

SESSION V

SAMPLING AND MONITORING

Tuesday, August 3, 1976

CHAIRMAN: H. Etinger

SELECTIVE SAMPLING OF HYPOIODOUS ACID

M. J. Kabat

AN ANALYSIS FORMAT AND EVALUATION METHODS FOR EFFLUENT PARTICLE SAMPLING SYSTEMS IN NUCLEAR FACILITIES

L. C. Schwendiman, J. A. Glissmeyer

THE USE OF A SINGLE PARTICLE INTRA-CAVITY LASER PARTICLE SPECTROMETER FOR MEASUREMENTS OF HEPA FILTERS AND FILTER SYSTEMS

B. G. Schuster, D. J. Osetek

OPENING REMARKS OF SESSION CHAIRMAN:

Most of the other sessions of this conference are concerned with the design and performance of control systems for nuclear facilities. The three papers this morning will consider different aspects of monitoring or evaluating the performance of some of these controls. As such, they constitute the documentation that these controls perform as advertised over the normal operating cycle, or during non-standard operating conditions; satisfy existing regulatory requirements; and satisfy public concerns, which, as Dr. First pointed out, is becoming more and more critical regarding potential health and environmental effects. These three papers are quite important to the overall air cleaning systems used in nuclear facilities.

The first paper will consider another aspect of the iodine problem which we heard a lot about yesterday. It describes a sampling system to distinguish between hypoiodous acid from other forms of iodine, since their effects are different. The second paper will detail a systematic approach to evaluating air sampling systems for nuclear facilities. This can be used to optimize design of new air sampling systems, as well as evaluating the limitation of existing systems. Since air sampling is one of the cornerstones of monitoring proper, for safe performance of these facilities, this is a very important topic. The last paper details a new technique and new instrumentation for in-place HEPA filter testing; especially when multiple HEPA filters are involved, or multiple layers of filter media are used. Since new emission restrictions and redundancy requirements are making the use of multiple HEPA filter banks more and more common, this subject has significant importance in terms of possible capital plant and operational cost savings.

14th ERDA AIR CLEANING CONFERENCE

SELECTIVE SAMPLING OF HYPOIODOUS ACID

M.J. Kabat
Ontario Hydro
Central Health Physics Services
Pickering Generating Station
Box 160
Pickering, Ontario, Canada.

Abstract

A new material has been developed which efficiently separates hypoiodous acid from a mixture of penetrative species of airborne radioiodine. This selective absorbent has high absorption and retention efficiency for hypoiodous acid under conditions of high relative humidity and short residence time for an extended sampling period. Very low absorption of methyl iodide in this absorbent has been measured under the same conditions.

A sampler, components of which selectively absorb particulate iodine, elemental iodine, hypoiodous acid and organic iodides, has been assembled and successfully applied for airborne radioiodine sampling in an operational field.

The method and equipment used for laboratory testing of the HOI absorbent and for field sampling of iodine species are described herein. Experimental results are also presented and discussed in this paper.

I. Introduction

It has been proven in many previous reports and publications that airborne radioiodine occurs in nuclear power station areas and effluents in three chemical forms: elemental iodine vapour, an inorganic compound - hypoiodous acid and organic forms - mostly as methyl iodide. These iodine species are of greatly different radiobiological significance, particularly for population exposure (via a food chain) evaluation. The deposition rate of elemental iodine on vegetation is reported to be approximately one thousand times greater than the deposition rate of methyl iodide. Hypoiodous acid deposition has not yet been measured because of difficulties experienced in its selective measurement under the practical range of atmospheric conditions and long time sampling. For the above reasons, a considerable effort has been made in our laboratory to develop an adequate system for selective sampling of the above airborne iodine chemical species.

II. Origin and Behaviour of HOI

A theoretical analysis of HOI chemistry and experimental investigation of its sorption on commercially available iodine absorbents was provided in the Central Health Physics laboratory and published in 1974⁽¹⁾. It was concluded in this paper that:

14th ERDA AIR CLEANING CONFERENCE

1. Iodine concentrations in all operational systems of CANDU nuclear power stations are extremely low, even when the radioiodine activity is close to the maximum permissible operational limits.

2. High yield of HOI is obtained from hydrolysis of I_2 in aqueous solutions of very low iodine concentrations ($\leq 10^{-8}$ M).

3. Besides elemental iodine, which is slightly volatile, HOI is the only inorganic iodine compound formed in water solutions under specified conditions which is considered to be highly volatile.

4. Hypoiodous acid has a low chemical reactivity in neutral aqueous solutions (probably because of low dissociation rate).

5. Charcoal is an efficient absorbent for airborne HOI.

6. The existing non-charcoal iodine absorbents are not as efficient for HOI removal, particularly at high humidity.

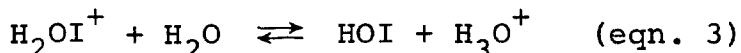
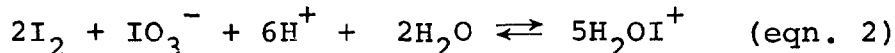
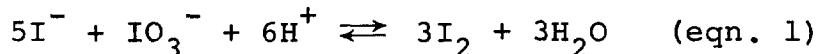
A method was developed, and it is being used in CANDU power stations, which eliminates the formation of HOI in the spent fuel storage bay (by addition of hydrazine to bay water).

Theoretical assumptions of HOI deposition on vegetation were derived from our theoretical and experimental studies on HOI chemistry, and were discussed at the 1975 annual meeting of ANS (2). These assumptions have now been confirmed by experiment, the results of which will be published in the near future.

III. Method of HOI Generation

The method described in (1) was used for short-term generation of HOI containing a minimum amount of organic species of iodine.

A long-term continuous supply of HOI was needed for the testing of selective HOI absorbents. Oxidation of $^{131}I^-$ with IO_3^- was applied for this purpose which provided a continuous supply of airborne HOI for a period of several days. The involved reactions can be expressed by the following chemical equations:



Practically all $^{131}I^-$ is transferred to H_2OI^+ through reactions (eqn. 1) and (eqn. 2) under conditions of low pH (pH ~ 2.0) and high excess of IO_3^- ($\sim 3 \times 10^{-4}$ M). The dissociation equilibrium in reaction (eqn. 3) forms HOI which is continuously stripped with a He-stream from the solution. Because of good stability of H_2OI^+ under the above conditions, HOI can be

continuously generated at a slowly declining rate through a period of several days.

Description of Testing Apparatus

The apparatus used for testing the selective HOI absorbents is illustrated in Figure 1.

HOI Generator. Hypoiodous acid formed in generator G through the above reactions was stripped with a He-stream (50 ml/min). The solution droplets were removed with a glass fibre filter F and dry helium (50 ml/min) added to reduce the relative humidity of the He-HOI stream. This was done in order to achieve efficient removal of elemental iodine with Cu screens. The He-HOI stream was then mixed with the air stream from the air supply system described below. The HOI generator is shown in Figure 2.

Air Supply System. Laboratory air was continuously saturated with steam in the heated drum humidifier DH. Partial condensation of the excessive water vapour was provided in condenser C1. The dew point of the air sample was controlled with condenser C2. Condensate was collected in a thermostatically cooled container. The air temperature (= dew point) was measured with thermometer T1 and the air stream carried into a heating coil HC, the temperature of which was kept constant with a thermostatically controlled bath TB2.

The relative humidity of air downstream of T2 was calculated from its dew point (= T1 reading) and its higher temperature at T2. The relative humidity was also continuously measured with monitor RH (Electro-Hygrometer, Lab-Line Instrument Inc., Model No. 2210) which was being recalibrated at regular intervals with an Abbeon Certified Hygrometer, Model ABl67B. A photograph of the air supply system is shown in Figure 3.

Absorbent Testing Columns. Column A1: Five rings (each 46 mm diameter x 10 mm deep) in series were filled with the tested absorbent. At the challenge gas flow of 25 lpm, its residence time in a 50 mm deep column of the HOI absorbent was 0.2 sec.

An additional ring with 25 mm of fresh HOI absorbent was applied in the cases when greater than 1% of HOI penetration through the original, 50 mm deep, absorbent bed was expected (i.e., during the absorbent aging experiments).

Column A2: Two rings (46 mm diameter x 10 mm deep and 46 mm diameter x 25 mm deep) in series, filled with a fresh TEDA impregnated charcoal, were used for absorption of organic species of iodine penetrating through the HOI absorbent.

Glass Fibre Filter: A glass fibre filter F2 was installed (on several occasions) downstream of column A1 in order to remove and measure particles of the HOI absorbent eventually stripped from column A1 with the challenge gas stream. No measureable ¹³¹I was ever identified on this filter. A disassembled set of testing columns is shown in Figure 4.

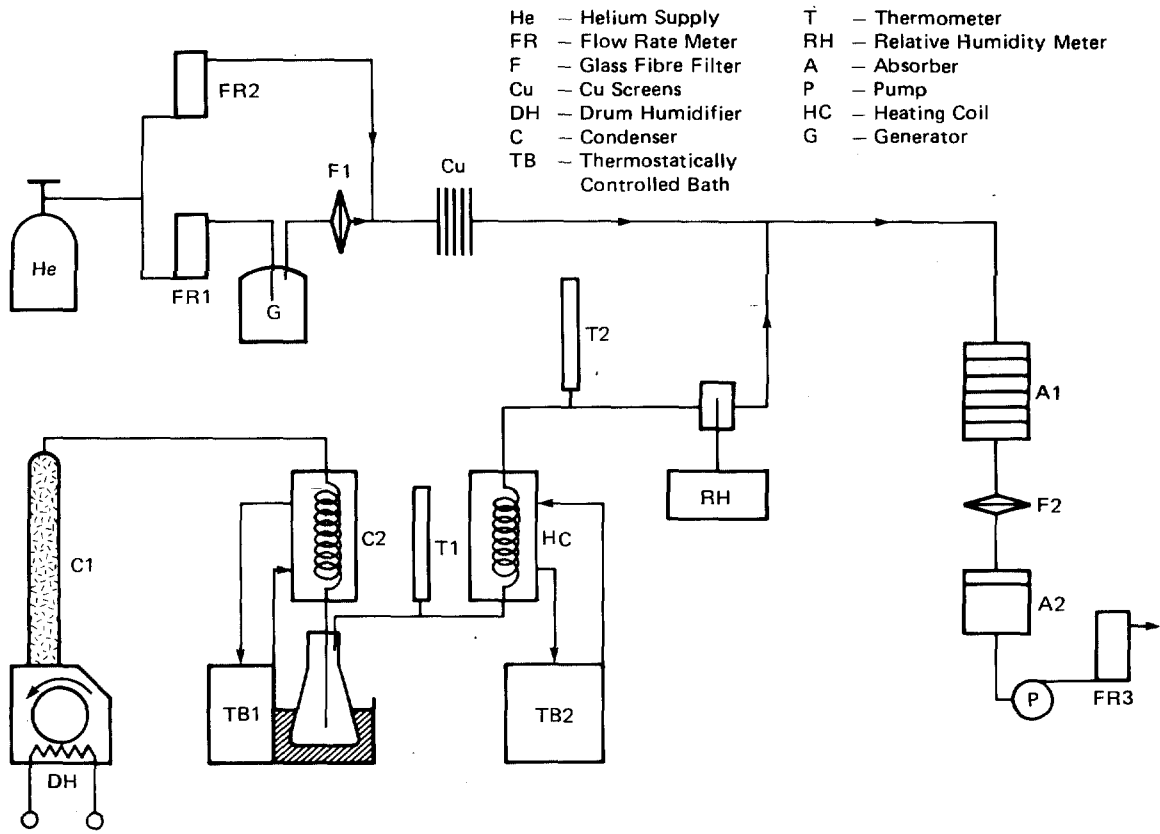


Figure 1 Test setup.

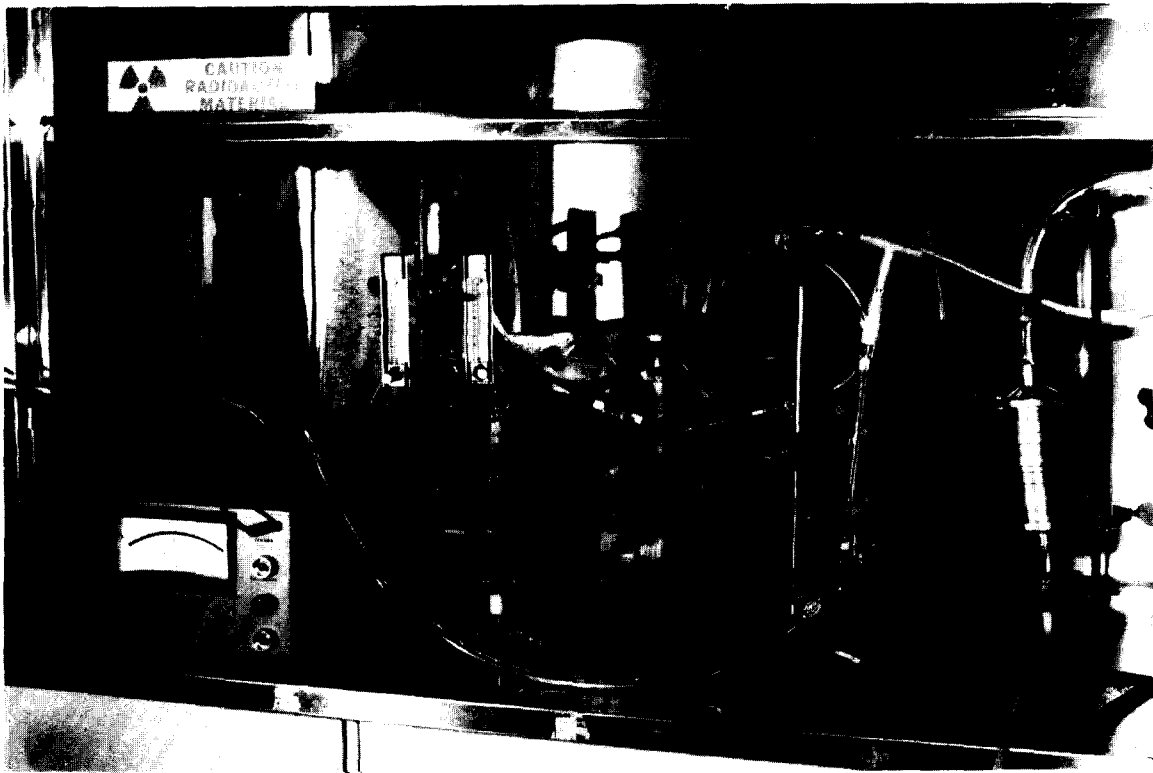


Figure 2 HOI generator.

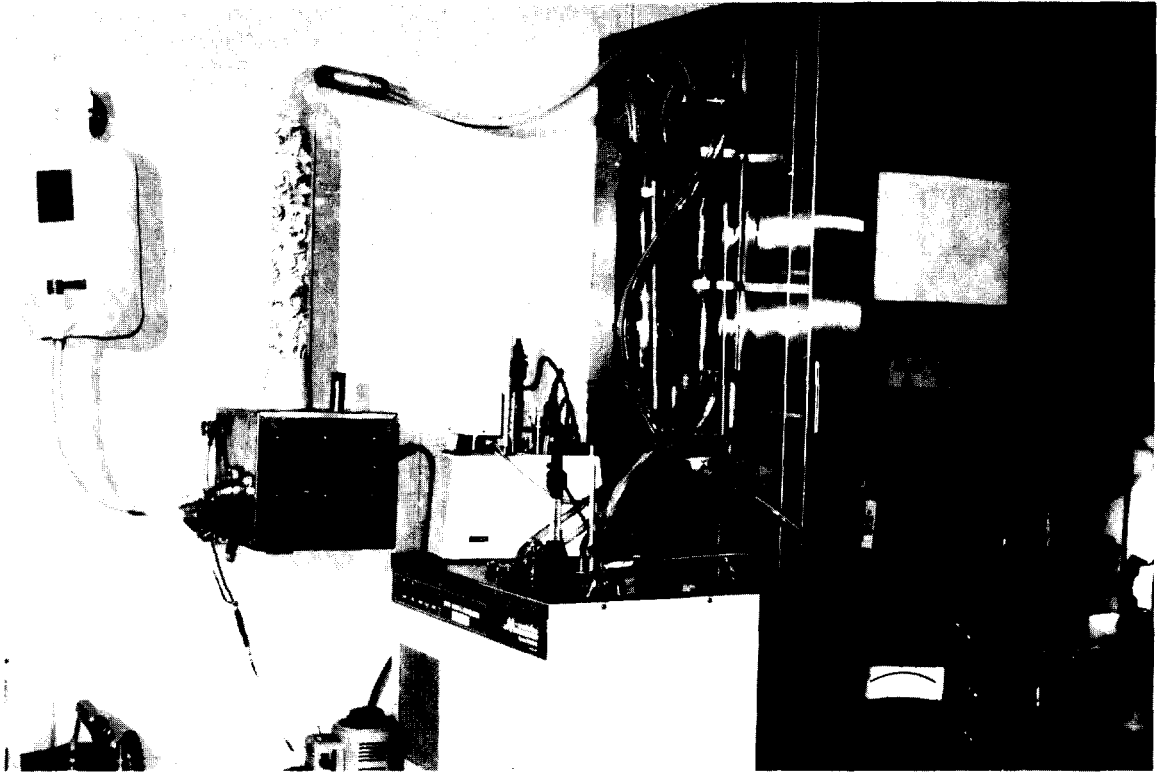


Figure 3 Air supply system.

