

# Measurement of the Particle Size Distribution and Concentration of Cigarette Smoke by the "Conifuge"

C. H. Keith and J. C. Derrick

Research Department, Liggett and Myers Tobacco Company  
Durham, North Carolina, U.S.A.

## Introduction

The smoke produced by a burning cigarette is a commonly encountered aerosol of considerable physiological significance. It also is an uncommonly concentrated aerosol which presents many of the various complications that make measurement of the particulate concentration and size distribution of aerosols quite difficult. These considerations have led a number of workers, particularly Sano *et al.* (1), Langer and Fisher (2), and Holmes *et al.* (3), to use it as a test aerosol with varying degrees of success. In this paper, a centrifugal sedimentation technique of general applicability to aerosols within a 0.05 to 10 micron size range is applied to tobacco smoke.

A centrifugal sedimentation technique is particularly suited to an aerosol such as tobacco smoke which combines a very high particulate concentration, a small and somewhat nonuniform particle size, and volatile and chemically unstable components (4, 5). The chief advantage arises from the indirect size estimation possible with such a technique, thereby avoiding the difficulties encountered with the use of microscopic, light-scattering, or electrostatic weighing techniques for size measurement.

Of the centrifugal techniques, impactors have been most commonly utilized for tobacco smoke (2-4).

Some difficulty has been encountered, particularly with multiple-stage impactors, in a considerable loss of material through re-entrainment of previously deposited smoke particles. A centrifugal instrument which avoids this problem and provides a continuously graded spectrum of particle sizes is the "Conifuge" developed by Sawyer and Walton (6). Their demonstration of the ability of this machine to collect and size grade a variety of liquid and solid aerosols made it apparent that this approach had distinct possibilities for the analysis of tobacco smoke.

The instrument developed by Sawyer and Walton collected particles larger than 0.5-micron in diameter. To accommodate tobacco smoke, a new instrument based on the same principles was constructed. The design was such that particles with diameters ranging from 0.05 to 10 microns could be sampled.

This paper describes this instrument and its application to the particle size analysis of cigarette smoke. The limited data included serve to demonstrate the usefulness of the equipment for this and other aerosol problems, and allow the estimation of some of the factors of importance in altering the concentration of smoke issuing from a cigarette.

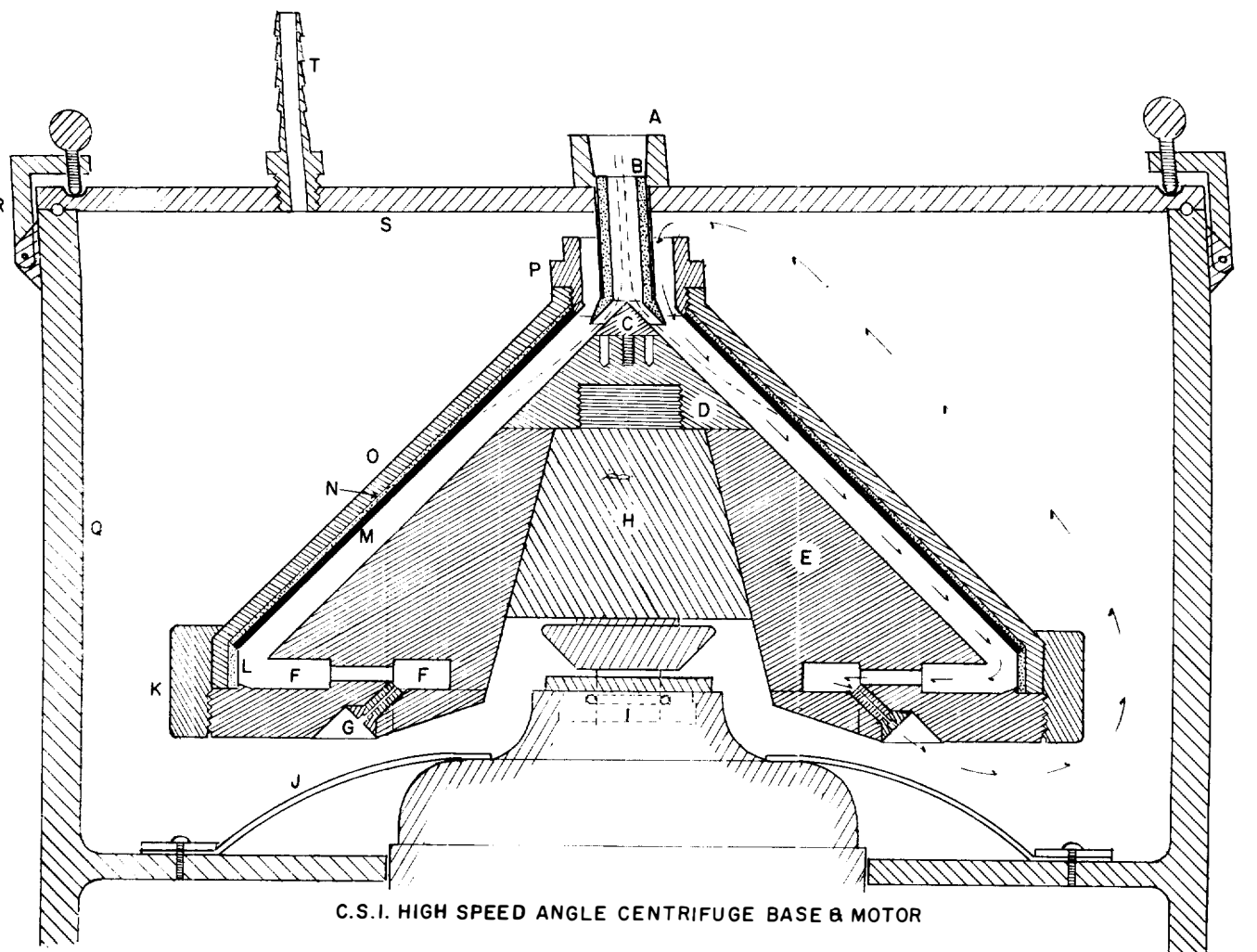
## Equipment

*The "Conifuge."*—A cross-sectional view of the centrifuge head and the surrounding air chamber is given in **Figure 1**. When rotating at high speeds, the head acts as a centrifu-

