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WORKSHOP ON

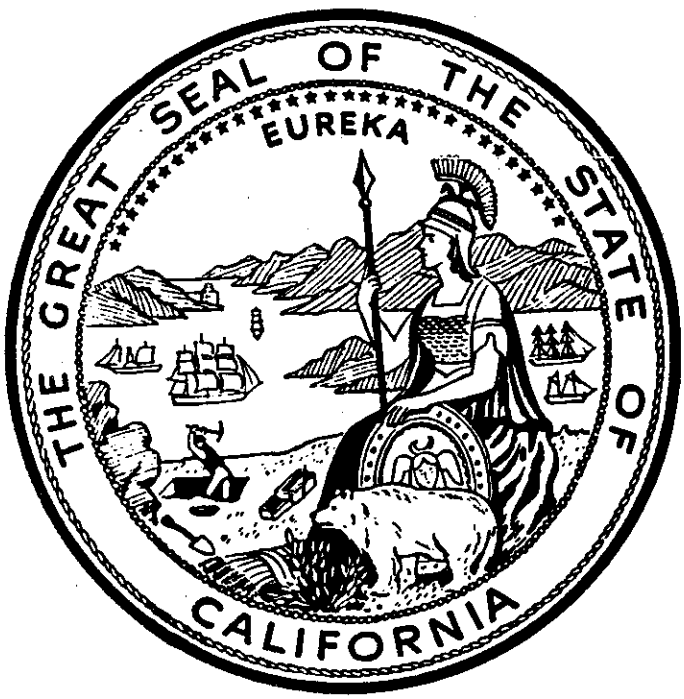
A CALIFORNIA AMBIENT AIR QUALITY STANDARD FOR INHALABLE PARTICLES

AIR RESOURCES BOARD

Final Report, Interagency Agreement (A8-094-31)

Prepared by:
Jerome J. Wesolowski

July 1979



Air and Industrial Hygiene Laboratory
Laboratory Services Branch
California Department of Health Services
2151 Berkeley Way, Berkeley, California 94704

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1979

A workshop on "A California Ambient Air Quality Standard for Inhalable Particles" was held May 10 and 11, 1979 at the Marriott Inn, Berkeley, California.

The purpose of the workshop was to bring together a small group of experts on health effects and monitoring of inhalable particles to draft a report concerning a possible California inhalable particle air quality standard to be sent to the Department of Health Services Air Quality Advisory Committee and the California Air Resources Board for their consideration.

There were 43 attendees at the workshop. They consisted of three groups:

1. A Working Committee on an Inhaled Particle Standard. The committee members were chosen by mutual agreement of the Department of Health Services and the Air Resources Board. The role of the working committee, and thus the output of the workshop, is the attached written report summarizing their conclusions concerning a possible California air quality standard for inhalable particles.

2. A group of experts on medical and monitoring aspects of inhaled particles chosen by mutual consent of the Department of Health Services and the Air Resources Board. The experts' role was to bring forward pertinent information and to answer specific questions posed by committee members.

3. Invited Auditors and Discussants

The attached report has been sent with appropriate cover letters to all attendees, members of the Department of Health Services' Air Quality Advisory Committee, members of the Air Resources Board, members of the Air Resources Board's Research Screening Committee, and copies are available to anyone interested in the workshop proceedings. The report of the working committee includes background information, recommendations, and various appendices, including the workshop agenda and a list of attendees.

Workshop On
A California Ambient Air Quality Standard for
Inhalable Particles
Berkeley, California
May 10-11, 1979

Report of the Working Committee

Background

Current Federal and California ambient air quality standards for particulates are stated in terms of mass of total suspended particulates (TSP) as collected by "Hi-Vol" samplers. A variable but substantial fraction of TSP, however, consists of particles of larger size than considered "inhalable" by humans. A major justification for an airborne particulate standard is to protect human health, particularly from damage to the respiratory system.

Accordingly, there has been a long felt need for an inhalable particle air quality standard to replace TSP. The development of such a standard has been impeded in the past by: 1) lack of agreement on size cut(s), 2) lack of availability of appropriate sampling equipment, and 3) inadequacy of data on which to base an air quality standard.

