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Particle Size Distribution of Automobile Paint Sprays

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Automobile paint spray operations produce a considerable amount of aerosol to which workers may be exposed. Different types of samplers may be used to distinguish the total amount of airborne solvent in droplets from that in vapor. For evaluation of total inhalation exposure from evaporating droplets the measurement of droplet size is also important. We found that the aerosols generated by base coat operations are generally large [geometric mean aerodynamic diameters (GMAD) range from 20 to 40 μm] and polydisperse [geometric standard deviations (GSD) range from 2 to 3]. Aerosols generated in clear coat operations are much larger and more polydisperse, with a GMAD greater than 50 μm and a GSD greater than 3. Measurements of aerosol size distribution demonstrated that inhalable particles are present near both workers and test stands. Thus, particles may contribute to overall solvent exposure. These results substantiate the need for periodic aerosol size distribution measurements and the use of prefiltered charcoal sorbent tubes for routine solvent exposure sampling in paint spray environments. Brosseau, L.M.; Fang, C.P.; Snyder, C.; Cohen, B.S.: Particle Size Distribution of Automobile Paint Sprays. Appl. Occup. Environ. Hyg. 7(9):607–612; 1992.

Introduction

Automobile paint spray operations result in worker exposures to both solvent vapors and solvent-containing droplets. Previously we described experiments in which solvent concentration from both vapor and droplets was shown to vary for three sampler types placed on test stands in a paint spray booth.[1] Charcoal sorbent tubes fitted with glass fiber filter precollectors (F-CST) showed significantly higher amounts of xylene vapor than plain (unfiltered) charcoal sorbent tubes (CST) and diffusion monitors (DM). These results indicated that solvent-containing droplets were present. The excess vapor measured by the F-CST must result from droplets that deposit on the filter and then evaporate into the air stream being drawn through the CST. These differences among samplers did not occur when they were used to measure individual worker exposures to base coat paint spray. We hypothesized that the disparity resulted from differences in particle collection by the samplers.

Efforts were undertaken to measure droplet size distributions of the evaporating airborne particles because droplet size is one determinant of sampling efficiency. This article reports the results. Specifically, these measurements determined whether aerosol size distributions differ between the test stand and worker locations. By comparing vapor measurements using prefiltered CSTs with unfiltered CSTs it is possible to estimate the amount of vapor associated with droplets; prefiltered CSTs should collect the solvent from both particles and vapor, while CSTs are thought to collect only the vapor.

It may not be correct to assume, however, that unfiltered CSTs collect only vapor, because they may capture particles as well. Aspirated particles may be trapped within the charcoal granule bed. In addition, collection efficiency may vary for different sized particles (i.e., collection efficiency may diminish with decreasing size). Thus, if F-CSTs and unfiltered CSTs are employed in locations with different aerosol size distributions (such as may occur between the personal sampling and test stand sites), greater vapor concentration differences between the two sampler types would be expected at the location (i.e., the test stand) with a relatively smaller droplet size distribution.

Experiments were designed to evaluate particle size–related collection efficiency of particles traveling through charcoal tubes and to measure the aerosol size distribution of spray paint in an automobile production facility. The latter was accomplished using cascade impactors located on test stands and in the breathing zone of workers.

A second goal of these experiments was to predict the inhalation hazards associated with solvent-containing paint spray droplets. The American Conference of Governmental Industrial Hygienists (ACGIH)[2] has recently suggested new Particle Size–Selective Threshold Limit Values (PSS-TLVs) for aerosol exposures. Such TLVs would be developed using one of three mass fractions, depending on