gal air pump. The centrifugal force imparted to the air in the inner chamber between cones (*E*) and (*O*) causes it to flow from the axially located entrance tube (*P*) to and out of the off-axis outlet jets (*G*). Since the rotating head is enclosed in an airtight chamber (*Q* and *S*), this flow is recirculated through the outer chamber back to the entrance tube, as indicated by the arrows of **Figure 1**. If a small portion of this circulating air is removed through the outlet tube (*T*), an equivalent volume of smoke is drawn in at (*A*). This incoming smoke is directed at the apex of the spinning head and spreads out in a thin film on the surface of the inner cone. As this film and the overlying clean, circulating air are carried downward and outward through the conical chamber an increasing centrifugal force is applied to the individual particles owing to their excursion away from the axis of rotation. In conjunction with the increasing centrifugal force, the forward velocity of the air stream in which the particles are embedded is decreasing as the annular area of the conical chamber increases. This combination of centrifugal force and stream velocity determines the trajectory of the individual particles. With a given set of dimensions and rotational velocity and assuming no significant slippage between the rotating head and incoming air stream, the trajectory of each particle is uniquely determined by its Stokes-Cunningham settling velocity. Since the trajectory of each particle determines its point of deposition on the

<sup>1</sup>Presented in part at the Southeastern Regional Meeting of the American Chemical Society, Durham, North Carolina, U.S.A., November 14, 1957.

<sup>2</sup>Reprinted with permission from the *Journal of Colloid Science*, 15:340-356, (1960).



C.S.I. HIGH SPEED ANGLE CENTRIFUGE BASE & MOTOR

- |                             |                                     |
|-----------------------------|-------------------------------------|
| A, SMOKE INLET TUBE         | K, CLAMPING RING                    |
| B, TEFLON GUIDE TUBE        | L, TEFLON CUSHION FOR BASE OF SLIDE |
| C, APEX CAP NUT             | M, DOVETAILED DARK FIELD SLIDE      |
| D, MOUNTING NUT             | N, DOVETAILED NYLON SLIDE HOLDER    |
| E, INNER CONE               | O, OUTER CONE                       |
| F, FLOW EQUILIZING CHAMBERS | P, AIR ENTRANCE TUBE                |
| G, OUTLET JET               | Q, CHAMBER WALL                     |
| H, C.S.I. ROTOR HUB         | R, CLAMP                            |
| I, "O" RING SHAFT SEAL      | S, CHAMBER TOP                      |
| J, COPPER CHAMBER SEAL      | T, OUTLET TUBE                      |

0 1 2  
INCHES

Figure 1. Cross sectional diagram of the "Conifuge."

outer wall of the chamber, a deposit is formed there which is continuously graded according to particle settling velocity. For spherical droplets of uniform density, such as cigarette smoke, the gradation may be expressed as a function of particle diameter. A representative sample of this graded size distribution suitable for counting numbers of particles is obtained from two dark-field microscope slides (*M*) embedded in the outer wall.

The conical shape of the chamber and the changing balance between stream velocity and centrifugal force are such that an extensive range of sizes are adequately represented on a relatively short collecting slide. The

easily centrifuged and relatively less numerous large particles are deposited near the apex of the cone or the head of the slide. The high stream velocity in this region fully sorts these particles, so that their distribution is opened out along the slide. However, an area deposit sufficient for counting purposes is maintained since the ring area in this region is relatively small. The more numerous smaller particles pass further down the conical chamber into a region of slower stream velocity and higher centrifugal force. This results in a compression of the range of sizes collected on a given length of slide, but the larger ring area in this region counterbalances this effect, so that

the area deposit is not excessive.

By equating the centrifugal driving force and Stokes' law resistive forces acting on a particle, Sawyer and Walton (6) formulated an equation for the trajectory of individual particles. This equation is given in their paper and is not reproduced here. The derivation contains a number of assumptions, so that their equation is at best only approximate, particularly for large particles. Because the integrated equation is not analytical, the deposition points for particles of given settling velocities are best obtained by superimposing these trajectories on an outline of the chamber dimensions. Such plots were utilized to design a chamber of

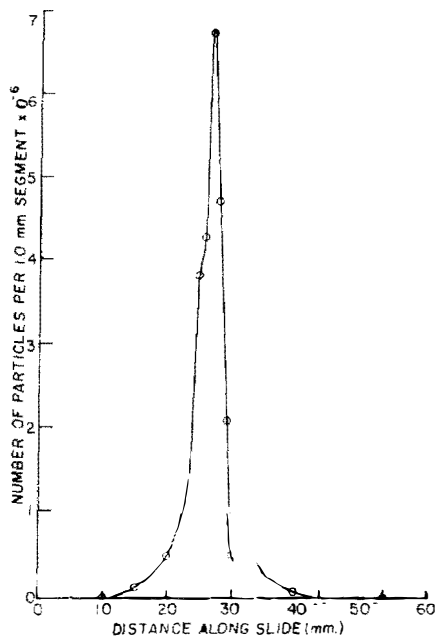


Figure 2. Deposition pattern of 0.814 micron polystyrene aerosol in the "Conifuge."

sufficient dimensions to capture the range of sizes thought to be present in cigarette smoke.

