}$ . Schuster and Osetek<sup>(2)</sup> placed the size of minimum DF near  $0.19 \mu\text{m}$  by a similar method under more controlled conditions.

#### IV. Summary

A laser fluorescent particle spectrometer has been developed and successfully used to measure decontamination factors of large two-stage HEPA filter systems. The fluorescent tag placed in the DOP test aerosol was detected with high efficiency and eliminated the background aerosol problem which interfered with earlier measurements. Measurements of DF as high as  $10^8$  were accomplished on a  $22.0 \text{ m}^3/\text{s}$  (48,500 cfm) system. This indicates the range of sensitivity provided by the fluorescent particle spectrometer method to be sufficiently broad to test most two-stage HEPA systems as single units. Filter loading by the test aerosol was shown to be a minor problem. Although the spectrometer and associated equipment are more complex, more expensive, and less rugged than photometer equipment typically used in filter testing, advantages of the method such as DF measurement more representative of actual filter performance and potential savings in testing costs make the fluorescent particle spectrometer method a useful test method.

#### Acknowledgements

The authors recognize and appreciate the cooperation and assistance of John Ortiz, Industrial Hygiene Group, in the performance of the field tests.

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DISCUSSION

STEINBERG: How much did the whole system you used for your tests weigh?

ELDER: The equipment that you saw on the cart weighs a little over 100 pounds.

STEINBERG: The next question is a little more important. Did you ever determine the significance of the filter leaks that you were finding? That is to say, what was your point of reference? For example, in DOP testing we use 100 micrograms per liter upstream and penetration of more than .01 or .05 micrograms downstream represents a significant leak.

ELDER: I don't think that we have what you call a point of reference. The upstream concentrations were on the order of 30 micrograms per liter, and the size of the gas-thermally generated DOP was about 0.2  $\mu\text{m}$  count median diameter with a geometric standard deviation of 1.5 to 1.6. I think this counting method comes as close to an absolute system as we can get; it is not a relative system. We just counted as long as we felt we needed to, to get sufficiently high numbers to give a reasonable result. Sometimes it might not be over 10 counts at the downstream point; that is, not more than 10 counts in the counting interval.

STEINBERG: You're familiar with the scanning of filters when you use DOP?

ELDER: Probing?

STEINBERG: Yes. Did you probe with this instrument, too, or did you just go to a point a certain distance downstream and then count everything that came up?

ELDER: The test program did not involve any kind of changes to the operating system. We did not coordinate the test program with a shutdown of the system. As far as I know, there has been no leak probing using the spectrometer as a testing device. It's really not for that purpose.

STEINBERG: Well, then, what is it for? Weren't you looking for leaks? Something wrong with the system?

ELDER: We sampled performance by conducting in-place testing, only, with an upstream and a downstream comparison of aerosol concentration. We did not do leak probing with it.

STEINBERG: So, it's just academic, then.

ELDER: No, it is not academic. It is an in-place testing procedure very commonly used at our site for showing that a system is maintaining its  $4 \times 10^6$  decontamination factor (DF) system integrity.

BERGMAN: I am curious about your dilution system. We have been trying to develop a dilution system for a few years for a smoke

aerosol without a great deal of success. We find the particular system you have is highly sensitive to aerosol size; that is to say, the effectiveness of the dilution system varies with different particle sizes. Would you comment on your experience with this?

ELDER: We were primarily interested in the fairly narrow size range that the thermal generator puts out. What you are saying is quite true for a broadly dispersed aerosol. We would not use our type of diluter for aerosol experiments. But for this particular purpose, i.e., measuring upstream and downstream particle size distributions with the spectrometer (which is a very handy device for this), we find that the particle size characteristics do not change. We know we lost a lot of particles in the small-diameter tube, but it seems to have been a quite evenly distributed loss.

BERGMAN: If you lose particles, for example, by a factor of 10 due to diffusion while traveling through a very small orifice tube, this could have a dramatic effect on the diluter performance. Unless these things are checked in independent tests, you really don't know what the true dilution factor is. This omission is relatively common.

ELDER: You are right. The dilution factor comes from two things: first, mixing a small volume of air with a very large volume of air, and second, with particles being lost in the tube. However, if you calibrate on that basis, and are reasonably satisfied that you are not losing a lot of important particles, perhaps only large ones that you are really not interested in, then I think that it's a reasonable way to go. With the narrow size distribution that we had, it worked quite well.

JOHNSON: You mentioned something about DOE validation of this technique. I assume that would be a requirement coming down the line if you are seeking validation. Could you explain or expand a little more on that.

ELDER: The method that I was talking about was the earlier method developed by Schuster and Osetek. It does not involve fluorescence at all. The method was introduced as a DOE nuclear energy standard. I am sure that a lot of you are familiar with it. It is a way of making available the method and everything that we have learned about it to people who have an immediate use for it. That's basically why it is being done this way. As I said, it has already gone through two drafts. It is being voted on right now by the Technical Review Committee.

## 16th DOE NUCLEAR AIR CLEANING CONFERENCE

### REPORT OF MINUTES OF GOVERNMENT-INDUSTRY MEETING ON FILTERS, MEDIA, AND MEDIA TESTING

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Over the past score years an informal working group has been concerned with solving the problems of high efficiency filters and their testing procedures. The existence of this group has now spanned nine air cleaning conferences and has shown successive growth and participation at each one. From the original handful of participants and their open and often argumentative mode of operation, the sessions have progressed to an invited audience with a permanent chairman and a prepared agenda.

The most recent session of this working group was held on Monday, October 20, 1980, just preceding the Department of Energy (DOE) Nuclear Air Cleaning Conference at San Diego, California, and was devoted to a series of discussions on subjects of current interest. At this, the 16th conference, over 200 people assembled in the meeting room to participate in the deliberations. Although the major portion were from industry, various government agencies, academic and contract investigators, and over seven international groups were represented. This session, following the precedents of earlier meetings, related the operating requirements placed on the high efficiency filter and the capacity of industry to meet them. To this end, the collected talents of the assembled body were unified toward the problems of the particulate filter, its components, and its methods of test. Representatives of all facets of the industrial complex were present, from the basic fiber suppliers through media producers, and finally to the filter unit fabricators. Various test station and evaluation groups also contributed to the overall process. Research organizations from government laboratories and academic institutions contributed status reports on work currently underway. Users at various levels expressed their problems and actively participated in the discussions.

The following review may seem to be an agglomeration of information and show little continuity of subject. It is the intent to review, in abstract form, the items of deliberation. The items will be addressed in order of their discussion and not in priority. At the recent session, the following seven separate subjects were discussed.

#### Round Robin Paper Test Program

At the 15th conference in Boston an action item was assigned to Rockwell-Rocky Flats to conduct a blind round robin filter exchange test program to ascertain the accuracy and reproducibility of the Q127 filter media tester. W. L. Anderson of the Naval Surface Weapons Center reported on these results.

Eight separate media from six different suppliers were utilized in the comparison study. Media were supplied by Flanders, Dexter, Lydall, Mine Safety (MSA), Hollingsworth and Vose (H&V), and Cambridge. Nine test facilities were included in the evaluation: Air Techniques (ATI), Cambridge, Dexter, Harvard University, Lydall, MSA, H&V, Rockwell, and Porton (England). A total of over 800 data points were accumulated on each media for both dioctyl phthalate (DOP) penetration and air resistance using the Q127 instruments at the listed sites.

On the first round of testing, six of the nine facilities showed complete agreement for both penetration and resistance. Resistance values of  $\pm 0.5\text{mm}$  and penetration values of  $\pm 0.002\%$  were obtained in each case. The three units failing to demonstrate agreement were sufficiently different to indicate major test variants beyond human error in interpretation. These three sites were included in a second round of testing to pinpoint their potential sources of error. Results of this testing confirmed the earlier results and gave indicators as to discrepancy source. Subsequent follow-up showed that one site was using a nonstandard-sized sample holder and thus was testing at a much lower linear test velocity, giving low values for both penetration and resistance. A second site failed to utilize their particle size meter to adjust the aerosol size to the correct value and hence their DOP penetration values were low. The third site detected an error in their air flow measurement system and thus reported low values for both resistance and penetration. Recognition and subsequent correction of these deficiencies appeared to give comparable results for the three instruments in question. A third round of test samples was subsequently evaluated on all nine test devices and complete agreement was obtained.

The ultimate conclusions indicated that the overall agreement between the widely dispersed instruments were much better than expected and previously determined. It appears that some uncertainties exist in the operator area and that further training in operational procedures and calibration are required to maintain reproducibility on a continuous basis.

#### DOP Carcinogenicity

H. Ettinger of the Los Alamos Laboratory reported to the group on the recent developments of the possible carcinogenicity of DOP. DOP was assigned for potential carcinogen testing because of the large amount produced and the widespread exposure of the general population to products containing the substance.

Earlier reports by the National Cancer Institute (NCI) suggested that DOP was a potential carcinogen and a later position by the Environmental Protection Agency (EPA) in moving against production of six related materials gave further impetus to this position. This early study showed a random effect of DOP, but gave statistical evidence that DOP was a potential carcinogen and that further confirmatory data were required. A second study is now complete and confirms the earlier data.

This later study was conducted by feeding diets containing 6,000 or 12,000 ppm of DOP to groups of 50 male or female F344 rats and 3,000 or 6,000 ppm to groups of 50 male or female B6C3F1 mice for 103 weeks. Matched controls of untreated animals were also included. Mean body weights of dosed animals were lower than those of the controls. Hepatocellular carcinomas or neoplastic nodules in high-dose rats of either sex and in low-dose females and hepatocellular carcinomas or adenomas in low- and high-dose mice of either sex occurred at incidences significantly higher than those in matched controls. Degeneration of the seminiferous tubules and atrophy of the testes and hypertrophy of cells in the anterior pituitary were also detected at increased incidences in high-dose male rats. A comprehensive draft bioassay report was issued October 15, 1980.

NCI has taken no specific official position at this time but it is expected that an intelligence bulletin will be issued in the near future; it probably will be in an alert format, pointing out that some carcinogenic effect was noted from bioassay data. It is anticipated that additional data will be obtained and an official position determined at a later date. At this time, industry has not taken a position to challenge the data. However, once the alert is issued, industry is expected to question the information and require further clarification. It is expected that industry will examine the issue from the general use technology first in a high priority setting, man testing second, and material/system testing programs third. Recommended action items were to keep well informed on the changing regulations and communicate on a regular basis to keep all organizations informed. A further action was initiated to find a substitute DOP replacement material that could be adapted as a direct substitute in existing instrumentation.

#### DOP Substitute (Thermal)

B. Gerber of the Army Chemical Corps, Edgewood, Maryland, reported on their efforts to find a replacement for DOP in the thermal generation or homogeneous aerosol mode. A comprehensive literature search was conducted for the substitute and a variety of preliminary experimental tests conducted. Working with the 3M Company, a mass of comparison data has been accumulated to show that a member of the polyethylene glycol family showed the most promise for the thermal DOP replacement. Toxicity data, physical/chemical parameters, operational characteristics, cost, and availability were obtained for the glycol materials. The Army has essentially completed the toxicity data for submission to obtain use clearance and endorsement is expected.

It was recommended that polyethylene glycol 400 be accepted as a substitute for DOP in thermal generators. Action items included the resolution of the operating conditions for the glycol substitute and to get further evidence to explain the discrepancies and inconsistencies of the quick look data presented. Although preliminary tests showed comparable data on test units, some concern was expressed on potential humidity effects on particle size homogeneity and distribution.

DOP Substitute (Air)

A similar but separate issue involving the determination of a suitable DOP substitute for the air-operated (polydispersed) generators was discussed by Dr. M. First from Harvard University. A determination of the physical parameters of DOP, together with the particle size characteristics of air-generated DOP aerosols was reported. Comparable data were obtained on a variety of materials. A general description of the experimental apparatus was outlined and physical data of the resultant aerosols were presented. Of the materials investigated, corn oil, mineral oil, polyethylene glycol 200 and 400, and dioctyl sebacate appeared to give the most promising results. It was concluded that several of the reported materials would make suitable direct substitutes in the Laskin-type air-operated generators. Action items were established to obtain a toxicity clearance from the appropriate regulatory agency for any material selected and to get as much operational data with the substitute material as possible so that direct comparisons with the DOP data base can be accomplished.

