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CHANGING METHODOLOGY FOR MEASURING AIRBORNE RADIOACTIVE DISCHARGES FROM NUCLEAR FACILITIES

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Changing Methodology For Measuring Airborne Radioactive Discharges From Nuclear Facilities

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ABSTRACT

The U.S. Environmental Protection Agency (USEPA) requires that measurements of airborne radioactive discharges from nuclear facilities be performed following outdated methods contained in the American National Standards Institute (ANSI) N13.1-1969 *Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities*. Improved methods are being introduced via two paths. First, the ANSI standard is being revised, and second, EPA's equivalency granting process is being used to implement new technology on a case-by-case or broad basis. The ANSI standard is being revised by a working group under the auspices of the Health Physics Society Standards Committee. The revised standard includes updated methods based on current technology and a performance-based approach to design. The performance-based standard will present new challenges, especially in the area of performance validation. Progress in revising the standard is discussed.

The U.S. Department of Energy recently received approval from the USEPA for an alternate approach to complying with air-sampling regulations. The alternate approach is similar to the revised ANSI standard. New design tools include new types of sample extraction probes and a model for estimating line-losses for particles and radioiodine.

Wind tunnel tests are being performed on various sample extraction probes for use at small stacks. The data show that single-point sampling probes are superior to ANSI-N13.1-1969 style multiple-point sample extraction probes.

INTRODUCTION

The standard governing air sampling in nuclear facilities for the past 26 years (ANSI N13.1-1969, *Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities*) has, since 1989, become part of USEPA requirements for the measurements of radionuclide emissions. Unfortunately, much of the standard has become obsolete because significant technical advances in the field have been made since 1969. The 1969 standard covers subjects that are not germane to the regulatory requirement and lacks performance standards. Consequently, the Health Physics Society was tasked by the American National Standards Institute to revise the standard. The revision is a major undertaking and progress is slow. This paper describes the significant changes being incorporated in the revised standard.

The U.S. Department of Energy's contractors have received permission to implement new sampling technologies at their facilities under certain circumstances. Wind tunnel tests of modern probe designs have been conducted by the Hanford contractors. The use of shrouded nozzle probes developed by McFarland, et al. (1) is being tested for application at certain facilities. The early results of those tests are described.

REVISION TO STANDARD

The standard governing air sampling in nuclear facilities for the past 26 years (ANSI N13.1-1969, *Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities*) covered air sampling in the workplace, in the environment, and in stacks and ducts. The scope of the revision has been narrowed to air sampling in stacks and ducts. This change was made because of significant

differences in air sampling objectives and to allow more thorough coverage of the latest developments in this technology. The current title for the revised standard is *Guide To Monitoring Releases of Airborne Radioactive Substances from the Ducts and Stacks of Nuclear Facilities.* The major change in the standard is the approach. Where the 1969 version was prescriptive in many ways, the 199x version will instead emphasize performance criteria.

Objectives and Approaches

The revised standard begins by emphasizing the importance of defining the objectives for a given sampling situation. Failure to understand the sampling objectives can lead to inappropriate or ineffective system design and implementation. While the typical objective is to monitor the performance of air treatment systems that ensure that people in the surrounding environment are not exposed to levels of airborne materials that are deleterious to their health, other objectives might be to:

- Assess the need for a permanent sampling or monitoring program;
- Aid detection of deteriorating equipment, faulty processes, or other conditions leading to the loss of effective control of airborne materials in an operation, and to subsequently determine the effectiveness of corrective measures; and
- Help assess the consequences of non-routine incidents and the selection of appropriate corrective actions.

The technical part of the revision provides the methodology for meeting the most stringent regulatory requirements; however, those requirements may not be applicable to all sampling programs, especially those with such limited objectives as process control. Therefore, the designer must exercise judgement in the application of these requirements and explicitly document the sampling objectives and the reasons for any exceptions to the requirements of the standard.

After defining the sampling program's objectives, the technical approach for meeting the objectives must be formulated. The factors that determine the approach to the sampling problem are 1) the potential emissions from the source, 2) the types of contaminants to be sampled, and 3) the desired detection limits.