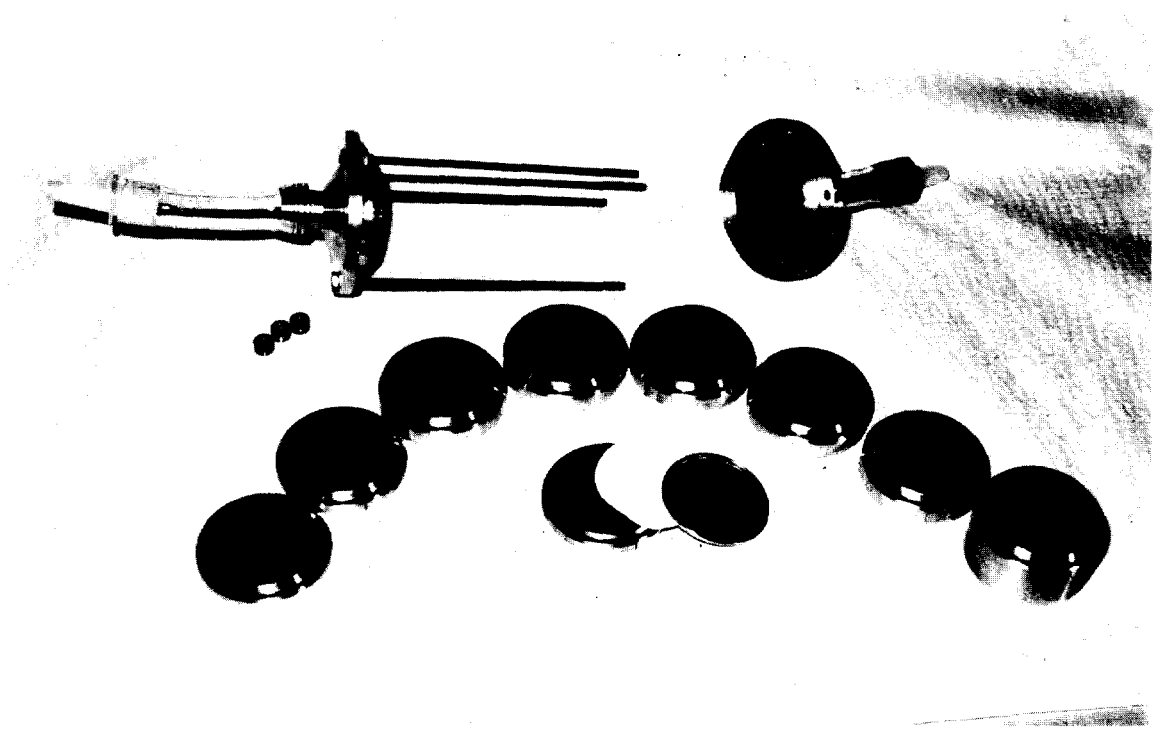


Figure 4 Disassembled set of testing columns.

IV. Experimental Evaluation of the Selective HOI Absorbent Performance

Several materials were tested for HOI absorption efficiency and reported in (1). All of the tested absorbents had high efficiency for CH_3I removal, therefore none of them were suitable for selective HOI sampling. A number of other solid and liquid absorbents (absorbents, metals, 4-iodophenol, solutions, oil) were tested subsequently which absorbed HOI efficiently and selectively, but their performance in long-term sampling at $\text{RH} > 70\%$ did not fulfill our requirements.

Finally, summarizing the practical experience from the HOI generation and absorption experiments, the physicochemical model of selective HOI absorption was established and an absorbent developed (a carbon based material) which gives optimum performance. The HOI absorption model and technical details on this absorbent will be published after submitting an application for a patent on the absorbent.

Absorption properties of the HOI absorbent are discussed below.

Experimental Conditions

The challenge gas (air + HO^{131}I with $< 1\%$ He) was passed through the adsorbent in columns A1 and A2 and each ring was separately measured with a calibrated Ge - Li detector coupled to a Canberra Multichannel Gamma Analyzer, Model 8180. Sufficiently long counting time was applied in measurement of the downstream side of the A1 column in order to reduce the statistical counting error below $\pm 10\%$ at 95% confidence level.

In experiments where the time interval between activity measurement on identical samples was longer than approximately five hours, the measured ^{131}I activity was corrected for decay and the integrated ^{131}I total in the columns was compared with the initial total from the first measurement in the set. This method was particularly useful in long-term desorption measurements.

The depth of the A1 column (HOI absorbent) was applied in all measurements that HOI penetration did not exceed 1%. Therefore, all ^{131}I identified in the A2 absorbent (TEDA impregnated charcoal) was considered to be an organic compound of iodine.

The amount of organic iodides varied within a range of 1% to 15% of the total ^{131}I absorbed in the columns. Low content of organic forms (1% to 3%) was present in HOI generated from freshly supplied ^{131}I solutions, and it was then continuously increased with the solution storage time up to approximately 15% after five weeks of storage.

14th ERDA AIR CLEANING CONFERENCE

A histogram of the typical absorption profile through the depth of the columns A1 and A2 is illustrated in Figure 5.

Absorption of HOI

The basic, non-treated carbon had high efficiency for HOI absorption at low humidity through short sampling intervals. It also absorbed a significant portion of CH_3I and retention of HOI was not satisfactory. The graph in Figure 6 indicates that a significant desorption of HOI with passing air occurs within a few hours.

Treated carbon (of the identical batch) was slightly less efficient for HOI absorption, but both CH_3I absorption and HOI desorption rates were substantially reduced. Figure 7 shows HOI absorption and desorption characteristics, measured under the same conditions as applied in the non-treated carbon testing (40% RH, 22°C).

The HOI absorbent was slightly less efficient at 98% RH as illustrated in Figure 8. Iodine retention in the absorbent was still very good under these conditions.

No concentration dependence in absorption of HOI in this material was determined as illustrated in Figure 9.

Dependence of HOI absorption efficiency on the challenge gas flow rate illustrated in Figure 10 indicates that the rate of HOI absorption was not constant when a different face velocity of the challenge gas was applied.

The HOI absorption rate is expressed by a quality factor K,

$$K = \frac{\ln D}{\Delta t}$$

where, D = Decontamination factor
 Δt = Residence time.

Then at $\Delta t = 0.1$ sec,

| <u>Sample Flow (lpm)</u> | <u>K</u> | <u>Depth of Absorbent that Removes 99% of HOI (mm)</u> |
|--------------------------|----------|--|
| 12 | 31.6 | 18 |
| 25 | 43.8 | 26 |
| 50 | 56.5 | 40 |

Then, the penetration P (%) after the challenge gas residence time Δt (seconds) can be calculated from,

$$P = e^{-K\Delta t} \times 100$$

497

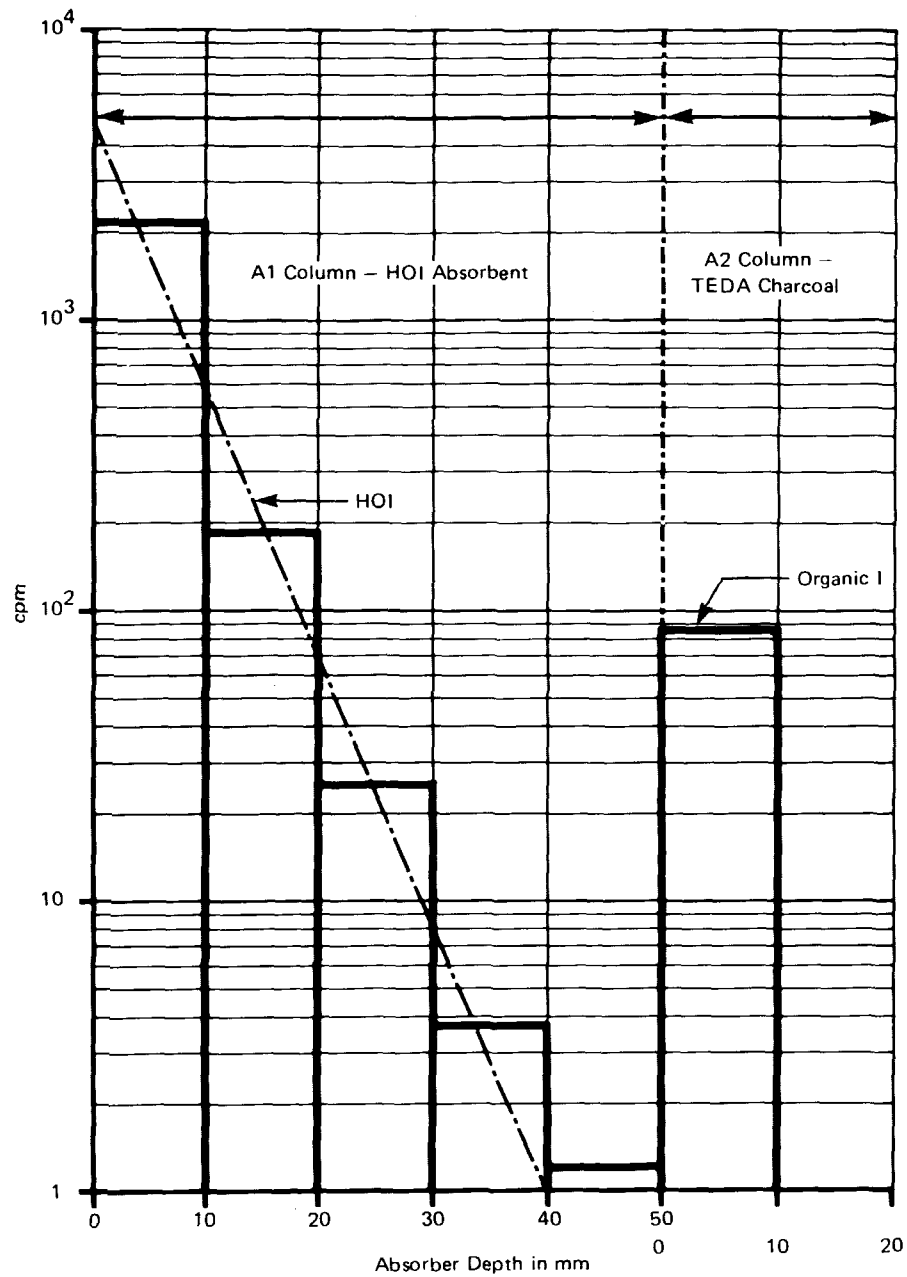


Figure 5 Typical ^{131}I profile through absorption columns.

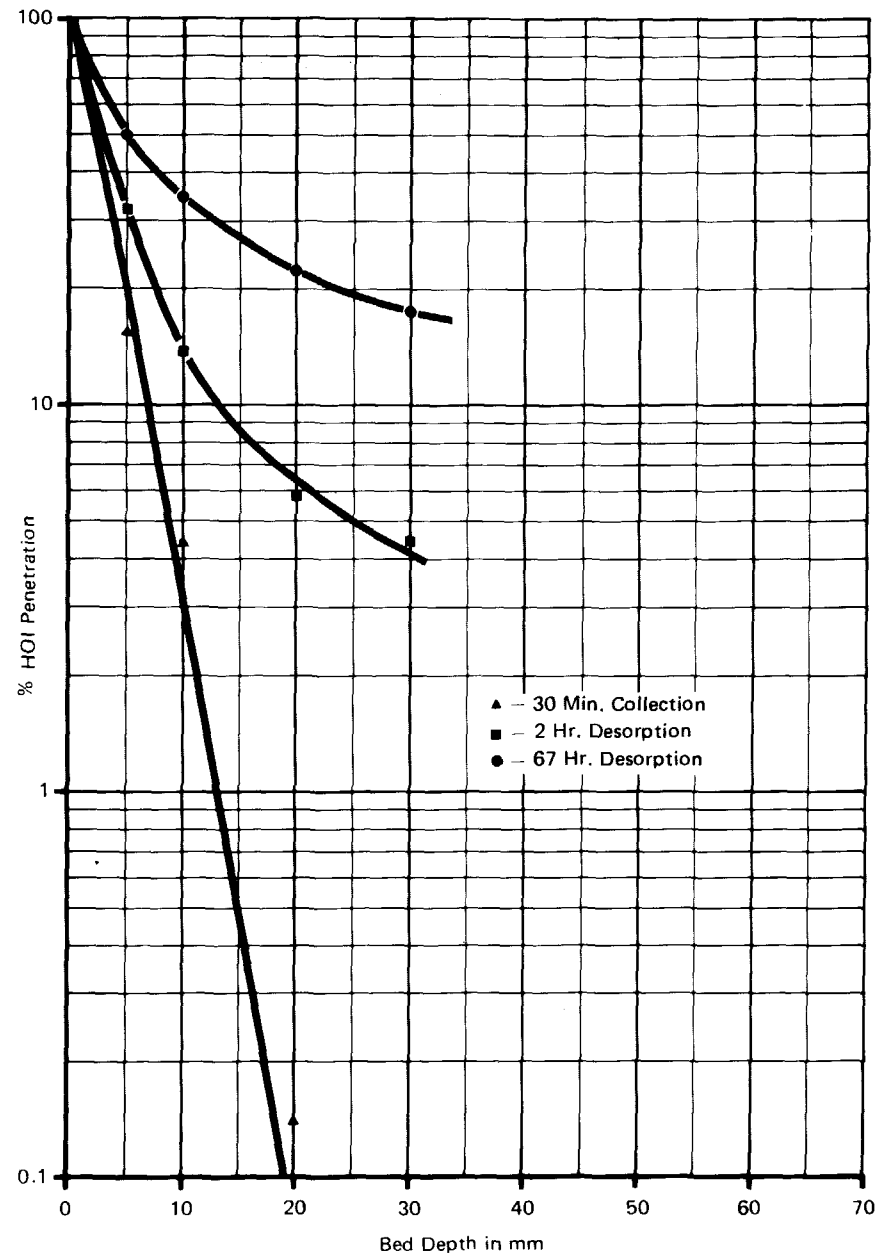


Figure 6 Absorption of HOI in a non-treated carbon; 40% RH, 22°C.

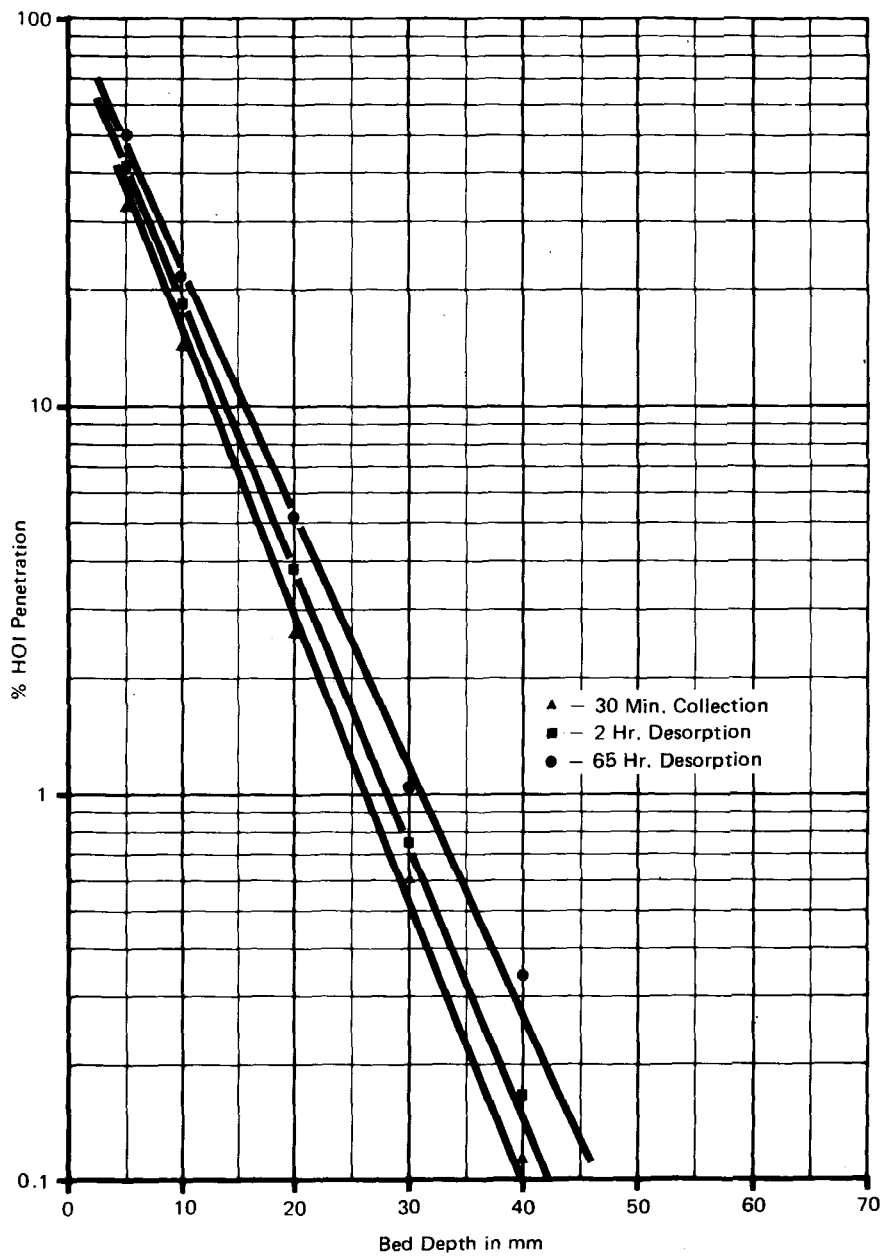


Figure 7 HOI absorbent; 40% RH, 22°C.