Filter Materials in Corrosive Environments

G. Brassell from Rockwell, Rocky Flats, discussed the compatibility of absolute filter materials in corrosive operational environments. Presentations at previous meetings were related to survival of the filter media itself under the strong acid aerosol attack experienced during industrial-type operations at Rockwell. From these presentations, acid resistant media were specified that exhibited extended life under these harsh operating conditions. Discussions at this meeting were directed toward the sealant materials utilized in attaching the media to the unit frame. More specifically, the problems related to the use of polyurethane foam were addressed.

Technical data were presented showing the drastic effect of the acid environment on the foam sealant. Pictorial evidence was given to demonstrate the magnitude of the thermal degradation of the foam with eventual complete charring and destruction of the sealant interface. An accelerated test program was developed to confirm the operational results and to screen existing or proposed sealants. A strong recommendation was made that all polyurethane foams be excluded from specifications and use where the filter unit will be exposed to acid environments.

Fire Retardant Filter Frames

H. Gilbert reported to the assembly on the replacement program for fire-retardant plywood and particle board frame materials. This program was necessitated by the lack of material availability from the single commercial source as a result of a fire that eliminated their total production capacity. He reported on recent tests at Underwriter Laboratories (UL) on untreated plywood as well as a proprietary particle board that successfully passed their qualification/certification tests. The UL-approved use of these materials for frame construction is pending. During the discussion it was pointed out that the use of metal flame arrestors upstream materially decreases the heat load and fire impact on the filter unit.

Filter Unit Leakage

A potential problem of aerosol leakage through selected filter units was brought to the attention of the group by Livermore Laboratories (LRL). This group pointed out that during extensive scan tests of newly installed filter units, they had observed leakage points at the media-sealant-frame interface. Although present in different makes and types, an increased frequency appeared to be present in the superflow groups. LRL attributed the leak to a lack of or insufficient adhesive and/or improper application of adhesive in the media-frame areas. Pictorial evidence was presented to document their position. A casual look at the pictures seems to indicate a lack of flow (wick effect) of the adhesive into the media structure. LRL stated that they had been working diligently with vendors, and feel that additional and properly applied adhesive bonds will solve the problem and prevent reoccurrence. It was pointed out to LRL that the scan test is a very rigorous test and that overall performance is more realistic for general system application and use. The practice of applying excessive adhesive or spot patching of filter units is not an acceptable practice for nuclear applications.

Several items were briefly discussed in response to questions from the floor. One related to the expected life of a filter unit. It was the consensus of the users that this time element is dependent almost entirely upon the environment in which the unit must operate. It was stated that the life is dependent upon the accumulation of filtered material (resistance increase) or the loss of filtration function (physical degradation). Times as short as several weeks in caustic or harsh environments or as long as 20 years in relatively clean atmospheres have been observed. In general, the life is determined to be the length of time determined by the system that will allow adequate air flows at specified air quality.

A second question from the floor inquired about the present status of the reference laboratory concept. H. Gilbert stated that the existence of and the identification of a specific reference laboratory is being held in abeyance for the moment pending further investigation, and that when a site is selected the interested groups will be informed.

In conclusion, it should be reemphasized that this informal working group, with its diversified representation, provides a means for a comprehensive and expedient solution to the problems of the high efficiency filtration industry. The total effort has proven invaluable because it permits the surfacing and exposure of problems that might otherwise be lost in the quagmire of bureaucracy and management. The meetings are intended to be, and actually are, a working level distribution of data and expertise as well as a progress report of ongoing projects in the particle filtration areas. To this end, the group feels they have been successful and future sessions are contemplated.

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## ADDENDUM 1

### SELECTED POLYETHYLENE GLYCOLS AS "DOP" SUBSTITUTES

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#### Abstract

The recommendation is made that Polyethylene glycol (PEG) 400 be considered as a substitute for "DOP" in aerosol generators producing a polydisperse distribution for testing the integrity of filters and filter assemblies and for testing respirator fit. Further, the recommendation is made that pentaethylene glycol (PTAEG) and possibly hexaethylene glycol be considered as a substitute for "DOP" in aerosol generators thermally producing "monodisperse" aerosol for quality acceptance tests according to US Federal specifications and standards. The toxicology data base available on the polyethylene glycol family of chemical compounds is discussed and the conclusion is drawn that the probability of approval and acceptance as a non-hazardous substance in the filter and filter media test role is high. Data and analysis supporting PTAEG performance equivalent to "DOP" in the filter and filter media test role are given or referenced. Cost and availability of the substitute materials is discussed. Conclusions based on the present data and information are given and recommendations for further work are made.

#### Objectives

The objectives of the effort herein described were (a) to conduct preliminary investigations and analyses of the use of certain polyethylene glycols as substitutes for di(2-ethylhexyl) phthalate (commonly known as "DOP") in both polydisperse aerosol generators and thermal ("monodisperse") aerosol generators as respectively used to determine the integrity of filters, filter assemblies, and respirator fit and the quality of filter media and filters, (b) to acquire and analyze information on the cost and availability of such substitute materials and (c) to acquire and analyze the available toxicology data base on these materials in order to assess the probability of approval and acceptance as non-hazardous materials in these testing roles.

#### Background

On October 15, 1980, the National Cancer Institute published a draft National Toxicology Program Technical Report<sup>(1)</sup> on di(2-ethylhexyl) phthalate, commonly known as "DOP". The report presents bioassay data on rats and mice and indicates that "DOP" causes increased incidence of hepatocellular carcinomas or neoplastic nodules in rats and hepatocellular carcinomas or adenomas in mice. Should the Secretary, US Department of Health and Human Services, or his designee, (OSHA), classify "DOP" as a Category I potential carcinogen, then the model standard set forth in CFR 190.151<sup>(2)</sup> will apply. The standard will set the lowest feasible concentrations and time limits for inhalation, dermal and eye exposures to "DOP".

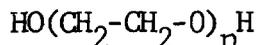
However, CFR 190.151 also states in paragraph (c)(1)(i) that, when it is determined by the Secretary that there are available substitutes for all uses or classes of users that are less hazardous to humans, the proposal shall permit no occupational exposure. The determination of the acceptability of substitutes includes consideration of availability, practicability, relative degree of hazard and the economic consequences of the substitution. It is therefore to be expected that use of "DOP" would be precluded for any specific role for which a less hazardous substitute is shown to be available and where practicability and

economic feasibility can be demonstrated. The Secretary must conduct public hearings on the establishment of any standard which is then published in the Federal Register. In the case of "DOP", it is reasonable to expect establishment of a quantitative standard and/or the acceptance of substitutes for use in filter testing by the summer or fall of 1981.

The present report addresses the possibility that members of the class of chemical compounds known as polyethylene glycols can substitute for "DOP" in the various modes of filter, filter media and respirator testing.

### Nature of the Polyethylene Glycols

The polyethylene glycol family has the following general formula:



where:  $n = 1, 2, 3, 4, 5, \dots$  defines a particular member of the family. The glycols are dihydroxy derivatives of the paraffins. Ethylene glycol ( $n = 1$ ) is the simplest member of the family. Ethylene glycol may be considered to be intermediate between the mono-hydroxy compound,  $\text{C}_2\text{H}_5\text{OH}$ , ethyl alcohol, and the trihydroxy compound, glycerol,  $\text{C}_3\text{H}_5(\text{OH})_3$ . As  $n$  increases, the members of the family change from liquids to waxy solids. The polyethylene glycols are prepared commercially by the condensation of ethylene oxide carried out in water or ethylene glycol made basic with sodium hydroxide. A mixture of polyethylene glycols results. In the United States, the major suppliers are the Union Carbide Corporation<sup>(3)</sup> and the Dow Chemical Company<sup>(4)</sup>. The polyethylene glycol mixtures are identified by their average molecular weight which is typically specified as 200, 300, 400, 600, 1000, etc. The polyethylene glycols are used in pharmaceutical preparations (ointments, lotions, suppositories, and tablet coatings) and cosmetics (lotions, creams, lip-stick, cake make-up, etc.). Some are used as direct food additives (coatings, flavorings). There are many other industrial and chemical uses<sup>(3,4)</sup>. The human ingestion or exposure to the polyethylene glycols has led to considerable study of their effects on biological systems.

The polyethylene glycols are presently under consideration by the US Army for use in generating smoke in which to conduct troop training exercises.

### Toxicology Data Base for Polyethylene Glycols

Table I is a summary checklist of available information on the toxicology data base of the polyethylene glycols. The information (with the exception of the current US Army studies) is available from the toxicology Information Response Center<sup>(5)</sup>. Other information sources are the literature of Union Carbide<sup>(3)</sup>, Dow Chemical Company<sup>(4)</sup>, and the Workplace Environmental Exposure Level Guide (Sept 1980)<sup>(6)</sup>.

The US Army ARRADCOM Chemical Systems Laboratory (CSL) has been investigating the inhalation toxicity of Polyethylene Glycol 200 (PEG 200) for some time. The experimental phase of the studies is complete as of October 1980. All bioassay studies should be complete and a report written by the spring of 1981<sup>(7)</sup>. So far it appears that the material will be judged suitable for use as a safe training smoke to which unprotected humans can be exposed using relatively high concentrations.

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Table I: Checklist Summary of Available Information, Polyethylene Glycol Family

Type of Information	Routes Other Than Inhalation	Inhalation Route
Acute Toxicity <sup>1</sup>	Some available information	In process by US Army CSL
Subacute Toxicity <sup>2</sup>	Some available information	In process by US Army CSL
Subchronic Toxicity <sup>3</sup>	Some available information	In process by US Army CSL
Chronic and Long-Term Toxicity <sup>4</sup>	Some available information	-
Human Tests <sup>5</sup>	Some available information	-

1. Acute Toxicity: Single exposure of up to 24 hours to a chemical by parenteral, oral, cutaneous, inhalation or other body routes of entry.
2. Subacute Toxicity: Any repeated exposure to a chemical for periods greater than 24 hours but usually within a time period of 21 to 30 days.
3. Subchronic Toxicity: Any repeated exposure to a chemical for periods greater than 30 days but usually for periods of 90 days or 13 weeks.
4. Chronic and Long-Term Toxicity:  
 Chronic: Any repeated exposure to a chemical for periods greater than 90 days but usually for periods of 1 or 2 years, i.e., 52 or 104 weeks.  
 Long-Term: Any repeated exposure to a chemical for periods greater than 104 weeks or for the lifespan of an animal species.
5. Human Tests: Any available information on human exposures.

Because of its intended use, the Army nominated polyethylene glycol (Chemical Abstract Services (CAS) Registry No. 25322-68-3) for carcinogenicity testing. On the basis of the available data however, the NCI Chemical Selection Working Group (CSWG) unanimously decided, in a meeting of June 28, 1979, that animal carcinogenicity testing was not required for the polyethylene glycols<sup>(8)</sup>. The NCI evaluation of the data base was performed by Dr. E. Weisburger.

In summary of this section, it is evident that an extensive published toxicology data base exists for the polyethylene glycols as a class of compounds. This data base will shortly (spring 1981) include detailed quantitative information on animal exposure via inhalation. Some longer term animal carcinogenicity studies.

have been performed in the past but no information considered adverse was found as of 1978.

### PEG Substitution in Polydisperse Aerosol Generators

"DOP" has been exclusively used to generate a polydisperse (relatively wide size distribution) aerosol for use in testing the integrity of clean rooms, clean benches, glove boxes, gas masks, HEPA filter banks and respirator fit.