One of the chief requirements for the satisfactory operation of the conifuge is a steady, nonturbulent air flow in the space between the cones. The design was made with this in mind, and sufficient flexibility was incorporated in the equipment so that turbulence might be avoided by an experimental choice of operating conditions. Extensive precautions were also taken in the machining of the head to eliminate any places which might cause turbulent mixing of the smoke and clean air streams. A set of operating conditions were experimentally chosen on the basis of excellence of size gradation, this being taken as an indication of lack of turbulence. These were a flow through the conical chamber of 54 c.c./sec. and a rotational velocity of 3000 r.p.m. Other chosen or fixed parameters were a horizontal cone separation of 1 cm., a semivertical angle of 45°, a 12.5-cm. slide length, and a smoke sampling velocity of 5 c.c./sec.

The design and construction of the equipment was considerably simplified and the cost reduced by adapting a commercial centrifuge to provide the driving mechanism. The machine used was a C.S.I. angle centrifuge (Custom Scientific Instruments, Arlington, New Jersey). The modifications chiefly consisted of replacing the manufacturer's safety shield with the airtight chamber and constructing the special centrifuge head, both of which are shown in Figure 1.

**Auxiliary Equipment.**—Several pieces of equipment were necessary for reproducibly obtaining and immediately diluting cigarette smoke for use in the conifuge. The latter was necessary because coagulation of the extremely concentrated raw smoke stream would significantly change its particle size distribution very rapidly. A several hundred fold dilution was made by surrounding the smoke stream as it issued from the cigarette with a high-velocity, clean air stream. Both streams were directly introduced into a 12-l. flask from which smoke samples could be drawn through a small sampling chamber into the conifuge. The sampling chamber was fitted at either end with plunger valves so designed that a small volume of smoke could first be drawn into the chamber and subsequently into the conifuge without interrupting the normal flow of clean air into the instrument.

Since immediate dilution of the smoke stream was necessary, it was not possible to smoke the cigarettes in the orthodox manner by applying a controlled vacuum pulse to the end of the cigarette. It was, instead, necessary to puff on the cigarette by forcing air through the burning cone. This was accomplished by placing a bell over the previously lighted cigarette just before a pulse of slightly compressed air was delivered from a 4-l. storage tank.<sup>3</sup> The duration and intensity of the puff were controlled by a clock-operated solenoid valve and an adjustable pressure drop in the supply line to the bell. The larger diluting air stream was supplied from the same tank and was similarly controlled so that the ratio of diluting air to smoke remained constant during the puff. The timing was such that the diluting air was flowing slightly before and after the puffing air stream. Save for the use of compressed air in place of vacuum, the mechanism employs the same principles as the smoking machine described by Keith and Newsome (7), and was adjusted to take a similar, reproducible 44-c.c., 2-sec. puff, every half minute.

A Bausch and Lomb research microscope equipped with a cardioid dark-field condenser, 20-power apochromatic objective, and a 25-power compensated eyepiece fitted with a standard Whipple disk was used for counting the number of particles collected on the dark-field slides. The

<sup>3</sup>In some early work, a puffing mechanism was used which utilized the suction induced by the high velocity diluting air stream to draw on the cigarette. This was discarded because of the extreme difficulty in controlling the volume and other characteristics of the puff.

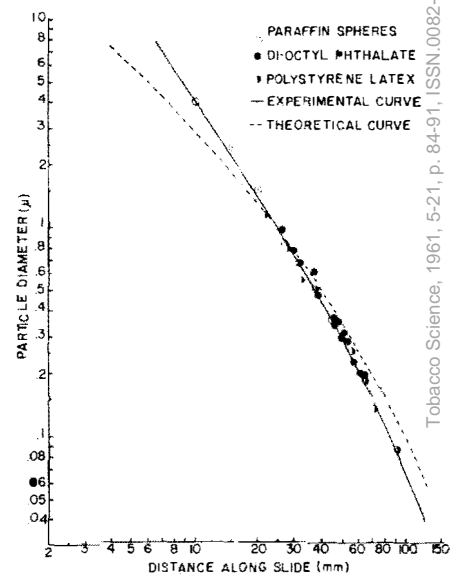


Figure 3. Theoretical and experimental calibration curves for the "Conifuge."

light source was a B and L Model 48 carbon arc microprojector. The slides require an excellent flame-polished optical surface for they must be examined with a dry upper surface to avoid possible shifting of the position of the captured smoke particles. Suitable slides were obtained from the Baltimore Instrument Company of Baltimore, Maryland. The slides were prepared for use by cleaning in nitric acid and rinsing with water and redistilled alcohol. Any residual dust was then removed by coating the slide with collodion and subsequently removing the collodion film. The final step consisted of rubbing on a thin film of silicone oil, which prevented the captured smoke particles from spreading.

### Experimental Methods

**Calibration of the Conifuge.**—In order to establish the relationship between particle size and place of deposition (as measured by distance from the head of the collecting slide), the conifuge was calibrated with several test aerosols, which were physically similar to tobacco smoke. These were atomized molten paraffin wax, homogeneous dioctyl phthalate aerosols obtained from a LaMer-Sinclair generator (8), and the homogeneous aerosols formed by atomizing greatly diluted samples of Dow monodisperse polystyrene latex by the method described by O'Konski and Doyle (9).