First, the potential emissions criterion for system design can be based on either risk, dose, or emitted concentration. An evaluation of potential emissions allows for a graded approach to the complexity or design of the sampling program. An example graded approach is shown in Table 1 where system complexity varies with offsite potential dose. The standard provides guidelines for estimating potential emissions when actual measurements are not available.

The second factor determining the sampling approach is the type of contaminants being sampled. The revision emphasizes the importance of determining the characteristics of the contaminants present in the effluent stream in its treated and untreated states. The differences in sampling for particles, gases, and vapors are discussed at a general level to draw attention to how they affect the choice of sampling methods. Problems associated with large particles, reactive gases, and off-normal conditions are discussed.

For example, the characteristics of the aerosol source terms (especially the particle size distribution for particles larger than 10 μm that contain radioactivity) is critical in planning a sampling approach. First, the presence or absence of such particles should be identified; if they are present, conventional sampling methods may not be effective, if they are not present, then conventional sampling methods could be used with confidence. If large particles with attached radioactivity are present in significant quantities, then they would need to be effectively sampled, or, they would need to be sampled at less than ideal recoveries, but with known recovery vs. particle size and known source term particle size distribution. This would allow a conversion from

measured to actual concentrations. If such particles do exist in off-gases, it would be important to develop non-conventional sampling strategies. Similarly, the presence or absence of reactive vapors and gases effect the sampling approach.

The third factor determining the sampling approach is the desired detection limits or action levels. Given a desired minimum measurable quantity, factors such as sample flow rate, required instrumentation accuracy, elapsed sampling time, and analytical method can be chosen accordingly. Measurement uncertainties also play a key role in determining practical action levels. The sources of uncertainty typical of air sampling and how they propagate are discussed. Guidance is then given to rank the sources of uncertainty and to control the most highly variable ones. Using this information, the designer can then set achievable action levels. When going through this process, the designer also sets performance criteria for various measurement uncertainties specific to the particular sampling problem.

Determining Representative Sampling Locations

The original ANSI standard required that the contaminant composition at the sample extraction location be representative of the average composition. That hasn't changed. However, because of the difficulties in showing that one point is representative, the fallback was to sample enough points simultaneously or sequentially so that the resulting combined sample represented the true average. Guidelines were then given for selecting sampling locations and the number of probes. Almost without exception, the prescription approach has been taken instead of the more rigorous airstream characterization studies. The original ANSI standard's requirement to locate the sampling plane at a certain distance from a flow disturbance or discharge point was never proven to be an effective means of ensuring sample extraction from a well-mixed system and has been shown in instances to be ineffective (Hampl, et al. (2) and Hanson, et al. (3)). Also, using multi-point probes was a means to compensate for poor mixing by obtaining a mixed sample from several locations in the cross section. The result was often poor sample penetration through complex designs and small probe inlets.

The revised standard will emphasize the original ANSI standard's first criterion that samples be extracted from a location where the contaminant concentration represents the true average and that the contaminant concentration in the sampling plane does not vary greatly from the average. The payoff of this strategy is that it enables the use of single-point sampling probes with very low internal losses for particles over a wide size range. The revision's default criterion to ensure that samples are representative of the airstream is to demonstrate that the coefficient of variation of the contaminant is within +/- 20% over the central 2/3 of the area of the sampling plane. The revision provides guidelines for qualifying the sampling location, which include characterizing the airstream, including composition, geometry, velocity mapping, and contaminant mapping.

In instances where the desired well-mixed condition is not achievable, modifications to the system are recommended to improve mixing. Only where this is impossible or impractical are multi-probe samplers recommended. Performance criteria are given for the transmission of particles through probes that are applicable to both single- and multiple-probe systems.

Design of Effective Sampling Systems

Several topics are dealt with in the sections covering designing effective sampling systems. First, the requirements for bulk stream-flow measurements are discussed. Guidance is given concerning when to use continuous or periodic flow measurements. The methods appropriate for both of these measurement modes are briefly described, and detailed procedures are referenced.