At least two aerosol generation methods are in use. One method is based on the Laskin submerged nozzle as described by Echols and Young<sup>(9)</sup>. The liquid physical properties controlling the resultant particle size distribution are probably viscosity, surface tension and density. The liquid atomization process is very complex and dependent on nozzle geometry. A priori theoretical predictions for a particular nozzle cannot be made. Considering only liquid substitution and the same atomization system, a first approximation is that the average particle size will be related to the liquid properties through the Ohnesorge number, such that:

$$\bar{d} \propto \mu_L^2 / \rho_L \sigma_L$$

where:

$\bar{d}$  is the average drop size

$\mu_L$  is the liquid viscosity

$\rho_L$  is the liquid density

$\sigma_L$  is the liquid surface tension

All of the liquid polyethylene glycols have similar values for density (1.127) and surface tension (44.5). The Ohnesorge Number approximation therefore results in a selection based on essentially viscosity. Using this technique, PEG 400 ( $\mu_L = 105$ ) is the liquid which matches the Ohnesorge Number for "DOP". A sample of PEG 400 was sent to the Harvard Air Cleaning Laboratory for controlled quantitative comparison (particle size distribution) with other competitive candidate "DOP" substitutes in this generation mode. The preliminary results are reported by M. First in another paper in these proceedings (16th DOE Nuclear Air Cleaning Conference, San Diego, CA, Oct 1980). The preliminary results show that the count median diameter and the geometric standard deviation, as measured with an aerosol laser spectrometer<sup>(10)</sup> were in the acceptable range for a "DOP" substitute. It should be pointed out however that, if needed, adjustments of particle size could probably be made by blending with PEG 300 (for smaller size) or PEG 600 (for larger size).

The second mode of polydisperse generation consists of discharging a regulated quantity of liquid onto a heated area<sup>(11)</sup>. The liquid vapors are picked up by an inert carrier gas (nitrogen or argon) and condensed to a polydisperse aerosol. The liquid physical properties most important in this generation mode are probably vapor pressure and thermal stability. Corn oil, another candidate "DOP" substitute, decomposed in use but PEG 400 did not and is apparently an acceptable substitute<sup>(12)</sup> (The flash point for PEG 400 by closed cup test is in excess of 350°F.)

The impact of the hygroscopicity of the polyethylene glycols on the stability and reproducibility of the test aerosol has been questioned<sup>(13)</sup>. Data are available<sup>(3)</sup> showing that, at equilibrium, the particle size because of water absorption

might increase 1% at 25% PH, 4% at 50% RH, and 11% at 75% RH. This factor does not appear to be significant but final determination should be made by extensive user test.

In summary to this section, PEG 400 (or a blend chosen for precise particle size distribution control) so far appears to be a good physical candidate as a "DOP" substitute in polydisperse aerosol generators. It is inexpensive and easily available and appears to reproduce the performance of "DOP" used presently in this aerosol generation mode. The extensive available toxicology data base, so far showing no adverse data, generates a high degree of confidence that this class of compounds will allow unprotected human exposure to reasonable and practicable concentration-time profiles.

### Substitutes For Use in Thermal ("Monodisperse") Aerosol Generators

#### Standard Methods

High efficiency Particulate Air (HEPA) filters manufactured for the US Government are tested in accordance with the quality acceptance test procedures for filters and media contained in MIL Std. 282. The industry standard method for evaluation of air assay media is given in ASTM Std. D2986 and is essentially equivalent to the standard military test procedure for filter media.

In all cases, the standards require the generation of "DOP" test smoke by a thermal evaporation/condensation process. The design goal of the test generator is to produce a cloud of essentially monodisperse particles having a diameter of  $0.3 \mu\text{m}$  and a mass concentration of  $100 \pm 5 \mu\text{g/l}$ . The particle size is monitored by an NRL particle size meter (owl) (14). However, as shown by Hinds, First, et. al. (15), the NRL owl indicates an average size (weighted by the eighth power of diameter) if a distribution of particle sizes is introduced. Using a laser aerosol spectrometer, they showed that the Q-127 generator available to them could not achieve the design goal of  $\pm 0.03 \mu\text{m}$ . In terms of the geometric standard deviation (GSD) they were able to achieve 1.15 but not the 1.1 desired. Arthur D. Little, Inc., under contract (16) to the US Army to improve the Q-127, also achieved a  $\text{GSD} \approx 1.15$  with a very considered and sophisticated approach but could do no better except at low concentrations and flow rates. They developed and used a real-time measurement device (which uses a HeCd laser) which measures the GSD by observing the polarization ratio at two fixed scattering angles ( $84^\circ$  and  $116^\circ$ ).

The capability of the thermal generation process to produce monodisperse particles deserves further study. However, for the present purpose and as a first step, a candidate substitute material need only demonstrate comparable behavior in a side by side comparison with "DOP" in equivalent thermal generators. The ability to produce monodisperse particles is confounded with generator design and operation and complete resolution cannot be expected at this time.

The critical liquid physical properties controlling thermal generation are vapor pressure and thermal stability. The desired goal of monodispersity dictates the use of a relatively pure compound. In order to obtain a monodisperse aerosol, all particle growth (condensation) must start simultaneously, proceed at the same rate and arrive at the same final size. This is clearly impossible with a mixture of compounds having different vapor pressures. The candidate "DOP" substitutes of relevant interest are therefore the pure polyethylene glycols which are closest to "DOP" in vapor pressure (measured at some reasonable reference temperature). Vapor pressure/temperature data have been published for a series of pure glycols up

to tetraethylene glycol by Gallagher and Hibbert<sup>(17)</sup>. Figure 1 below is a plot of estimates made from their data for the temperature achieving 5mm Hg vapor pressure for members of the series. A linear projection appears reasonable and indicates that a match of "DOP" performance should lie between pentaethylene glycol as a lower bound and hexaethylene glycol as an upper bound.

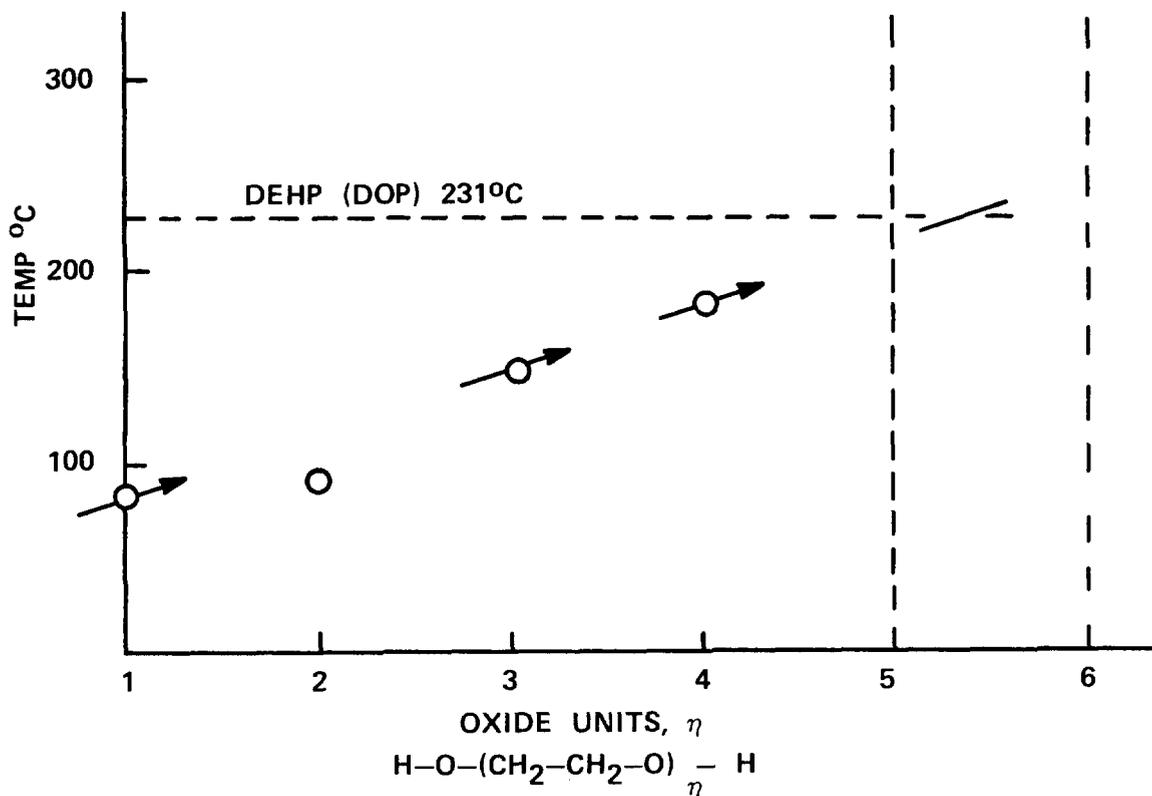


Figure 1: Oxide Units in Polyethylene Glycol vs. Estimated Temperature Achieving 5mm Hg Vapor Pressure

Either liquid should be useful if the appropriate generator controls can be varied to compensate for the differences. Pentaethylene glycol should make a test aerosol of acceptable concentration at a lower reservoir temperature than "DOP". Hexaethylene glycol will undoubtedly require a higher reservoir temperature. The count median diameter (CMD) and the geometric standard deviation (GSD) will be dependent on the control of the temperature gradients achievable in the condensation region of the specific generator in question.

Both pentaethylene glycol and hexaethylene glycol are difficult to obtain at present. Pentaethylene glycol purported to be 97% pure was procured from the Columbia Organic Chemical Company, Columbia, SC and used in the tests described herein. (Hexaethylene glycol was not received in time to be included in the tests.)

### Experimental Procedure

#### Preparatory Tests (Sept 23, 1980)

Tests were conducted with the cooperation and facilities of the Occupational Safety and Health Products Division of the Tape Group of the 3M Company, St. Paul, MN.

The quality assurance section there has had several years experience with the Q-127 type machine made by ATI, Inc., Baltimore, MD. They have many machines at various plant sites including those at the St. Paul laboratory. They test the filter media and their product line of disposable respirators as part of the R&D process and for quality control and acceptance in the manufacturing process.

Two side-by-side Q-127 type machines of very recent manufacture were used for the present work. The machine used for testing the pentaethylene glycol was drained of "DOP" while still hot and allowed to cool. The reservoir was then flushed with methyl-ethyl-ketone, drained and blown dry with compressed air. Next, the reservoir was filled with tetraethylene glycol and drained. The reservoir was then filled with pentaethylene glycol and brought up to 170°C. ("DOP" is generated at 167°C in the St. Paul laboratory). All other machine controls were set at the standard levels for "DOP". Dense smoke was immediately made but initial indications from the owl and the aerosol laser spectrometer were that the particle size was much larger than desired. It was therefore decided to chill the compressed air (incoming dew point - 20°C) to lower the temperature in the condensation region of the machine. This was accomplished by constructing a jury-rigged heat exchanger using a length of compressed air hose coiled into a 5 gallon pail containing dry ice. Modifications were made to the Q-127 to allow insertion of thermometers into both the vapor pick-up and diluent air lines. Although the expedient dry ice technique appears drastic, the temperatures resulting in the vapor pick-up and diluent air lines are not unreasonable and appear easily achievable by more conventional laboratory and plant practices. As a further precaution, the machine was again drained and refilled with fresh pentaethylene glycol. At a reservoir temperature of 170°C the concentration was found to be in excess of 185 µg/ℓ. (Concentration was measured conventionally by weighing a timed accumulation on an absolute filter pad with knowledge of the volume flow rate.) The reservoir temperature was therefore reduced and variations in flow rates and temperatures of the vapor pick-up and diluent air were tried in order to bring the apparent particle size (as indicated by the owl and the laser aerosol spectrometer) into the desired size range region.