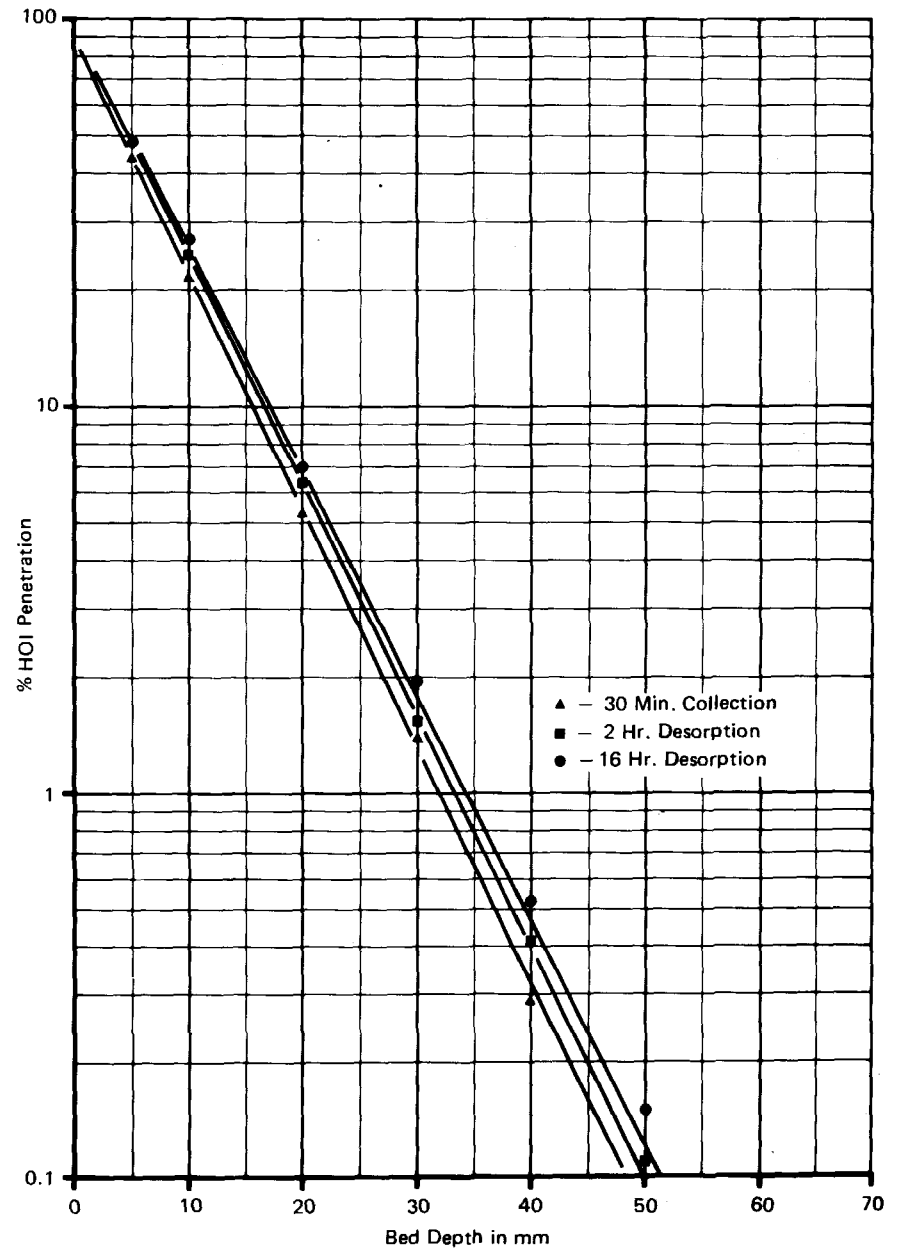


Figure 8 HOI absorbent; 98% RH, 22°C.

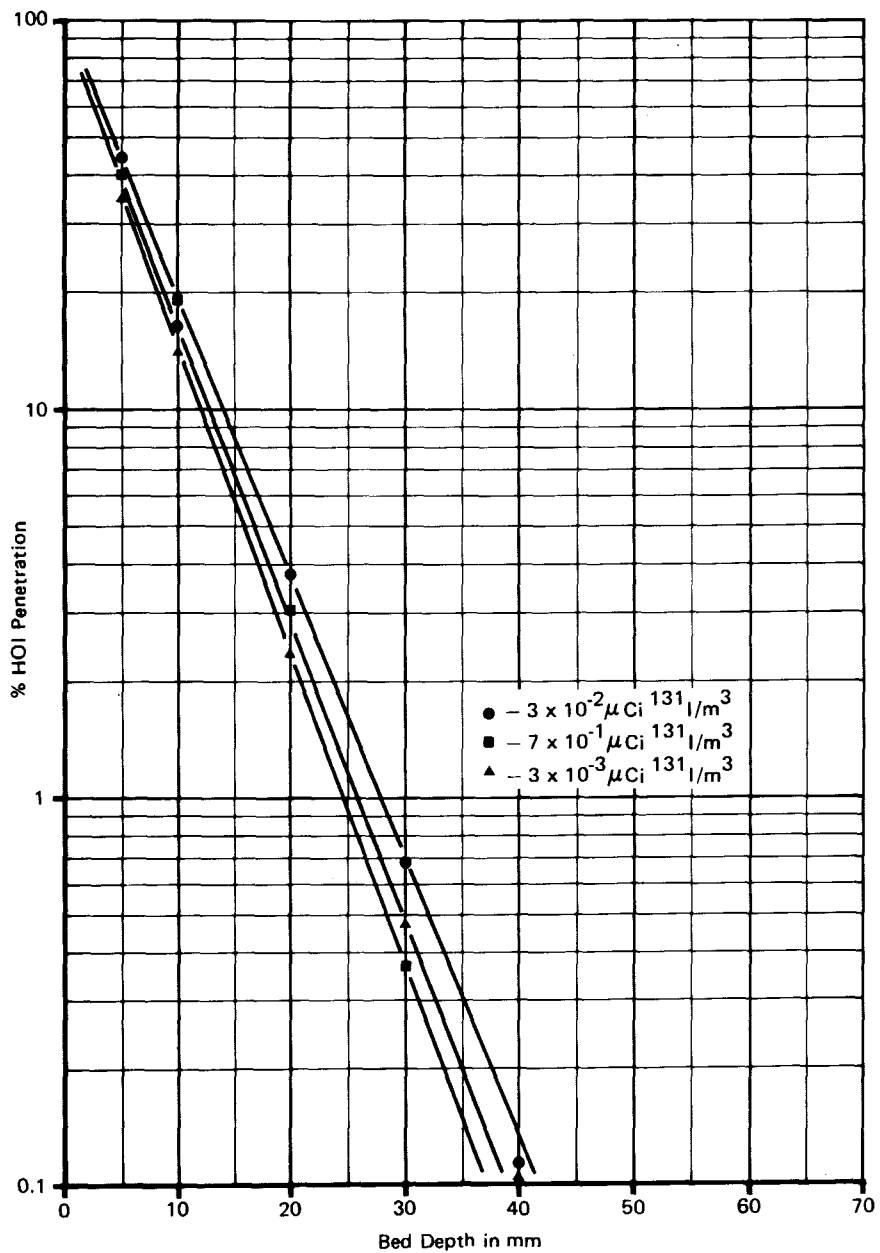


Figure 9 HOI absorption at different airborne concentrations; 40% RH, 22°C.

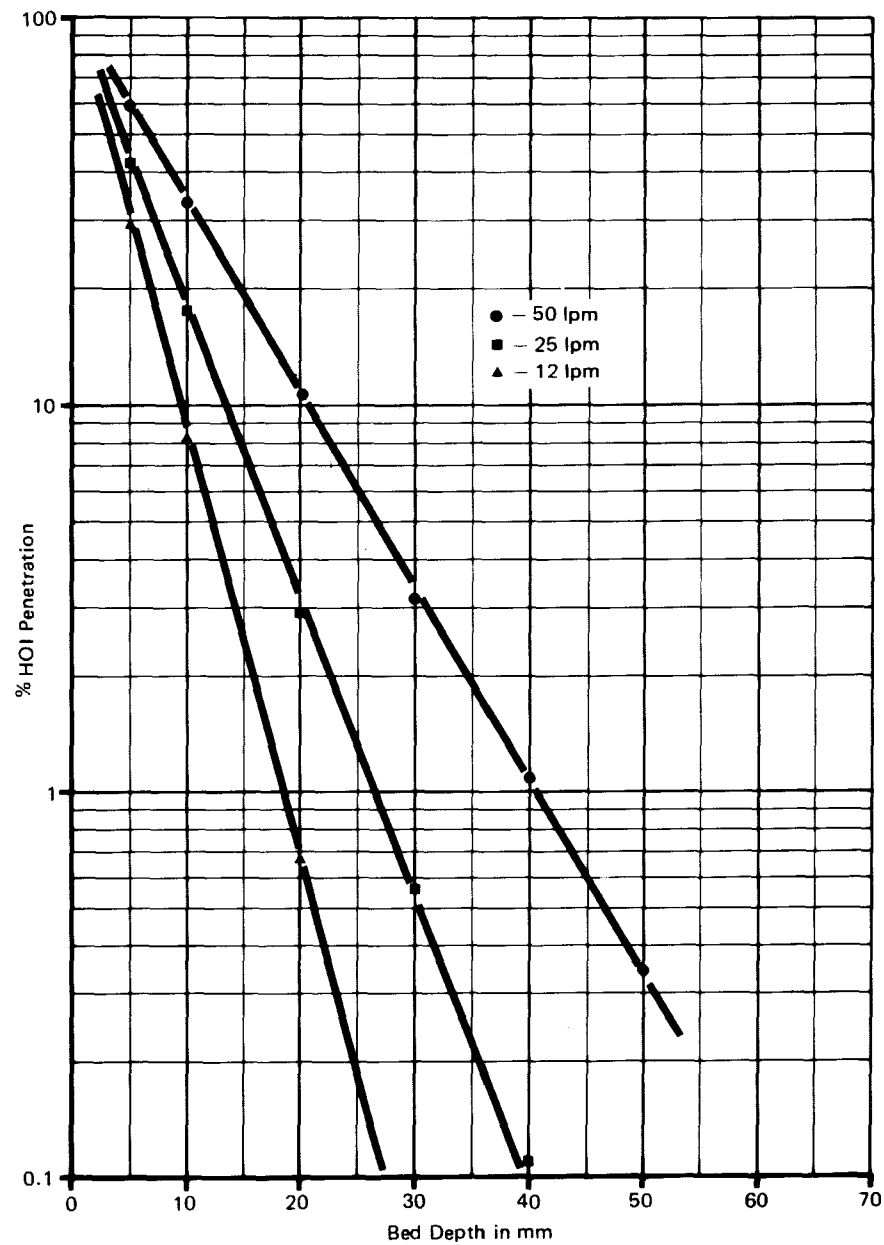


Figure 10 Dependence of HOI absorption efficiency on the sample flow; 40% RH, 22°C.

14th ERDA AIR CLEANING CONFERENCE

The practical implication of this observation was that the sampling column depth did not increase proportionally with the sampling flow increase at the same decontamination factor. This is an advantageous feature of the HOI adsorbent for its use in high volume sampling applications.

Further, the HOI adsorbent was tested under long-term sampling conditions. The adsorbent column A1 was exposed to a continuous air flow under the most common sampling conditions (50% RH, 22°C), and its efficiency for HOI absorption was measured after six days and twenty-one days of continuous aging.

Graphical results in Figure 11 show that the adsorbent retained its good efficiency (in ≥ 50 mm deep bed) for a period of one week under these conditions. After three weeks of aging, the efficiency was reduced to approximately 80% with low desorption rate, which can be classified as good under the applied conditions.

The adsorbent efficiency for HOI was reduced more drastically (as expected) during the high humidity aging process. Figure 12 shows that good absorption efficiency remained after four days of continuous aging at 98% RH, 22°C conditions.

Results of the adsorbent testing under extreme conditions, illustrated in Figure 13, indicate that the adsorbent can be used for HOI sampling under fog conditions when water condensation occurs in the adsorbent column. The suggested factor for sampling efficiency correction is approximately 1.4, if the sampling or aging interval does not exceed twenty-four hours under these conditions. The graph also shows that retention of the absorbed iodine is still very good under such extreme conditions.

Absorption of I_2 and CH_3I in the HOI Adsorbent

The HOI adsorbent was highly efficient for elemental iodine absorption. Therefore, when iodine species were to be sampled selectively, I_2 had to be absorbed in a preceding adsorbent which had a minimum affinity to HOI and CH_3I .

Absorption of CH_3I on the HOI adsorbent was measured under 30%, 50%, 60% and 98% RH conditions. The results presented in Figure 14 show that the fraction of the absorbed CH_3I varied with the relative humidity of the challenge gas from about 4%/25 mm at low humidity to only 1%/25 mm at 98% RH.

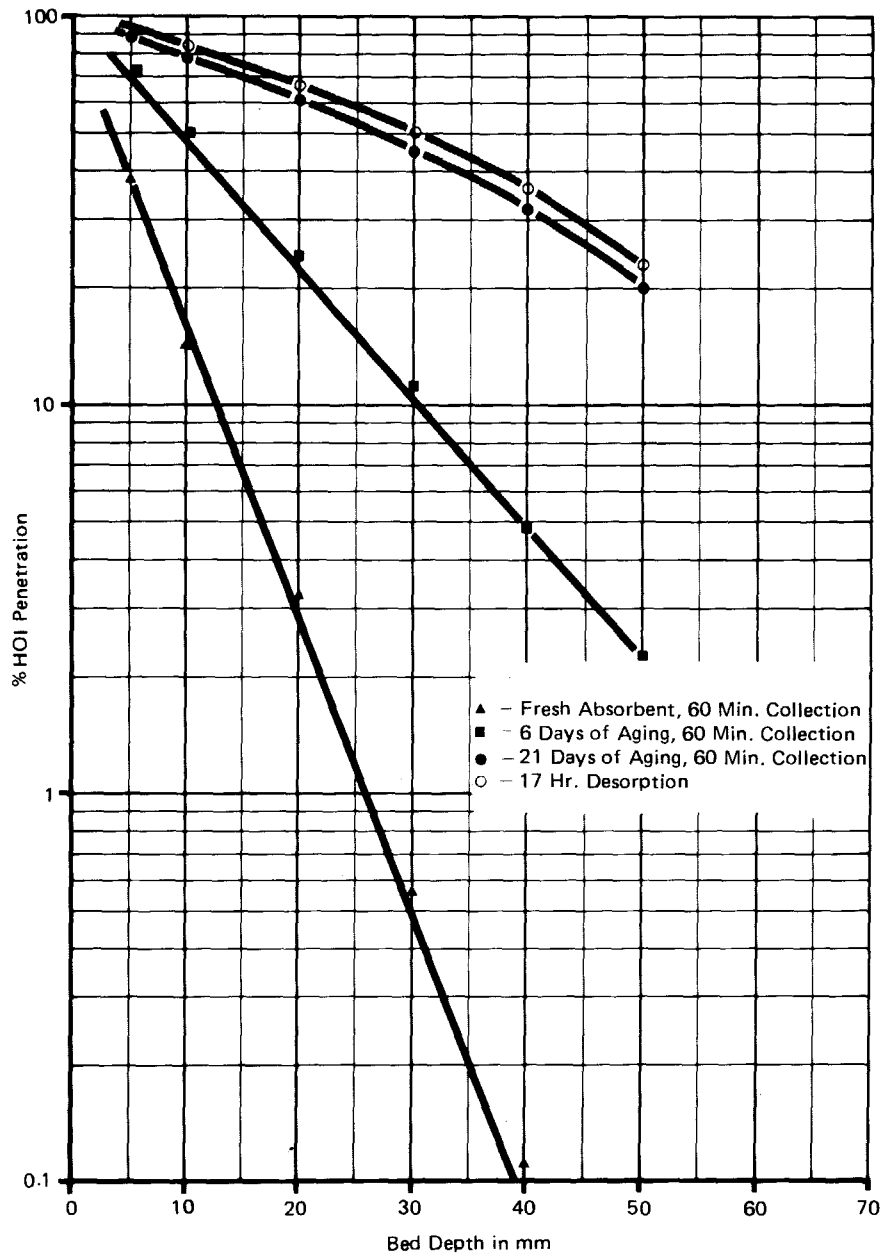


Figure 11 HOI absorbent aging at 50% RH, 22°C.

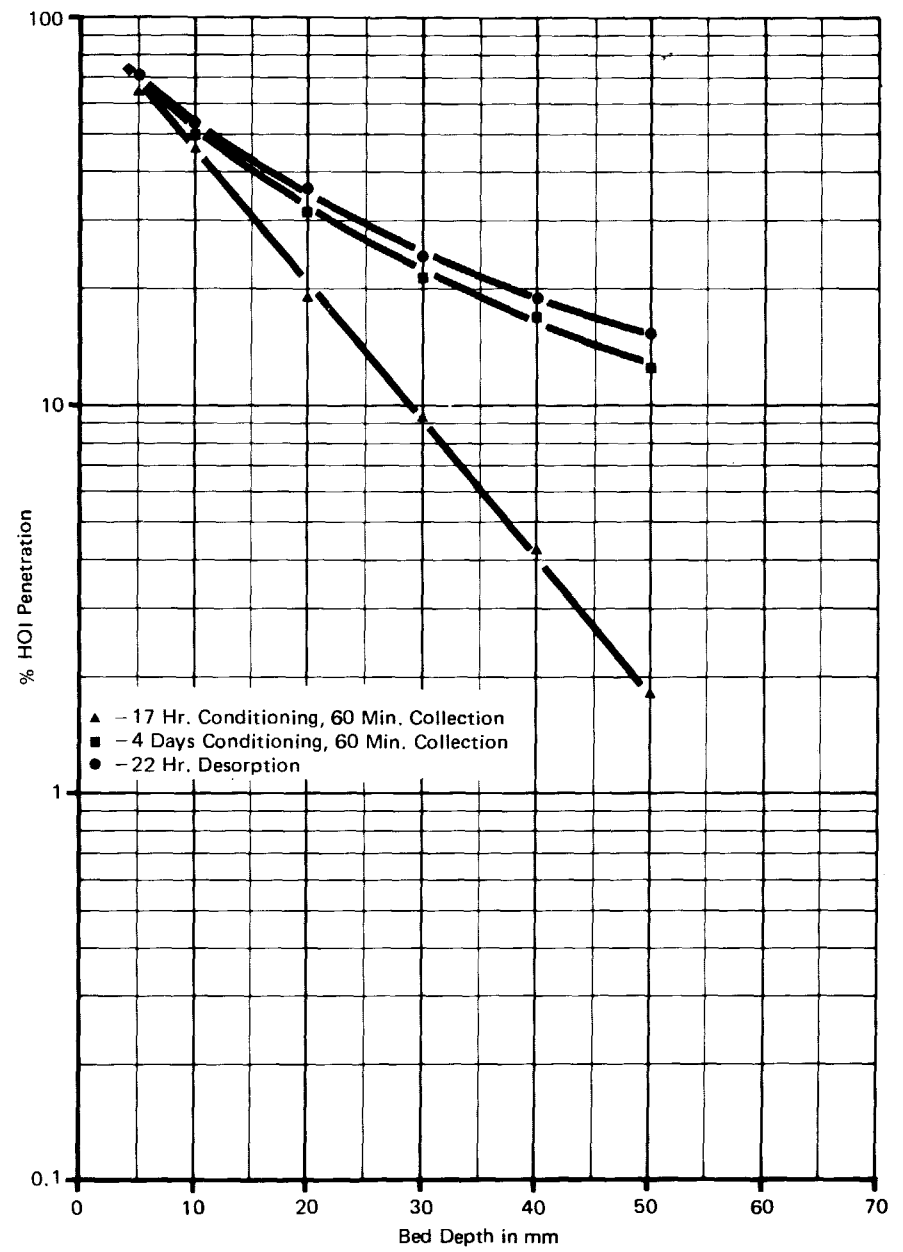


Figure 12 HOI absorbent aging at 95% RH, 22°C.