The discussion of probe designs begins with the relative merits of single inlet and multiple-inlet probes and classical isokinetic or non-isokinetic types. The performance criterion for any style of probe is a demonstrated transmission of 80 - 130% of 10 micrometer aerodynamic equivalent diameter (AED) from the free stream to the end of the probe. The need for tracking proportional or fixed sample flow rate control is addressed. Periodic inspections of probe condition are emphasized.

Optimizing and upgrading existing and new systems are discussed. Bringing existing systems into compliance with new standards is a major challenge, and each emission point's dose potential and the contaminant characteristics will be factors to consider in upgrade decisions. In analyzing the priority of an existing system's upgrade, the objective and approach issues described above must be re-evaluated. The use of airstream mixing and characterization methods and other performance testing methods are tools for determining whether upgrades are necessary.

Other topics covered in this section are briefly described below.

- The minimization of sample line-loss for particles and reactive gases is discussed. The performance criterion is the delivery of at least 50% of the sampled contaminants from the free stream to the collector/analyzer.

- Current data and references are given on selecting filters and gas collection devices.

- Guidelines are given on sample flow rate measurements and when automatic control is necessary.

- Sampling system failure alarms and emission exceedance action levels are addressed.

Quality Control and Quality Assurance

The final section of the revision addresses quality control and assurance issues. The system documentation requirements include the QA plan, effluent stream characterization studies, detailed design drawings, vendor information, operating procedures, training records, and maintenance data. Requirements are given for regular inspections of probe condition and the functioning of flowmeters, pumps, controls, heat tracing, detectors, and other instruments. The performance standards found throughout the standard are summarized at the end of this section.

Appendices

The general guidelines and requirements are presented in the main body of the standard. The appendices contain information for which there is a likelihood of continued technical development and are not considered part of the standard's requirements. The appendices include:

- Techniques for the measurement of flow in a stack or duct;
- Modeling particle losses in probes and transport systems;
- Special considerations for sampling radioiodine;
- Selecting filters for particle collection and radionuclide analysis resolution;
- Contaminant mapping; and
- Methods for performance testing or sample validation with examples.

Progress

The first complete draft of all sections was assembled in March 1995 for internal review by the working group. That review will be followed by a peer review. The group's goal is to submit the reviewed draft to the Health Physics Society in August 1995.

ALTERNATIVE APPROACH

In 1994 the U.S. Department of Energy (DOE) petitioned the USEPA for an alternate approach to complying with ANSI N13.1-1969 requirements. Approval was received to implement alternate sampling methods. Approval was granted to extract samples from nonstandard locations provided that the contaminants were shown to be well mixed. The use of single probes for sample extraction from well-mixed locations was permitted. The use of shrouded non-isokinetic probes that meet certain performance requirements was approved. Finally, the use of the DEPOSITION

computer code (Anand, et al. (4)) and equivalent codes could be used for the design and estimation of particle line-loss in sample transport systems. This approval covers several of the key methods in the revised ANSI N13.1 and will provide a bridge to using these approaches until the revised standard is adopted.

TESTS OF PROBE TYPES

Wind tunnel tests are being performed at Pacific Northwest Laboratory on various sample extraction probes for use at small stacks with diameters less than 37 cm. The tests on two of the types will be described here -- the shrouded and standard isokinetic types. The four probes discussed here were of the simplest possible configuration for their size, with one nozzle and a short transport line leading to the filter holder mounted just outside the stack.

The probes designated C and D both have a single inlet nozzle of the "shrouded probe" type, shown in Figure 1, as described by McFarland et al. (1) . Probe C was designed for a sample flow rate of 57 lpm at an airstream velocity of 2.5 - 8.5 m/s. Probe D was designed for the same flow rate at an airstream velocity of 8 - 16 m/s. The main difference between the two is the diameter of the nozzle inlet. These probes do not follow the usual convention of isokinetic sample aspiration, but are designed to operate at a fixed flow rate over a range of airstream velocities. The shroud aligns the air velocity with the axis of the nozzle, and decelerates the velocity of the approaching airstream. The large diameter nozzle aspirates the sample from the central portion of the airstream, well away from the turbulence caused by the shroud's leading edge and walls. The transport line consisted of a large radius 90-degree bend, a short, straight section and internally tapered adapters to accommodate the nozzle and filter holder. These nozzles were tested both in their normal fixed-flow rate mode and in a mode where the flow rate was varied in proportion to the air velocity.