#### Laser Aerosol Spectrometer

The Laser Aerosol Spectrometer Model ASAS-300A system made by Particle Measuring Systems, Inc., Boulder, CO (PMS) was used to determine particle size. The system had been calibrated the previous week with Dow polystyrene latex microspheres of known size. The validity of the calibration procedure and the insensitivity of the instrument to refractive index changes is discussed by Hinds, First, et. al.,<sup>(15)</sup> and by Knollenberg<sup>(18)</sup>. The ASAS-300 probe can measure particle diameter down to 0.15 µm in the lowest range. Procedures exist to merge the data when the measurements involve overlapping ranges.

For the present tests the instrument was modified for "in-line" sampling. The normal inlet horn was removed and replaced with a PMS supplied stream "focuser" with tubing connector. The normal integral suction fan was removed and replaced with a plug and tubing nipple. An aerosol "clean up" filter and rotameter was placed downstream of the instrument. A Gast Model 1531 vacuum pump was used to pull the smoke sample from the chuck of the Q-127 through the instrument train. Surgical tubing (3/8" OD) was used throughout for connections.

The validity of measurements using the laser aerosol spectrometer is very dependent on particle number concentration. A "by-pass" diluter was therefore used between the Q-127 chuck take off and the PMS instrument to achieve a count rate of about 100 counts per second. In the actual test, count data were

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accumulated for about 30 seconds. The "by-pass" diluter splits the sampled stream into two fractions. One of the fractions is filtered and then remerged with the unfiltered fraction. The count rate can be varied using throttling valves and controlling the fraction of the flow which is filtered in relation to the unfiltered fraction.

Proof Testing (Sept 24, 1980)

Stable operation of the Q-127 with pentaethylene glycol was achieved in the desired concentration range. Mass concentrations in the range of 72-113  $\mu\text{g}/\ell$  were achieved over the reservoir temperature range of 145-150°C. The operating conditions are shown in Table II.

Table II: Q-127 Conditions Using Pentaethylene Glycol

Reservoir Temperature .....	145-150°C
Temperature Vapor Pickup Air .....	~23°C
Temperature Diluent Air .....	~17°C
Flow Rate Vapor Pickup Air .....	~12 lpm
Flow Rate Diluent Air .....	~85 lpm
"Heater" Variac Setting .....	~34 div
"Particle Size" Variac Setting .....	~17 div
Owl Setting .....	28-29°

Three sets of representative pentaethylene glycol particle size measurements as made with the PMS ASAS-300A laser aerosol spectrometer system are given in Table III and compared with two sets of representative "DOP" data.

Table III: Particle Size Count Comparison  
Pentaethylene Glycol (PTAEG) vs. Di(2-ethylhexyl)  
Phthalate Using PMS ASAS 300A Instrument

<u>Instrument Channel</u>	<u>Instrument Range</u>	<u>Calibrated Midpoint Size, <math>\mu\text{m}</math></u>	<u>"DOP" A</u>	<u>"DOP" B</u>	<u>PTAEG A</u>	<u>PTAEG B</u>	<u>PTAEG C</u>
1	3	0.155	180	189	267	375	285
2	3	0.165	185	210	294	408	301
3	3	0.175	209	233	329	428	339
4	3	0.185	251	206	274	384	355
5	3	0.195	241	286	263	379	291
6	3	0.205	220	208	241	333	297
7	3	0.215	209	225	194	279	304
8	3	0.225	204	160	169	207	288

Table III: (Cont.)

9	3	0.235	197	180	133	203	255
10	3	0.245	170	180	150	139	207
11	3	0.255	166	189	104	176	182
12	3	0.265	138	125	97	113	154
13	3	0.275	131	122	70	75	135
14	3	0.285	108	110	40	65	101
15	3	0.295	126	110	54	51	89
4	2	0.318	68	77	16	21	12
5	2	0.343	54	60	8	10	13
6	2	0.368	36	43	5	6	7
7	2	0.393	19	34	2	3	3
8	2	0.418	12	14	1	2	1
9	2	0.443	5	7	0	1	0
10	2	0.468	7	6	0	0	0
11	2	0.493	3	7	0	1	0
12	2	0.518	4	4	0	0	0
13	2	0.543	1	4	0	0	0
14	2	0.568	2	2	0	0	0
15	2	0.593	2	1	0	0	0

### Particle Size Data Reduction

The PMS ASAS 300A probe can only sense particle diameter as low as 0.15  $\mu\text{m}$ . It is evident from Table III that the size distribution was not completely determined since the instrument was not capable of measuring the lower end of the size spectrum. Initially, an heuristic method was developed to try to infer the missing data. That method was based on the premise that the mode of the distribution was contained in the measured data. It was clear that calculations based on the truncated data overestimated the count median diameter (CMD) and underestimated the geometric standard deviation (GSD). This is to be expected when the contribution of the smaller particle diameters are not included in the population. It is of interest to note that a linearized graphical estimation procedure using log-probability paper was more reliable in estimating the GSD than a purely computational procedure. The linearized graphical procedure is more satisfactory than computation when the distribution deviated from the assumption that the logs of the diameters are normally distributed. The standard computational procedure gives all points equal weight whereas in the linearized graphical procedure one ignores the deviations at the tails of the distributions. (The need for a weighting factor scheme similar to that used in some types of statistical bioassay is apparent.)

A search of the statistical literature uncovered the Pearson-Lee-Fisher (PLF) method (19) of extrapolating singly truncated normal distributions. The method was validated and programmed for the present purpose for the computer by Pennsyle (20).

Table IV presents the results of computation for the two sets of "DOP" size data and the three sets of PTAEG size data. The CMD and the GSD are given for both the measured truncated population (raw data) and the P-L-F corrected estimation.

Table IV: Count Median Diameter (CMD) and Geometric Standard Deviation (GSD) Estimates

Test	Based on Raw Data		P-L-F Corrected	
	<u>CMD</u> μm	<u>GSD</u>	<u>CMD</u> μm	<u>GSD</u>
"DOP" A	0.222	1.26	0.210	1.31
"DOP" B	0.223	1.27	0.206	1.34
PTAEG A	0.202	1.20	0.188	1.26
PTAEG B	0.200	1.20	0.186	1.26
PTAEG C	0.209	1.21	0.201	1.24

Owl Interpretation of Particle Size Distribution

Hinds, First, et. al. (15) showed that the polarization-ratio owl, when challenged with a distribution of particle sizes, indicates a weighted average particle size. The specific polarization ratio was estimated to vary with particle diameter to the 8.1 power. A numerical integration scheme was employed to compute values for the average polarization ratio, the components of which were calculated from the Mie theory. They showed that, for a true log-normal distribution, an infinite number of GSD-CMD pairs would yield an owl setting of 29°. However, it can be shown for log-normal distributions of particle size that simple relationships exist between the weighted average particle size and the CMD and GSD(21). In particular, the average particle size weighted to the eighth power is related to the CMD and GSD as follows:

$$\ln \bar{d}^8 = \ln \left[ \frac{\sum n_j d_j^8}{\sum n_j} \right]^{\frac{1}{8}} = \ln [\text{CMD}] + 4 \ln^2 [\text{GSD}] \tag{1}$$

Table V shows the relationship between the GSD and CMD pairs used by Hinds, First, et. al. (15), (all of which yield an average particle size ( $d_{PR}$ ) of 0.3 μm "seen by the owl at setting of 29°) and the value of  $\bar{d}^8$  computed from equation (1) using the same pairs.

Table V:  $\bar{d}^8$  vs.  $d_{PR}$

<u>GSD</u>	<u>CMD</u>	$\bar{d}^8$	<u><math>d_{PR}</math></u>
1.00	0.300	0.300	0.300
1.05	0.294	0.297	0.300
1.10	0.286	0.297	0.300
1.15	0.274	0.296	0.300

Table V: (Cont.)

1.20	0.260	0.297	0.300
1.25	0.246	0.300	0.300
1.30	0.230	0.302	0.300

It is concluded that the weighted average  $\overline{d^8}$ , gives results practically equivalent to  $d_{\overline{PR}}$  and may be used to infer the average size "seen" by the owl when sampling a polydisperse aerosol whose diameters are log-normally distributed. Table VI gives the estimations made for the two "DOP" tests and the three PTAEG tests previously tabulated:

Table VI: Average Particle Size  $\overline{d^8}$  "Seen" by Owl  
P-L-F Corrected

<u>Test</u>	<u>CMD</u> $\mu\text{m}$	<u>GSD</u>	<u><math>\overline{d^8}</math></u> $\mu\text{m}$
"DOP" A	0.210	1.31	0.281
"DOP" B	0.206	1.34	0.290
PTAEG A	0.188	1.26	0.233
PTAEG B	0.186	1.26	0.230
PTAEG C	0.201	1.24	0.242

#### Filter Penetration Comparisons

Several disposable half-face respirators manufactured by the 3M Company were tested on the side by side Q-127 machines respectively filled with pentaethylene glycol and "DOP". Two glass filter pads were also tested. The results are shown in Table VII.

Table VII: Comparative % Penetration Tests

<u>Item</u>	<u>"DOP" % PEN.</u>	<u>PTAEG % PEN</u>
Respirator # 1	24	23
Respirator # 2	32	27
Respirator # 3	10	10
Respirator # 4	10	10
Respirator # 5	1.27	1.1
Respirator # 6	24	24
Respirator # 7	1.37	1.24
Respirator # 8	1.26	1.11
Respirator # 9	4.44	4.0
Glass Fiber Filter A	3.7	3.1
Glass Fiber Filter B	0.08	0.06

Tetraethylene Glycol

Tetraethylene glycol is commercially available and relatively inexpensive. Time did not allow testing it during the program of September 23-24. However, it was later determined (22) that it would not make aerosol within the operating range of the Q-127 with any control over particle size and concentration.

Discussion of Results

Although additional tests are required before unequivocal conclusions can be drawn, it appears that pentaethylene glycol (PTAEG) shows good promise as a substitute for "DOP" in the thermal generation mode. Use of neither "DOP" nor PTAEG in the Q-127 machines at the 3M Company in St. Paul achieved the design goals of a count median diameter of 0.3  $\mu\text{m}$  and a geometric standard deviation of 1.10. The lowest geometric standard deviation reported by others as achievable in the Q-127 is 1.15. However, the lowest (best estimate) during this test series was 1.24 using PTAEG (Table IV). The 3M Company standard operating procedures are scrupulous in attention to detail. The machines are even kept in a temperature controlled environment at 72°F. The compressed air is oil-free and has a dew-point of -20°C. Passing the compressed air through a particulate filter rated to retain sub-micron particles does appear to affect the resultant particle size distribution (23). Means of controlling the GSD, therefore, remain a mystery. It is conceivable, however, that lack of control of condensation nuclei too small to be filtered by the particulate filters normally employed for compressed air service cause a variation in particle growth rate which then affects the GSD. The only known way of removing these nuclei is to condense liquid upon them and cause them to grow to a physically removable size. This is demonstrated in variable volume cloud chambers by saturating with water vapor and then cooling by sudden expansion of the volume. The water droplets condense on the nuclei and are removed by settling and/or filtration. The process is repeated ("pumping") until no nuclei are present. It is conceivable that this nuclei removal process or an equivalent process applied to the compressed air used for vapor pick-up and diluent air in the Q-127 might resolve the GSD problem. Control of the CMD, on the other hand, is much easier. Covariation of the mass concentration (reservoir temperature) and the temperature gradient in the condensation region of the Q-127 machine can usually result in the desired owl reading. However, the GSD is also affected by this procedure and without a real-time GSD "meter" one would not be aware of this. It is conceivable that with a real-time meter the GSD might be improved through proper setting of the Q-127 controls. The Arthur D. Little GSD meter (16) would be a very valuable addition to the Q-127 system for such a purpose. The manual owl version allowing observation over a scan of scattering angles to pick up the Higher Order Tyndall Spectra (24) produced by monodisperse aerosols might also serve the purpose. It would be slower in use than the A. D. Little instrument but would be much cheaper to make. Conceivably its operation could also be made faster by automating the coupled electro-optical and mechanical functions.

