### Title
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### Authors
Phalen, RF
Cannon, WC
Esparza, D



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# COMPARISON OF IMPACTION, CENTRIFUGAL SEPARATION AND ELECTRON MICROSCOPY FOR SIZING CIGARETTE SMOKE[1]

R. F. Phalen, Department of Community and Environmental Medicine, College of Medicine, University of California, Irvine, California 92664

W. C. Cannon, Biology Department, Battelle Northwest Laboratories, Richland, Washington 99352

D. Esparza, Aerosol Physics Department, Inhalation Toxicology Research Institute, Albuquerque, New Mexico 87108

## ABSTRACT

Determination of the size distribution of aerosols composed of small liquid droplets involves special problems, especially when the particles are numerous per unit volume of carrier gas. Electron microscopy, useful in examining most solid aerosols, is not generally applicable to liquids. Using unfiltered smoke from research cigarettes as a test aerosol, three techniques for obtaining a size distribution were compared. The aerosol, having a mass median aerodynamic diameter of about 0.7 micrometers, was examined with a seven-stage cascade impactor, a centrifugal separator, and by rapid fixation of the aerosol with methyl 2-cyanoacrylate vapor followed by electrostatic precipitation and electron microscopy. The resulting size distributions were compared and found to be in fairly good agreement. Rapid fixation in the airborne state appears to be a useful technique for preparing certain liquid aerosols for electron microscopy.

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## INTRODUCTION

The objective of this study was to compare three methods of obtaining size distributions of main-stream cigarette smoke. No attempt was made to reproduce cigarette smoke as it exists in the human smoking situation. Instead, laboratory methods of puffing, diluting, mixing and sampling were used so that the three sizing methods could be compared and so that other laboratories could repeat the studies. The methods for sizing were: a) use of a cascade impactor, b) use of a centrifugal, spiral aerosol separator, and c) coating of the smoke with methyl 2-cyanoacrylate, collecting with an electrostatic precipitator and sizing from electron micrographs. The latter method of sizing is not considered a standard laboratory technique.

Fresh cigarette smoke has several properties that make it difficult to characterize; it is chemically complex, initially highly concentrated and not in equilibrium with its surroundings with respect to moisture content, temperature, or vapor pressures of volatile components. In its fresh state it consists primarily of aqueous, spherical droplets with dissolved and suspended solid materials. Unless it is diluted, coagulation proceeds rapidly, and depending on the factors of dilution, the median size of a cloud may increase or decrease with time.

A great deal of research has been and is being done with respect to the health effects of cigarette smoke and there is a need for standardization of sizing methods. The methods compared in this study are all relatively straightforward and one or more probably could be made available to the majority of those investigating cigarette smoke.

## MATERIALS AND METHODS

### CIGARETTES

The Kentucky Reference IRI cigarette, stored at 60% RH and 20-23°C was used throughout.

### SMOKING PARAMETERS

The smoking machine (Figure 1) was recommended by C.H. Keith (Celanese Fibers Company, Charlotte, N.C.). It consisted of a short glass cigarette holder connected to a 3-necked 500 ml glass flask fitted with a metal stirring paddle and a critical orifice through which a timed flow of air could be drawn. The cigarette holder was removed to permit sampling.

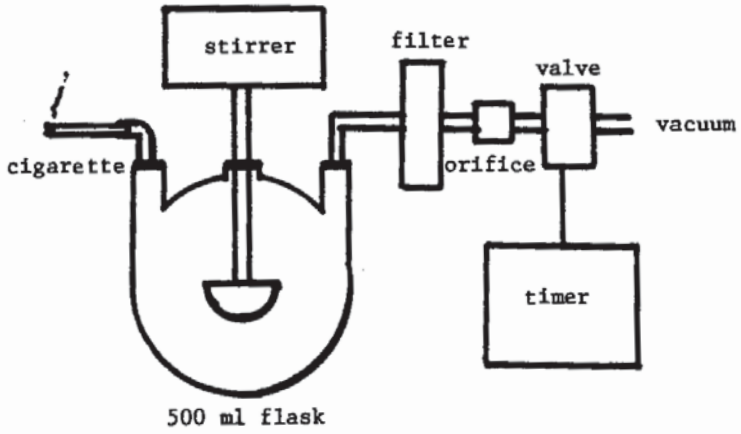


Figure 1. Machine used to generate cigarette smoke for comparison of three methods of sizing sub-micron droplets.

The smoking regime began with lighting the cigarette during a 35 ± .5 ml puff of 0.94 seconds duration. Two subsequent puffs of the same size were taken at 1 min. intervals. After the third puff the smoke was purged from the flask. The fourth puff, 1 min. after the previous puff, was sampled for 15 seconds, 5 seconds after it was drawn into the flask.