Considerable progress has been made recently on at least the first two impediments. Accordingly, the California Air Resources Board and Department of Health Services convened a workshop, involving technical experts on monitoring and health effects of inhalable particles, to examine the feasibility and desirability of an inhalable particle standard. Funding for the workshop was provided by the Air Resources Board; it was organized and conducted by the Air and Industrial Hygiene Laboratory of the Department of Health Services.

The Workshop

Participants in the Workshop (see Appendix A) included an 8 member Working Committee, a 13 member Advisory Panel, and 22 invited auditors and discussants.

The Workshop agenda is included as Appendix B. The first day was devoted to formal presentations, with discussion after each paper, to expose all participants to the most up-to-date knowledge regarding fine particle sampling and health effects. A selection of these papers are included, for background purposes, as Appendices C, D, and E. The second morning was devoted to a series of panel discussions, during which the implications of the information presented the day before were explored. That afternoon the Working Committee convened to deliberate on its recommendations.

Conclusions and Recommendations of the Working Committee

The Working Committee wishes to make clear that this report does not represent a critical review of specific studies; it is not intended as a Criteria document. Members of the committee, however, were thoroughly familiar with the relevant background material such as the recent reviews:
1) B. G. Ferris, "Health Effects of Exposure to Low Levels of Regulated Air Pollutants", J. Air Poll. Control Assoc. 28:482 (1978); and 2) National Research Council, "Airborne Particles", University Park Press, Baltimore. 1979. 343p. Moreover, the present state of knowledge was presented and extensively discussed for a day and a half prior to committee deliberations.

The Working Committee conclusions and recommendations will be presented in three parts:

1. Particle Sampling and Size Cuts

In order to establish an inhalable particle air quality standard it is first necessary to have available appropriate, reliable, size-selective sampling equipment and procedures. The conclusion contained in Appendix C is that such equipment and procedures are indeed available and adequate to support an inhalable particle standard. Either the virtual dichotomous or cyclone type sampler will suffice.

The Environmental Protection Agency has examined intensively the issue of appropriate size cut(s) for an inhalable particle standard. The results are summarized in Appendix D. The issues were explored during several sessions of the workshop and considered by the Working Committee. It was the unanimous opinion of the committee to recommend size cuts of 15 and 2.5 μm aerodynamic equivalent diameter. This coincides with the EPA proposals. Although favoring the notion that there be nationwide uniformity, the committee concluded that there is compelling justification for these size cuts even without considering such uniformity.

In a variety of urban atmospheres, there has been shown to be a bimodal distribution of mass or volume vs particle size, with a trough centering around 2 μm. There is a fundamental difference between the two fractions thus divided. They originate separately, are transformed separately, are removed from the atmosphere by different mechanisms, have different chemical composition, have different optical properties, and differ significantly in their deposition patterns in the respiratory tract. Moreover, the fine particles are responsible for almost all visibility reduction (see Appendix C, Figure 1).

It is recommended that inhalable particles be defined as those less than 15 μm aerodynamic equivalent diameter, based on considerations of particle deposition in the respiratory tract. No more than about 5% of particles larger than 15 μm reach the trachea during mouth breathing.

The 2.5 μm diameter size cut is based on considerations of penetration of particles to the gas exchange region of the lung, on the differences in chemical composition from larger particles, and on differences in origin from larger particles - important from the standpoint of controlling emissions.

There have been proposals for making the small size cut-off variously between 1 and 5 μm. Actually, only a small fraction of the total ambient aerosol mass

occurs in this size range. The choice of 2.5 μm appears to be the most reasonable compromise, inasmuch as the minimum mass concentration based on aerodynamic particle size tends to center around 2.5 μm.

There was discussion about the desirability of consistency with the 3.5 μm cut-off for "respirable dust" as used in industrial hygiene. The Working Committee concluded that the two are used in different ways for different purposes, and cannot be considered as analogous.

Two other comments need to be made about sampling. The two size cuts just discussed are intended to relate to routine, area monitoring. It is recognized that for various research projects, other size cuts - particularly more fractionation - might be appropriate. Second, it is important for historical comparison and other reasons to continue "Hi-Vol" sampling for some time into the future.

