Los Alamos
NATIONAL LABORATORY
memorandum

Environment, Safety and Health Division
Industrial Hygiene and Safety Group (ESH-5)

To/MS: Eric McNamara, ESH-17 (J978)

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Phone/FAX: 7-7783/5-3689

Symbol: ESH-5:95-714

Date: August 11, 1995

LA-SUB--96-115

SUBJECT: FINAL REPORT OF EXPERIMENTS REQUIRED BY LANL
SUBCONTRACT 9-X73-4229J-1

← Required by 0028N0014-3C, Task # 5

The attached report, "A Laboratory Comparison of Two Methods of Characterizing Exhaust Stack Emissions," describes the performance and results of Task 2 of the reference contract between Texas A & M University (TAMU) and LANL. The task, entitled "Comparison of Optical and Fluorometric Methods for Characterizing Concentration Profiles," was included in the contract because the optical method for characterizing exhaust stack aerosol concentration uniformity is being used at Los Alamos at present. Because another method has been used at TAMU and elsewhere, it was prudent to establish the equivalency of the two methods. Experiments were conducted at the TAMU Riverside Campus during the period of March 28-31, 1994 to ascertain this equivalency. Statistical analysis of the data was provided by Dick Beckman of TSA-1.

The report will be used as a draft to be finalized and submitted to a peer reviewed journal to achieve wider knowledge of, and acceptance of the light scattering method by others involved in sampling of radioactive emissions.

CIF:MLL

Cy: Bruce D. Reinert, ESH-5 (K553)
ESH-5 Group Files (K486)
R&D Section Files (K553)

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A LABORATORY COMPARISON OF TWO METHODS OF CHARACTERIZING EXHAUST STACK EMISSIONS

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Los Alamos National Laboratory

Introduction

Concern for the environment and public health, and compliance with DOE and EPA regulations require that representative sampling be conducted on exhaust stacks that emit radioactive materials. In order to design and install particulate samplers, EPA Regulation 40CFR61, Subpart H (NESHAP) specifies that particle concentration profiles be determined, in addition to velocity profiles, at the sampling cross section of all stacks requiring sampling. Neither the NESHAP regulation nor ANSI standard N13.1-1969, A3.2, p27, which is incorporated into NESHAP by reference, specify detection or analytical methods for determining effluent concentration uniformity in stacks that may emit radioactive gases or particles. Methods are described for stacks emitting nonradioactive materials, but these are not suitable for radioactive emissions, nor do the regulations specify any tolerances on the concentration uniformity for exhaust stacks.

Background

There is one major problem in the sampling of radioactive emissions that does not normally occur with nonradioactive emissions. The solution to this problem introduces additional problems into the sampling. The contaminant concentration is normally extremely low in the effluent emitted because excellent filtration is employed. Although this is essential insofar as emissions are concerned, it complicates efforts to determine that the effluent is being sampled properly. In such an environment even the meaning of concentration uniformity is unclear. In nonradioactive emissions the concentration uniformity is the steady state spatial gradient of particle concentration across a diametrical plane of the duct or stack at some defined position along the stack axis. But in the effluent under consideration there is no steady state gradient because particles pass through the HEPA filter banks only occasionally and usually follow very different paths up the stack.

However, to satisfy the EPA mandated requirements, an aerosol environment that will allow characterization of the flow up the stacks can be created by introducing a large number of particles into the stack. This provides "tracer particles" that mix throughout the stack to allow uniformity determination. Thus, an innocuous surrogate aerosol with a particle size distribution similar to the expected contaminant aerosol may be injected immediately downstream of the filters to increase the concentration. The surrogate aerosol must be injected in several places to assure that the particle distribution at the sampling station is not sensitive to injection position.

Even the added aerosol may be insufficient for accurate detection in a reasonable sampling time by many instruments if the stack effluent flow is large. Long collection times at many positions across the stack diameter may be required to fully define the uniformity.

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Mass Tracer Detection

This method, which is used at Texas A&M University (TAMU), may be termed the traditional method. It is reliable and accurate but requires a wet chemical procedure for analysis that is inconvenient for field tests, and it has sample collection limitations for large volume flow. The method can be used in combination with a surrogate aerosol to increase particle concentration in a nuclear facility effluent. However, at attainable concentrations long sampling times may be required, even though the aerosol is doped with a small amount of fluorescent tracer to enhance detection sensitivity. The injected aerosol must be monodisperse to avoid errors due to size distribution variation at different positions. Filters are used for collection at each required position across two perpendicular diameters of the stack sampling plane. If the number of positions is small, a well designed rake or jig can be used to allow simultaneous sampling at all points; however, the total area of the rake and filter holders must be a small fraction of the stack cross sectional area to avoid major disturbance of the flow. For many stacks, samples must be taken consecutively along the perpendicular traverses. Isokinetic inlets are attached to the filter holders and collection flow rates up to 84.9 L/min (3 cfm) are used. Collected samples are subsequently analyzed fluorimetrically for mass to provide a concentration profile.