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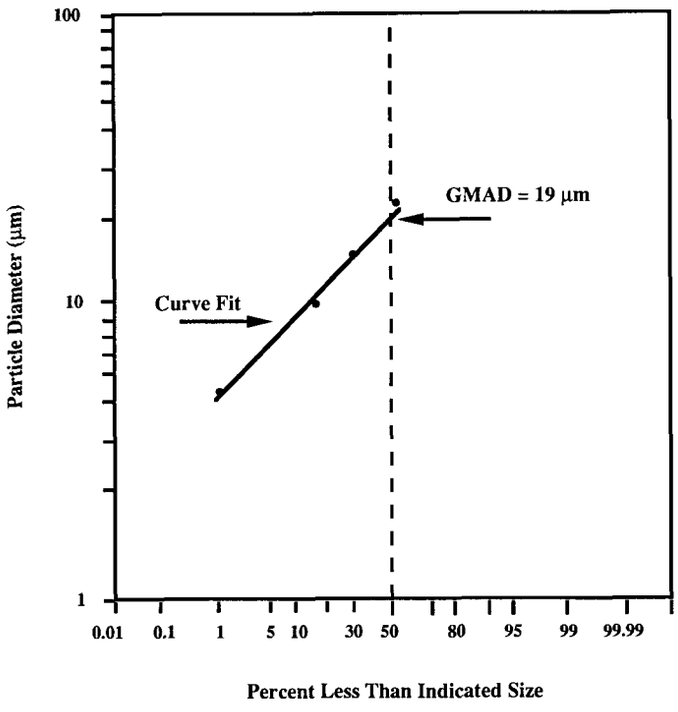

FIGURE 1. Example of Kaleidagraph® log probability plot used to determine paint spray size distribution, showing the GMAD and curve fit.

the health effects of a particular material. The ACGIH criteria for inspirable particulate mass indicate that about 80 percent of all particles with aerodynamic diameters (d_{ae}) less than 20 μm and about 50 percent of all droplets between 20 and 100 μm will penetrate into the head airways region. Once inhaled, tissue absorption of the solvent will probably be complete regardless of the site of particle deposition in the lung.

The size distribution measurements using cascade impactors, in combination with measurements of vapor downstream of the impactors, were used to predict whether workers would experience enhanced exposure from the presence of solvent-containing paint spray droplets.

Experimental Methods

Charcoal Sorbent Tube Particle Collection Efficiency

Collection efficiency of CSTs was examined over a range of flows (100–400 ml/min) for tubes selected randomly from a commercial lot (SKC Inc., Eighty Four, PA, lot 200). Arizona road dust (ARD) dispersed by a Wright dust feeder (BGI, Waltham, MA) was used to measure CST collection efficiency of particles of 0.5 to 5 μm diameter. These measurements were performed by determining particle concentrations upstream and downstream using an optical particle counter (Climet 208, Redland, CA) coupled to a multichannel analyzer (Canbera, Meriden, CT). In addition, Tc^{99m}-labeled sodium chloride particles (0.1 μm diameter) generated by an atomizer and electrostatic classifier were used to measure collection of an ultrafine aerosol. These latter measurements were made by counting individual sections of the charcoal tube in a gamma well scintillation counter and comparing activity to that collected on filters when no CST was present. Collection efficiency of the charcoal bed (E) was determined as the fraction of entering material retained by the CST:

$$E = \frac{N_{in} - N_{out}}{N_{in}} \quad (1)$$

where N_{in} and N_{out} are the number of entering and exiting particles, respectively.

Particle Size Distribution

Base Coat

Four- and six-stage cascade impactors (Series 290, Andersen Samplers, Inc., Atlanta, GA) were used to measure particle size distributions on both test stands and workers. Cut points for the four stage impactors were d_{ae} = 21.3, 14.8, 9.8, and 3.5 μm, while those for the six-stage impactors were d_{ae} = 21.3, 14.8, 9.8, 6.0, 3.5, and 1.55 μm, when these impactors are operated at 2 L/min. These cut points change somewhat for lower sample flows, and adjustments were made when actual flows were less than 2 L/min.[3] In addition, some of the impactors were operated with "visors" over the impactor entry, which change the impactor collection efficiency for particles greater than about 5 μm. Adjustments for visors were made in sampler "effectiveness" when necessary.[3]