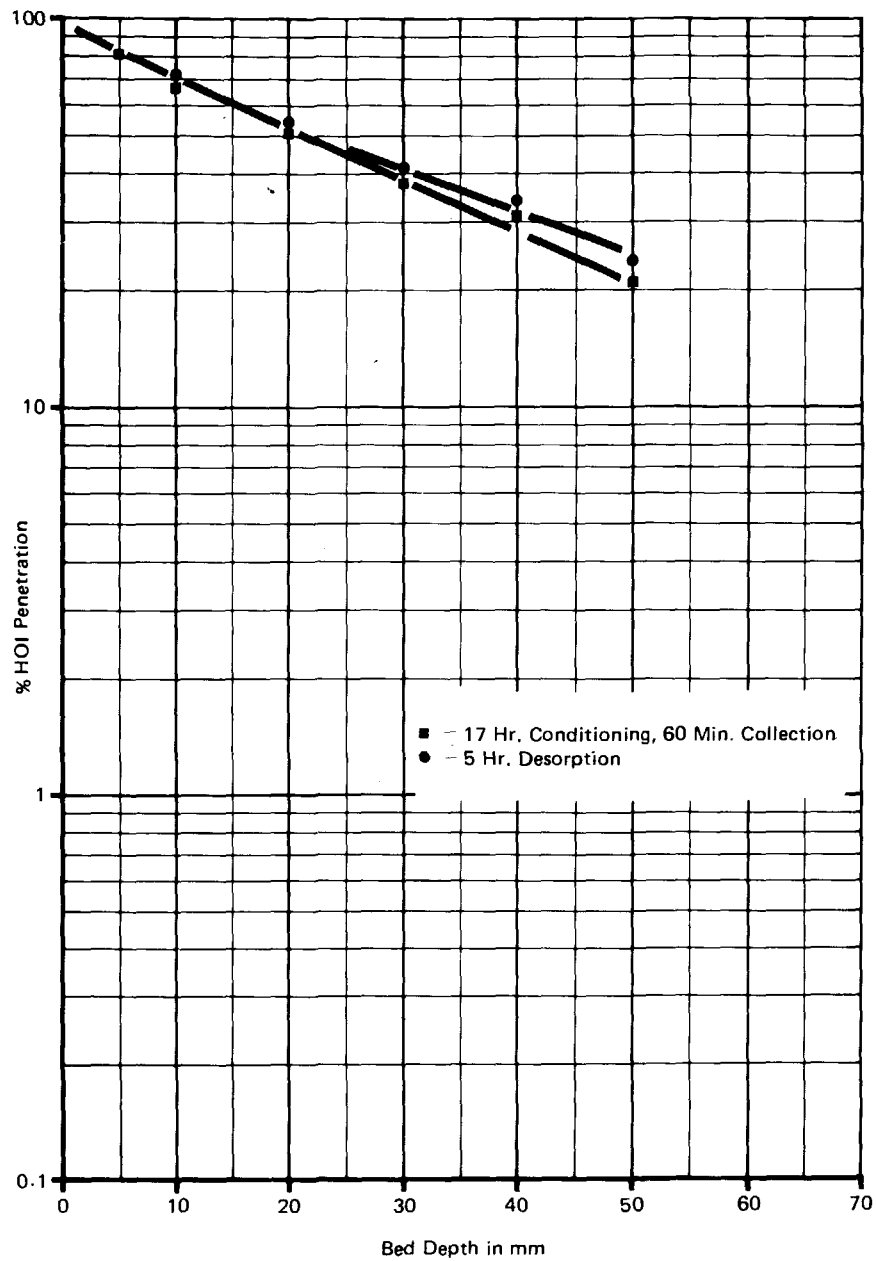


Figure 13 HOI absorbent aging; slight condensation, 22°C.

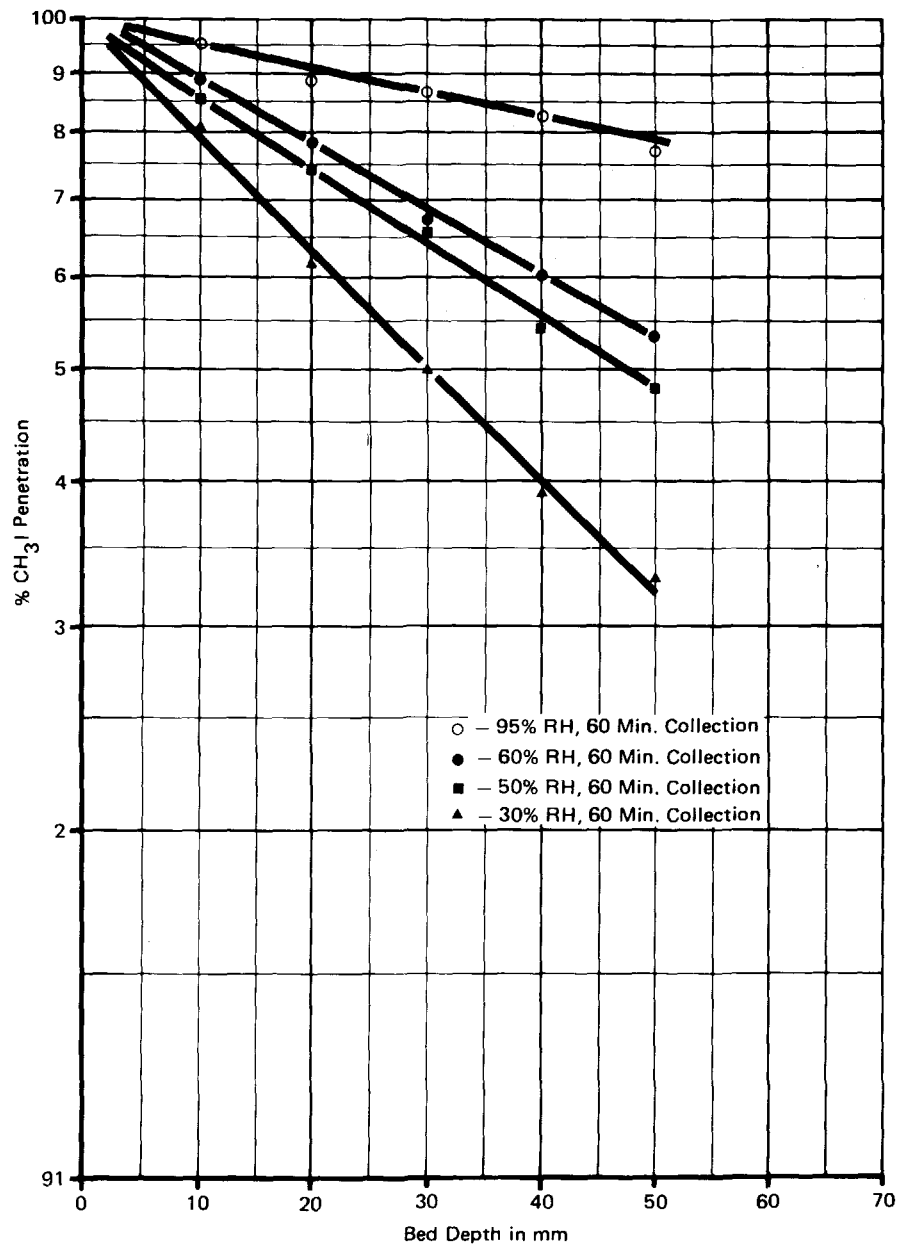


Figure 14 CH₃I absorption in HOI absorbent.

V. Selective Sampling of Airborne Iodine SpeciesSampling Equipment

A RadēCo H809V Sampler with a modified head, as illustrated in Figure 15, was used for the sampling of iodine species in the CANDU station areas.

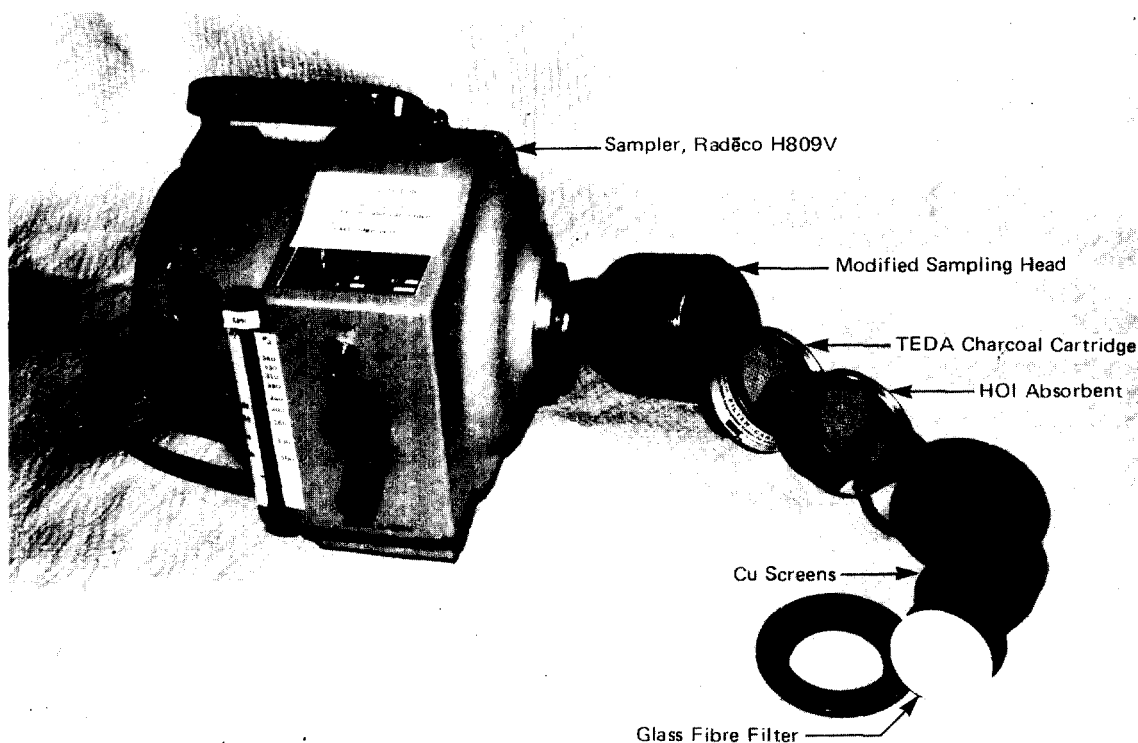


Figure 15 Sampling equipment.

The function of the sampler is as follows:

- Particles are removed from the passing air sample with a Gellman A filter (pure glass fibre, diameter 50 mm).
- Elemental iodine is absorbed in Cu screens (approximately 10 screens, mesh 100, 50 mm diameter).
- HOI is absorbed in a metal cartridge filled with HOI absorbent.
- Organic compounds of iodine are absorbed in a Scott No. 4235-TA cartridge (filled with TEDA impregnated charcoal).

Cuprum screens were the only components in the samplig system which were suitable only for short-term I_2 sampling (maximum of thirty minutes at 30 lpm [~ 1 cfm] air flow). After this time their efficiency for I_2 absorption started to decline. Further, Cu

screens must be etched with HI just before their use. The etching procedure is given in ⁽³⁾.

In spite of the above complications, Cu screens are being used in our laboratory for specific sampling of I₂ and its removal from gas streams, because of the high efficiency for I₂ absorption and the minimum absorption of HOI (<5%) under the applied conditions. All of the other tested I₂ absorbents absorbed significant portions of HOI; iodine retention was not satisfactory.

Procedure for Selective Sampling and Evaluation of Airborne Radioiodine Species

A sampling flow rate of 30 lpm (~1 cfm) corresponds to the residence time of 0.1 seconds in both the HOI and organic I absorbents. This gives ≥97% absorption efficiency for all three iodine components at 98% RH. Absorption of both HOI on Cu screens and CH₃I in the HOI absorbent does not exceed 5% within the RH range of 30%³ to 98%. A sampling time of up to thirty minutes can be applied when I₂ concentration is to be selectively measured.

It is recommended that in long-term HOI and organic I sampling, two or more HOI absorbent cartridges be applied in series, or the air sampling flow be adequately reduced.

The last cartridge (TEDA charcoal) remains at >98% efficiency for a period of several weeks under the above conditions.

When a dry air (<30% RH) sample is taken, which contains a major portion of organic I, its estimated amount (5%) may be subtracted from iodine activity measured in the HOI absorbent and coincidentally added to the TEDA cartridge results as follows:

$$\text{cpm (org. I)} = \text{cpm (TEDA cartridge)} + 5\%$$

$$\text{cpm (HOI)} = \text{cpm (HOI absorbent)} - 5\% \text{ of cpm (TEDA)}$$

In most practical situations this correction can be neglected.

VI. Conclusions

1. The new absorbent described above has high efficiency for selective HOI absorption within a wide range of sampling conditions. The absorbent retains its good efficiency for a period of several weeks in a continuous sampling regime.

2. The described assembly provides selective sampling for particulate forms of iodine, elemental iodine vapour, hypoiodous acid and organic species of radioiodine.

3. The sampler was successfully applied in identifying the ratio of HOI to organic iodine species in the Pickering NGS spent fuel transfer room atmosphere, after a significant release of fission products had occurred from a defective fuel bundle.

4. For long-term sampling of all radioiodine species in nuclear stations stack effluents and in the environment, an adequate elemental iodine absorbent still has to be developed.

Acknowledgement

The author wishes to express his appreciation to Mr. J. Hartwell for his contribution to the experimental program.

VII. References

1. M. Kabat. "Testing and evaluation of absorbers for gaseous penetrative forms of Radioiodine." Paper presented at the 13th AEC Air Cleaning Conference, San Francisco, August 1974, Proceedings pp.765-800.

2. Panel discussion: "Atmospheric dispersion and nuclear power plant dose assessment - II." ANS annual meeting, New Orleans, June 1975, p.524.

3. P.J. Barry. "Sampling of airborne radioiodine by copper screens." Health Physics, 15, pp.243-250. (1968)

DISCUSSION

LORENZ: I have two questions. (1) Will your adsorber work at higher temperatures, such as 125°C, in either steam and air or dry steam? (2) Have you tried silver plated screens to see if they are specific for elemental iodine?

KABAT: The sampler can be used under the same conditions as other carbons. The impregnant is not volatile. I tested silver plated screens for both elemental iodine and HOI. Silver is very efficient for elemental iodine but absorbs a significant fraction of HOI.

DIETZ: Can the HOI adsorbent be regenerated by heating?

KABAT: Yes, this material can be regenerated, but it is not practical because of its low cost.

WILHELM: What is the chemical reaction that traps HOI?

KABAT: I am sorry, I cannot release this information now. I am going to apply for a patent on the HOI adsorbent. The physiochemical model of the selective absorption process and detailed information on this material will be published after submitting the patent application.

KABAT: The limit on sampling time is related to the limited lifetime of the copper screen which is used in the sampler for elemental iodine. The other components, HOI and methyl iodide adsorbents, have a much longer life; at least one week, as discussed in more detail in the paper. HOI is analyzed on the adsorber by radiometric evaluation of the column components to determine the amount of radioiodine absorbed in the HOI and methyl iodide adsorbents.

14th ERDA AIR CLEANING CONFERENCE

AN ANALYSIS FORMAT AND EVALUATION METHODS FOR EFFLUENT PARTICLE SAMPLING SYSTEMS IN NUCLEAR FACILITIES*

L. C. Schwendiman, J. A. Glissmeyer

Battelle, Pacific Northwest Laboratories
Richland, Washington

Abstract

Airborne effluent sampling systems for nuclear facilities are frequently designed, installed, and operated without a systematic approach which discloses and takes into account all the circumstances and conditions which would affect the validity and adequacy of the sample. Without a comprehensive check list or something similar, the designer of the system may not be given the important information needed to provide a good design. In like manner, an already operating system may be better appraised. Furthermore, the discipline of a more formal approach may compel the one who will use the system to make sure he knows what he wants and can thus give the designer the needed information. An important consideration is the criteria to be applied to the samples to be taken. This analysis format consists of a listing of questions and statements calling forth the necessary information required to analyze a sampling system. With this information developed, one can proceed with an evaluation, the methodology of which is also discussed in the paper. Errors in probe placement, failure to sample at the proper rate, delivery line losses, and others are evaluated using mathematical models and empirically derived relationships. Experimental methods are also described for demonstrating that quality sampling will be achieved. The experiments include using a temporary, simple, but optimal sample collection system to evaluate the more complex systems. The use of tracer particles injected into the stream is also discussed. The samples obtained with the existing system are compared with those obtained by the temporary, optimal system.