For the paraffin aerosol, the place of deposition of 0.5-micron or larger particles was correlated with particle size, as measured by ordinary microscopic techniques. For the homogeneous test aerosols the position of maximum deposition was correlated with

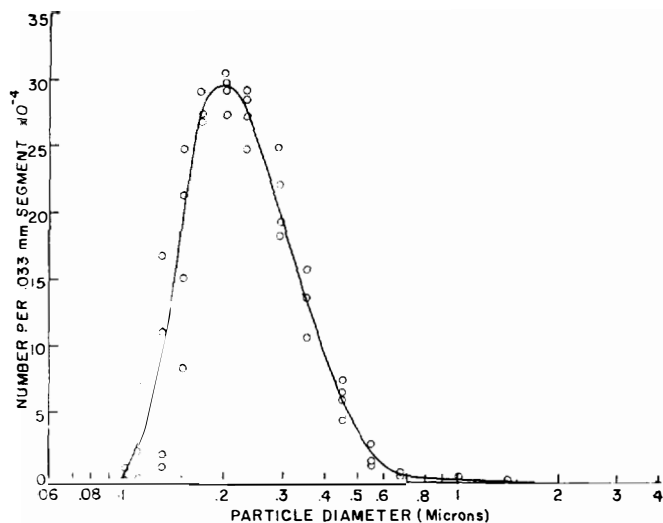


Figure 4. Reproducibility of the particle size distribution of cigarette smoke.

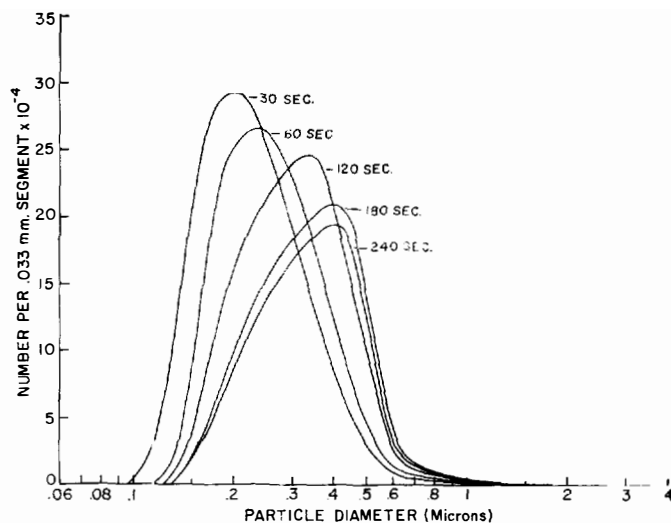


Figure 5. The effect of aging on diluted cigarette smoke.

the previously determined particle size. These sizes were determined by the red band structure of the D.O.P. aerosol and from the given particle size of the polystyrene dispersion, the measurement of which has been described by Bradford and Vanderhoff (10).

Figure 2 illustrates the distribution of particle counts obtained from 0.814-micron polystyrene particles. The ordinate of this curve is the calculated number of particles present in a conical segment of the outer cone of 1 mm. slant height, the calculation being based on the average slide count in that region. The abscissa is the distance from the top of the sampling slide in millimeters. The extreme sharpness of the maximum deposition peak is an indication of the quality of the size gradation obtained in this machine. The slight inflection on the left side of the curve and the somewhat slower rise on this side are thought to be caused by the presence of double and larger aggregate polystyrene particles which were formed either during the atomization process or by subsequent coagulation. The rapid fall of the curve on the right side, where the choice is between one or no polystyrene particles per atomizer droplet, is more representative of the deposition of a truly monodisperse aerosol.

The calibration curve for the instrument is given in Figure 3. The separate calibrations form a continuous curve for particles between 5 and 0.08 microns, indicating an acceptable size grading action for liquid and solid aerosols over this range of particle size. The deposition curve predicted by Sawyer and Walton's equation (6) is given by the

dashed curve of Figure 3. The departure of the two curves in the region of large particles is not unexpected in light of the assumption made in their development. In the small particle region the two curves again depart. This occurs because the equation predicts the maximum distance at which deposition of a particle of a certain size would occur. The experimental points are, however, based on the position of maximum deposition of a given particle size. From the distribution of deposition distances of the homogeneous aerosols, such as that illustrated in Figure 2, it is found that the points of furthest deposition agree well with the theoretical curve.

*Size Distribution of Cigarette Smoke.*—Uniformly blended cigarettes of known weight and resistance to draw were conditioned to an equilibrium moisture content by storage in an atmosphere of constant humidity and temperature. In the smoking operation, the cigarette was lighted by the operator, and the routine was continued in the previously described equipment. After sampling and collection of the smoke, the slides were removed and the numbers of particles counted<sup>4</sup> at 5-mm. intervals along the slides. In general for each slide, 5 Whipple disk squares, each representing a field 0.033 by 0.033 mm. and containing from 1 to 30 particles, were counted. The average of these counts from both slides was multiplied by the appropriate factor to obtain a count of the number of

<sup>4</sup>Although there was a generally low and somewhat variable background count for clean slides, no correction was applied to the observed counts. During the counting operation particles and slide imperfections which obviously appeared to be foreign to the sample were not included in the count

particles in the frustrum of a cone of 0.033 mm. slant height and with a radius equivalent to that of the outer cone of the conifuge at the counting position. Plotting these counts against a particle diameter as obtained from the calibration curve<sup>5</sup> formed a size distribution curve. A logarithmic scale was used for the diameter axis to open out the smaller particle end of the scale.