The probes designated E and F consist of a single nozzle of the type shown in Figure 2. Probe F was designed to extract samples isokinetically at 42.4 lpm from an airstream velocity of 11.5 m/s. Probe E was identical except it had a larger inlet diameter so the intake-to-airstream velocity ratio would be 0.59. Thus probe E would be operated sub-isokinetically, which would theoretically cause the sample airstream to be enriched in particles larger than around 2 μm AED. The nozzles have an internal expansion taper of 8°. Each nozzle is mounted perpendicular to and welded to a larger pipe through which the sample stream exits the stack. To maintain a constant airstream-to-nozzle-opening velocity ratio, the flow rate through both of these probes must be varied in proportion to the airstream velocity.

Method

The testing was conducted in a recirculating wind tunnel with a test section that is 0.61-m square and 6.1-m long. The test aerosol was sodium fluorescein tagged oleic acid generated with a vibrating orifice aerosol generator. Each probe was tested at air velocities from 3 to 15 m/s and particle sizes from 1 to 15 μm. A filter holder was attached to the end of the transport line just outside the wind tunnel wall and 47-mm diameter glass fiber filters were used for collecting particles that were transmitted through the probes. The particle collection on the filter was dissolved in methanol and the fluorescence content of the solution was analyzed with a spectrofluorometer. Aerosol collected on the inside surfaces of the probes was removed with the same solution and then analyzed in the spectrofluorometer.

Prior to the tests, probe mounting positions were selected and the spatial distribution of aerosol between those positions was determined at different particle sizes and velocities using reference isokinetic samplers, each consisting of a straight nozzle and a filter holder. Spatial distribution was also monitored at the beginning and end of most tests with reference samplers placed in the probe positions used during the test.

Two types of data were calculated for the tests. The key datum for each test was the concentration ratio, or ratio of aerosol concentration determined at the test probe's filter to that of the reference sampler, adjusted for the spatial effects. This is the key result because actual sampling

probes are rarely removed for analysis of internal collection. Also, because the shrouded probe types were designed to maximize quantitative sample delivery to the filter, accounting for probe deposition is unnecessary (and in fact, is an error). The other datum was the concentration ratio accounting for the deposition of aerosol inside the probe. This was determined for a portion of the tests to ascertain its significance.

Results

Figures 3 and 4 show the resulting concentration ratios, measured at the collection filters, as a function of particle size for the probes at 5 m/s and 15 m/s respectively. Because the data analysis is in progress, the data points are shown without error bars. Third-order polynomial fitted lines are shown to clarify the trends for each probe. Particle diameters shown are aerodynamic equivalent diameters (AED), or the diameter of a spherical water droplet that has the same settling velocity in quiescent air as the actual particle. The most desirable result would be a flat line at 100%, indicating quantitative aerosol delivery to the filter. The shrouded type probes designed for the particular velocity range had the most favorable results, Probe C at 5 m/s and Probe D at 15 m/s. At 15 m/s, Probe C (well outside of its design range) exhibited erratic results, but they were always higher than 100%. At 5 m/s, Probe D (somewhat below its design range) exhibited steadily declining concentration ratios with increasing particle size. The probe with a nozzle operated isokinetically, Probe F, exhibited generally declining results with increasing particle size and had a concentration ratio greater than 90% only for particles smaller than around 5 μm AED. The subisokinetic Probe E exhibited concentration ratios above 90% out to 9 or 10 μm AED particles.

Figure 5 shows the results for 10 μm aerosol as a function of air velocity. This was the only particle size for which the response versus velocity was investigated in detail. While Probe D at its fixed design flow rate showed excellent concentration ratio characteristics at 15 m/s in Figure 4, that did not appear to be the case at lower velocity. Instead, Probe D operating in the proportional flow rate mode showed a fairly favorable flat response over the entire velocity range. On the other hand, Probe C exhibited erratic results in proportional-flow mode compared to fixed-flow mode. Probes E and F exhibited variable results as a function of velocity; however, the subisokinetic Probe E showed concentration ratios nearly 30% higher compared to the isokinetic Probe F.