Measurements were made in the manual spray booths (where base coat is applied) in plant 2 on two separate dates (trips 5 and 6), as described in a previous paper.[1] During the first sampling trip six size distribution measurements were made with 4-stage impactors (with visors) worn by workers. The impactors were loaded with preweighed stainless steel substrates and a final PVC filter. On the second trip, ten 6-stage and four 4-stage personal impactors (some with visors) were employed on both workers and test stands. Both Mylar® and stainless steel substrates were used, and glass fiber filters were employed on the final stage. To min-

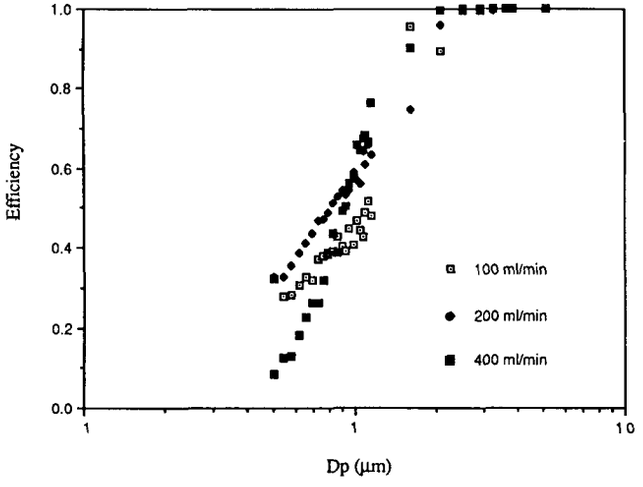

FIGURE 2. CST collection efficiency.

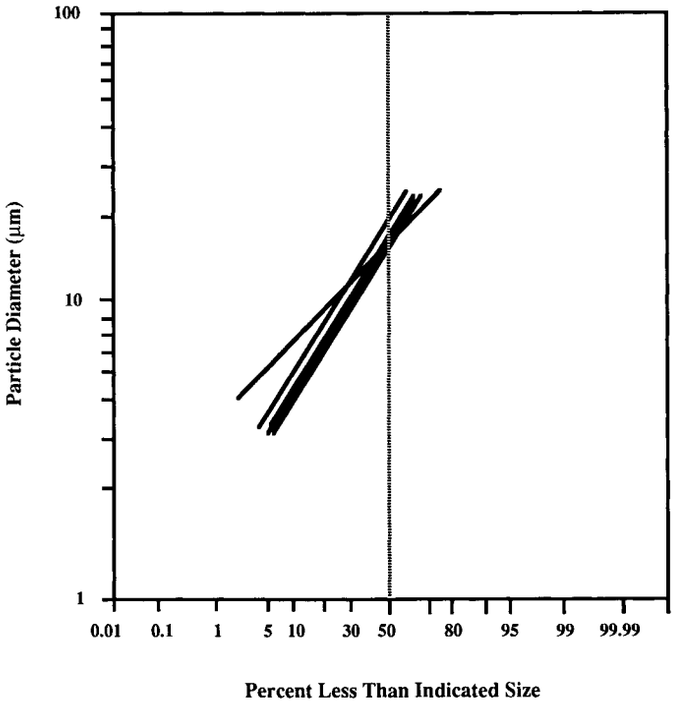

FIGURE 3. Size distribution measurements for all personal samples from trip 5.

imize solvent evaporation from the stages, the impactors were wrapped in foil and placed on ice immediately after sampling. They were removed from ice and unwrapped immediately before final stage weighing. Eight measurements of size distribution were made at each of the two sampling locations, for a total of 16 size distribution measurements.

Particle size distributions were calculated by a method described by Hinds.[4] Data were plotted using a computer spreadsheet program (Kaleidagraph by Abelbeck Software, Synergy Software, Reading, PA) showing particle d_{ae} (cut point) on a logarithmic scale versus percentage weight less than each size (on the probit scale). Curve fits were generated by the program and used to determine the GMAD and GSD of each sampled distribution. An example is shown in Figure 1.