2. An Inhalable Air Particle Standard?

The state of current knowledge concerning the health effects of airborne particles is summarized in Appendix E. Based on this, the discussions at the Workshop, and on the intimate knowledge of the relevant literature on the part of the members, the Working Committee concluded that there is insufficient information at hand on which to base a recommendation for an inhalable particle air quality standard at this time.

As pointed out in the Preface to the NRC report, Airborne Particles:

> "Particles come in all shapes and sizes. They may be of animal, vegetable, or mineral origin. They may be living or inanimate. They can affect plants, animals, or materials. Particles may affect a persons health in a variety of ways: some are inert, producing changes

in the body only by their passive accumulation and inducing little or no tissue reaction; others are intensely irritant or toxic, causing changes that may result in serious illness - even death when inhaled in sufficient quantity. Some particles are known to produce cancer. Evidence indicates that the action of particles may be modified by the presence of other particles. Particles may also interact with gases that may be present in the air. These interactions may either enhance or moderate the effect of either substance when inhaled alone".

The foregoing provides a graphic illustration of the complexity of approaching a single inhalable particle standard. Moreover, there are relatively few size-segregated sampling data available relating ambient aerosol exposures to health effects.

EPA is under Congressional mandate to undertake a thorough review of existing air quality criteria documents and associated standards by the end of 1980. EPA scientists who participated in the Workshop indicated that it is the Agency's intent to issue a criteria document relating to an inhalable particle standard ready within about a year, with the first draft expected to be completed in May or June 1979.

Some members of the Working Committee expressed the view that California has led the way in setting air quality standards in the past and might wish to proceed with an inhalable particle standard. Others concluded that it would be a wasteful duplication of effort for California to parallel the work that EPA is doing in preparing a criteria document. The Committee concensus was to recommend that California defer action in setting an inhalable particle air quality standard, pending review and evaluation of the forthcoming EPA Draft Criteria document.

The committee further recommends that there be instituted, as soon as practicable, a program of particulate monitoring in representative locations around the State, size segregated at 2.5 and 15 μm. This should be supplemental to and not replace "Hi-Vol" monitoring. To the extent possible, samplers should be designed to collect specimens suitable for chemical and physical analyses. Consideration should also be given to the use of preliminary treatment of the sampled air to remove reactive gases, which can change the collected particles.

The data thus obtained will be extremely useful as background information in the ultimate determination of an air quality standard, and will insure that the standard is appropriate for California conditions.

3. Research Needs

As has been the case in the initial stages of almost all past efforts to develop new environmental health standards, the issues are complex and the data ambiguous. The issue of fine particles is no exception. As one Workshop participant phrased it, we are being asked to propose a mass standard for inhalable particulates in the face of great uncertainty. Research will lessen that uncertainty. Research needs are of two kinds.

First, there is need for better information about the health effects of air pollutants generally, not just particles. Examples of this type of research include:

A. Much better information is needed about actual exposures of people to air pollutants. There is widespread agreement that existing air monitoring networks are inadequate to characterize human exposures. The degree of inadequacy is uncertain; it needs urgently to be determined, and overcome, to the extent feasible.

B. Animal studies play an important role in elucidating mechanisms of air pollution damage. The extent to which the results of such studies can be extrapolated to the human condition is always uncertain. A great deal of work has been done

in this area, but more is needed to attempt better definition of the utility of animal exposure data.

C. Typically, the first instance of possible harm to a human from air pollutants is sought by measuring functional changes in the respiratory system. A variety of lung function and related tests are used for this purpose. Currently, these tests lack sufficient sensitivity to measure subtle or gradual changes in function. One member of the Working Committee, a pathologist, pointed out that substantial sections of a lung can be virtually destroyed and go undetected by lung function tests. More sensitive damage indicators are badly needed, perhaps structural and functional tests at the cellular level.