Line-loss, as a percentage of the sum of probe deposition and filter collection, is plotted in Figures 6 and 7 for the 5-m/s and 15-m/s tests. The line-loss was only significant for particles larger than 10 μm, and was considerably more so at the higher velocity. Further analysis will be needed to determine if the deposition occurred throughout the probe or was mostly in the nozzle.

CONCLUSION

The revised ANSI N13.1 emphasizes setting performance objectives and verifying that they are met. Look-alike cookbook design is deemphasized. The structure of the original standard made it easier to use the standard as a source for a default design than to embrace the performance-based design concepts it did contain. The major challenges of the revised standard are requirements to demonstrate that the contaminants are well mixed in the sampling plane and that the system extracts and delivers a representative sample. The impact of the revised standard on some existing systems may be significant, but careful consideration of the system objectives and the potential and real effluent dose impacts of the source will focus any upgrade efforts where they are most needed.

The wind tunnel tests indicated that the modern shrouded nozzle type of probes have the potential to deliver samples with much less particle-size bias (over the range tested, up to 15 μm AED) than the isokinetic nozzle probes tested. The use of the "shrouded" type probe operated in proportional flow shows even lower sensitivity to particle size and velocity than the fixed-flow mode; however, further verification over a wider range of particle sizes would be beneficial.

ACKNOWLEDGMENTS

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Table 1. Sample graded approach to application of sampling and monitoring.

Potential Effective Dose Equivalent Category	Required Monitoring and Sample Analysis Procedures	Potential Effective Dose Equivalent Range (mrem/y)
1	Continuous extractive sampling for a record of emissions, and in-line, real time monitoring with alarm capability; consideration of separate accident monitoring system	>1
2	Continuous extractive sampling for record of emissions, with retrospective, off-line periodic analysis	>0.1 and <1
3	Periodic confirmatory extractive sampling and off-line analysis	>0.001 and ≤ 0.1
4	Annual administrative review of facility uses to confirm absence of radioactive materials in forms and quantities not conforming to prescribed specifications and limits	≤ 0.001

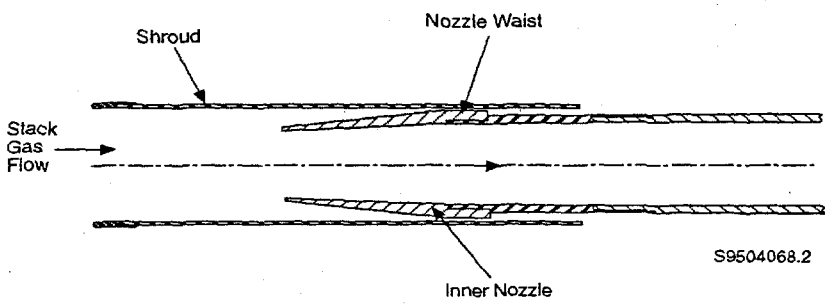

Figure 2. Features of Shrouded-type Nozzle Of Probes C and D.