Laser Light Scattering Detection

This method, developed at Los Alamos National Laboratory (LANL), also utilizes a surrogate aerosol. The method consists of injecting a well characterized monodisperse or polydisperse aerosol into the stack and sampling the aerosol at selected traverse points at a sampling station. Because the concentration profile is a comparison between samples across the stack, it is sufficient that the aerosol injection, sampling train, and detector operate reproducibly during the traverses, but not necessarily accurately. It is necessary that a size distribution be determined for each sample if a polydisperse aerosol is used. Generally, this requires the use of a single probe designed to sample near isokinetically, and ducting designed to transport a maximum fraction of the sampled particles to a particle spectrometer. Sampling time at each traverse point is short, usually one minute or less.

Experimental

The two methods for determining concentration profiles were compared at the Texas A&M University Riverside campus in a wind tunnel laboratory of the Mechanical Engineering Department. Both methods were used to measure the number (LANL method) or tracer mass (TAMU method) of monodisperse 11.2 μm diam spherical oleic acid particles doped with sodium fluorescein (NaFl) and injected into a wind tunnel.

The monodisperse aerosol was generated by a Berglund-Liu generator (TSI Model 3050). To continually monitor the size distribution of the test aerosol, two Aerodynamic Particle Sizers (APS), (TSI Model 3310A) were used. Velocity profiles at the tunnel exit were measured for each set of experiments (Fig. 1) with a calibrated TSI velocity meter, and velocity pressure was also monitored at a single point during all tests by a pivot tube connected to an electronic manometer.

The wind tunnel used to provide a high velocity aerosol flow had a 33 by 33 cm (13" by 13") square cross section initially, but could be easily converted into a 43.2 by 48.3 cm (17" by 19") cross section by removing a full length insert. The tunnel was an open cycle, forced draft type of 8 m (24 ft) length. All sampling was performed at the exit of the tunnel. Due to the large diameter of the filter holders used in the TAMU method, space restriction prevented sampling by both methods simultaneously.

Sampling grids were outlined by threads attached to the tunnel exit face. A 3 by 3 grid (Fig. 1 inset) was used to sample the 33 cm square tunnel and a 4 by 3 grid (Fig. 4 inset) for the larger 48 cm rectangular tunnel. In the LANL method, samples were taken consecutively at grid positions 1 through 9 for the 33 cm, and 1 through 12 for the 48 cm tunnel. However, because of space (flow blockage) and pumping limitations TAMU sampled simultaneously in a cruciform array from 5 cells only, numbers 2, 4-6, and 8, in the 33 cm tunnel. Similar considerations limited the TAMU array to 4 horizontal filters when sampling from the 48 cm tunnel, so that the cells were sampled consecutively 4 at a time in the order 1-4, 5-8, 9-12. Each group of 5 or 4 TAMU samples required ~30 minutes from start to the start of the next group, so that sample sets of three "traverses" required 90 minutes and 270 minutes in the 33 cm and 48 cm tunnels respectively. During this time the airflow and particle generation conditions had to be constant.

Small angle expansion nozzles with inlets sized to be isokinetic at 56.6 L/min (2 cfm) were attached to each TAMU filter holder. In the LANL measurements, subisokinetic sampling nozzles sampling at 56.6 L/min were used to enrich the sample in the large particles of interest. Because the measurements were comparative within the same profile, and because an independent means of measurement was used as a size distribution control, it was necessary only to sample consistently from point to point, not isokinetically.

Two laser light scattering spectrometers (MET-1 Model A2420) were used in the LANL method to measure particle concentration profiles. These instruments sampled at 56.6 L/min at rates up to 7000 particles/L ($2 \times 10^5/\text{ft}^3$) and classified them into 6 size ranges from 0.3 to $>10\mu\text{m}$. Both sampled the aerosol at the exit of the tunnel. One spectrometer was used as a stationary reference to eliminate temporal variation from the data and the other was the traversing instrument that obtained data at each grid position. Although the instruments sampled for a selectable duration, generally 1 minute samples were taken simultaneously by each instrument. In a sampling traverse, 9 positions could be completed in less than 15 minutes. A computer was used to control the simultaneous sampling of the two spectrometers and acquire and store data from each.