Introduction

Compliance with Nuclear Regulatory Commission, Environmental Protection Agency requirements, and state regulatory limits on the release of airborne radioactive particles and gases mandates that airborne effluents discharged to the atmosphere be sampled and measured. Although information is available to assist the specification and design of samplers and monitors for particles in an effluent stream, there is no comprehensive analysis format which can be used to assure that a sampler design will achieve its stated objective. Similarly, the evaluation of a system already installed against a stated performance criteria can be accomplished efficiently only when such an analysis format or required data sheet is available. Once the criteria are stated, and the analysis has been

* Work performed under U.S. ERDA Contract E(45-1)-1830.

14th ERDA AIR CLEANING CONFERENCE

completed, using the questions of the analysis format, a sampling installation can be designed, or one already installed can be evaluated.

The evaluation and validation process may require field measurements and demonstrations to support the theoretical evaluation, depending upon the degree of compromise made with good sampling practices.

Objective

The objectives of this paper are: (1) to present an analysis format for a nuclear facility particulate effluent sampling system and (2) to describe some methods for the evaluation of such a system against performance criteria.

Establishing the Sampling and Monitoring Criteria

Those responsible for reporting effluent concentrations to demonstrate compliance with regulatory requirements must determine exactly what the sampling and monitoring system is to accomplish. At first glance this may seem a trivial requirement, but what is required may be a very simple system or a very complex one. The sample may be an inventory sample which will integrate over a 24-hour or a one-week period. On the other hand, the sample may be a monitor-type sample for unusual releases and require a much larger or smaller sampling rate than the inventory-type sample. Are both types needed? The question of the range of particle sizes to be sampled in a representative way should be decided. Even though effluents from nuclear facilities are efficiently filtered, over long periods of operation, the ducts downstream of the filters may accumulate contaminated dust which may have entirely different characteristics than the primary released materials. The question of particle sizes present during a malfunction in the system yielding off-standard conditions must likewise be addressed. The sensitivity of the radiochemical analysis methods for the sample must be carefully considered taking into account the length of time available for analysis and counting the sample for the required accuracy.

Following is a list of statements which taken together will establish the criteria for which a sampling system can be designed or against which an installed sampling and radiochemical analysis system can be evaluated:

1. Name the isotopes to be sampled and measured. (Specific or general such as gross beta, gross alpha emitters.)
2. State the purpose of the sampler installation.
 - a. To obtain an integrated release of radioactivity associated with particles.
 - b. To obtain a sample for continuous monitoring by a detector "looking" at the collected material.

3. State the minimum quantity (μCi) of the isotope(s) in question which must be measured to a given \pm on a stated confidence interval, in a stated period of time. For example:

"The inventory sampler must provide a ^{239}Pu sample over a 24-hour period such that $0.01 \mu\text{Ci}$ can be measured by direct counting a filter in a 10-minute count to $\pm 25\%$ at the 90% confidence level. Detector efficiency is 0.40."

or

"The monitor sample must be withdrawn in a representative way at such a rate that $1 \times 10^{-12} \mu\text{Ci/cc}$ of ^{239}Pu will trip an alarm within 15 minutes."

The statement of sensitivity and accuracy must be consistent and realistic with:

- The total stack flow of effluent and the release limit in terms of curies per day or per week, etc.
- The efficiency of the detector to be used.
- The background counting rate, taking into account energy discrimination in the system, and naturally occurring interferences.
- The counting statistical reliability within the time constraints imposed.

4. State overall accuracy required of sample extraction and delivery to the collector. This statement would cover the acceptable deviation from complete representativeness of the sample extraction and any distortion resulting from line losses, etc. The accuracy may be related to the fraction of the release limit experienced during the sampling period. For example:

at 10% of permitted release, Accuracy: $\pm 100\%$
 at 80% of permitted release, Accuracy: $\pm 20\%$

Compliance under these criteria statements would be determined only after the sampling. A system designed for $\pm 20\%$ at the outset would not fall short even when very low emission levels were experienced.

5. State the range of particle sizes for which the system is to be designed. (Comment: This statement may permit all or some fraction of particles above a certain size to be very poorly sampled. A statement such as "At least 95% of particles $\leq 5 \mu\text{m}$ AED to be extracted and 75% of particles $\leq 10 \mu\text{m}$ and $\geq 5 \mu\text{m}$." Since many sampling systems' efficiencies are highly dependent on particle size, it is necessary to set down a particle size criteria.)

14th ERDA AIR CLEANING CONFERENCE

6. State whether the inventory sample is also to be the continuous monitor sample which would be analyzed in a laboratory after the fact.
7. State the upper limit of the range of the monitoring system. This will help define the electronics of the system to prevent overloading during release incidents.
8. State the emergency electrical back-up system required to assure continuity during power failure.
9. State the requirements for "fail safe" monitoring. Malfunction of sampler air mover, detector failure, etc., should activate an alarm.
10. State required detector calibration frequency and methods.

The Analysis Format and Check List

With the sampling and monitoring criteria established, the design of a new system, or evaluation of an existing sampling system can be undertaken. In either situation, the same information is required. In the case of the new designs the designer has flexibility and many options to adjust the system parameters to meet the design criteria. The format and check list developed below is one to be applied to the evaluation of an already-installed system. It is the purpose of the analysis format and check list to acquire the needed data to insure that the system can be designed or evaluated properly.

Sampler Location and Physical Description

- A. Building number and name, sampler designation.
- B. Ventilation component or exhaust to be sampled. State anticipated radionuclides in exhaust.
- C. Description. Brief description with good, understandable sketch of ventilation system and sampler. Current engineering drawings. (Insure that actual system conforms to drawings.)
- D. Specific information needed.
 1. Air flow rate in stream sampled.
 2. Velocity distribution at section sampled.
 3. Dimensions of duct.
 4. Location of sample withdrawal point(s) with respect to nearest fan, bend, transition, breeching, air filter.
 5. Sampling system materials in direct contact with sampled air.
 6. Inlet sample probe dimensions.
 7. Bends in lines between probe inlet and collector media.

14th ERDA AIR CLEANING CONFERENCE

8. Horizontal delivery lines - lengths, dimensions.
9. Vertical lines - lengths, dimensions.
10. Design sampler flow rate. Velocity in the sampling probe inlet.
11. Is a manifold and secondary sample probe used? Describe.
12. Location and description of sample flow measuring device and recorder, if used. Calibration, or other confirmation of flow rate accuracy taking into account humidity, temperature, and pressure.
13. Provision for eliminating moisture condensation in sampling line.
14. Sample temperature measurement.
15. Measurement or knowledge of temperature and humidity of effluent exhaust air.
16. Description and location of sample pump or aspirator.
17. Are there any provisions for measuring radioactivity associated with different particle sizes? If so, describe.

Sampler Collector Media and Support

- A. Media - glass fiber filters, cellulose, membrane, etc.
- B. Dimensions of filter.
- C. Design face velocity.
- D. Assumed filter efficiency, references.
- E. Description of filter holder.

Sampler Operation

- A. Provide written procedure for start-up and operation of sampler.
- B. Provide written procedure for shut-down and collector removal.
- C. How are collectors protected in transport to counting room?
- D. State sampling time.
- E. Are checks made of equipment and flowmeter during the run? Describe. Are alarms or indicators showing pump failure incorporated in the system? How soon can corrective action be carried out? Is emergency power available in case of power failure?
- F. Is visual appearance of collector recorded, such as color, wet, dry, torn, sound, etc.?
- G. How is filter or other collector identified?

Measurement of Radioactive Constituents on Collector Media (Sample Assayed After Collection)

- A. Sample removal processing.
 1. Collector media sectioned?
 2. Chemical processing, leaching, extraction?
- B. Describe detector system used.
 1. Alpha particle counter, beta particle counter, gamma detector, other? Describe.

14th ERDA AIR CLEANING CONFERENCE

2. Geometrical arrangement for counting radioactivity on collectors.
- C. Describe counter operation.
 1. Give time-length of count, or other index such as total counts taken.
 2. State statistical quality controls on counting system. Describe calibration procedures for specific isotopes and total α , β , and γ counting.
 3. Show how error and confidence limit of sample count are calculated and expressed, taking into account the background of the counter, and length of count. State how "minimum detectable" quantity is determined.
- D. Calculation of 24-hour release to the environment, or other reporting basis.
 1. Define elements of calculation, and the formula used.
 2. Show error estimate for each term in calculation and how determined.
 3. Is an error range determined for the 24-hour release of radionuclides? If so, how is it determined, and what confidence level is used? For example, is the error based on the most significant error in the several elements of the determination, or is the error estimated taking into account propagation of errors in separate, several elements of the calculation?
 4. State the minimum "detectable" 24-hour release, and state the confidence interval with such statements as "...There is only 1 chance in 10 that a release twice this value will go undetected." Convert the release limit into equivalent concentration in the effluent stream sampled in $\mu\text{Ci/cc}$. Explain how the detection limit is determined (Anticipated statistical fluctuations in the background count will provide one reference point for estimating detection sensitivity.)

Records

- A. State how sample results are entered into the official record of released radioactive materials, and the procedure for maintaining this record.
- B. Describe format of this record.

The following section is the analysis format for the monitoring function of the sampling system. In general, the above sections are applicable to the constant monitoring function of a sampling system. Frequently, but not necessarily, the sampler fills a dual role of an inventory of the 24-hour release and a monitor. A detector is located in close proximity to the collector, and the counting rate or accumulated count is recorded at a point in a control room, or other frequently occupied space. Appropriate alarms are provided.

Monitoring Function

- A. Description -- Describe the collector-detector installed in the system.

14th ERDA AIR CLEANING CONFERENCE

1. Geometrical arrangement of collector and detector.
 2. Written and graphic description of pathway of gas and particles through the system. Drawings, sketches, etc.
 3. Describe radioactivity detectors. (Whether α , β , or γ , solid state, GM counters, sodium iodide, etc.) Is discrimination provided to permit only selected radio-nuclides to be sensed?
 4. Flow rate, if not included in section titled "Measurement of Radioactive Constituents on Collector Media (Samples Assayed After Collection)."
 5. Describe signal generated and how displayed. (Count rate instrument, scaler, other integrating system. Is background automatically subtracted, etc.)
- B. Response to radionuclides for which system is designed.
1. State whether total alpha particle, total beta, total gamma, photon detector, or whether discrimination is provided for radiation type and energy.
 2. State method for detector calibration.
 - a. Source used - certification, standardization.
 - b. Frequency (built-in checks, etc.).
 - c. Adjustment to predetermined limits? Discriminator setting, etc.
 - d. Linearity at "high" levels of release. Calibration under this circumstance.
 3. State dynamic range of the instrumentation.
 - a. Anticipated maximum rate of release during accident or other inadvertent releases.
 - b. System response at important action levels. Describe the release levels prompting various actions. State how and how often the monitor is functionally tested at the various levels of concern.
 4. State sensitivity of the system. Provide statement of minimum radioactivity detectable and support the statement in a discussion of background routinely experienced, the detector-collector geometry and intrinsic detection efficiency of the radiation sensor, the sample flow rate and the time required to reach a stated level above background. State the results in terms of the release in μCi , which would be detected above background with a 90% probability. For the design sampling time between filter or other collector replacement, state the minimum detectable release rate and equivalent concentration $\mu\text{Ci}/\text{cc}$ in the effluent stream sampled.

Reliability

- A. State whether a redundant monitor is provided.
- B. Describe provisions for insuring that failure of any critical feature will be signaled.
 1. Sample flow.
 2. Detector components.
 3. Charts.
 4. Alarm circuits.

14th ERDA AIR CLEANING CONFERENCE

- C. Describe availability of emergency power and time sequence to activate.
- D. Describe the time sequence of actions to correct a monitor malfunction. Is the time required to restore the monitor to normal operation consistent with technical specifications or other licensing requirement for operating the facility with continuous monitoring of effluents? Explain the rationale leading to current operating practice.
- E. Describe preventive maintenance procedures which assure reliability.
 - 1. Component functional tests.
 - 2. Parts replacement program.
 - 3. Spare parts and components availability.
 - 4. Other.

Evaluation of Particle Sampling and Monitoring Systems

With the Sampling Criteria defined and with the data supplied from the Analysis Format and Check List, one can proceed to evaluate the system as designed or as it exists. The compliance with many of the sampling criteria can be assessed by simple inspection; however, determining the compliance with criteria governing performance and representativeness requires more detailed attention. In this section are discussed the elements of theoretical and experimental evaluation procedures which will lead to decisions regarding compliance with sampling criteria.

Theoretical Evaluation

With the complete data about the configuration and operation of a sampling system one can proceed with a theoretical (or paper study) evaluation of the system's compliance with the performance and sample representativeness requirements of the sampling criteria. Topics that can be addressed in a paper study include: (1) sample aliquot sizes as regards monitor sensitivity and inventory sample analysis accuracy; (2) extraction caused sample bias; (3) fractional delivery of particulates to the collector; (4) collection efficiency; and (5) the combining of these factors to determine if the samples collected are likely to be representative* of the particulate effluent.

Aliquot Size - Inventory Sample Accuracy. The size of the sample aliquot is an important element to consider when evaluating ability to determine integrated releases with a given accuracy. Some of the factors which determine a minimum sample aliquot (assuming representative samples are collected) are:

* A representative sample is one withdrawn from the bulk stream and delivered to the collector in such a way that the collected sample will have the same radioactivity (particulate) per unit volume sampled as exists in the bulk stream. The degree to which the sample may fail to be representative should be defined in the criteria.

14th ERDA AIR CLEANING CONFERENCE

1. Radionuclides of concern.
2. Minimum concentration of isotope which must be measurable.
3. Time interval over which the concentration is to be averaged.
4. Desired accuracy for the analysis of the collected sample at the minimum required concentration.
5. Analytical techniques.

The requirements of the first four factors are usually specified in the sampling criteria and the last is defined by existing laboratory techniques. A common equation relating many of these factors is:

$$\frac{\mu\text{Ci}}{\text{cc}_{\text{effluent}}} = (\text{net c/min})_{\text{sample}} \times \frac{1}{\text{eff}} \times \frac{1}{\text{sample flowrate, ft}^3/\text{min}} \times \frac{\text{ft}^3}{28,320 \text{ cc}} \times \frac{1}{\Delta t, \text{min}} \times \frac{\mu\text{Ci} \cdot \text{min}}{2.22(10^6)\text{d}} \quad (1)$$

where eff is the counting efficiency in count/disintegration and Δt is the sample collection period. The equation is easily rearranged to express the sample flowrate as a function of the other parameters.

A common equation which relates analysis accuracy to the net counting rate is

$$\text{95\% Confidence Interval Limit} = 1.96 \sqrt{\frac{\text{net c/min} + \text{background c/min}}{\text{sample counting time}} + \frac{\text{background c/min}}{\text{background counting time}}} \quad (2)$$

Given a desired \pm accuracy at the 95% confidence level and knowing the details of the analytical technique, the required net count rate for that accuracy and technique can be calculated. The required net count rate can then be inserted into the rearranged equation (1), along with the collection period, minimum measurable concentration and counting efficiency to calculate a minimum required sample flow rate.

To illustrate the use of these equations an example case will be considered with the following assumptions and specifications:

1. Principal isotope present in particulate form and of concern is ^{239}Pu .
2. Minimum measurable concentration is $6 \times 10^{-14} \mu\text{Ci/cc}$.
3. Sample collection time is 24 hours.
4. Detector counting efficiency is 0.40 c/d.
5. Sample counting time is 15 minutes, and background counting time is 5 minutes.
6. Normal background for detector is 0.2 c/min.
7. Desired count accuracy for measuring the above release is $\pm 10\%$, 95% confidence level.

14th ERDA AIR CLEANING CONFERENCE

Taking the last three assumptions and substituting into equation (2) it can be shown that a net count rate of 27 c/min is needed to achieve the desired counting accuracy. Rearranging equation (1) and substituting in the first four assumptions, the minimum required sample flow rate is:

$$\begin{aligned} \text{Sample flow rate} &= 27 \text{ net c/min} \times d/0.4 \text{ c} \times \frac{1}{1440 \text{ min}} \\ &\times \frac{\text{cc}}{6 \times 10^{-14} \text{ } \mu\text{Ci}} \times \frac{\text{ } \mu\text{Ci min}}{2.22 (10^6)d} \times \frac{\text{ft}^3}{28,320 \text{ cc}} \quad (3) \\ &= 12.4 \text{ ft}^3/\text{min} \end{aligned}$$

The existing sample flow rate or the design sample flow rate can then be compared to the calculated flow rate. In some cases, the required flow rate is impractical, this indicates that the specifications of the sampling criteria need to be reconsidered or improvement in the analytical technique needs to be sought. It should be remembered that the calculated sample flow rate is a minimum value because only counting errors are considered and the collected samples are assumed to be representative.

Aliquot Size - Monitor Sample Sensitivity. Many of the factors which affect the monitor sensitivity are the same as those which affect the inventory sample accuracy. They are:

1. Radionuclides of concern,
2. Minimum concentration which must be detected,
3. Time interval within which the minimum concentration must be detected,
4. Counting time (if not continuous),
5. Background level at monitor,
6. Monitor counting efficiency,
7. Sample aliquot size.

Again, the first three factors are usually specified in the sampling criteria and the second three are usually dictated by the available monitoring equipment. Thus, the size of the sample aliquot again becomes an important element in monitor sensitivity and is dependent on the other six factors. With the assumption that ideally representative samples are collected, a minimum required sample flow rate can be calculated once the other factors are specified. The existing sample flow rate or proposed flow rate can then be compared to the calculated minimum.