Graphical integration of the size distribution curves was used to compute the total number of particles collected in the conifuge. This figure divided by the known volume of the sample gave the concentration of diluted smoke, which in turn was converted to a concentration of raw smoke by multiplication with the dilution ratio. This ratio was generally 295 volumes of diluted smoke to 1 volume of raw smoke.

As was pointed out by Sawyer and Walton (6), the size distribution curves can be readily transformed into mass distribution curves, and from these the total mass of the sample may be obtained by integration. Computations of these quantities were not extensively used in this work, since the results are generally imprecise. This arises from the relatively few counts for large particles, which contribute heavily to the mass

<sup>5</sup>The diameters, as read from the calibration curve, are those for unit density particles, and may be called a "settling diameter" for particles of other densities. The "settling diameter" is defined as the product of the actual particle diameter and the square root of the density of the particle. These diameters were used throughout this work for lack of exact data on the density of cigarette smoke particles. Sam (4) measured a value of 0.98 g./c.c. and Sano (1) obtained values ranging from 0.92 to 1.00 g./c.c. Using Sam's value or Sano's highest values causes no sensible change in converting from settling to actual diameters. Using Sano's lower value would increase the listed diameter values by a factor of 1.27.

**Table 1. Effect of Smoking and Cigarette Variables.**

Cigarette conditions	Smoking conditions <sup>a</sup>	Most freq. occurring diameter <sup>b</sup> ( $\mu$ )	Mean <sup>b,c</sup> diameter ( $\mu$ )	Standard deviation ( $\mu$ )	No. of particles <sup>d</sup> per c.c. $\times 10^{-11}$
Blended, 1.08 g., 11% H <sub>2</sub> O	4th, 44-c.c. puff	0.21	0.23	0.14	3.01
Blended, 1.08 g., 11% H <sub>2</sub> O	4th, 35-c.c. puff	0.23	0.23	0.14	2.97
Blended, 1.08 g., 11% H <sub>2</sub> O	4th, 55-c.c. puff	0.21	0.21	0.14	2.89
Blended, 1.08 g., 11% H <sub>2</sub> O	2nd, 44-c.c. puff	0.22	0.21	0.14	2.24
Blended, 1.08 g., 11% H <sub>2</sub> O	5th, 44-c.c. puff	0.23	0.23	0.14	4.13
Blended, 1.08 g., 11% H <sub>2</sub> O	7th, 44-c.c. puff	0.23	0.21	0.14	4.66
Blended, 1.08 g., 11% H <sub>2</sub> O	10th, 44-c.c. puff	0.22	0.21	0.14	4.31
Blended, 1.00 g., 11% H <sub>2</sub> O	4th, 44-c.c. puff	0.23	0.22	0.14	3.57
Blended, 1.13 g., 11% H <sub>2</sub> O	4th, 44-c.c. puff	0.20	0.21	0.13	2.86
Blended, 1.08 g., <sup>d</sup> 3.6% H <sub>2</sub> O	4th, 44-c.c. puff	0.22	0.22	0.14	4.66
Blended, 1.08 g., <sup>d</sup> 6.2% H <sub>2</sub> O	4th, 44-c.c. puff	0.23	0.21	0.14	4.54
Blended, 1.08 g., <sup>d</sup> 20.4% H <sub>2</sub> O	4th, 44-c.c. puff	0.20	0.22	0.14	1.65
Bright, 1.09 g.	4th, 44-c.c. puff	0.20	0.21	0.14	3.25
Burley, 0.90 g.	4th, 44-c.c. puff	0.20	0.20	0.14	3.01
Turkish, 1.15 g.	4th, 44-c.c. puff	0.20	0.20	0.14	3.07

<sup>a</sup>All puffs of 2 seconds' duration taken at 1/2 minute intervals.

<sup>b</sup>Extrapolated values at zero aging time.

<sup>c</sup>Estimated from a logarithmic-probability plot of particle size against cumulative frequency, the geometric mean being the size at which 50% of the particles are greater and less than that size. The standard deviation is taken as the ratio of the 84% size to the 50% size adjusted to the size range under consideration (13).

<sup>d</sup>Weight of the cigarettes at 11% moisture.

distribution and total mass of the smoke deposit. Calculations of the total mass of smoke were, however, found to be in reasonable agreement with estimates of this quantity obtained by direct weighing of the total particulate matter collected in a suitable trap.

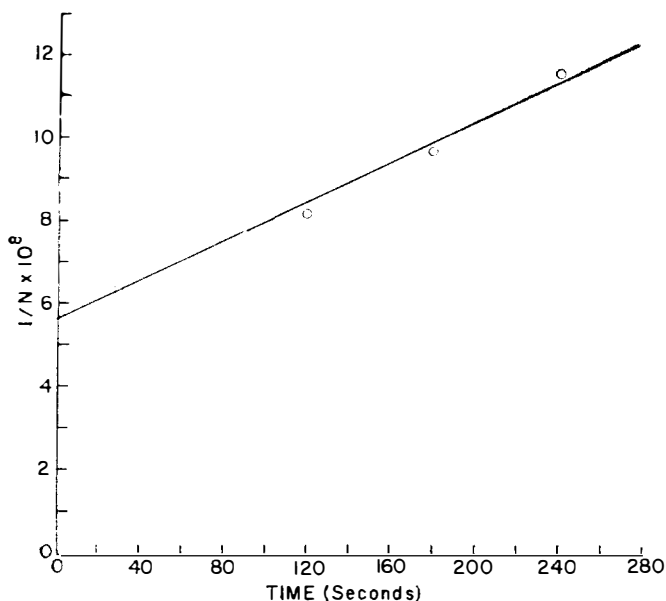
Measurements of the size distribution of side stream smoke were obtained by allowing a cigarette, selected as above, to freely burn without puffing in the 12-1. collection flask for 1 min. The resulting sample was collected and counted in the same manner as main stream smoke samples.