In perspective, it appears that the pentaethylene glycol shows excellent promise of substituting for "DOP" in thermal generation. In the side-by-side comparison with "DOP" the GSD was smaller and although the CMD was also smaller than desired, no difficulty is anticipated in increasing it. The limited schedule available for this program simply did not permit further work at this time and future tests are planned to acquire the additional information. The results of the side-by-side filter penetration tests are encouraging. There is an unexplained enigma, however, in that the smaller average particle size of the PTAEG always gave a somewhat lower penetration value than the larger average particle size of the "DOP". The only explanation so far conceived is the possible existence of a

bimodality (skewed toward larger particles) in the particle size distribution which might not have been observed with the laser aerosol spectrometer. It would be very interesting to check the particle size distribution with another type of instrument having a larger range such as the quartz crystal micro-balance cascade impactor (25).

From a qualitative point of view, PTAEG is relatively odorless even when hot. "DOP" is odorless when disseminated cold but has a distinctive odor when disseminated hot. The smoke observed in the owl seemed brighter than "DOP" smoke at equivalent concentrations. Changes in the aerosol characteristics (size and concentration) seemed very responsive to changes in the Q-127 control settings. Except for an initial stabilization period (in which impurities were perhaps volatilized and purged), the PTAEG operation appeared more stable over time than the "DOP" operation. One unknown factor at this time is the stability of PTAEG under prolonged heating in the reservoir. The initially water-white material was slightly yellow when looked at after two days of operation. This must be investigated for longer periods in future tests.

#### Cost and Availability

Neither pentaethylene glycol nor hexaethylene glycol is easily available at present. PEG 200 which could be the feed-stock source of the pure compounds is available and very inexpensive (about \$7/gallon in 55 gallon lots). Pentaethylene glycol represents 21% of the total weight in PEG 200 but requires extreme measures such as high vacuum distillation to effect separation. The Inland Vacuum Company of Rochester, NY, made a preliminary feasibility study of high vacuum distillation for recovery of PTAEG. They reported (26) that about 25% of the 21% PTAEG in PEG 200 might be recovered. However, two passes through their equipment are required at a feed rate of about two gallons/hour. Product purity has yet to be determined. Taking all factors into consideration, they believed that they could initially supply PTAEG at a cost of \$500/gallon but that further process refinement and larger scale operation might conceivably lower the cost to the neighborhood of \$200/gallon (27).

An alternative process which may be more efficient and thus faster is plant-scale liquid chromatography (28). PEG 200 has been supplied to the domestic representative of the Elf Aquitaine Company of France for a feasibility determination and preliminary cost estimate. Results are not yet available.

Another avenue not yet explored is the possibility that pentaethylene or hexaethylene glycol might be produced by some optimized chemical reaction which would enable simple and inexpensive purification. Further exploration of this possibility is required.

In perspective, one must note that total present usage in the US of "DOP" for filter media and filter quality acceptance testing is probably less than 5000 gallons/year. Considering the limited alternatives (expensive ventilation equipment, personnel protection, environmental pollution control equipment) the additional operating cost may not be unreasonable. The cost of labor to operate and support the quality control and acceptance facilities will probably be a much higher element of operating cost than the cost of the pentaethylene glycol.

It should be noted that the comments made above are relevant only to operation of thermal generation ("monodisperse") equipment. The PEG 400 recommended for polydisperse aerosol generators is relatively inexpensive and cost should not be an important factor.

Conclusions

1. Based on the available toxicology data base for the polyethylene glycols, the probability appears high that PEG 400 and pentaethylene glycol (PTAEG) could receive approval by the Secretary, US Department of Health and Human Services or his designee (OSHA) as substitutes for "DOP" in the various filter, filter media, and respirator test roles.
2. Based on comparative tests performed (Sep 80) at Harvard Air Cleaning Laboratory, it appears that PEG 400 or a related mixture or blend acceptably matches "DOP" performance in polydisperse aerosol generators employing submerged atomization nozzles.
3. Based on comparative tests performed (Sep 80) at the 3M Company, it appears that PTAEG acceptably matches "DOP" performance, controllability, and stability in the Q-127 thermal generator and filter penetration measurement system.
4. PEG 400 and related mixtures or blends are relatively inexpensive (about \$7/gallon) and widely available.
5. PTAEG is not easily available at present.
6. Although the present estimated cost of producing PTAEG is high (\$200-\$500/gal) its use might not be unreasonable considering the limited and expensive alternatives known at present.
7. Alternative production methods for PTAEG have been conceived and may result in a more reasonable cost.
8. Tetraethylene glycol which is widely available and relatively inexpensive could not be made to work in the Q-127.

Recommendations

1. Investigation of the use of pentaethylene glycol (PTAEG) as a "DOP" substitute should be expanded and continued. In particular the effects of continuous heating should be determined.
2. The need for "monodisperse" filter and media testing should be re-examined. New specifications and standards should be developed as appropriate.
3. New equipment should be developed to implement new standards as appropriate. Shortcomings of present equipment; e.g., continuous heating, should be eliminated.
4. If "monodisperse" testing continues to be the standard method, efforts should be made to determine the causes of GSD variations and control them with the hopeful outcome of achieving a GSD closer to 1.1.
5. If "monodisperse" testing continues to be the standard method, real-time GSD meters should be incorporated into the test systems.
6. The possibilities of producing PTAEG at lower cost should be intensively explored.

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7. The capabilities of hexaethylene glycol as a "DOP" substitute should be explored.
8. Requests should be submitted to the Secretary, US Department of Health and Human Services and to OSHA asking approval for the use of PEG 400 and PTAEG in their respective test roles.

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## ADDENDUM 2

### SIZE DISTRIBUTIONS OF AEROSOLS PRODUCED FROM SUBSTITUTE MATERIALS BY THE LASKIN COLD DOP AEROSOL GENERATOR

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#### Abstract

Test aerosols of di(2-ethylhexyl)phthalate (DOP) produced by Laskin nozzle aerosol generators are widely used for in-place filter testing and respirator fit testing. Concern for the health effects of this material has led to a search for substitute materials for test aerosols. Aerosols were generated with a Laskin generator and diluted 6000-fold with clean air. Size distributions were measured for DOP, di(2-ethylhexyl)sebecate, polyethylene glycol, mineral oil, and corn oil aerosols with a PMS ASAS-X optical particle counter. Distributions were slightly bimodal with count median diameters from 0.22 to 0.30  $\mu\text{m}$ . Size distributions varied little with aerosol material, operating pressure, or liquid level. Mineral oil and corn oil gave the best agreement with the DOP size distribution.

#### Introduction

The Laskin aerosol generator is used to produce test aerosols of di(2-ethylhexyl)phthalate (DOP) for respirator fit testing, in-place filter testing of nuclear and clean room facilities and biological safety cabinets, and to produce theatrical fogs. The aerosol produced for these applications is a high concentration (about 0.5  $\text{g}/\text{m}^3$ ) submicrometer, stable aerosol. The generator uses a submerged compressed air Laskin nozzle<sup>(1)</sup> having four jets that require a total flow rate of 75 L/min at 140 kPa (20 psig). All of the applications cited above may involve some exposure of personnel to submicrometer aerosols of DOP. Recent studies at NCI have found an association between exposure to 3,000 to 12,000 ppm DOP in the diet and the development of hepatocellular carcinomas or neoplastic nodules in mice and rats.<sup>(1)</sup> Concern about the potential health effects to people working with DOP test aerosols has led to a search for substitute materials.

Over 180 million kilograms of DOP are produced each year in the United States, 95% of which is used as a plasticizer primarily for PVC plastics. The properties of DOP that make it useful as a plasticizer, e.g., low vapor pressure, chemical stability, and insolubility in water, also make it a desirable material for use as a test aerosol. DOP is a material of low acute toxicity and prior to recent findings of its carcinogenic activity was considered an ideal test aerosol material.

The objective of this study was to compare the size distributions of aerosols produced from substitute materials by the Laskin nozzle aerosol generator with that produced from DOP. The substitute materials were chosen to match the properties of DOP as closely as possible. Properties considered were density, viscosity, non-hygro-

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scopcity, flash point, refractive index, surface tension and commercial availability. The substitute materials tested were corn oil, di(2-ethylhexyl)sebecate (DOS), mineral oil, and polyethylene glycol. Relevant properties of DOP and the substitute materials are summarized in Table 1.

The DOP was manufactured by the Hatco Chemical Corporation, Fords, N.J. It has FDA approval for use in polymers that contact food of high water content. The corn oil was a laboratory grade equivalent to USP or NF, obtained from Fisher Scientific Co., Medford, MA. The DOS was manufactured by Union Camp Corp., Wayne, N.J. under the designation Uniflex DOS. It meets requirements of CFR 176.180 and 176.170 for use as a food additive. The mineral oil was ARCoprime 200 manufactured by the Atlantic Richfield Company, Houston, TX. It is a white paraffinic-base food grade mineral oil that meets FDA food additive regulations, sections 122.1146 and 121.2589(a) and meets USP and NF listings. Polyethylene glycol was manufactured by Union Carbide Corporation, New York, N.Y., under the designation PEG 400. It is soluble in water and hygroscopic.

The size distribution of the aerosol produced by the Laskin nozzle aerosol generator has been measured previously by two investigators. A report by Echols and Young<sup>(3)</sup> gives the dimensions of the Laskin nozzle and describes the characteristics of a six nozzle generator that was the prototype for commercial DOP generators. They measured the size distribution of the DOP aerosol produced by this generator with a combined light scattering and impactor method and found the light scattering median size at 175 kPa (25 psig) to be 0.82  $\mu\text{m}$ . The best fitting log normal distribution, based on data between 10 and 90 cumulative percent, had a GSD (geometric standard deviation) of 1.4. The light scattering median diameter (LSMD) decreased from 0.94 to 0.80  $\mu\text{m}$  and GSD decreased 1.44 to 1.37 as operating pressure was increased from 70 to 210 kPa (10 to 30 psig).

Ettinger, et al.<sup>(4)</sup> measured the size distribution of the aerosol produced by the Echols and Young generator using three grades of DOP. They measured the distribution by count using a Royco model 200 optical particle counter. Their instrument classified particles according to their light scattering properties into seven categories over the range 0.3 to 1.5  $\mu\text{m}$ . They reported the CMD (count median diameter) and GSD of the best fitting log normal distribution based on a nonlinear least squares technique. An industrial grade of DOP used at a generator pressure of 70 kPa (10 psig) gave a CMD of 0.83  $\mu\text{m}$  and a GSD of 1.65 in a single nozzle generator. Octoil, a pure DOP, gave a CMD of 0.54  $\mu\text{m}$  and a GSD of 1.43 under the same conditions, and 0.56 CMD and 1.43 GSD for the six nozzle generator. Changes in the size distribution parameters were small over the range of operating pressures of 35 to 170 kPa (5 to 25 psig) and there was no clear pattern to these changes.

### Experimental System

Our test aerosols were generated with a single Laskin nozzle manufactured to the specifications given by Echols and Young<sup>(3)</sup>. Three of the four compressed air jets were blocked off with solder to reduce the air flow and mass output of the generator to one fourth

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Table 1. Properties of DOP and Substitute Materials.

Material	Specific Gravity	Viscosity	Refractive Index
Di(2-ethylhexyl)phthalate Hatcol DOP	0.983	82 cps @ 20°C	1.485
Di(2-ethylhexyl)sebacate Uniflex DOS	0.915	17.4 cps @ 25°C	1.448
Mineral Oil Arcoprime 200	0.8608	52.4 cps @ 100°F	1.471
Corn Oil Fisher USP	0.918	-	1.464
Polyethylene glycol Union Carbide PEG 400	1.127	105 cps	1.455

Table 2. Effect of Immersion Depth of Laskin Nozzle on Particle Size Distribution of DOP Aerosol.

Immersion Depth (above collar) cm	CMD μm	$\frac{D_{84\%}}{D_{50\%}}$ *
0.64	0.25	1.59
1.3	0.25	1.58
1.9	0.25	1.58
2.5	0.26	1.58

\*84% percentile size divided by the 50% percentile size; approximately equal to the geometric standard deviation (GSD).