To give an example of such a calculation the following assumptions will be made:

1. The radionuclide to be monitored is ^{239}Pu .
2. A minimum average concentration of $10^{-13} \text{ } \mu\text{Ci/cc}$ must be detectable above background within a 24-hour collection period.
3. The monitor functions continuously and uses energy discrimination to analyze for ^{239}Pu .

4. The monitor counting efficiency is 0.1 c/d, the background level is 1 c/min, and the detectable response above background is 5 c/m.

The required sample flowrate is then calculated as follows:

$$\begin{aligned} \text{Sample Flowrate} &= \frac{\text{cc}}{10^{-13} \mu\text{Ci}} \times \frac{\text{ft}^3}{28,320 \text{ cc}} \times \frac{\mu\text{Ci min}}{2.22(10^6)\text{d}} \times \frac{\text{d}}{0.1 \text{ c}} \\ &\times \frac{1}{1440 \text{ min}} \times 5 \text{ c/min} = 5.5 \text{ ft}^3/\text{min}. \end{aligned} \quad (4)$$

Again, if in some cases the required minimum flow rate is impractical to achieve, then the specifications of the criteria and the monitoring instrumentation need to be reconsidered.

Extraction-Caused Sample Bias. A bias in collected particulate samples can be caused by the process of sample extraction.* The magnitude of the bias is dependent upon the different sampling conditions, effluent conditions, and particle sizes. Extraction-caused bias can be the result of nonisokinetic extraction or a poor choice of extraction location(s).

Nonisokinetic Extraction Bias. Because particulates do not exactly follow the flow of the gas, it is desirable to minimize flow disturbances and direction changes caused by extraction nozzles. Sample bias caused by the sample gas velocity being different from the approaching gas velocity has been observed by several workers.^[1,2,3,4] Matching the sample airstream velocity at the nozzle to the approaching airstream velocity is known as isokinetic sampling. Several models have been developed to correlate bias, degree of isokinesis, and particle size.^[2,5] The following model by Davies^[4] relates the bias or ratio of sample particle concentration to approach particle concentration, C/C_a , with a function of velocities and particle characteristics,

$$\frac{C}{C_a} = 1 + \left[\frac{V_a}{V_s} - 1 \right] \left[\frac{2 \text{ Stk}}{1 + 2 \text{ Stk}} \right]. \quad (5)$$

In this equation:

- C = concentration in the sample,
- C_a = actual concentration in the stream,
- V_a = gas velocity approaching the nozzle,
- V_s = velocity in the inlet of the sample probe,
- Stk = Stokes' number, the ratio of twice the stopping distance to the nozzle inlet diameter.

* Extraction is herein defined to be the process of separating the sample aliquot from the effluent.

Stokes' number is calculated from the expression:

$$\text{Stk} = \frac{v_a d_p^2 \rho_p}{9 \mu d_o} \quad (6)$$

in which

- d_p = diameter of the particle in cm,
- ρ_p = density of particle in g/cm³,
- μ = viscosity of air in poise,
- d_o = diameter of inlet nozzle in cm.

It can be shown from the above equations and for a given extraction configuration that the bias becomes more significant as the particle size increases.

Location Caused Extraction Bias. The magnitude of location caused extraction bias is not predicted by calculations or theoretical considerations; however, guidelines exist which can indicate whether or not representative samples can be extracted from a given location.^[6] In general the ideal sampling location in a stack or duct is in a vertical run 10 duct diameters and 5 duct diameters from the nearest significant upstream and downstream flow disturbances respectively. It is theoretically possible to obtain a representative sample from a single location; however, it is difficult to find that location, especially if the velocity profile is at all complicated. Instead of finding a single ideal location it is recommended to make up the total sample from samples extracted at various points on the location cross section^[5], or to use some sort of moving probe.

Fractional Delivery of Particles to the Collector. The concentration of sampled particulates is further altered by the deposition and possible later resuspension of particulates in the piping and fittings through which the sample passes before reaching the collection point. The effects of particulate deposition in fittings, valves, transitions, cavities, and later particulate resuspension upon the sample representativeness cannot be predicted by present models. However, there are some models that can be used to estimate the magnitude of concentration modification in smooth horizontal and vertical tubes and in bends. With these models one can find indications, at least, of portions of sample delivery systems where sample modification occurs. The following are some of the available models to aid in the evaluation. (The reader is referred to the references cited for the detailed development of the models.) It can be shown with all the following models that the concentration modification is strongly dependent on the particle sizes that are being considered.

14th ERDA AIR CLEANING CONFERENCE

Gravity Settling in Laminar Flow. The following model was developed [7, 8] for calculating the fractional penetration (or concentration modification) of aerosols in laminar flow in horizontal tubes.

$$C/C_a = 1 - \frac{2}{\pi} \left[2Z\sqrt{1 - Z^2/3} + \arcsin (Z^{1/3}) - Z^{1/3}\sqrt{1 - Z^2/3} \right] \quad (7)$$

$$Z = 0.75 \frac{V_{gs} L}{V_a D} \quad (\text{dimensionless}) \quad (8)$$

V_{gs} = terminal gravitational settling velocity

V_a = velocity in tube (laminar flow conditions)

L = length of duct

D = diameter of duct,

and

$$V_{gs} = \frac{g d_p^2 (\rho_p - \rho)}{18\mu} \quad (9)$$

g = acceleration due to gravity

d_p = particle diameter

ρ_p = particle density

ρ = gas density

μ = gas viscosity.

When $Z \leq 0.2$, equation (6) is approximated within 2% by: [10]

$$C/C_a = e^{-1.7Z}. \quad (10)$$

Bend Losses in Laminar Flow. A model for predicting impaction efficiency of aerosols in laminar flow in bends has been proposed by Cheng and Wang. [9] Adapting the model for the calculation of particulate concentration modification for aerosols with Stokes' Number < 0.1 yields;

$$C/C_a \approx 1 - \left(1 + \frac{\pi}{2R_o} + \frac{2}{3R_o^2} \right) Stk, \quad (11)$$

where R_o is the radius of curvature of the bend divided by the tube radius.

Gravity Settling-Turbulent Flow. The models for calculating the concentration modification in turbulent flow and for various particle sizes are of the general form^[10]

$$C/C_a = e^{-AL^*} \quad (12)$$

where A is a dimensionless loss factor and L* is the dimensionless tube length. Generally, the dimensionless loss factor is a ratio of settling velocity and gas velocity. For the gravity settling problem

$$A_g = \frac{4}{\pi} \frac{V_{gs}}{V_a}, \quad (13)$$

where V_{gs} is the settling velocity which can be calculated by equation (9).

The dimensionless tube length, L*, is given by

$$L^* = L/D \quad (14)$$

Turbulent Impaction. Turbulent impaction is generally taken into account for turbulent flow in both horizontal and vertical tubes. From the empirical correlation proposed by Sehmel^[11] the following equation for the dimensionless loss factor is derived^[10]:

$$A_t = 1.169 \times 10^{-16} \text{ sp.gr.}_p^{1.01} \text{ Re}^{2.9} R^{2.10} \quad (15)$$

Again, the concentration modification is calculated by equations (12) and (14).

For horizontal tubes the gravity settling and turbulent impaction models are combined to calculate the concentration modification. The two models are simply combined as follows:

$$A_{\text{total}} = A_t + A_g \quad (16)$$

and

$$C/C_{a(\text{combined})} = e^{-A_{\text{total}}L^*} \quad (17)$$

For vertical tubes the turbulent impaction model alone is used to calculate a concentration modification.

Bend Losses in Turbulent Flow. For the case of turbulent flow through bends the following simple model is proposed to give an indication of concentration modification.^[10] The model is essentially similar to that for the gravity settling problem. The dimensionless loss parameter is defined as:

$$A_b = \frac{4}{A} \frac{V_{cs}}{V_a} = \frac{2}{9} \frac{\rho_p d_p^2 V_a}{\mu R_b}, \quad (18)$$

where V_{cs} is the centrifugal settling velocity. The centrifugal settling velocity is similar to that for gravity settling except that the acceleration of gravity is replaced by the centrifugal acceleration which is approximated by V_a^2/R_b .

Again the concentration modification is calculated by equation (12) where,

$$L^* = \frac{\pi}{180^\circ} \frac{\theta R_b}{D} \quad (19)$$

Sample Collection Efficiency. The efficiency of the collection filter is an important factor to be considered in the evaluation of sample representativeness. Included in the consideration of efficiency is the effect of self absorption of the filter material upon the counting efficiency. Unfortunately, the collection efficiency and self absorption have to be determined experimentally; however, the collection efficiencies for several filters and particle sizes have been reported in the literature, and can be factored into the theoretical evaluation.

Combining Concentration Modifications. Many sample delivery systems include one or more bends and horizontal and vertical runs of tubing. The calculated concentration modifications for each segment of the system plus the nonisokinetic extraction bias can be combined in most cases (except when extraction occurs more than once) by a simple calculation. The total system concentration modification can be calculated as follows:

$$C/C_a \text{ (total system)} = \prod_{i=1}^{n \text{ segments}} (C/C_a)_i \quad (20)$$

The combined concentration modification can indicate whether or not the samples collected by an existing or proposed system will be representative of the particulate effluent to the extent desired.

Experimental Methods for Evaluation of Sampling Systems

A second approach to the evaluation of an existing sampling system is to conduct a series of experiments to demonstrate the representativeness of the collected samples. A reasonable way to do this is to collect samples of the effluent with a standard sampling device while at the same time samples are being collected with the existing system. The concentration data from the standard and routine samples can then be compared to give an indication of the representativeness of the samples collected with the existing system. The following discussions will address some of the preparations necessary to collect the standard samples and offer some suggestions as to what can be done with the samples to allow comparisons of the data.

Preparations for Obtaining Standard Samples. One of the major preparations for standard sampling is to choose as ideal a sampling location as possible. The guidelines mentioned earlier under the heading "Location Caused Extraction Bias", can be helpful in finding a satisfactory location. The access at the chosen location must be large enough for the insertion of sampling probe and/or filter support.

Once the location is chosen, it is necessary to learn as much as possible about the condition of the effluent at that location. A complete velocity profile of the duct cross section is necessary and a temperature and pressure profile will be helpful as well (for determining mass flux). It is also valuable to determine the effluent humidity and dewpoint so steps can be taken to avoid moisture condensation in sampling lines and on the collected samples.

After the above information is obtained, it is necessary to determine the concentration profile of the particulate effluents in the available cross section. This can be found by simultaneously collecting samples from several points in the cross section. With the data from several sets of simultaneous samples, significant differences from point to point may be determined by constructing a two-way classification analysis of variance table. Then one or more "standard" sampling points can be selected.

The sampling configuration for the concentration profile experiment and for the following comparison experiments should be as simple as possible. Each sample can be collected by fixing a tapered nozzle to the front of a filter holder and suspending the assembly in the duct. The inside and outside tapers of the nozzle should be no more than 15° to minimize the airflow disturbance and particle deposition on the inside wall. The filter should be chosen to have a high collection efficiency for the particulate sizes expected and to permit a sufficient sample flow rate. The vacuum pumps and flow control equipment can be located exterior to the duct. The flow rate and nozzle inlet size should be adjusted so each sample can be extracted isokinetically for the

velocity at each sampling point. Prior to analyzing the concentration profile data the data must be normalized for differences in sample flow rate and any other differences at each location.

Information about the size distribution of the particulates of interest is also helpful for interpreting the results of the comparison tests because the performance of a sampling system is highly dependent upon the sizes of the particles it is expected to sample. Particle size data can be obtained with the use of light scattering devices for particle sizing, microscopic analysis of particles collected on filters, or cascade impactors. The cascade impactor is the preferred device because the aerosol size fractions are collected on separate samples which can then be analyzed for the element, compound, or radionuclide of interest. The particle sizing samples should be extracted from the same location as the standard samples.

Comparison of Sampling Data. Once a location is found and equipment assembled to obtain the standard samples the performance of the existing sampling equipment can be tested. The collection intervals for the standard samples should be the same as for the sampling system being tested. Also, the samples collected by both systems should be analyzed for the particulates of concern in the same laboratory and with the same procedure, and the data should be normalized for differences in sample flow rates.

There are several ways the two sets of data can be analyzed and the sampling criteria that are being used will influence the choice of method. One method is to perform a linear regression on the data to see if a constant or percent bias exists. If the regression line does not pass through the origin, a constant bias exists between the routine and "standard" samplings. If the regression line passes through the origin, but with a slope other than one, then a percent bias is indicated. A second method is to calculate the difference in each sample pair and then calculate the mean difference. Hypotheses on the mean difference can then be tested. Another possibility is to perform statistical manipulation on the ratios of each sample pair if the ratios can be shown to be normally distributed. Methods other than these three may also be appropriate to test if the samples collected with an existing system comply with the chosen sampling criteria.

Use of Tracer Aerosols. As is common for the effluents of some nuclear industry facilities, the concentrations of the particulates of concern are too low to permit the collection of samples and the completion of such an experimental program as the above in a reasonable length of time. In such instances a tracer aerosol injected into the effluent may help to expedite the experiments. Some of the desirable characteristics of the tracer aerosol are:

1. Nontoxic in the quantities used,
2. Inert to the effluent and sampling system environments,
3. Not already present in the effluent or collection media to any appreciable extent,

14th ERDA AIR CLEANING CONFERENCE

4. Same particle size distribution upon generation as the particulates of concern or of the same size which corresponds to the requirements of the sampling criteria,
5. Easy to analyze on collected samples with an available technique.

The tracer aerosol should be injected into the effluent stream at a point far enough upstream of both the existing and standard sampling systems to allow for thorough mixing of the tracer with the effluent.

Also, it is important that the device used to generate the aerosol can be used in the field and can generate the required amount and required rate reproducibly.

Summary and Conclusions

An evaluation of an existing or proposed particulate effluent sampling system includes the following elements.

- It is essential to decide what function the sampler is to perform and what defines successful performance of that function.
- Information must be gathered concerning all the factors affecting sampler performance. Some of the factors mentioned in the paper are: sampler location, sampler environment, sample collection medium, sampler construction, and sample analysis and monitoring.
- The evaluation then proceeds as either or both a theoretical or experimental study. In the theoretical study the effects of aliquot size, extraction bias, particle deposition, and other factors upon both the representativeness of collected samples and accuracy of sample analysis or monitoring can be estimated. In the experimental study the samples collected with the existing system are compared with simultaneously collected standard samples to estimate the performance of the existing system. Tracer aerosols have been found to be a useful tool in the experimental study because of the cleanliness of nuclear facility effluents.

Many of these elements of the evaluation method can also be useful to the designer of sampling systems because the effects of design upon the sampler performance can be predicted.

Nomenclature

- A = dimensionless loss factor
- A_b = dimensionless loss factor for bends
- A_g = gravity settling dimensionless loss factor
- A_t = turbulent deposition dimensionless loss factor

14th ERDA AIR CLEANING CONFERENCE

C = aerosol concentration
C_a = original aerosol concentration
D = tube inside diameter
L = length
L* = dimensionless length
R = ratio of particle size in microns to tube diameter in centimeters
R_b = bend radius
R_o = ratio of bend radius to tube inside diameter
Re = tube Reynolds' Number
Stk = particle Stokes' Number
V_a = average gas velocity
V_{cs} = particle settling velocity in bends for centrifugal acceleration.
V_{gs} = particle terminal settling velocity
V_s = sample airstream velocity inside nozzle orifice
Z = dimensionless settling factor for laminar flow
c = counts
cc = cubic centimeter
d = disintegration
d_p = particle diameter
d_o = probe nozzle orifice diameter
eff = counting efficiency, c/d
ft³ = cubic feet
g = acceleration due to gravity
min = minute
sp.gr._p = particle specific gravity
Δt = elapsed sample collection time

14th ERDA AIR CLEANING CONFERENCE

θ = angle of bend, degrees

μ = gas viscosity

μCi = microcurie

ρ = gas density

ρ_p = particle density

14th ERDA AIR CLEANING CONFERENCE

References

- [1] S. Badzioch, "Collection of Gas-Borne Dust Particles by Means of an Aspirated Sampling Nozzle," British Journal of Applied Physics, Vol. 10, January 1959.
- [2] H. H. Watson, "Errors Due to Anisokinetic Sampling of Aerosols," American Industrial Hygiene Association Quarterly, Vol. 15, March 1954, pp. 21-25.
- [3] S. P. Belyayev and L. M. Levin, "An Experimental Investigation of Aerosol Aspiration," translated by J. Findlay from Izv. Akad. Nauk. Ser., Atmospheric and Oceanic Physics, Vol. 6, No. 11, 1970, p. 1137.
- [4] G. A. Sehmel, "Particle Sampling Bias Introduced by Anisokinetic Sampling and Deposition Within the Sampling Lines," American Industrial Hygiene Assoc. Journ., Vol. 31, Nov.-Dec., 1970, pp. 758-771.
- [5] C. N. Davies, "The Entry of Aerosols into Sampling Tubes and Heads," Brit. J. Applied Physics, Ser. 2, 1 p. 921-932.
- [6] American National Standards - ANSI N13.1-1969, Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities, American National Standards Institute, New York, NY, (1969).
- [7] G. Natanson, Quoted by N. A. Fuchs, The Mechanics of Aerosols, Pergamon Press, Oxford, England, 1964, p. 112.
- [8] J. W. Thomas, "Gravity Settling of Particles in a Horizontal Tube," J. Air Pollution Control Association, Vol. 8, pp. 32-34, 1958.
- [9] Yung-Sung Cheng and Chiu-Sen Wang, "Inertial Deposition of Particles in a Bend," Aerosol Science, Vol. 6, pp. 139-145, 1975.
- [10] L. C. Schwendiman, G. E. Stegen, and J. A. Glissmeyer, "Methods and Aids for Assessing Particle Losses in Sampling Lines," presented at the American Industrial Hygiene Association Meeting, Minneapolis, Minnesota, 1975. BNWL-SA-5138, Battelle-Northwest, Richland, Washington, 1975.
- [11] G. A. Sehmel, "Particle Deposition from Turbulent Air Flow," Journal of Geophysical Research, Vol. 75, No. 9, pp. 1766-1781, 1970.