**Results and Discussion**

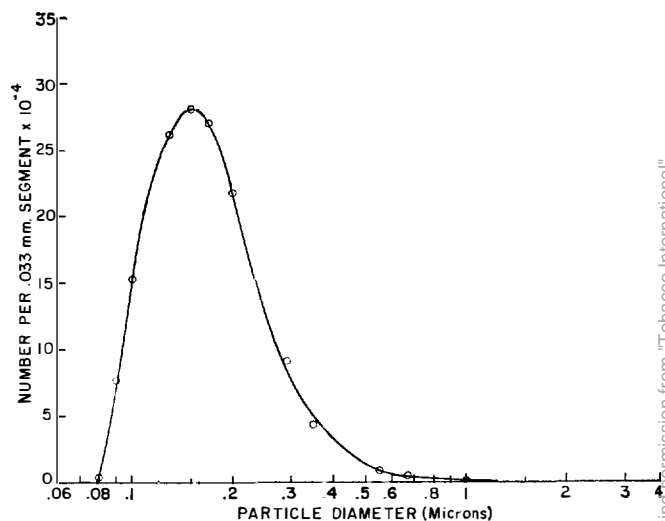
*Reproducibility of Collection of Cigarette Smoke.*—In order to test the reproducibility of the particle size distribution obtained from selected cigarettes, four separate cigarettes were smoked and the smoke from the fourth puff was diluted, aged for 30 sec., and collected in the confuge. The results of these measurements are illustrated in **Figure 4**, where the circles represent the measured counts and the smooth curve represents the average size distribution. Although there is considerable scatter of the points, the

figure shows that it is possible to reproduce the particle size distribution from one cigarette to the next. A subsequent series of ten measurements, each on a separate cigarette, gave essentially the same results for diluted smoke aged 60 sec., the most frequently occurring diameter being 0.21 micron, the geometric mean diameter being 0.23 micron, and the standard deviation being 0.14 micron, the latter being obtained in the manner outlined in **Table 1**.

*Aging Studies on Cigarette Smoke.*—Although the previously described puffing mechanism provides a large



**Figure 6.** Relation between particulate volume and aging time.



**Figure 7.** Particle size distribution of side stream cigarette smoke.

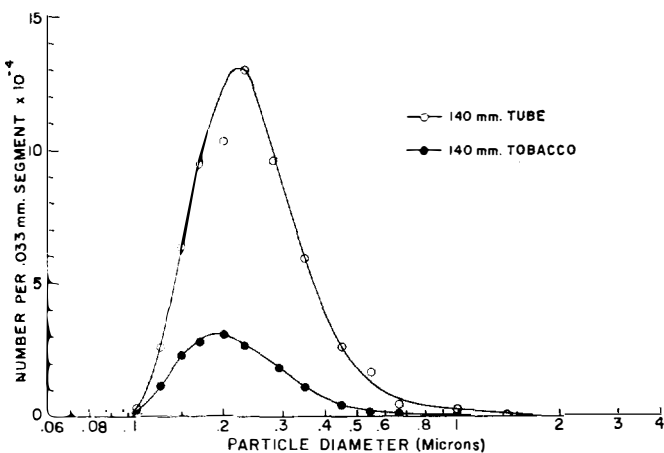


Figure 8. Particle size distribution of tobacco smoke after coagulation and filtration in a 140-mm. tube and tobacco column.

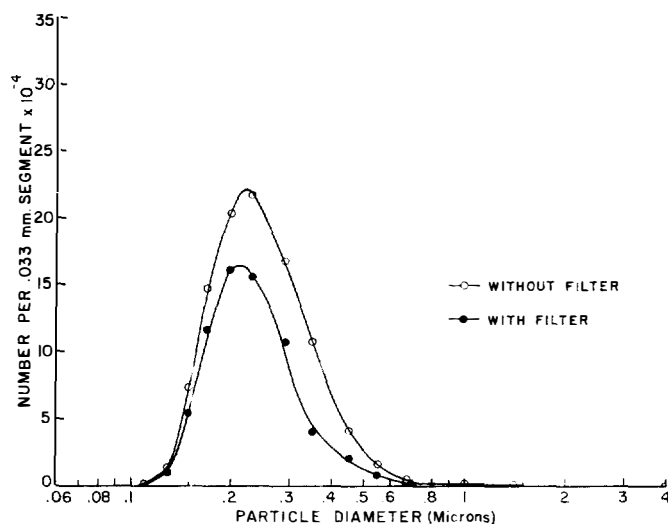


Figure 9. Particle size distribution of the smoke from filtered and unfiltered cigarettes.