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the normal value. The nozzle was mounted off-axis in a 14 cm diameter lucite cylinder, 24 cm high, shown in Fig. 1. The aerosol flowed through a vertical splash partition and out the top via  $\frac{1}{2}$  inch copper pipe fittings. The generator air flow rate was 21 L/min at 140 kPa (20 psig). The generator output to the dilution system was reduced still further by diverting approximately 18 L/min to exhaust, via a 20 x 25-cm filter, leaving a 3 L/min aerosol stream to travel to the dilution system.

The dilution system shown in Fig. 2 consists of a test loop of 20 cm (8 in) diameter pipe with a 25 x 25 cm square cross section test section. The 3 L/min aerosol stream was introduced into a flow of 0.35 m<sup>3</sup>/s (730 cfm) of clean air along the duct centerline. The combined streams passed through a mixing section containing a centrally located mixing disk having approximately one half the cross sectional area of the duct. This system provided an overall dilution of 6000 to 1. The system flow rate was monitored by an orifice meter which was calibrated by a hot wire anemometer traverse of the test section.

The aerosol was sampled isokinetically into an ASAS-X optical particle counter manufactured by Particle Measuring Systems, Inc., Boulder, CO. The distance from the sampling probe inlet to the sensing volume of the optical particle counter was about 30 cm and the sampling flow rate was 1.0 cm<sup>3</sup>/s. The optical particle counter uses a parabolic mirror to collect all the light scattered over an angular region of 35° to 120°. The sensing volume is approximately 4 x 10<sup>-6</sup>cm<sup>3</sup>. The system yields particle size by count in 45 contiguous channels over the range of 0.09 to 3.0  $\mu$ m. The width of the size channels ranges from 0.007  $\mu$ m for the smallest sizes to 0.160 for the largest sizes. The accumulated information in all 45 channels is transferred every 10 seconds to a Wang 2200 minicomputer for data analysis, calculation, and output.

The size calibration of the optical counter was checked with four sizes of polystyrene latex spheres that had been measured by Porstendorfer and Heyder<sup>(5)</sup> and found to have diameters of 0.151, 0.206, 0.318, and 0.494  $\mu$ m. Particles were nebulized from dilute water suspensions with a DeVilbiss number 40 nebulizer and passed through the dilution and sampling system described above but at a lower dilution air flow. For each calibration aerosol the measured size distribution showed a pronounced peak within  $\pm 10\%$  of the size found by Porstendorfer and Heyder.

### Results

A preliminary set of DOP size distribution measurements was made to determine how much dilution was required to provide negligible coincidence error in the optical particle counter. Size distributions were measured at five dilutions to give aerosol concentrations of 10<sup>3</sup> to 10<sup>5</sup> particles/cm<sup>3</sup>. The effect of concentration on CMD is shown in Fig. 3. The measured CMD increases gradually as aerosol concentration increases above 10<sup>4</sup>/cm<sup>3</sup>. To minimize coincidence error, all subsequent measurements were made at a number concentration of 3500  $\pm$  500 cm<sup>3</sup>. This concentration corresponds to a theoretical coincidence loss of about 1%(6).

Another preliminary set of measurements was made to determine the effect of the immersion depth of the nozzle on the particle size distribution. Echols and Young<sup>(3)</sup> recommended that the liquid feed collar be immersed to a maximum depth of 2.5 cm. The DOP size distributions were measured for nozzle immersion depths of 0.64, 1.3, 1.9, and 2.5 cm. The results given in Table 2 show little variation and no trend of the particle size distribution with immersion depth. All subsequent measurements were made at a nozzle immersion depth of 2 cm.

One set of measurements was made with a standard four jet nozzle to determine if the use of only one jet modified the size distribution. No difference was found between the two and subsequent measurements were made with the modified single jet nozzle.

Mass distributions were determined by numerical integration of the count data. In all cases, the calculated mass distribution, shown in Fig. 5, was slightly bimodal. For comparison purposes, size data are reported as CMD, MMD, and the ratio of the 84 percentile diameter to the 50 percentile diameter (CMD). Aerosol size measurements were made at four pressures, 70, 105, and 175 kPa (10, 15, 20, and 25 psig). Results are summarized in Table 3 and Fig. 6.

#### Discussion

In general, the substitute materials gave the same size distribution as DOP within  $\pm 20\%$ . All showed a trend toward a decrease in CMD with increasing operating pressure. In part, this is because high operating pressures resulted in a greater air flow rate through the generator and there was less time for coagulation prior to dilution. The CMD's in Table 3 and Fig. 6 have been corrected for this coagulation effect. Monodisperse coagulation theory was used to estimate the CMD at the time of formation. The corrections were on the order of 10%. Of the materials tested, the mineral oil and corn oil gave the closest match to the size distribution of DOP. Mineral oil is also the closest match in refractive index and has a flash point only slightly lower than DOP, i.e., 410°C versus 420°C.

These comparative results give CMD values that are substantially smaller than reported previously. The reasons for this difference have to do with the measurement methods of previous investigators, which were insensitive to the particle size range below 0.3  $\mu\text{m}$  where the greatest number of particles are. The method used by Echols and Young<sup>(3)</sup> relies on measurements of light scattering of the aerosol stream after aerodynamic separation by impaction. Light scattering decreases rapidly with particle size for particles less than 0.5  $\mu\text{m}$ .<sup>(7)</sup> For example, a similar type of photometer gives a response that is 170 times greater for a 0.5  $\mu\text{m}$  particle than for a 0.2  $\mu\text{m}$  particle. This leads to an underestimation of the contribution of these small particles and a LSMD that is substantially greater than the CMD.

Ettinger, et al.<sup>(4)</sup> measured the size distribution of the DOP aerosol with an instrument that did not respond to particles less than 0.3  $\mu\text{m}$ . As shown in Figs. 4 and 5, approximately 70% of the particles are less than this size. Furthermore, the optical particle counter that was used required a 700,000:1 dilution to eliminate the effect of coincidence error. From the qualitative description of

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Table 3. Size Comparison of Generator Output at 140 kPa (20 psi).

Material	CMD	$D_{84\%}/D_{50\%}$ *	MMD
DOP	0.25 $\mu\text{m}$	1.53	0.70 $\mu\text{m}$
DOS	0.22	1.53	0.57
Min. Oil	0.27	1.52	0.70
Corn Oil	0.25	1.53	0.77
PEG	0.30	1.48	0.83

\*84% percentile size divided by the 50% percentile size; approximately equal to the geometric standard deviation (GSD).

the dilution procedures used, it seems likely that a dilution of this order was not achieved and their method overestimated particle size.

Because of the very low vapor pressure of the materials used,  $6 \times 10^{-8}$  mm Hg for DOP, a significant size reduction due to evaporation could not have occurred during the few seconds required to travel from the generator to the optical particle counter.

Observation of the very large numbers of microscopic bubbles that were formed in the generator reservoir suggests that there were two mechanisms of aerosol production, 1) conventional nebulization, i.e., shearing of a liquid filament drawn through the liquid feed collar hole by the low pressure of the high velocity air stream issuing from the radial jet and 2) bursting of large numbers of fine bubbles at the liquid surface. A separate experiment was conducted to determine the relative importance of these two mechanisms. The nozzle was operated in the normal way but with the liquid feed collar hole blocked so that the first mechanism would be inoperative. No significant difference in the size distribution was found between the two operating modes. Echols and Young<sup>(3)</sup> found that increasing the size of the liquid feed holes increased the mass output of the generator so conventional nebulization must be responsible for the larger particles that contain most of the mass. The bursting bubbles are apparently responsible for the smaller particles that represent most of the numbers. All the materials showed similar bubble production in the reservoir except for the PEG which produced fewer bubbles and also produced the largest particle size distribution.

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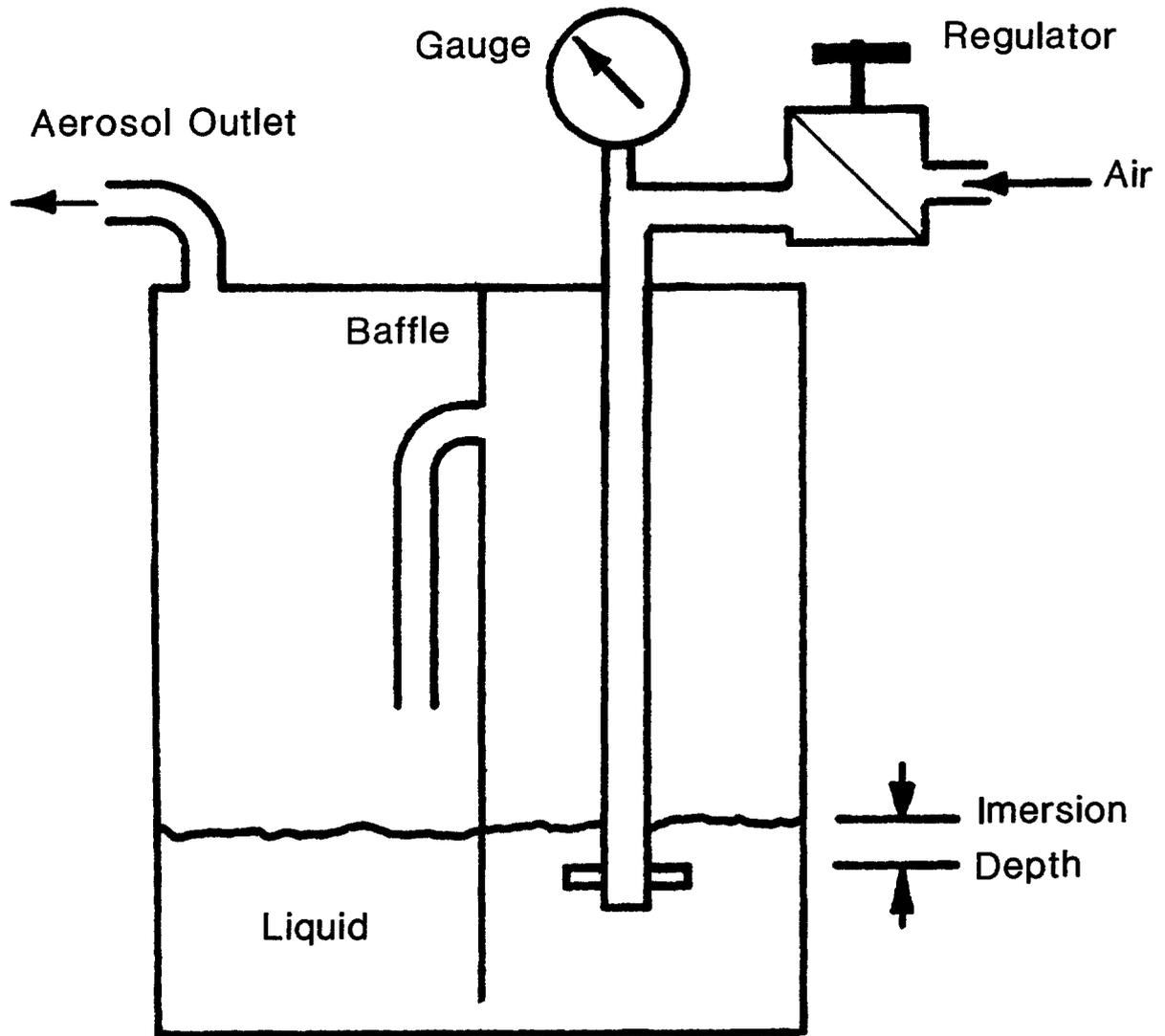