Clear Coat

An additional sampling trip was made to evaluate the particle size distribution of aerosols generated during clear coat spray operations (trip 7).[1] Impactors were worn by workers and placed on test stands.

Other Samples

All personal sampling by impactors was accompanied by sampling with filtered and unfiltered CSTs, as well as DMs. The filter cassette inlets were enlarged to 15 mm to match that of the Institute of Occupational Medicine (IOM) Personal Sampler, the inlet efficiency of which matches the inspirable mass fraction recommended by the ACGIH Air Sampling Procedures Committee.[2,5] In addition, these three sampler types were matched to every impactor placed on the test stand. A CST was placed after each impactor to capture airborne vapor as well as vapor from evaporating particles (I-CST). Solvent collected on charcoal was desorbed and analyzed as described previously.[1]

Vapor on Impactor Substrates

Several additional impactor samples were used to analyze for solvent contained in the deposited particles. For these samples, the Mylar substrates were removed immediately after sampling and sealed into glass vials. They were stored on ice until analysis for xylene by carbon disulfide desorption and gas chromatography.

Results and Analysis

The collection efficiency of CSTs ranged from 10 percent for 0.5-µm particles at 400 ml/min to 100 percent for particles greater than about 2 µm (Figure 2). To be more accurate, a correction should be made for the differences in refractive indices and densities of experimental (ARD) and calibration (latex sphere) aerosols. The particle size should be corrected to d_{ae} by a factor of 1.62 (the square root of ARD density, 2.61 g/cm^3), which would cause a slight shift to the right of all data points. However, the general trends remain similar, where (1) smaller particles are collected with less efficiency than larger ones, (2) collection efficiency increases with increasing particle size, and (3) 50 percent collection efficiency occurs at or slightly above the 1-µm particle size. The collection efficiency of 0.1-µm particles (not shown) was found to be about 20 percent, which reflects the expectation that collection efficiency of smaller particles should increase as diffusion plays a stronger role in deposition on the charcoal bed.

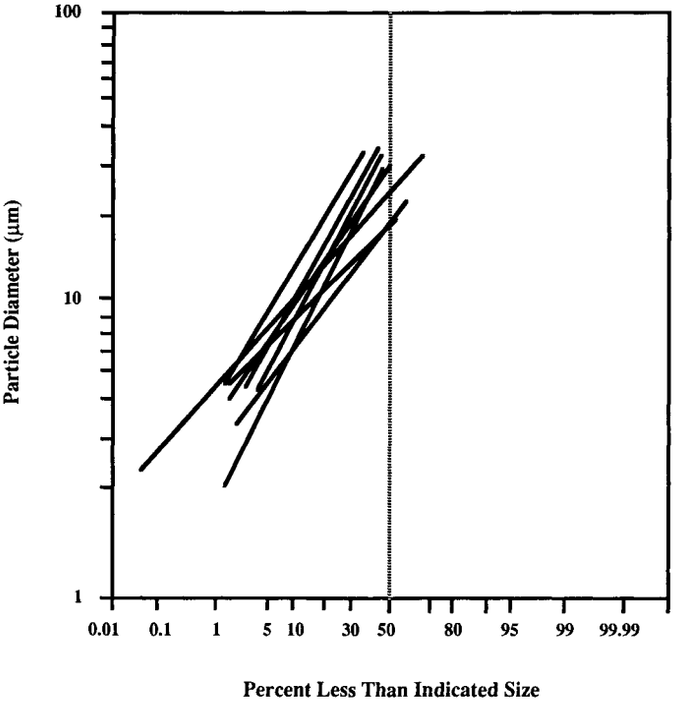

FIGURE 4. Size distribution measurements for all personal samples from trip 6.