### SAMPLING DEVICES AND METHODS

A seven stage cascade impactor of the type designed by T. T. Mercer[2] was used. When operated at 500 cc/min. the effective cut-off diameters of the impactor used were:

| | |
|---|---|
| Stage 1 | 6.7 μm |
| 2 | 4.3 μm |
| 3 | 3.0 μm |
| 4 | 2.3 μm |
| 5 | 1.6 μm |
| 6 | 1.0 μm |
| 7 | 0.46μm |
| filter | 0.0 μm |

Impactor stages were covered with glass cover slips coated with a silicone-oil spray. After sampling, the deposit on each impactor stage was dissolved in a fixed volume of isopropyl alcohol and aliquots were assayed with a spectrophotometer at a wavelength of 350 nm. The relative distribution of material on each stage was fitted to a log-normal distribution and the mass median diameter and geometric standard deviation determined. Two cigarettes were used for each impactor run.

Smoke was drawn into a Lovelace Aerosol Particle Separator (LAPS) (P. Kotrappa and M.E. Light[3]) for 15 seconds at 310 cc/min. The device had been calibrated with monodisperse polystyrene spheres and found to collect particles from 0.54 μm to 5.1 μm along a 40 cm-long metal foil strip. After sampling, the strip was cut into 22 segments and assayed photometrically by the same method used with the impactor. The exit filter was also assayed. Six cigarettes were used for each run.

Samples for electron microscopy were collected using a point-to-plane electrostatic precipitator operated at 50 cc/min. for 15 seconds (P.E. Morrow and T. T. Mercer[4]). Before these sampling runs 3 drops of methyl-2-cyanocrylate (Polysciences, Inc., Warrington, Pa.) were placed in the 500 ml smoke chamber and the flask was heated to 70°C (this method is similar to that described by G.P. Morie, C.H. Sloan and V.G. Peck[5]). Samples were collected on carbon-coated diffraction grating replicas with line spacings of 0.833 μm. Sizing was performed using a Zeiss TGZ3 Analyzer (Carl Zeiss, Inc., N.Y., N.Y.) from photomicrographs with a magnification factor of about 30,000. An estimate of mass median aerodynamic diameter was made by assuming a density of unity and that the aerodynamic diameter of agglomerates containing n primary particles was equal to $n^{1/3}$ times the average primary particle diameter.

Impactor and spiral-centrifuge samples were taken at two locations, the Inhalation Toxicology Research Institute (ITRI), Albuquerque, New Mexico, and at Battelle Northwest Laboratories in Richland, Washington (BNW) using essentially identical smoking machines and sampling methods. The precipitator samples were taken only at ITRI.

## RESULTS AND CONCLUSIONS

The agreement between all sampling methods and, in fact, all sample runs was within the experimental errors of a given technique. The table gives the results of each sampling run. In each case, the size distribution was approximately log-normal and the mass median and geometric standard deviation ($\sigma g$) were used to describe the results.

| run | method | average MMAD | $\sigma g$ | location |
|---|---|---|---|---|
| 1,2 | Impactor | 0.74 μm | 1.4 | ITRI |
| 3-7 | Impactor | 0.71 μm | 1.4 | BNW |
| 8 | LAPS | 0.79 μm | 1.3 | ITRI |
| 9-11 | LAPS | 0.72 μm | 1.4 | BNW |
| 12 | EM | 0.57 μm | 1.5 | ITRI |

The precipitator sample contained spherical particles, many of which were in cluster or chainlike agglomerates (Figure 2). Primary particles, sized from electron micrographs (n=191),were log-normally distributed with a count median diameter of 0.29 μm and a $\sigma g$ of 1.4. A background (no cigarette) precipitator sample yielded some spherical particles, negligible in quantity when compared to samples of cigarette smoke.

Since results from each of the three methods were in close agreement, it is concluded that any of the three techniques could be used for describing the size distributions of cigarette smoke except when extreme accuracy is required. Furthermore, the fixation by coating with methyl-2-cyanoacrylate may be of use in electron-microscopic studies of other aqueous aerosol particles.

## ACKNOWLEDGEMENTS

This study was suggested by Dr. Charles H. Keith and performed with the invaluable assistance of Robert Yarwood.