Discussion

Criteria for sampling equivalency was that the mean difference between the overall results as indicated by the coefficients of variation (CoV) differed by no more than 10%. Thus a CoV was calculated for the three traverses for each velocity set in the two tunnels (Figs. 2-4 and Table 1). Sampling was conducted alternately by LANL and TAMU investigators so that even though not simultaneous, the sampling was in a similar environment. Prior to starting tests in each tunnel (33 cm or 48 cm) and environment (11 or 22 m/s) velocity profiles were measured. The tunnel environment was monitored at all times by velocity pressure measurement for velocity and by aerodynamic size distribution measurement to assure that testing conditions did not change during or between tests. Indeed, occasionally the aerosol generator would stop or change its output size distribution significantly, and any test in progress at the time would be restarted when conditions were corrected.

Results

Table 1 shows that the results obtained by both methods were equivalent in each tunnel and for each velocity. The repeatability with the 33 cm tunnel running at 22 m/s was determined to be $+0.068$ for three replicate samples at each of the 12 positions of one LANL traverse, whereas, it was $+0.043$ for four TAMU sample sets.

TABLE 1

Comparison of Sampling Results

<u>Sample Description</u>	<u>TAMU</u>		<u>LANL</u>	
	<u>No.</u>	<u>CoV</u>	<u>No.</u>	<u>CoV</u>
33 cm Tunnel, 22 m/s	5	0.21	9	0.21
33 cm Tunnel, 11 m/s	5	0.19	9	0.19
48 cm Tunnel, 11 m/s	12	0.14	11*	0.14

*Post analysis of the traverse, and visual inspection revealed that the reference sampling probe had rotated out of alignment with the airstream at the no. 12 position on one traverse. The two reference values, which were >50% smaller than the average reference value, were discarded as outliers at this one position.

The wall losses in the sample collection nozzles were analyzed by TAMU in special experiments. The losses in both the TAMU and LANL inlet nozzles were comparable and reproducible (Table 2) in 4 and 3 tests of the TAMU and LANL nozzles respectively. However, the overall sampling losses in the TAMU sampling train (nozzle to collector) were probably much less than those in the LANL sampling train. The TAMU nozzle was connected directly to the filter holder, whereas the LANL nozzle was connected to a 1" diameter transfer tube having one 90° bend, a 24" long horizontal transfer tube, and a converging, tapered transition tube into each MET-1 instrument. Nozzle and transfer losses were immaterial in this comparison because, so long as a sufficient mass (TAMU) or number (LANL) of particles reached the filter or instrument, and the losses were consistent from sample to sample, the concentration profile being determined was not affected by sampling losses.

TABLE 2

Collection Nozzle Particle Loss

<u>Tunnel Velocity</u>	<u>No.</u>	<u>TAMU</u>		<u>No.</u>	<u>LANL</u>	
		<u>Loss</u>	<u>CoV</u>		<u>Loss</u>	<u>CoV</u>
11 m/s	4	0.36	.02	3	0.29	.01
22 m/s	4	0.41	.04	3	0.43	.01

Conclusions

1. The sampling comparison shows that under controlled conditions both methods produce equivalent results in the measurement of aerosol concentration profiles in the tunnels used.
2. The TAMU method required at least 4 times longer to sample and analyze the tunnel profiles than did the LANL method. The disparity in time would increase as the number of sampling positions increased. Also the difficulty of designing and fabricating an efficient multipoint probe for the TAMU method would increase with the increasing number of sampling positions, e.g. a larger stack diameter.
3. The TAMU mass based method is restricted to the measurement of monodisperse aerosols, which could require extremely long collection time because of the limited generation rate of monodisperse particles. The LANL method has an included capability for determining aerosol size distribution, therefore polydisperse aerosols may be measured and analyzed. Polydisperse aerosols are much easier to generate in large concentrations than are monodisperse aerosols.
4. The LANL method has been proven comparable to the mass based TAMU method in accuracy and reproducibility. Qualitatively, it is also more suitable for use in field testing of most exhaust stacks for aerosol concentration uniformity and installed sampler efficiency. Much less sampling and analysis time is required for the LANL method and the equipment is more suitable to the rugged conditions encountered in field testing. These considerations make the LANL method more cost effective in all testing of exhaust stacks and ducts.

LANL/TAMU COMPARISON

33 cm Tunnel Velocity Profile at Exit

LANL/TAMU COMPARISON

33 cm Tunnel, 11 m/s, 3 Reps

LANL/TAMU COMPARISON

33 cm Tunnel, 21 m/s, 3 Reps

LANL/TAMU COMPARISON

48 cm Tunnel, 11 m/s, 3 Reps

TEXAS A&M UNIVERSITY
Department of Mechanical Engineering

June 26, 1994

Mr. James Bland
ESH-17
Los Alamos National Laboratory
Los Alamos, NM 87545

Dear Jim:

Attached herewith are the performance data for the probes that we designed for the CMR facility. Also enclosed, for Quality Assurance purposes are detailed drawings of the probes. No laboratory data have been acquired on the "Super low velocity probe," because we had earlier agreed to substitute a flow splitter for the third probe. A design of that flow splitter was provided to you on May 4, 1994.