sample of diluted tobacco smoke, this smoke is still relatively concentrated in comparison to other aerosols and therefore would be expected to coagulate at an appreciable rate. In order to study this process, the particle size distribution of cigarette smoke was determined at varying times after the puff. These distributions are presented in Figure 5, and as expected for coagulation of small particles into larger aggregates there is a progressive decrease in the numbers of particles and an increase in mean particle size with aging. In Figure 6 it is apparent that the particulate volume or reciprocal of the particulate concentration is a linear function of aging time, as has been observed for many coagulating aerosols (11). From this plot, the initial number of particles in the sample,  $N_0$ , was estimated to be  $1.8 \times 10^7$  particles per cubic centimeter of the diluted smoke sample, or 5.3 billion particles per cubic centimeter in the original smoke. The slope of the curve, or coagulation constant,  $K$ , was found to be  $2.4 \times 10^{-10}$  c.c./sec. Further measurements of these quantities yielded an average coagulation constant of  $3.4 \times 10^{-10}$  c.c./sec. and 3 billion particles per cubic centimeter for the fourth puff on a regular length, blended cigarette of average weight and moisture content. This coagulation constant is somewhat less than the values obtained by others for a variety of considerably more dilute aerosols. Using Whytlaw-Gray's (12) modification of the Smoluchowski coagulation equation, a slightly curved relation between particulate volume and time was calculated with slopes ranging from  $4.2$  to  $4.5 \times 10^{-10}$  c.c./sec.,

which are in reasonable agreement with the experimentally determined slope of  $3.4 \times 10^{-10}$  c.c./sec., especially since the theoretical slope is for an ideal monodispersed aerosol, whereas the experimental value was obtained with a concentrated heterogeneous smoke.

*Effect of Various Smoking and Cigarette Variables.*—The data reported in earlier sections were obtained from fourth, 44-c.c. puff on an unfiltered blended cigarette of normal weight and moisture content. Some data obtained from cigarettes in which one or more of these variables were altered are presented in Table 1.

In Table 1 it is apparent that the particle size distribution is a relatively stable property of cigarette smoke over a considerable range of variations of cigarette and smoking conditions. It is also apparent in Table 1 and the previous distributions that cigarette smoke is a relatively homogeneous aerosol with regard to particle size. This stable and narrow distribution of sizes suggests that the distribution is formed by a combination of removal processes common to all cigarettes. At the small particle end of the spectrum, it is probable that processes such as coagulation, diffusional capture by the tobacco strands, and growth by condensation of vaporized materials would tend to eliminate particles less than 0.1 micron diameter. With regard to the coagulation process, in Figure 5 it was shown that the peak diameter of diluted smoke would double within approximately 180 sec. In raw smoke, with its three hundred fold greater particulate concentration, this should

occur in 0.6 sec., which is of the same order of magnitude as the 0.1 sec. residence time of the smoke within the cigarette. Thus it is apparent that this process would significantly contribute to the removal of small particles.

Another process which probably controls the particle size distribution of the exit smoke stream is removal of the larger particles through filtration by the tobacco strands in the cigarette butt. It is known from other work (14) that appreciable amounts of smoke are collected by this portion of the cigarette. In a later section this process will be considered in more detail.

Although the particle size distributions in Table 1 are relatively stable, there is a considerable variation in the total number of particles per cubic centimeter of smoke gasses for these cigarettes. In general these variations are in line with the effect of these parameters on the weight of smoke (14). Increments in puff volume have a nearly linear effect on the weight of smoke, which is consistent with a constant particulate density, the increment coming from the increased total volume. Later puffs on a cigarette have been found to produce more smoke, chiefly because less is filtered out by the stub, and such an increment is detected in Table 1. With the smoking method employed herein the cigarette tends to become extremely moist after the seventh or eighth puff. This is thought to cause the observed decrease in numbers of particles between the 7th and 10th puff. The variables of cigarette weight and moisture content show the expected increase in numbers of particles with

decreasing weight and moisture content. Bright, burley, the Turkish tobaccos appear to give slightly anomalous results as they produce nearly the same number of particles, although these tobaccos give somewhat different weights of smoke.

*Side Stream Measurements.*—The smoke rising from the burning cone of a cigarette during the interval between puffs should have a different size distribution than the smoke drawn from the cigarette during a puff since it is not subjected to the same size-determining processes as the latter. **Figure 7** shows the distribution obtained for this smoke, which was collected by the previously described method. Larger numbers of small particles, down to 0.08 micron, were observed and the most frequently occurring size was 0.15 micron as compared to 0.20-0.23 micron for main stream smoke. It was estimated from this curve that free burning produces particles at the rate of 6.3 billion per second. Since the sample was obtained over a period of 60 sec., it is likely that the original smoke contains greater numbers of still smaller particles.

*Filtration of Tobacco Smoke.*—To investigate the role of coagulation and filtration in the removal of larger tobacco smoke particles, the effect of drawing the smoke stream through additional cigarettes, open tubes, and efficient cigarette filters was studied. To isolate the effects of coagulation and filtration two assemblies were prepared by taping together three 70-mm. cigarettes and by attaching a 140-mm. long, 8-mm. glass tube to a single cigarette. The assemblies are thus equivalent cigarettes with an open 140-mm. mouthpiece and with a 140-mm. tobacco filter. The distributions obtained from the fourth puff on these cigarettes are illustrated in **Figure 8**.

In **Figure 8**, it is readily apparent that the tobacco filter removed considerably greater numbers of particles than the equivalent mouthpiece and also that this removal was selective for larger particles since the distributions are slightly displaced. The raw smoke coming from the mouthpiece was found to contain 2.1 billion particles per cubic centimeter, whereas the tobacco filter delivered only 0.5 billion. From previous data it was estimated that 3 billion particles entered both assemblies, so that 0.9 billion particles were removed by the mouthpiece and 2.5 billion by the filter. From the residence time of the smoke in the mouthpiece and the coagulation curve, it was calculated that 0.5 billion particles were removed by coagulation, and that 0.4

billion remained in the tube at the end of the puff or were lost at the entrance or walls of the tube.