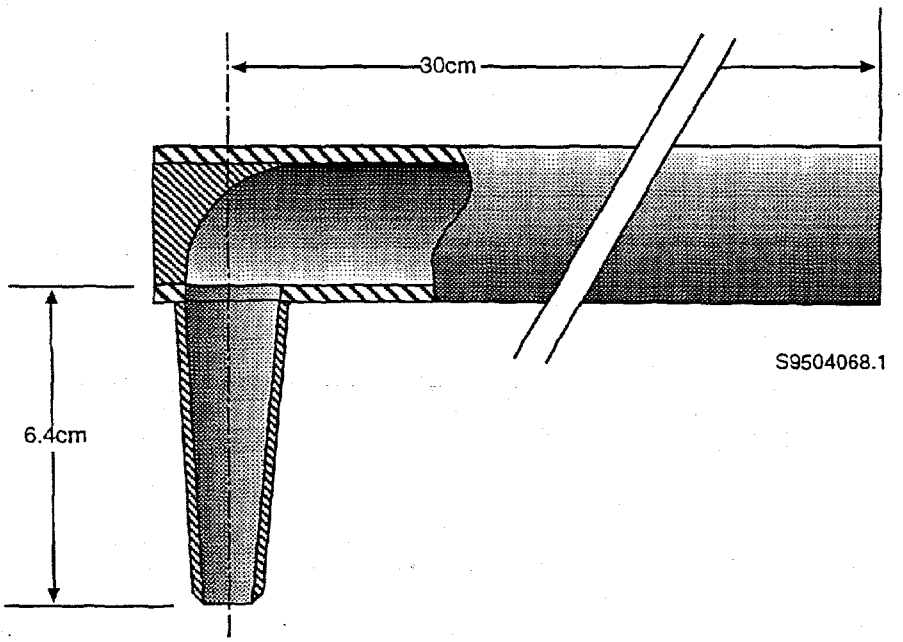

Figure 1 Nozzle Design For Probes E and F

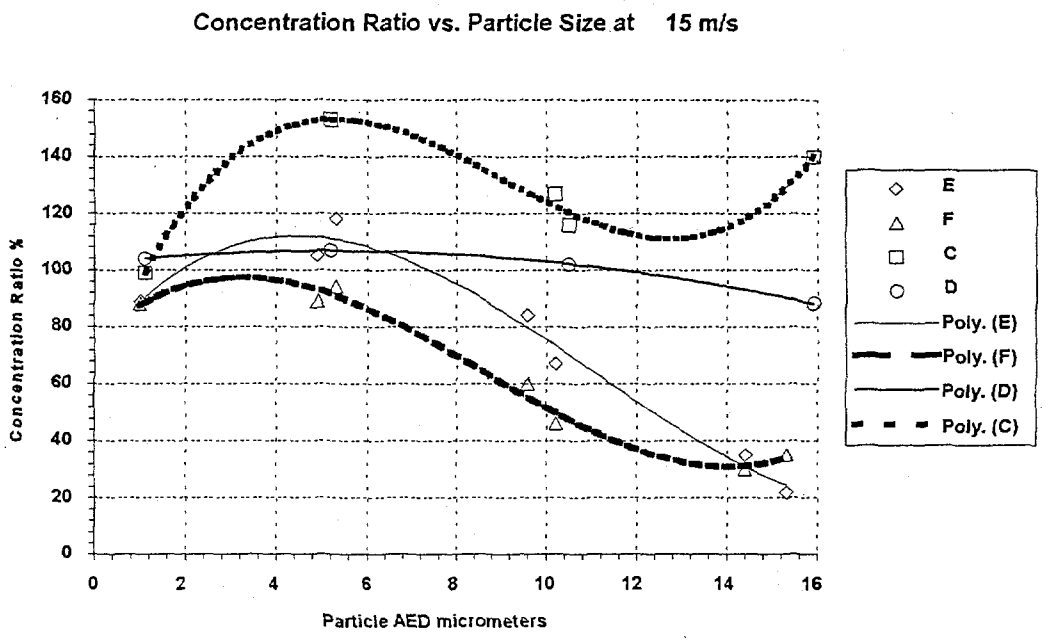

Figure 3 Concentration Ratios versus Particle Size at 15 m/s