14th ERDA AIR CLEANING CONFERENCE

THE USE OF A SINGLE PARTICLE INTRA-CAVITY LASER PARTICLE SPECTROMETER FOR MEASUREMENTS OF HEPA FILTERS AND FILTER SYSTEMS*

B.G. Schuster and D.J. Osetek
Los Alamos Scientific Laboratory
University of California
Los Alamos, New Mexico 87545

Abstract

Current tests of HEPA filters and/or filter installations using DOP aerosols and conventional forward light-scatter photometers are limited to measuring protection factors of 10^4 to 10^5 . In addition, forward light-scattering photometers have markedly decreased sensitivity to $<0.3 \mu\text{m}$ particles and basically measure only a scattering signal which is not uniquely related to any given concentration or size distribution of scatterers. These limitations require that high efficiency systems, such as multiple stage HEPA filters, be evaluated one stage at a time, a procedure which is quite often impractical for many existing air cleaning systems and which may be in error.

In order to obviate these difficulties, a single particle intra-cavity laser particle spectrometer has been used to measure protection factors of up to 2.4×10^8 for multiple HEPA systems and individual double ply HEPA filters. Because of the instrumental size resolution, protection factor as a function of particle size can be determined from $.06 \mu\text{m}$ to $2.9 \mu\text{m}$. The lack of background enables single counts to be statistically significant. Since coincidence errors occur at particle concentration $>10^6/\text{liter}$, a known dilution must be introduced to measure challenge concentrations greater than this. The dilution measurement may be accomplished with the aid of a forward light-scattering photometer.

I. Introduction

Test methods for HEPA filter installations currently consist of testing individual stages of multi-stage systems. The method currently in use consists of introducing a challenge aerosol of DOP upstream of the stage to be measured and making light-scattering measurements with a forward light-scattering photometer upstream and downstream of the filter bank. The ratio of the two scattering signals is then used to determine the protection factor (PF) or penetration of the stage. Light-scattering photometers currently in use have an effective dynamic range limited, primarily by sensitivity, to 10^4 , hence limiting the measurable PF to this range. The PFs thus measured are simply a ratio of light-scattering signals which are not uniquely related to either the concentration or size distribution of the population of scatters. Since the filtration process changes the size distribution, the PFs determined in the above fashion are strictly

*Work supported by the U.S. Energy Research and Development Administration.

14th ERDA AIR CLEANING CONFERENCE

valid only for monodisperse aerosols, and then only if size changes due to coagulation and/or evaporation do not occur. High efficiency systems such as multi-stage installations can at best be evaluated only on a single stage basis, a procedure, which, because of some present designs as well as new design criteria,¹ is quite often impractical and may result in defining an incorrect PF.

A method for accurate testing of PFs to 10^8 , e.g., two-stage systems, would obviate the above difficulties and in addition decrease operational downtime and worker exposure. The design of new facilities, e.g., installations using doubleply HEPA filters, should also become simpler and less expensive. The following results, on a laboratory scale, show the feasibility of one such method.

II. Design Criteria and Apparatus

The design criteria for the laboratory scale experiment were predicated on the method to be used for measurement of downstream aerosol concentration. Considering the protection factor desired an initial challenge of 10^9 particles/liter would be attenuated, after passage through 2 HEPA filters, to as little as one particle/liter. Greater challenge concentrations (of approximately $0.3 \mu\text{m}$ CMD) would load the first filter so excessively that the testing procedure would be destructive. A conventional forward light-scattering photometer² is incapable of measuring the low concentrations anticipated downstream of two HEPA filters. Filter sampling, even with a tracer aerosol such as uranine, would require impractically long sampling periods to obtain a detectable concentration. The required sensitivity dictated a device capable of counting and measuring single particles.

A sensitive, laser operated, optical single particle counter and spectrometer was chosen as the detector. This device relies on the scattering of laser light at 633 nm within the optical cavity of a He-Ne laser. The He-Ne laser, being a low gain device, has an intracavity radiation field some 500 times as great as the rated output power. It is the scattering³ of this intense field by a particle, which is detected and measured. The detector signals are sorted and stored in a 16-channel multi-channel analyzer. By use of a range selection switch, a size spectrum from $.06 \mu\text{m}$ to $2.9 \mu\text{m}$ may be measured in concentrations as high as 10^7 /liter with less than 1% coincidence error.

A laboratory wind tunnel was constructed for testing two $20.3 \text{ cm} \times 20.3 \text{ cm} \times 10.2 \text{ cm}$ ($8'' \times 8'' \times 4''$) HEPA filters in tandem (Fig. 1). This system is simply a scaled-down version of a typical HEPA industrial system, the flow volume per unit filter surface area being the same in both cases. In this manner, the feasibility of the measurement could be verified without the necessity of first building a high volume challenge aerosol generator. Clean air was provided by filtration of room air through a $30.5 \text{ cm} \times 30.5 \text{ cm} \times 15.3 \text{ cm}$ ($12'' \times 12'' \times 6''$) filter. The design airflow was 25 cfm and appropriate orifice plates for flow measurements, isokinetic probes for sampling, and pressure gauges at each filter station were incorporated into the system. In order to enable measurements of the challenge concentration upstream of the first filter, a diluter stage consisting of a second wind tunnel was also constructed so that 0.2% of the challenge aerosol could be sampled by the diluter and measured by the optical

single particle counter. The dilution ratio is variable and is confirmed by either filter sampling or photometer sampling of the challenge and diluted aerosol. A six stem (24 jet) Collison type aerosol generator was constructed to provide the challenge aerosol.

III. Challenge Aerosol Characteristics

Two different aerosols have been characterized for their use in laboratory experiments. Both of these were generated by the six stem Collison generator described above. The solid aerosol was produced from a 5% solution of NaCl in water. Its characteristic spectrum (Fig. 2) is quite different than would be expected from a log-normal distribution; in particular, the low end of the spectrum shows no tendency to return to zero, i.e., the distribution is monotonically decreasing.

The spectral characteristics (Fig 3) of an aerosol generated from a 5% DOP in ethyl alcohol solution are very similar to those produced by the NaCl solution. Both distributions may be quite well approximated by the function

$$\frac{\ln N(D)}{\ln N(D_0)} = \exp \{-k(D-D_0)\} \quad \text{where,} \quad (1)$$

$N(D_0)$ is the count at smallest size D_0 , and $N(D)$ is the count at some other diameter, D . The constant, k , may be obtained from fitting the curve, and in the case of the DOP aerosol, is equal to 3.21.

These distributions are different from those measured with an earlier model spectrometer. This discrepancy is presumably due to the much higher resolution and sensitivity of the present detector, which does not decrease at the small end of the spectrum.

The aerosol generator design figure of approximately 10^9 particles/liter was verified with the single particle spectrometer.

IV. Single HEPA Filter Characteristics

Sodium Chloride Aerosol

One of the problems anticipated in challenging HEPA filter systems with extremely high challenge aerosol concentrations was destruction of the performance of the first filter due to loading, particularly with a solid aerosol. In order to determine HEPA performance under these conditions, a filter with DOP quality control protection factor of 8.3×10^3 was monitored for 11 sequential 2-min periods, for a total of 22 min, using the aerosol spectrometer. The penetration of this filter decayed exponentially with a half-life of 4.5 min. This same penetration decay was consistent for all size ranges, i.e., penetration was not size dependent for particles between 0.1 and 1.0 μm (Fig. 4). Extrapolation to zero time sampling yielded a count protection factor of 9.8×10^4 , an order of magnitude greater than determined by DOP quality control testing.

A similar experiment was conducted with a second filter with a DOP quality control determined protection factor of 7.1×10^3 . The

penetration for this filter remained constant for 8 min and then assumed an exponential decay with half-life of 1.9 min as shown in Fig. 5. Spectral decomposition of the decay rates indicated some variation as a function of size, but probably within the figure of merit associated with the data. The initial constant penetration was probably due to a leak which finally was plugged by the challenge aerosol. Even so, the measured protection factor was 1.1×10^5 , more than an order of magnitude greater than the DOP figure. Eight sets of filter decay data provide widely scattered penetration decay rates with a range of half-lives of 1.6 min to 13.5 min, with an average of about 4 min.

While performing these and subsequent tests, substantial leaks were discovered in the cases of several of the test filters. A coating of RTV cement was sufficient to stop the leaks.

One area of interest in multiple filter test studies is the variation of efficiency with size of the challenge aerosol. If such a variation exists, a challenge aerosol distribution will be modified so that the least efficiently collected part of the aerosol spectrum will be the challenge for the second filter in a tandem configuration. Hence, if a gross measurement of PF is made for each of two filters separately (as is now the case), the product of these measurements will overestimate the system PF.

Four individual filters were challenged with the aerosol distributions displayed in Figs. 2 and 3. The quality control penetration data for these four filters were within a factor of two of each other. Three of the filters were challenged with NaCl aerosol, and one with DOP aerosol. Protection factors per unit size interval were obtained by dividing the challenge aerosol count/unit time/size channel by a similar count in the downstream flow. The results of these measurements are plotted as PF against size (Fig. 6.). It is immediately manifest that these filters are extremely efficient for particles less than $0.10 \mu\text{m}$. Protection factor reaches a minimum at about $0.19 \mu\text{m}$ and begins to increase again beyond $.23 \mu\text{m}$. These results are in consonance with theoretical arguments which predict enhanced collection of very small particles and large particles due to Brownian diffusion and direct impaction respectively. The inference to be drawn from these results is that for test results to be at least grossly reliable, a substantial portion of the test aerosol must be in the range $0.1 \mu\text{m}$ to $0.23 \mu\text{m}$ for particle count/size measurements. For nonsize discriminating instruments such as a forward light-scattering photometer, where the signal may be proportional to the second or third moment of the diameter, the challenge aerosol should not exceed $0.23 \mu\text{m}$ and preferably should be within the limits of $0.1 \mu\text{m}$ to $0.23 \mu\text{m}$.

DOP Aerosol

Similar HEPA filter performance experiments were conducted using the DOP spectrum illustrated in Fig. 3. The loading characteristics of these filters challenged by DOP are very much different than for the NaCl aerosol. Referring to Figs. 7 and 8, penetration initially drops, then slowly climbs to near its starting value. The system is flushed with clean air for 30 min before DOP is re-introduced, at which time the penetration increases. It was

14th ERDA AIR CLEANING CONFERENCE

initially thought that liquid DOP, blown off the backside of the filter was responsible for the first increase in penetration. However, the subsequent drying period (DOP vapor pressure is 8% that of H₂O at STP) and further increase of penetration discounts this supposition. Examination of the filter media under high magnification before and after exposure to DOP indicated no obvious change in structure.

The variation of protection factor with size is displayed in Fig. 9. The performance against DOP is very similar to that of NaCl, with the minimum occurring for the same size. The minimum value is again approximately that given by the quality control test.

V. Multi-Ply HEPA Filter Tests

Several filters constructed with a multi-ply HEPA filter medium were supplied for evaluation. The utility of these filters would appear to be in their use in compact filtration systems, in reducing down time for changes (fewer filters), reducing potential personnel exposure (fewer changes), and decreasing total number of potential leaks. A problem would exist with conventional testing since the PF for two plies would be expected to be approximately equal to the product of the PFs per individual ply, i.e., in excess of 10⁶, and this could not be quantitated with existing test techniques.

The filters were tested with NaCl or DOP aerosols. Loading of the filter, particularly by NaCl, changes the characteristics of the filter sufficiently so that a subsequent test with DOP would not be meaningful.

The test results for the filters are summarized in the first two rows of Table I.

TABLE I
PENETRATIONS OF SELECTED FILTERS

| Filter No. | Pressure Initial | Drop Final | Tested Penetration (50 CFM) .3 μm QC(DOP) | Measured Penetration @LASL (25CFM) |
|------------|------------------------|------------------------|--|---------------------------------------|
| A452698 | .6" H ₂ O | .8" H ₂ O | .002% | (.000011 ± .000003)% (NaCl) |
| A452696 | 1.39" H ₂ O | 1.40" H ₂ O | .004% | (.0000014 ± .0000005)% (DOP) |
| 34652 | .72" H ₂ O | .76" H ₂ O | .008% | (.0031 ± .0002)% (DOP) |

The initial and final pressure drops indicate the loading effects of the aerosol over the 15 min test period. The loading is substantial for NaCl. For comparison purposes, the data for a conventional single ply filter tested with DOP is shown in the third row of Table I.

The gross discrepancy between these results and the QC test results are principally due to the lack of sensitivity of the forward scattering photometer used in the DOP QC test. Readings of 10⁻⁵% simply get lost in the noise of the forward scattering photometer.

14th ERDA AIR CLEANING CONFERENCE

VI. Tandem HEPA Filter Tests

A series of measurements was performed on two stage HEPA filter configurations using both 5% NaCl in water and 5% DOP in alcohol to generate the challenge aerosols. The procedure used is as follows. Background counts downstream of each filter section were made with the aerosol spectrometer and external leaks sealed until the count rate was less than 1 per 10-min interval. This was done with a 25 cfm airflow with no aerosol generation. With the generator turned on, a 1-min sample was taken downstream of the first filter. A subsequent 10-min sample was then taken downstream of the second filter. Samples were then obtained downstream of the first filter for 10 more 1-min sample periods so that an interpolated (back to time zero) penetration decay curve could be obtained. A 1-min sample was taken at the diluter stage and the dilution factor was determined by photometer and/or filter samples obtained between the diluter and the main wind tunnel.

Table II summarizes the results of the 6 sets of filters measured.

P is the protection factor, the subscripts refer to the position of the filter (1, 2 or 2 + 1), and whether this is a DOP quality control determined (s) or measured (m) value. The values of P_{21m} are actual measured operational values integrated over a 10-min interval and have not been corrected for the penetration decay of the first filter. Such a correction would decrease P_{1m} and would be applied to P_{21m} hence decreasing it. The values of P_{2m} are derived from the ratio of P_{21m}/P_{1m} . To reiterate, P_{1m} is the zero-time first filter protection factor, and P_{21m} is the overall factor for a 10-min period during which the penetration of the first filter is constantly decreasing. The product of P_{1s} and P_{2s} yields P_{21s} . During an 11-min period of testing, the pressure drop across the first filter increases by about 15%. This loading makes it mandatory that, at least with a NaCl challenge aerosol, the testing period be kept as short as possible. It may be that a DOP challenge aerosol will not present this problem. This hypothesis will be checked.

These measurements indicate that overall protection factors as high as 2.5×10^8 can be measured during a 10-min integration period. A physically less dense aerosol coupled with a spectrometer sensitive to $0.05 \mu\text{m}$ particles may extend the measurements to 1×10^9 .

Testing of similar configurations with DOP provided more information because the tests could be run for a longer period of time due to the different loading characteristics produced by DOP. Table III provides histories of the PF as a function of time. Unlike the systems tested with NaCl, where the loading on the first filter continually increased the PF to a value greater than would be expected from the quality control data, the DOP tested systems usually display a PF an order of magnitude less than inferred from the quality control data. The variation with time is not monotonic. The figure of merit associated with the counts determining P_{21m} in both tables are typically $\approx 25\%$. Note the value of 3.03×10^7 in the fourth row.

14th ERDA AIR CLEANING CONFERENCE

TABLE II

TANDEM PROTECTION FACTORS (NaCl)

| <u>Quality Control (0.3 μm DOP)</u> | <u>Measured (NaCl)</u> |
|--|---|
| I | |
| P _{1s} 7.2 x 10 ³ | P _{1m} 1.1x 10 ⁵ |
| P _{2s} 1.3 x 10 ⁴ | P _{2m} 1.5 x 10 ³ |
| P _{21s} 9.4 x 10 ⁷ | P _{21m} 1.6 x 10 ⁸ |
| II | |
| P _{1s} 1.3 x 10 ⁴ | P _{1m} 1.7 x 10 ⁴ |
| P _{2s} 1.0 x 10 ⁴ | P _{2m} 8.2 x 10 ² |
| P _{21s} 1.3 x 10 ⁸ | P _{21m} 1.4 x 10 ⁷ |
| III | |
| P _{1s} 1.0 x 10 ⁴ | P _{1m} 3.2 x 10 ⁴ |
| P _{2s} 4.2 x 10 ³ | P _{2m} 6.6 x 10 ³ |
| P _{21s} 4.2 x 10 ⁷ | P _{21m} 2.1 x 10 ⁸ |
| IV | |
| P _{1s} 1.3 x 10 ⁴ | P _{1m} 2.8 x 10 ⁵ |
| P _{2s} 4.2 x 10 ³ | P _{2m} 1.5 x 10 ² |
| P _{21s} 5.5 x 10 ⁷ | P _{21m} 4.1 x 10 ⁷ |
| V | |
| P _{1s} 8.3 x 10 ³ | P _{1m} 4.7 x 10 ⁴ |
| P _{2s} 4.2 x 10 ³ | P _{2m} 5.1 x 10 ³ |
| P _{21s} 3.5 x 10 ⁷ | P _{21m} 2.4 x 10 ⁸ |
| VI | |
| P _{1s} 1.3 x 10 ⁴ | P ₂₁ >1.3 x 10 ⁵ |
| P _{2s} 4.2 x 10 ³ | P _{2m} >1.8 x 10 ³ |
| P _{21s} 5.5 x 10 ⁷ | P _{21m} >2.4 x 10 ⁸ |

14th ERDA AIR CLEANING CONFERENCE

Under conditions in which long integration times are required to obtain a statistically significant count downstream of a tandem system, DOP appears to be a better challenging agent than NaCl in that it produces less of a change in the system during the duration of the test and also provides more conservative test data. The initial pressure drop of about .75 inches H₂O across the first filter would rise to a maximum of 1.23 but then decrease to 1.00 after a short period of clean air flushing. This was not true with filters challenged by NaCl. The excessive upstream concentrations required give rise to a concern over agglomeration processes that may occur for either aerosol between the upstream measurement point and the first filter face.