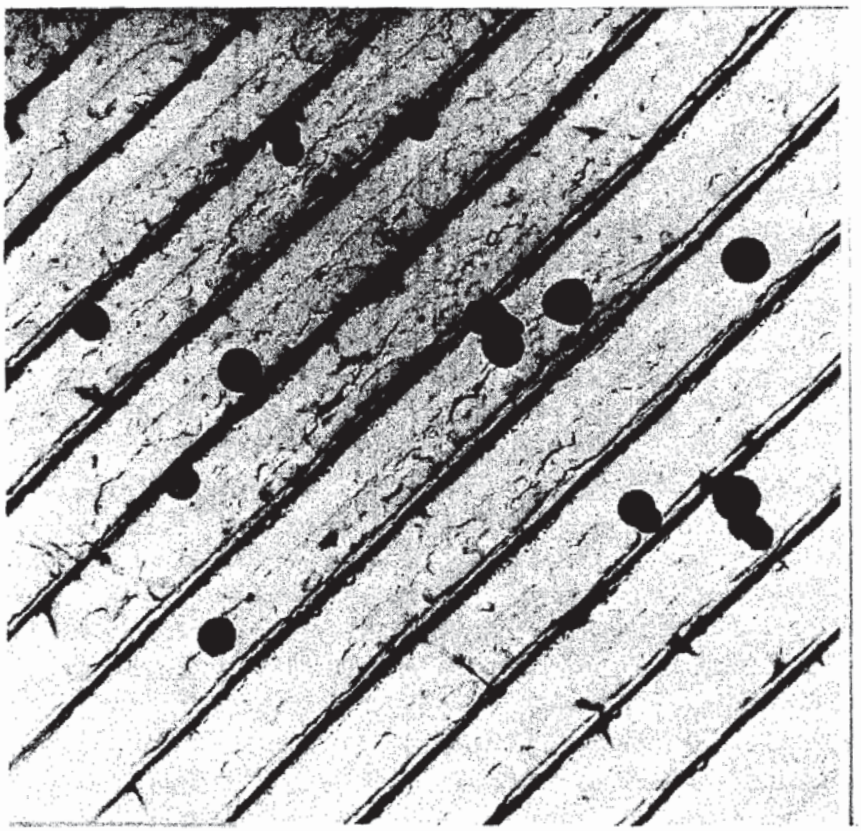

Figure 2. Electron micrograph of cigarette smoke particles fixed by coating with methyl-2-cyanoacrylate vapor. Diffraction-grating-replica spacing is 0.833 μm.

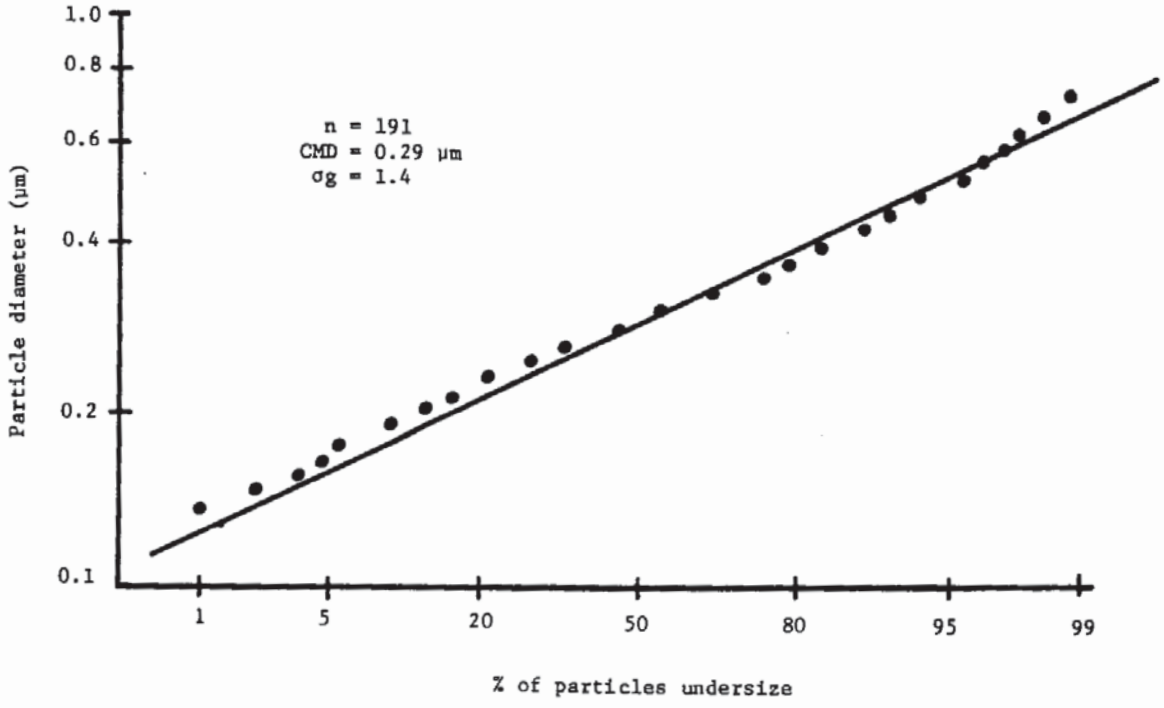


Figure 3. Size distribution of cigarette smoke primary particles after coating.

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