Please call me if you have any questions.

Sincerely,

A handwritten signature in cursive script, appearing to read "Andy", written over the printed name.

Andrew R. McFarland

*CMR Super Low Velocity Probe
Numerical Data*

Mid Velocity Range Probe For CMR

High Velocity Shrouded Probe For CMR

DATE	SYM	REVISION	RECORD	AUTH	DR.	CHK.

ASSSEMBLY 4 CFM PROBE

SHROUD

HI 4 CFM PROBE

MI 4 CFM PROBE

TITLE: 4 CFM PROBE

MATL: STAINLESS STEEL

TEXAS A&M
MECHANICAL ENGINEERING
AEROSOL TECHNOLOGY LAB

DRAWN BY	APPROVED BY	DRAWING NUMBER	DATE	SCALE
M.G.	S.C./C.O.	3CMRP14B	06-13-94	1:3

DATE	SYM	REVISION	RECORD	AUTH	DR	CK

NOZZLE HI 4 CFM

COLLAR AND FLANGE HI 4 CFM

OUTLET TUBE HI 4 CFM

FIN HI 4 CFM

TITLE: PROBE HI 4 CFM

MATL: STAINLESS STEEL

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DRAWN BY	APPROVED BY	DRAWING NUMBER	DATE	SCALE
M.G.	S.C./C.O.	3CMRP15B	06-14-94	1:1

TITLE: PROBE MI 4 CFM
MATL: STAINLESS STEEL

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DATE	SYN	REVISION	RECORD	AUTH	DR.	CK.

TITLE: NOZZLE MI 4 CFM		TEXAS A&M		DRAWN BY	APPROVED BY	DRAWING NUMBER	DATE	SCALE
MATL: STAINLESS STEEL		MECHANICAL ENGINEERING AEROSOL TECHNOLOGY LAB						
				M.G.	C.D.	3CMRPR3	05-17-94	1:1

DATE	SYM	REVISION	RECORD	AUTH	DR.	CK.

TITLE: COLLAR & FLANGE MI 4 CFM
MATL: STAINLESS STEEL

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DRAWN BY	APPROVED BY	DRAWING NUMBER	DATE	SCALE
M.G.	C.D.	3CMRPR4	05-17-94	1:1

DATE	SYM	REVISION	RECORD	AUTH	DR.	CK.

TITLE: OUTLET TUBE MI 4 CFM

MATL: STAINLESS STEEL

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AEROSOL TECHNOLOGY LAB

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M.G.

APPROVED BY

C.D.

DRAWING NUMBER

3CMRPR5

DATE

05-17-94

SCALE

1:1

DATE	SYN	REVISION	RECORD	AUTH	DR.	CK.

TITLE: NOZZLE HI 4 CFM

MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPR6	05-17-94	1:1

DATE	SYN	REVISION	RECORD	AUTH	DR.	CK.

2.25 SCH. 40 PIPE

TITLE: COLLAR & FLANGE HI 4 CFM
MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPR7	05-17-94	1:1

DATE	SYM	REVISION	RECORD	AUTH	DR.	CK.

TITLE: OUTLET TUBE HI 4 CFM

MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPR8	05-17-94	1:1

DATE	SYM	REVISION	RECORD	AUTH	DR.	CK.

TITLE: ASSEMBLY

MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPRI	05-17-94	1:3

DATE	SYM	REVISION RECORD	AUTH	DR.	CK.

TITLE: SHROUD

MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPR2	05-17-94	1:3

DATE	SHEET	NO.	OF	TOTAL	REV.	BY	DATE

TITLE: PROBE HI 4 CFM

MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPR9	06-01-94	1:3

DATE	SYN	REVISION	RECORD	AUTH	DR.	CK.

TITLE: FIN HI 4 CFM

MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPRII	05-17-94	1:1

DATE	SYM	REVISION	RECORD	AUTH	DR.	CK.

TITLE: PROBE MI 4 CFM

MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPR10	05-17-94	1:3

DATE	SYM	REVISION	RECORD	AUTH	DR.	CK.

TITLE: FIN MI 4 CFM

MATL: STAINLESS STEEL

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M.G.	C.D.	3CMRPR12	05-17-94	1:1