Of additional concern, in the case of DOP, is a possible change in size distribution due to evaporation between the two measurement stations. The severity of such a process would depend on the time in transit and DOP vapor supersaturation. The smallest droplets observable are still too large to be greatly influenced by the Thompson effect⁴ so that the loss would manifest itself in the large particle part of the spectrum as the entire observable spectrum uniformly evaporates.

VII Field Tests

One single stage 20,000 CFM system has been tested using the single particle laser spectrometer and a cloud maker thermal DOP generator. This test yields a P.F. of $5 \times 10^4 \pm 16\%$ for a 30 second integration time. A forward light scattering photometer usually used for this test was unable to provide a reading out of the background noise. Extrapolating the requirements for a similar two stage system results in 3 aerosol generators, 20 min. integration time and on accuracy of $\pm 40\%$. A 200,000 CFM single stage system would require only that the integration time be extended to 5 min to provide the same accuracy as for the 20,000 CFM stage system. Measurements of both two stage and large single stage systems will be made in the near future.

VIII. Conclusion

The use of a single particle laser particle spectrometer has proven to be remarkably successful in the characterization of HEPA filter performance against NaCl and DOP aerosols. Single particle counting statistics and total absence of detector background noise make it possible to detect and measure particle concentrations below the detection level of other detectors. Total counts as low as 4 are significant and yield a figure of merit of $\pm 50\%$.

TABLE III

TANDEM PROTECTION FACTORS (DOP)

| QUALITY CONTROL | MEASURED (DOP) | | | |
|------------------------------|------------------------------|--------------------|--------------------|--------------------|
| | PERIOD #1 | PERIOD #2 | PERIOD #3 | PERIOD #4 |
| | 1 - 13 minutes | 13 - 36 minutes | 37 - 62 minutes | 63 - 96 minutes |
| P_{1s} 2.50×10^4 | P_{1m} 8.21×10^3 | 1.27×10^4 | 1.01×10^4 | 2.84×10^3 |
| P_{2s} 2.50×10^4 | P_{2m} 2.24×10^3 | 3.73×10^3 | 9.71×10^4 | 1.10×10^4 |
| P_{21s} 6.25×10^8 | P_{21m} 1.84×10^7 | 4.74×10^7 | 9.81×10^7 | 3.12×10^7 |
| | 1 - 11 minutes | 12 - 17 minutes | 18 - 23 minutes | 24 - 27 minutes |
| P_{1s} 2.50×10^4 | P_{1m} 5.80×10^3 | 6.80×10^3 | 5.05×10^3 | 4.24×10^3 |
| P_{2s} 2.50×10^4 | P_{2m} 4.35×10^3 | 7.42×10^3 | 5.56×10^3 | 4.76×10^3 |
| P_{21s} 6.25×10^8 | P_{21m} 2.53×10^7 | 5.05×10^7 | 2.81×10^7 | 2.02×10^7 |
| | 0 - 1 minute | 2 - 11 minutes | 12 - 23 minutes | 24 - 60 minutes |
| P_{1s} 2.50×10^4 | P_{1m} 2.87×10^3 | 2.87×10^3 | 2.87×10^3 | 2.87×10^3 |
| P_{2s} 2.5×10^4 | P_{2m} 7.58×10^3 | 3.52×10^4 | 7.58×10^3 | 8.44×10^3 |
| P_{21s} 6.25×10^8 | P_{21m} 2.18×10^7 | 1.01×10^8 | 2.18×10^7 | 2.42×10^7 |
| | 4 - 9 minutes | 10 - 40 minutes | 41 - 45 minutes | 46 - 50 minutes |
| P_{1s} 5.00×10^4 | P_{1m} 2.44×10^4 | 6.29×10^4 | 3.14×10^3 | 5.22×10^3 |
| P_{2s} 5.00×10^4 | P_{2m} 2.07×10^4 | 4.82×10^4 | 1.79×10^4 | 2.42×10^4 |
| P_{21s} 2.50×10^9 | P_{21m} 5.05×10^8 | 3.03×10^9 | 5.02×10^7 | 1.26×10^8 |
| | 0 - 5 minutes | 6 - 10 minutes | 11 - 15 minutes | 16 - 20 minutes |
| P_{1s} 1.67×10^4 | P_{1m} 3.22×10^3 | 7.86×10^3 | 5.09×10^3 | 5.59×10^3 |
| P_{2s} 5.00×10^4 | P_{2m} 1.57×10^4 | 6.42×10^3 | 2.48×10^4 | 9.03×10^4 |
| P_{21s} 8.33×10^8 | P_{21m} 5.05×10^7 | 5.05×10^7 | 1.26×10^8 | 5.05×10^8 |

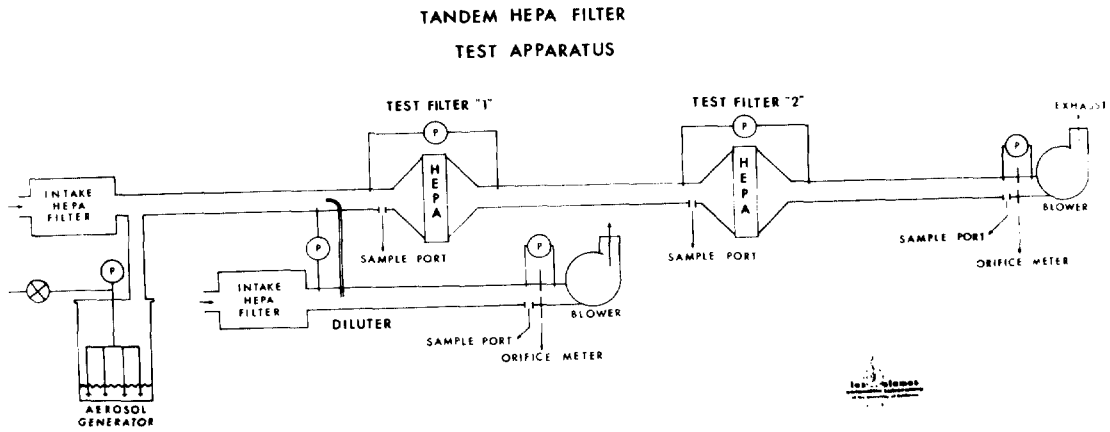


Fig. 1.
HEPA Filter Test Apparatus.

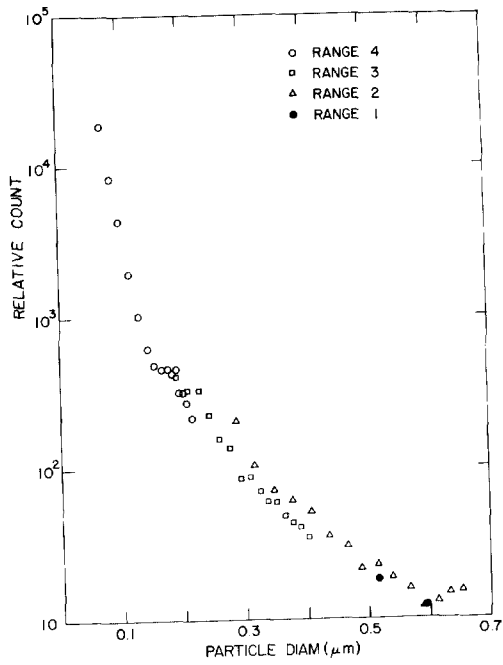


Fig. 2.
Size Spectrum of
NaCl in H₂O.

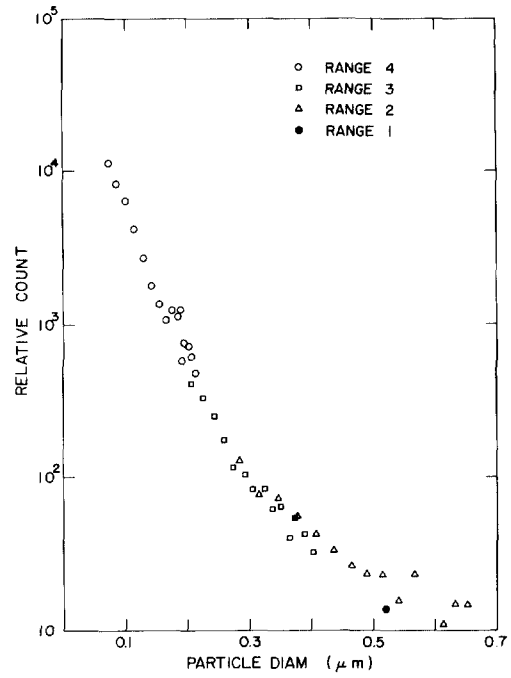


Fig. 3.
Size Spectrum of
5% DOP in alcohol.

NaCl TEST RESULTS

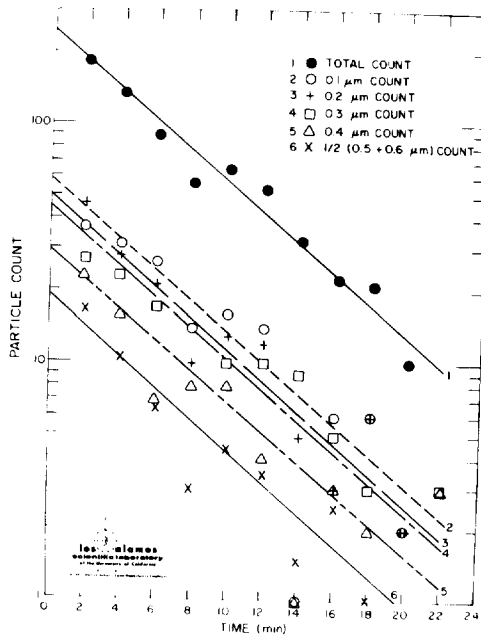


Fig. 4.

Size Dependence of Penetration Decay, NaCl Challenge.

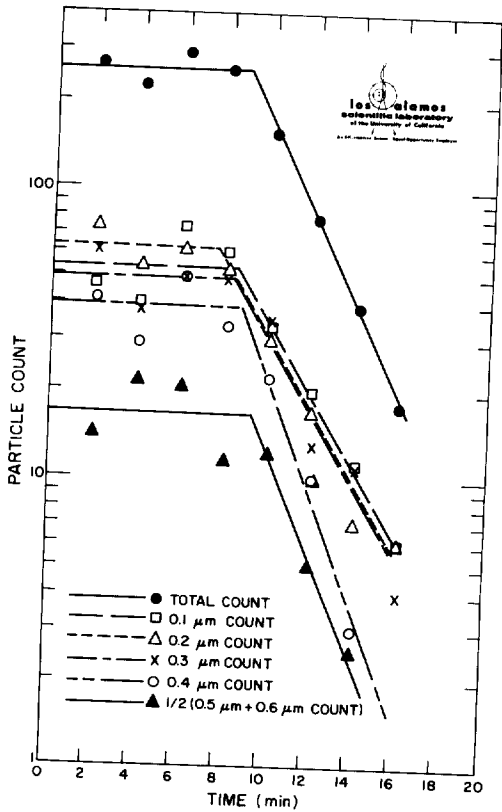


Fig. 5.

Size Dependence of Penetration Decay, NaCl Challenge.

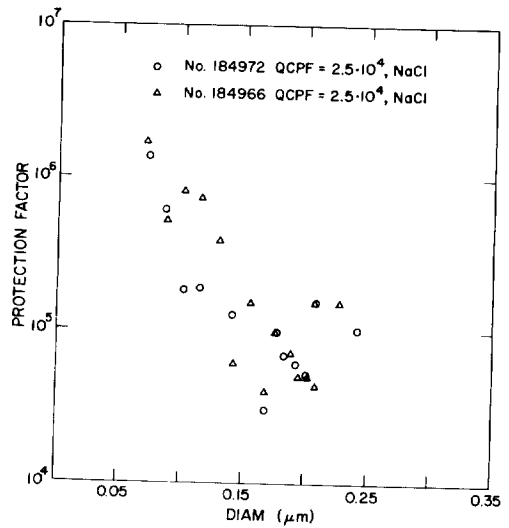


Fig. 6.

Protection Factor as a Function of Particle Size.

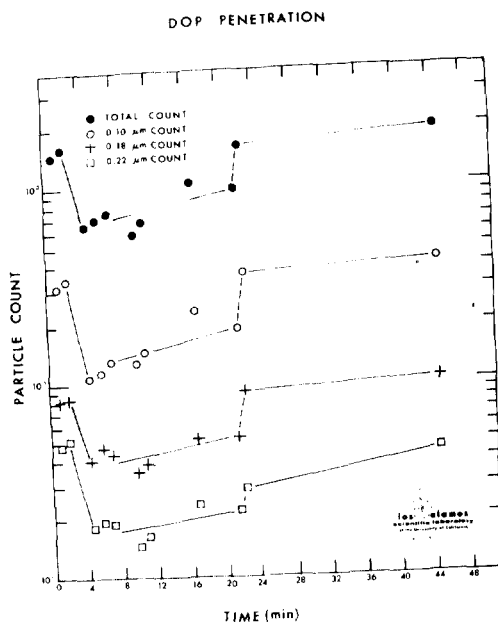


Fig. 7.
Size Dependence of
Penetration Decay,
DOP Challenge.

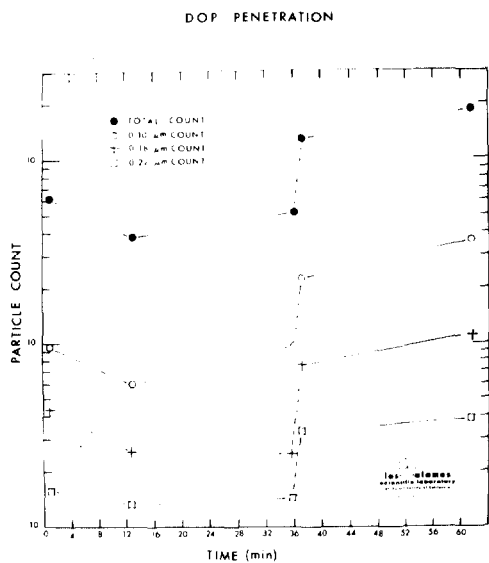


Fig. 8.
Size Dependence of
Penetration decay,
DOP Challenge.

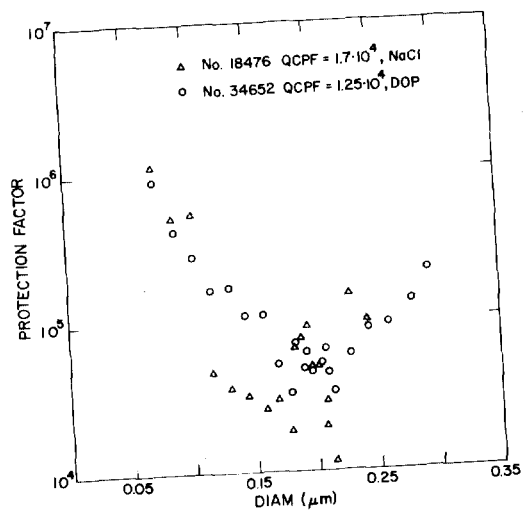


Fig. 9.
Protection Factor as a
Function of Particle Size.

14th ERDA AIR CLEANING CONFERENCE

References - HEPA Paper

1. A.E.C. Division of Operational Safety and Division of Engineering Construction, "HEPA filtration guidelines (draft)" (1971).
2. R. Hiebert, R. Stafford, and L. Pollat, "Improved light-scattering photometer for air filtration studies", Los Alamos Scientific Laboratory Report LA-4627-MS (1971).
3. B.G. Schuster and R. Knollenberg, "Detection and sizing of small particles in an open cavity gas laser", Appl. Optics, II, No. 7 PP 1515-1520, (1972).
4. J. Frenkel, Kinetic Theory of Liquids, Dover Publications, Inc., New York, 1955, P366.

DISCUSSION

SCHURR: Is your laser spectrometer applicable to on-line particle size analysis of general particle composition?

SCHUSTER: No. We have used polystyrene latex but the instrument certainly has been used for collecting all sorts of atmospheric dust. The gentleman who now builds this instrument is an atmospheric scientist and has a wide variety of these instruments. He has them on aircraft and has measured a wide variety of atmospheric dust as well as some of the industrial dusts.

CHEEVER: Are there plans to make the laser particle spectrometer commercially available?

SCHUSTER: It is already commercially available.

RIVERS: Your Figures 4 and 5 show a very rapid improvement in particle penetration over a period of about 20 minutes exposure to the sodium chloride aerosol. In both cases, regardless of particle size, penetration declined by a factor of about 200 with this exposure. Table 1 shows that pressure drop increases for these exposures were in the order of 0.1 in. W.G., or even less. The concentration, by my calculations, was about 31 mg/m³, in contrast to the usual DOP concentration of about 80 mg/m³. We have never observed or read about such improvements using either DOP smoke, NaCl, or ambient dusts. Do you think the effect could be due to your measurement technique? If the effect is real, and sustained, we probably ought to treat every filter bank with a 20-minute dose of NaCl smoke.

SCHUSTER: The single filter test displayed in Table I is for a DOP challenge and so cannot be correlated with Figures 4 and 5 which illustrate NaCl penetration. I'm curious as to how you arrived at your figure for mass challenge concentration since there is not sufficient information in the paper to make such a computation. I would not advocate filter bank pre-treatment with a 20 minute challenge of NaCl smoke.