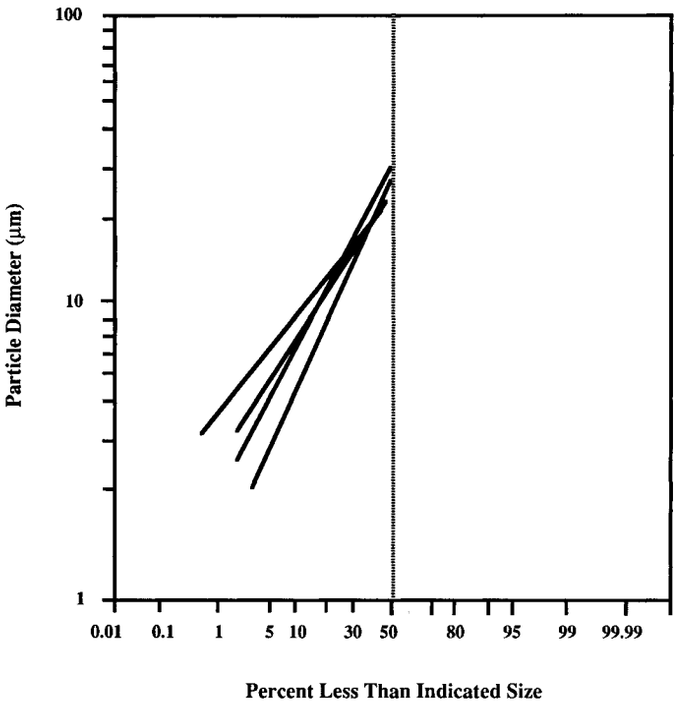

FIGURE 5. Size distribution measurements for all test stand samples from trip 6.

These results agree with those of Fairchild *et al.*,[6] where collection efficiency of monodisperse latex sphere and Di-sec octyl phthalate aerosols was measured over time for several CSTs. However, it is important to note that we did not examine the variability of collection efficiency between CSTs and, as noted by Fairchild *et al.*, this variability could be quite important, because CSTs are not designed as particle collectors.

The particle size distributions measured on the first sampling trip showed GMADs ranging from 15.7 to 20.5 μm and GSDs of 1.9 to 2.7 (Figure 3). A single four-stage Mercer impactor (In-Tox Products, Albuquerque, NM), with cut points ranging from 0.7 to 7 μm, was placed on the test stand. This showed more than 80 percent of the aerosol deposited on the top stage. It is difficult to ascertain aerosol size distribution when so much of the aerosol is larger than the largest cut point, but by plotting the points on lognormal probability paper and extending the line beyond the data, we extrapolated a GMAD of around 15 μm.

Results of the second sampling trip showed that GMADs for personal samples ranged from 19 to 46 μm, with an average GMAD of 23.9 μm (Figure 4). The aerosols were polydisperse with an average GSD of 2.7. The GMAD for the test stand samples ranged from 22 to 26 μm, with an average GMAD of 21.2 μm, and an average GSD of 2.5 (Figure 5).

Determinations of particle size from personal impactor samples of the clear coat aerosol were generally unsuccessful, because we could not obtain enough mass on the impactor substrates during allotted sampling periods. The one sample that did record enough mass to allow extrapolation showed that the median size of the clear coat aerosol was very large (53 μm) and that the aerosol was quite polydisperse, with a GSD of 3.6. This particle size distribution suggests that these impactors were not appropriate samplers for the clear coat operations, as the largest cut diameter is 20 μm at the recommended flow rate of 2 L/min.

Statistical analyses were performed to determine (1) if the base coat aerosol size distributions differed from one sampling trip to the other and (2) if these size distributions were different for test stands compared to workers. For these comparisons, the GMADs were evaluated using *t*-tests (two-sided with a 0.05 level of significance), as were the GSDs (Table I).

There was no difference between the personal aerosol size distributions for the two sampling trips ($0.5 > p > 0.2$ for GMADs and $p > 0.5$ for GSDs). Thus, all personal sample aerosol size distributions were tested for differences between the aerosol size distributions at the two sampling locations; no significant difference was found in the size distributions at the test stands versus the personal samples ($p > 0.5$ for both GMADs and GSDs).