FIGURE 1a. A Crossection of the Laskin Nozzle Aerosol Generator

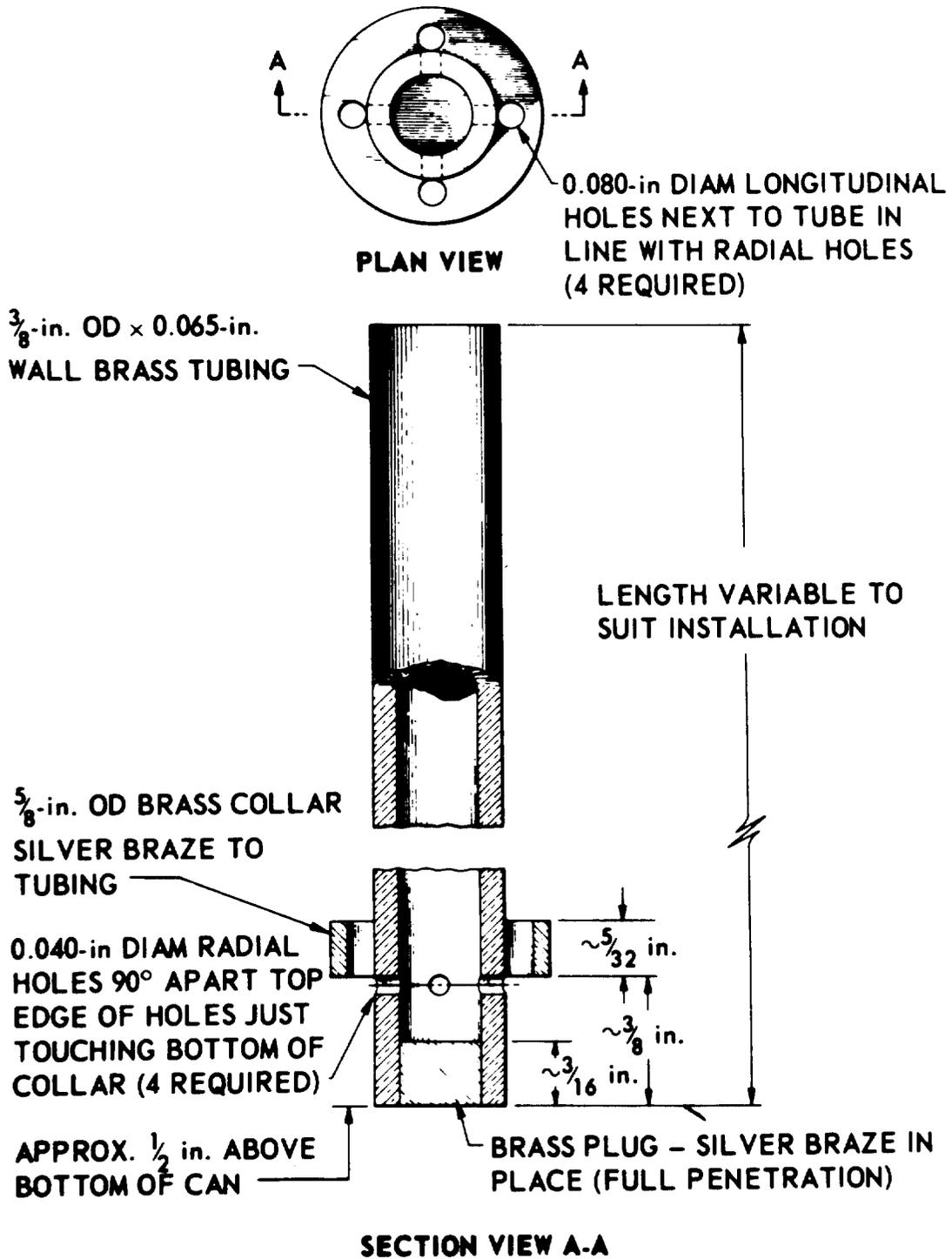


FIGURE 1b. Details of Laskin Nozzle

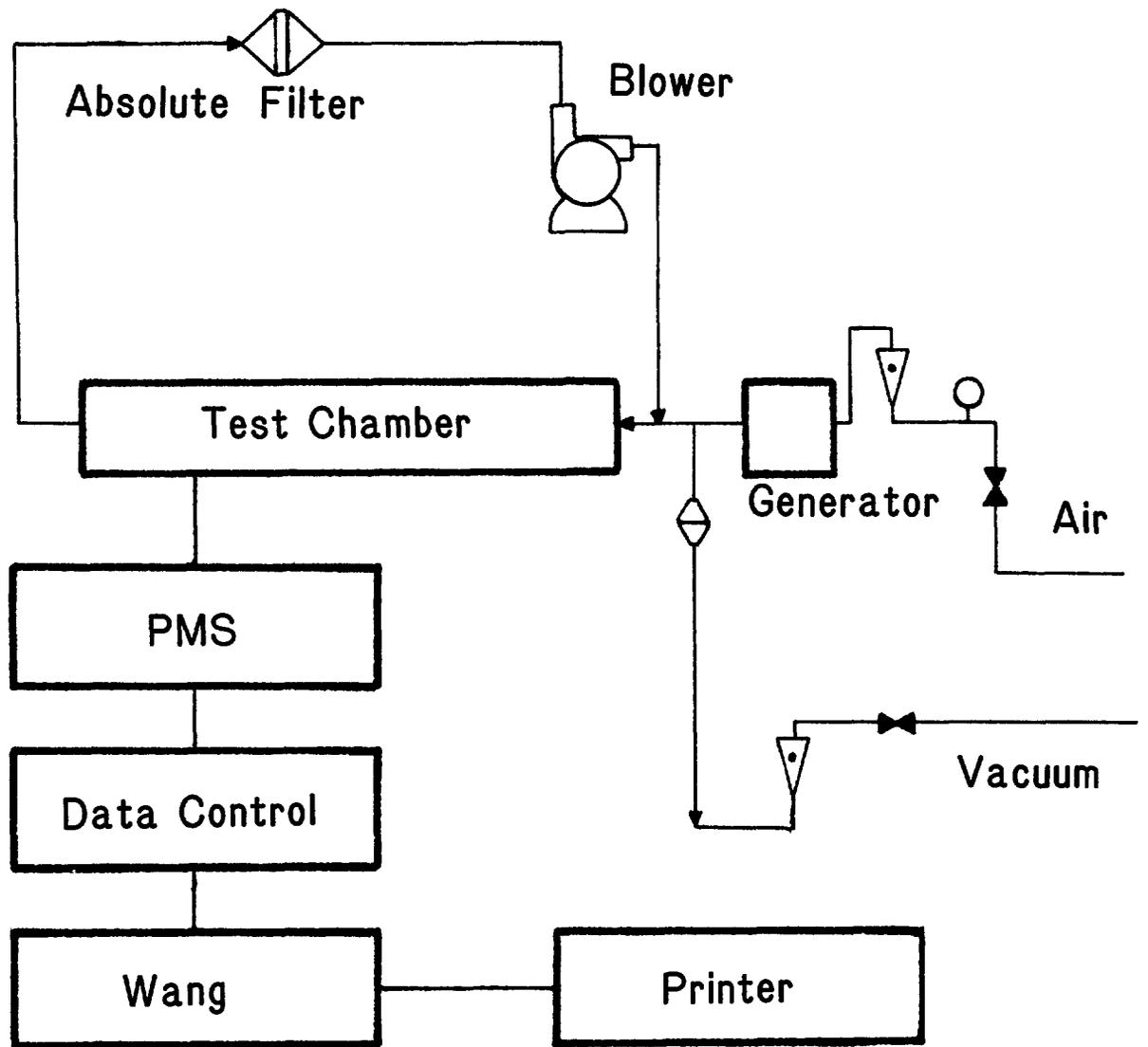


FIGURE 2. Schematic Diagram of the Test System

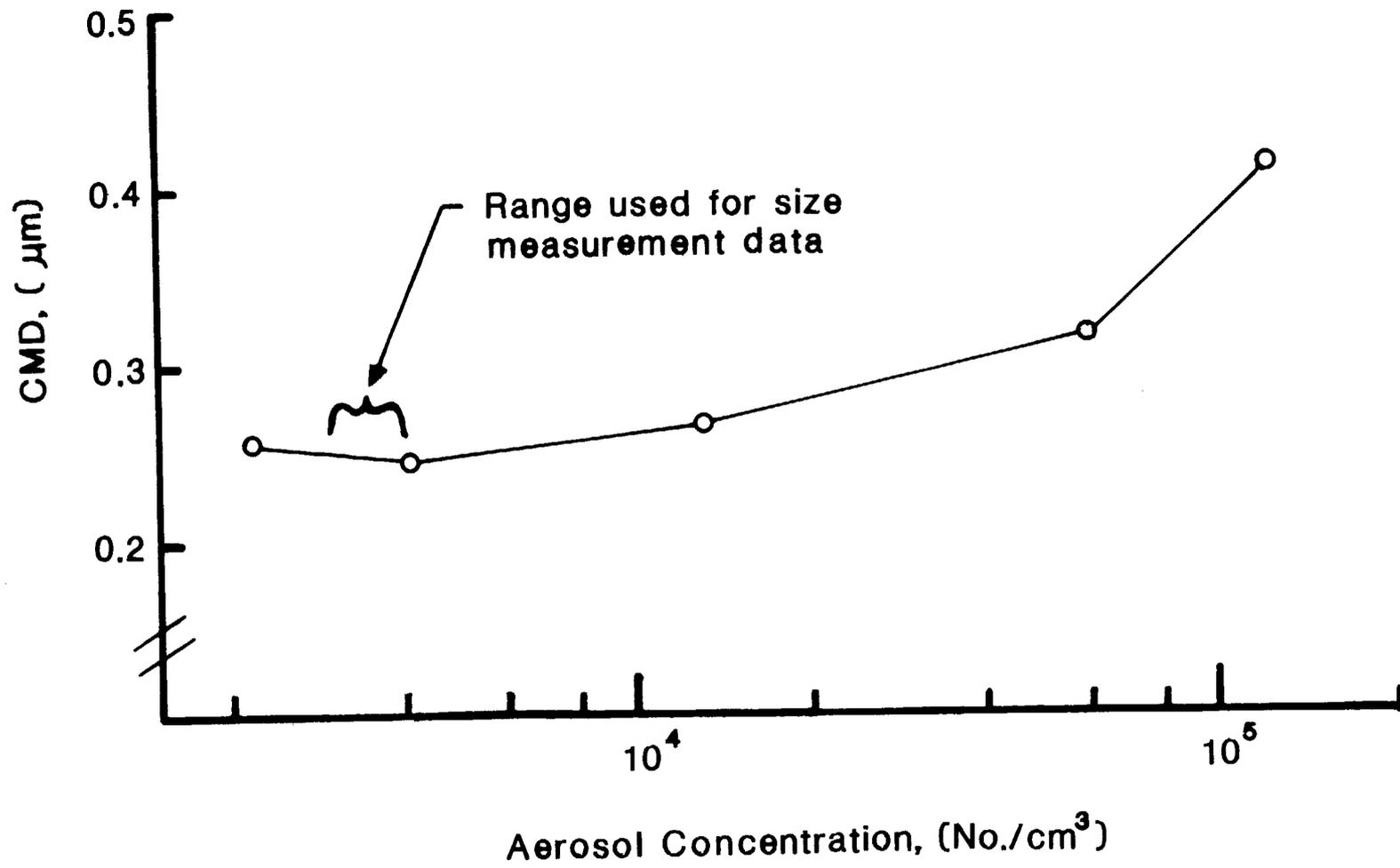


FIGURE 3. Effect of Concentration on Measured Count Median Diameter

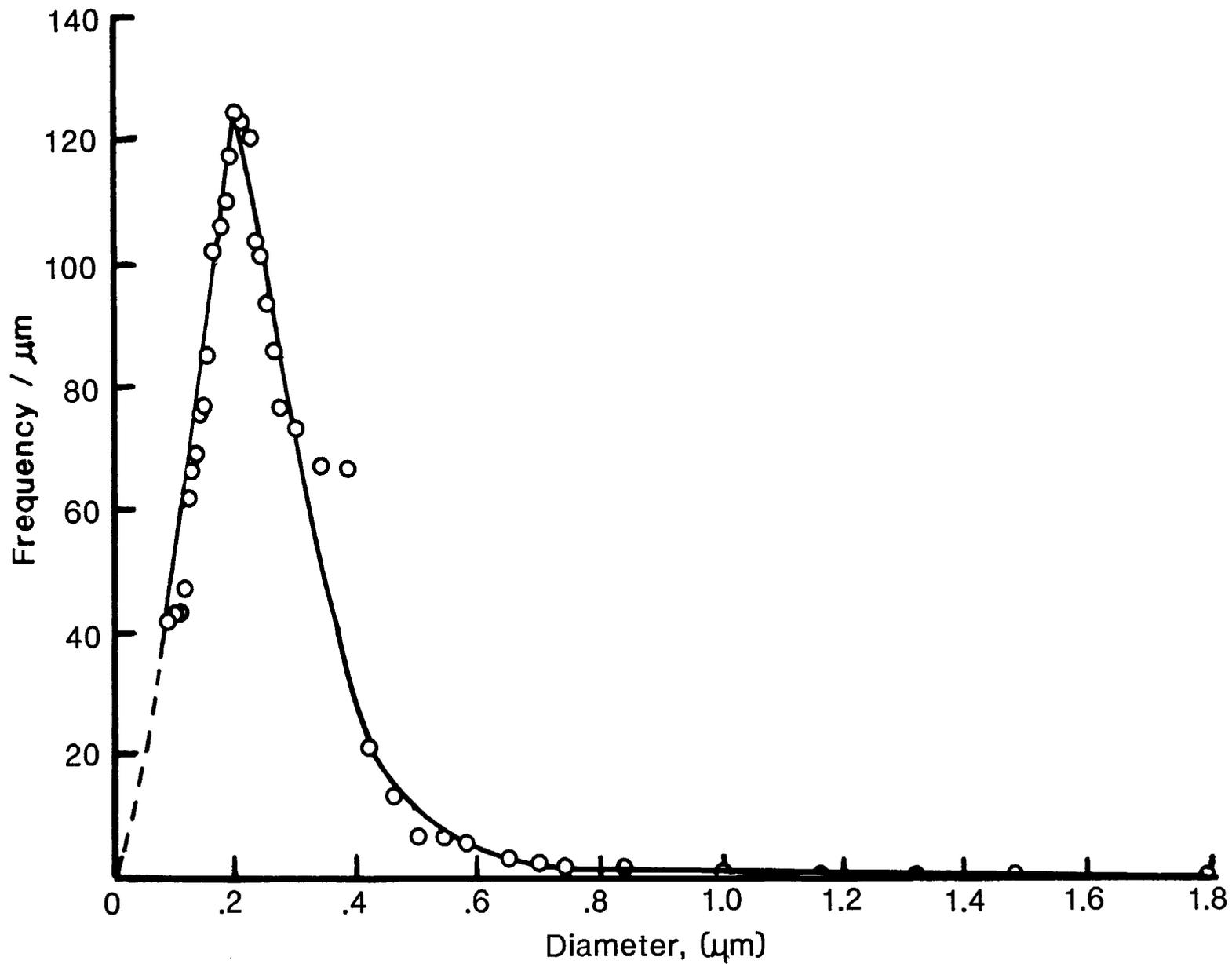
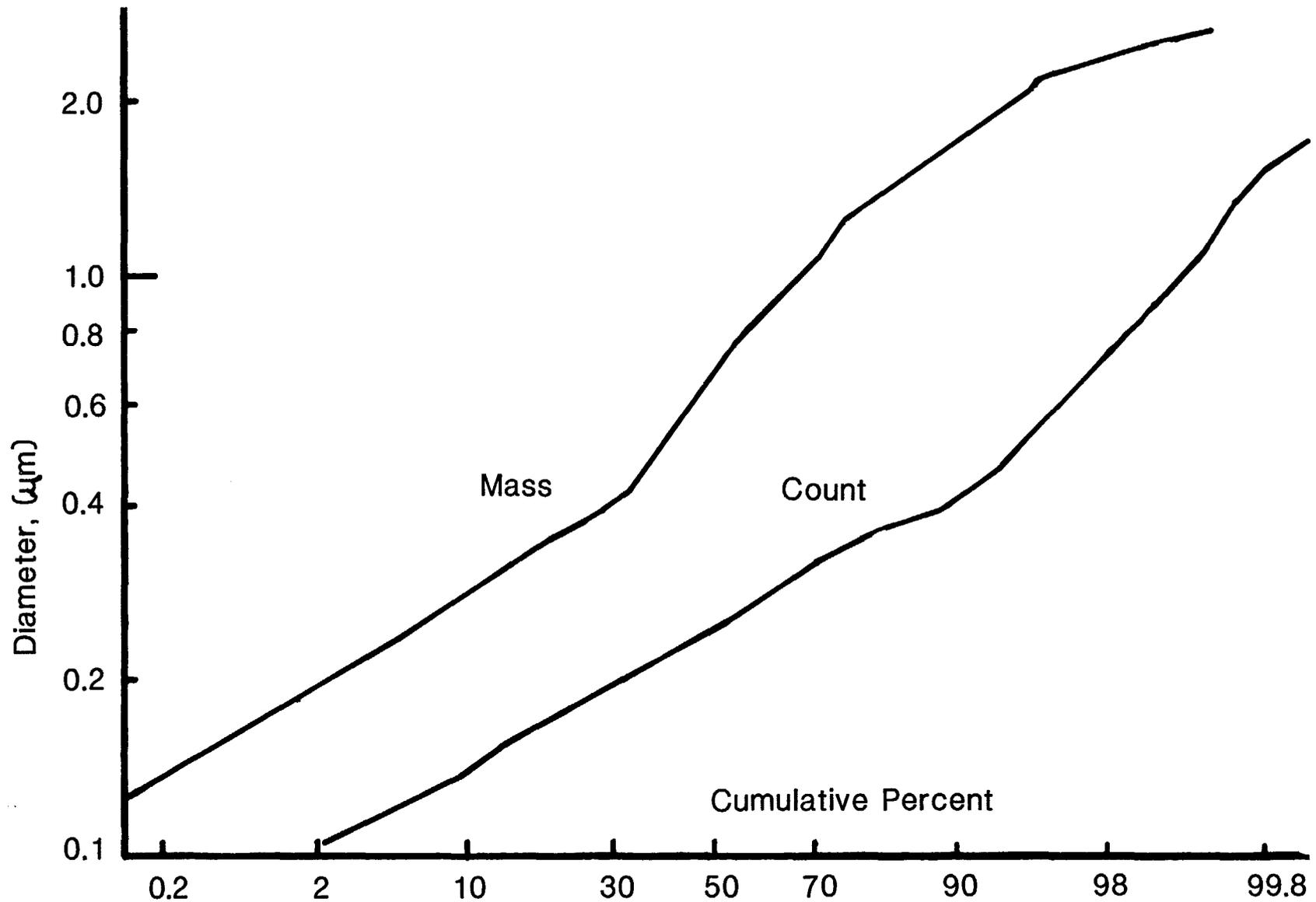


FIGURE 4. Particle Size for DOP at a Nozzle Pressure of 140 kPa (20 psig)



**FIGURE 5. Cumulative Particle Size Distributions for DOP at a Nozzle Pressure of 140 kPa (20 psig). (Mass Data Calculated from Count Data)**

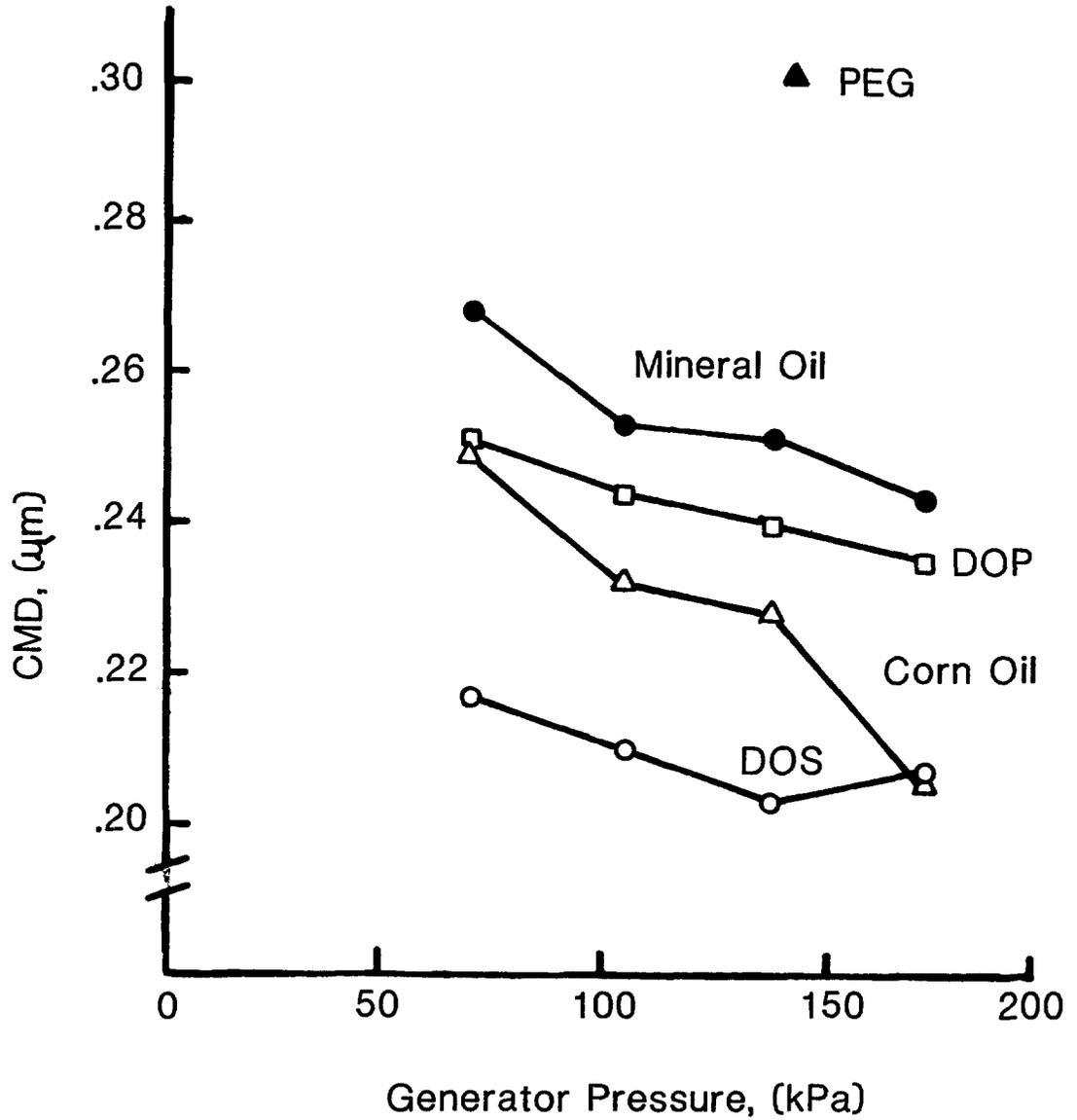


FIGURE 6. Effect of Generator Pressure on CMD for 5 materials

CLOSING REMARKS OF SESSION CHAIRMAN:

It is my prerogative now to try to summarize the session and I think I will do it by making one or two very brief remarks.

From a position of relative innocence, the first few papers led me to believe that perhaps at last we are beginning to see the problems of testing filters both on rigs and in-situ being solved, and even, hopefully, progress being made toward producing international standards on perhaps just one or two methods and techniques. But shortly after that, my confidence began to suffer as Mr. Elder and Mr. Murphy pointed out that no sooner can people do routine measurements by a fairly straightforward method than someone is going to come along and want some really difficult problems solved. Taking Mr. Anderson's point, I am fairly confident now, if of nothing else, that the bright young men he referred to are going to have plenty to keep them going. And so, they will become the "grand old boys" as time goes on.