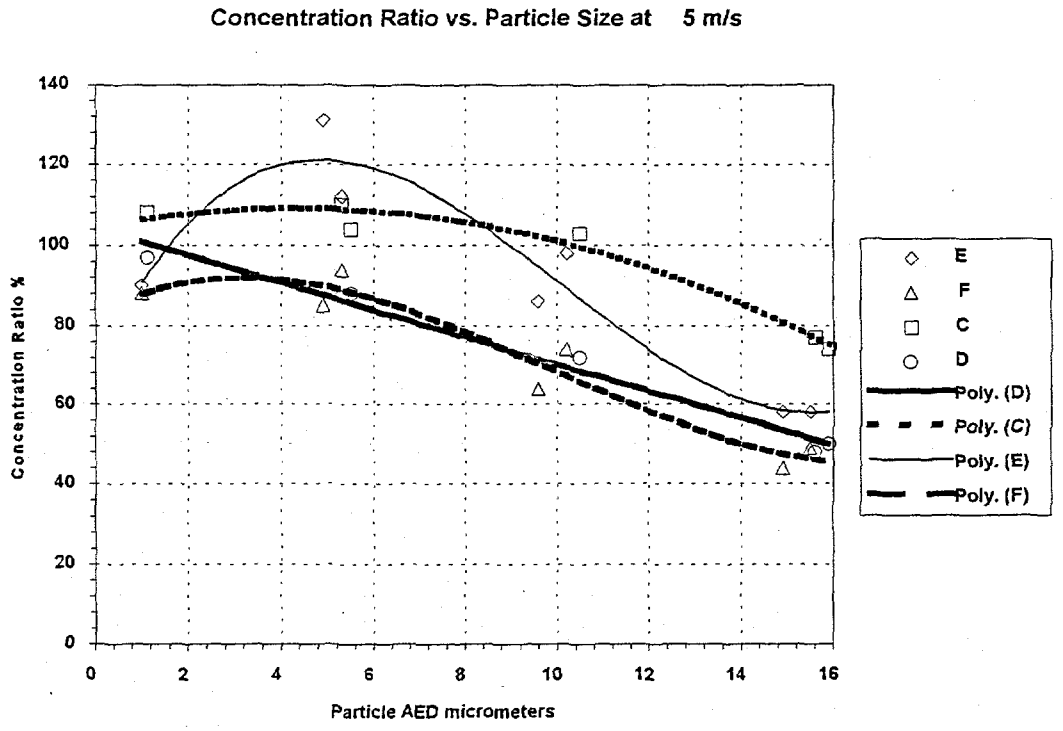

Figure 4 Concentration Ratios versus Particle Size at 5 m/s

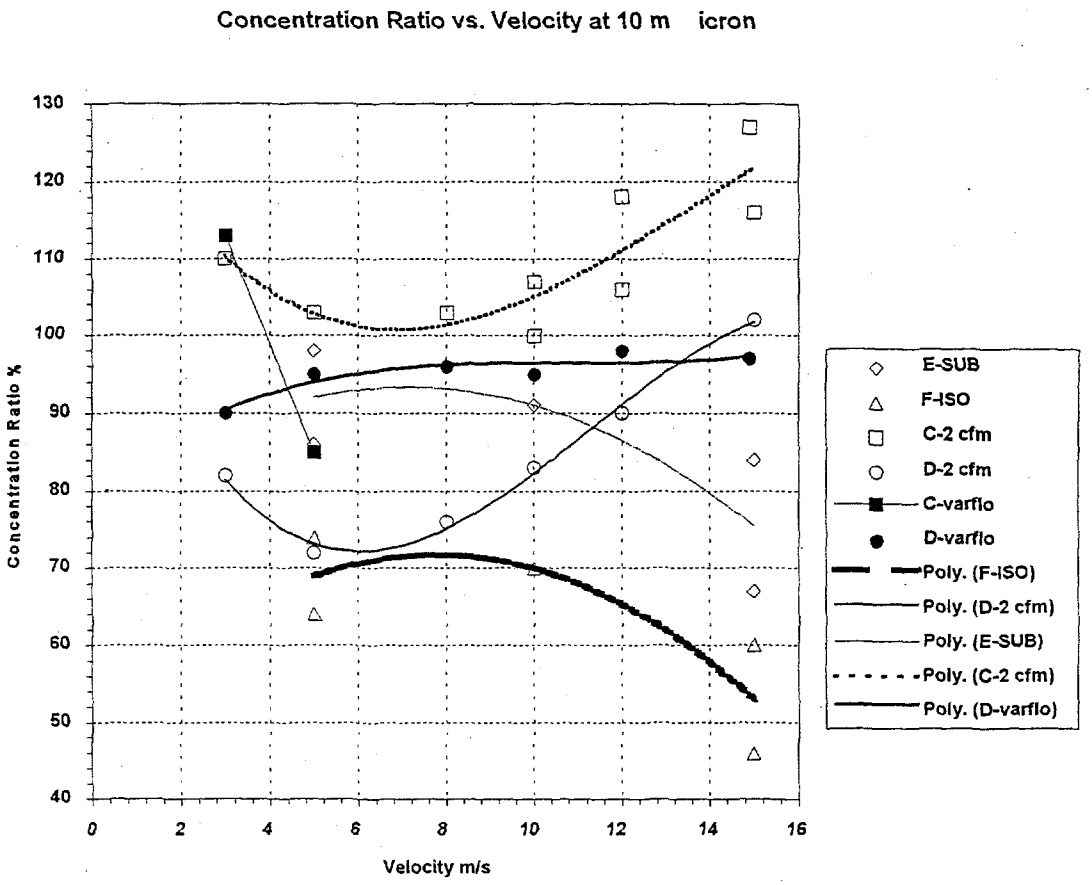

Figure 5. Concentration Ratio