Xylene vapor concentrations from sampling trip 6 were examined for differences among several factors by use of analysis of variance. This test indicated that sample type (CST, F-CST, DM, and I-CST) and location (test stand and workers) were significant sources of variation (Table II). Further analysis of the data by use of a Newman-Keuls multiple range test[7] showed that, for all samples, (1) F-CSTs were significantly higher than CSTs and DMs, (2) CSTs and DMs showed similar vapor concentrations, and (3) the I-CSTs were significantly lower than the other three sampler types. A similar analysis of the data after separation into test stand and personal samples showed no differences in any of the vapor concentrations measured by the personal samplers, as was found on previous sampling trips.[8] The differences described above were due entirely to differences in the test stand sampler results, with F-CST ≥ CST = DM ≥ I-CST (Table III).

The I-CSTs showed the lowest vapor concentration of all samplers. This was unexpected, because I-CSTs should col-

TABLE I. Geometric Means (GM) (μm) and Geometric Standard Deviations (GSD) for Personal and Test Stand Paint Aerosol Size Distributions

Personal Samples				Test Stand Samples	
Trip 1		Trip 2		Trip 1	
GM	GSD	GM	GSD	GM	GSD
18.8	2.5	19	1.8	23	2.7
15.7	2.6	39	3.0	26	2.4
20.5	2.7	36	3.2	26	2.3
17.8	2.5	46	2.7	26	2.4
17.0	1.9	31	3.3	25	2.6
		24	2.0	24	2.6
		19	2.3	22	2.4
		30	2.5		

TABLE II. Analysis of Variance for Measured Xylene Concentrations from Trip 6

Source	DF	MS	F	p
Time	1	180.7	21.3	0.0001[A]
Location[B]	1	746.2	87.8	0.0001[A]
Sample type[B]	3	62.2	7.3	0.0001[A]
Time × Location	1	63.6	7.5	0.0090[A]
Time × Sample type	3	13.2	1.6	0.2140
Location × Sample type	3	29.3	3.4	0.0240[A]
Time × Location × Sample type	3	8.2	1	0.4150
Error	48	8.5		
Total	63	27.6		

[A]Statistically significant ($p \leq 0.05$)
[B]Location = personal or test stand; Sample type = F-CST, CST, or DM

lect vapor plus evaporated solvent from particles and results should be comparable to F-CSTs. A fitting of unknown makeup (probably polystyrene) had been used to connect the impactor to the charcoal tube and was suspected of having adsorbed xylene vapor. To investigate the possibility we performed an experiment using a vapor generation technique described by Thomas.[8] Solvent was deposited onto a filter connected to a follow-up CST and air was then drawn through the system. Comparison of CSTs connected to filter cassettes showed that, when the fitting was in place, 21–27 percent less vapor was found on the CST than when no fitting was in line. If the vapor concentrations measured by the I-CSTs are adjusted for this adsorption effect by the fitting, the results are consistent with values for the CSTs and DMs on the test stand samples, although the results are still significantly lower than for the F-CSTs.

No xylene was detected on any of the impactor substrates. As in previous research, no xylene was found on any of the filters preceding CSTs. These results suggest that either particles were essentially dry when collected, or that they dried during sampling.

Discussion

The aerosols generated by automobile manual spray paint operations are large, with GMADs ranging from 20 to 40 μm and GSDs from 2 to 3. We hypothesized that the differences we originally found in solvent concentrations between the filtered and unfiltered CSTs on test stand and personal samples were due to particle size distribution differences at the two locations. This would have led to variations in CST collection efficiency, E, which differs for different-sized particles. For this hypothesis to have been correct, it would have been necessary to measure GMADs ≤ 1 μm near the test stands and about 10 μm near the workers. That this did not occur suggests that our hypothesis is incorrect; other factors probably caused the observed differences.