For the tobacco filter equivalent losses would be expected from coagulation and smoke remaining in the filter, so that the net removal of particles through filtration is 1.6 billion per cubic centimeter, amounting to a better than 50% removal by this mechanism.

From the distributions of **Figure 8**, a figure for the weight filtration efficiency of the 140-mm. tobacco filter may be obtained and compared with an independent estimate of this quantity. Converting the number distributions of **Figures 4 and 8** to mass distributions and integrating in the region 0.1-1.0 micron gave a mass of raw smoke coming from the cigarette of 32  $\mu\text{g}$ . per cubic centimeter, while that issuing from the tube and tobacco filter was 30 to 5  $\mu\text{g}$ . per cubic centimeter, respectively. These yield an overall filtration efficiency for the tobacco filter of 84%. From measurements on shorter tobacco filters, an efficiency of 82% is calculated for a 140-mm. tobacco column; this agrees well with the efficiency calculated from the particle size distributions.

**Figure 9** illustrates the effect of a relatively efficient 17-mm. cellulose acetate cigarette filter on the particle size distribution of cigarette smoke. The distributions illustrated were the average for diluted smoke aged for 60 sec. from 4 cigarettes with, and 4 cigarettes without, filters. As in the case of the tobacco filter, a preferential reduction in the numbers of large particles is apparent in the shift of the filter distribution towards smaller sizes in addition to an overall lowering of the curve.

The total number of particles was found to be 2.3 and 3.6 billion per cubic centimeter of raw smoke for the filtered and unfiltered smoke, respectively. From these figures it is calculated that the filter removes 36% of the numbers of particles from the smoke stream. Converting the curves to mass distributions for particles less than 1.0 micron in diameter and again computing the filtration efficiency yields a figure of 39%, the increase stemming from the greater contribution of the larger particles to the total mass. Considering the fact that separate samplings of cigarettes were necessary for each determination, these figures agree well with the measured filtration efficiency of 37%.

These experiments with tobacco filters and relatively efficient cigarette filters indicate that the filtration mechanism is effective in re-

moving larger particles. This in conjunction with the coagulation and other growth processes produce an essentially stable and narrow distribution of particle sizes in cigarette smoke as it comes from the cigarette.

## Summary

The particle size distribution and particulate concentration of cigarette smoke has been measured using a centrifugal collection instrument called the "Conifuge." The instrument avoids the difficulty of direct size measurement by continuously grading the particles according to settling velocity or size. It is capable of collecting particles ranging in diameter between 0.05 and 10 microns. For these reasons the instrument is well suited for measurement of the size distribution of cigarette smoke and other aerosols.

The size distribution and particulate concentration of smoke were found to be reproducible quantities for similar cigarettes, but were found to vary according to the age of the smoke sample, even after considerable dilution. The rate of decrease of the number concentration of smoke particles was found to agree reasonably well with the rate predicted by the modified Smoluchowski coagulation equation.

Alteration of a number of smoking and cigarette variables was found to have no detectable effect on the particle size distribution, but changed the concentration of particles in a manner similar to the effect of these variables on the weight of smoke.

A preferential removal of larger particles was observed for high-efficiency tobacco and cigarette filters, in addition to a considerable decrease in particulate concentration.

The essentially constant size distribution appears to arise through the removal of small particles through coagulation and larger particles through filtration by the tobacco strands.

## Acknowledgment

The authors are indebted to Mr. Henry Pierce for his considerable assistance in the design and construction of the conifuge and accessory equipment.

## Literature Cited

1. Sano K., Fujiya, Y., and Sakata, S., *J. Chem. Soc. Japan* **74**, 666 (1954).
2. Langer, G., and Fisher, M. A., *Am. Med. Assoc. Arch. Ind. Health* **13**, 373 (1956).
3. Holmes, J. C., Hardcastle, J. E.

- and Mitchell, R. I., *Tobacco Sci.* **3**, 148 (1959).
4. Sam, A., "A Study of Particle Size Distribution and Dilution of Cigarette Smoke." Thesis, Duke University, Durham, North Carolina, 1956.
  5. Kahler, H., and Lloyd, B. J., Jr., *J. Natl. Cancer Inst.* **18**, 217 (1957).
  6. Sawyer, K. F., and Walton, W. H., *J. Sci. Instr.* **27**, 272 (1950).
  7. Keith, C. H., and Newsome, J. R., *Tobacco Sci.* **1**, 51 (1957).
  8. Sinclair, D., and La Mer, V. K., *Chem. Revs.* **44**, 245 (1949).
  9. O'Konski, C. T., and Doyle, G. J., *Anal. Chem.* **27**, 694 (1955).
  10. Bradford, E. B., and Vanderhoff, J. W., *J. Appl. Phys.* **26**, 863 (1955).
  11. Whytlaw-Gray, R., and Patterson, H. S., "Smoke: A Study of Aerial Disperse Systems," Chapter 5. E. Arnold & Co., London, 1932.
  12. Whytlaw-Gray, R., and Patterson, H. S., "Smoke: A Study of Aerial Disperse Systems," Chapter 6. E. Arnold & Co., London, 1932.
  13. Drinker, P., and Hatch, T., "Industrial Dust," pp. 144-149. McGraw-Hill, New York, 1936.
  14. Newsome, J. R., and Keith, C. H., *Tobacco Sci.* **1**, 58 (1957).

---