Versus Velocity With 10 μm Particles

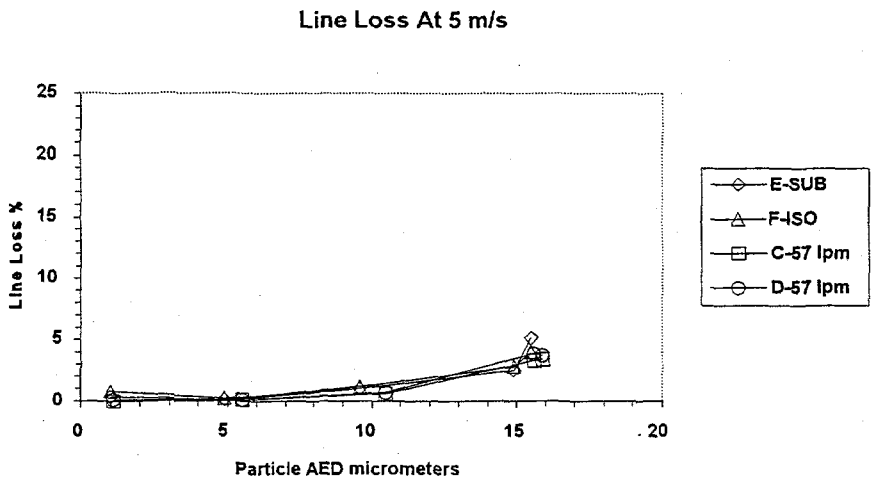

Figure 6 Line Loss at 5 m/s

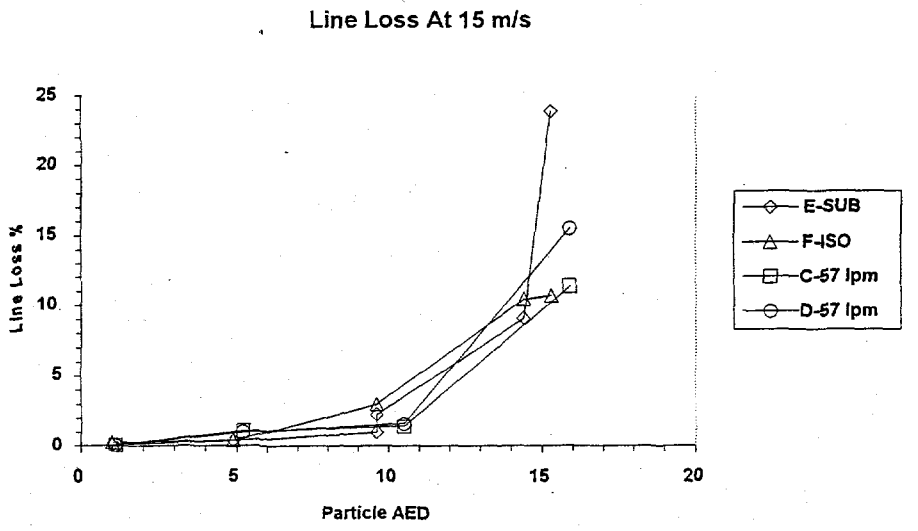

Figure 7 Line Loss at 15 m/s