Total sampling efficiency of a device depends on both aspiration (entry) and collection efficiencies. Collection efficiency relies on particle deposition behavior within the sampler (i.e., the filter or charcoal bed for our experiments) and is relatively simple to predict or measure. The aspiration (entry) efficiency of a sampler also depends on particle size, generally decreasing for larger particles. Since it is reasonable to assume that droplet size decreases with distance from the source, resulting variations in entry efficiency would not explain the differences originally observed.

Aspiration efficiency can be predicted for relatively simple and stable situations, but is difficult to evaluate for unstable or unpredictable airflow conditions. When still air conditions exist, theoretical models can be used to predict a sampler's entry efficiency. For example, the Yoshida criterion[9] suggests that particles less than 35-μm diameter will be collected with 90 percent efficiency by the CSTs under our operating conditions. However, for our measurements, with air moving vertically downward at 37 m/min (120 ft/min), the conditions cannot be described as "still air." A model developed by Vincent *et al.*[10] suggests that, if the CST is treated as a thin-walled probe oriented at a 90° angle to the predominant wind direction, the entry efficiency will be 100 percent for 5-μm diameter particles and will decrease to about 20 percent for particles of 30-μm diameter. Thus differences in entry efficiency would have played an opposite role to collection efficiency in paint spray droplet collection by CSTs. While it is possible that our size distribution measurements are not representative of conditions during earlier sampling trips, we do not believe that substantial changes in aerosol generation occurred in the interim period.

One essential difference between the two sampling locations concerns the proximity of the worker to a hand-held spray nozzle. A reduced pressure zone in front of the worker may cause an airflow pattern that may give rise to nonideal sampler behavior. There are at present no theoretical models of sampler entry efficiency in the more complex environments found in workplaces. The IOM inhalable sampler, which we attempted to mimic by modifying the filter cassette entry, was evaluated under a variety of wind speeds and directions in a laboratory wind tunnel.[5] It was found to exhibit minimal deviance from the inhalable efficiency curve suggested by the ACGIH.[2] However, while this sampler may be adequate for many complex sampling situations, it is also possible that the filter cassette's entry efficiency could have been adversely affected by the presence of a hand-held spray nozzle.

It is important to note that the differences in sampler

TABLE III. Newman-Keuls Multiple Range Ranking of Mean Xylene Concentrations (mg/m³) for Four Sample Types by Location, Sampling Trip 6

	Sample Type			
Test Performed	I-CST	DM	CST	F-CST
All samples combined[A]	10.2	12.3	12.8	15.0
Test stand	11.6	16.2	16.7	19.5
Personal	8.4	8.8	9.0	10.5

[A]Lines indicate ranking whereby means not found to be significantly different are grouped and those significantly different are separated.

performance at the two sampling locations demonstrated in initial field experiments[1] were not borne out by our subsequent vapor measurements. Rather, a significant difference was found between F-CST and CST samplers at both locations. This agrees with the particle size measurements reported here, which demonstrated that particles of similar size were present at both locations. (Note: average xylene concentrations were also similar for all sampling trips.)

Conclusions and Recommendations

Paint spray aerosols with size distributions similar to those we measured will contain about 70 percent (by mass) inhalable, or inspirable, particles.[2] While we found no solvent on impactor substrates, any solvent on inhaled and deposited paint droplets will probably be absorbed within the respiratory tissue. The median size of these sprays was very large: most of the penetrating droplets would probably deposit in the nasal passages and upper airways. Since solvents are essentially a systemic toxin, both larger droplets deposited in the nasal region and smaller ones collected in the more distal respiratory tract will contribute to a worker's overall solvent exposure.

Both components (particle and vapor) of a paint spray aerosol should be assessed. In routine sampling, particles should be captured by filters preceding charcoal sorbent tubes, using a sampler that matches the ACGIH inhalable particulate mass criterion. To identify the important sites of deposition and to predict long-term health effects, size discrimination techniques should be employed periodically. Particle size distribution measurement of spray aerosols with impactors has an important drawback, however, because solids or pigments must be treated as surrogates for actual droplets. Better methods should be developed that would permit more accurate measurement of evaporating droplet size distributions.

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