D. Epidemiological studies represent the only feasible means for assessing the health effects of air pollution in association with all of the other variables of the real world. Epidemiological studies, however, are expensive, time consuming, and variables are difficult to sort out. But such studies, particularly longitudinal studies, are clearly needed. They should be carefully designed and managed, and given the long term committment necessary for effectiveness. Future epidemiological studies involving particulates as a variable should have associated size segregated monitoring with the size-cuts recommended earlier.

Second, there is need for research specifically relating to inhalable particles. Questions posed at the workshop included: How serious and of what nature are the health effects of airborne particles? How are these effects related to particle mass, size, surface area, chemical composition? Are the effects independent of other pollutants? To what extent are the effects influenced by age, pre-existing disease, nutritional status, mental state, or climatological factors? None of these questions can be fully answered on the basis of existing information. Among research needs are:

1. More development of fine particle sampling, from design of individual samplers to schemata for a complete monitoring system. Important considerations

are filters, filter artifacts, and the effects of gaseous pollutants continually "bathing" particles on a filter.

2. Determinations of particle size distribution, with adequate size resolution and chemical analysis, taken under a variety of conditions to verify the details of the bimodal distribution. More short term sampling is also needed to evaluate the intensity of exposures.

3. Animal toxicological, human clinical, and epidemiological studies relating to health effects of inhaled particles. EPA is supporting, or planning to do so, a number of such studies. An important and desirable feature is that EPA is stressing uniformity of technical approach to these studies to maximize comparability. It is recommended that related studies, supported from other sources, attempt to coordinate with the EPA studies and, to the extent feasible, utilize the same technical approaches.

LIST OF PARTICIPANTS

Working Committee

John Heslep (Chairman)	California Department of Health Services
Rodney Beard	Stanford University Medical Center
Timothy Crocker	University of California, Irvine
Robert Frank	University of Washington
James Pitts	University of California, Riverside
Ian Higgins	University of Michigan
Stanley Rokaw	Air Quality Advisory Committee
Russ Sherwin	University of Southern California

Advisory Panel

Jerome Wesolowski	California Department of Health Services
Karl Bell	University of California, Irvine
Robert Phalen	University of California, Irvine
Judy Graham	Environmental Protection Agency
Lester Grant	Environmental Protection Agency
Fred Miller	Environmental Protection Agency
William Belser	University of California, Riverside
Thomas Cahill	University of California, Davis
Walter John	California Department of Health Services
Stanley Dawson	Harvard University
Bruce Appel	California Department of Health Services
Walter Tyler	University of California, Davis
Peter Flessel	California Department of Health Services

Invited Auditors and Discussants

Olav Bjorseth	California Department of Health Services
Jan Bush	Ventura County Air Pollution Control District
Richard Carrigan	National Science Foundation
Daniel P. Y. Chang	University of California, Davis
William Clark	California Department of Health Services
Rodney Cummins	Environmental Protection Agency
Warren Galke	Environmental Protection Agency
Simon Goren	University of California, Berkeley
John Holmes	California Air Resources Board
Paul Husman	California Air Resources Board
Edward Jeung	California Department of Health Services
John Kinosian	California Air Resources Board
Kenneth Kip	Chevron Research
Kiyoshi Nishikawa	California Air Resources Board
Lawrence Perry	California Department of Health Services
Brian Smith	Energy Resources Conservation & Development Commission
Susan Solarz	California Air Resources Board
Albert Starr	California Department of Health Services
Dane Westerdahl	California Air Resources Board
Jack Suder	California Air Resources Board
Peter Venturini	California Air Resources Board
Steve Wall	California Department of Health Services

LIST OF WORKING COMMITTEE AND ADVISORY PANEL MEMBERS

WORKING COMMITTEE

John Heslep (Chairman)	Cal. Dept. of Health Services
Rodney Beard	Stanford U. Medical Center
Timothy Crocker	UC Irvine
Robert Frank	University of Washington
James Pitts	UC Riverside
Ian Higgins	University of Michigan
Stanley Rokaw	Air Quality Advisory Comm.
Russ Sherwin	USC

ADVISORY PANEL

Jerome Wesolowski	Cal. Dept. of Health Services
Karl Bell	UC Irvine
Robert Phalen	UC Irvine
Judy Graham	EPA
Lester Grant	EPA
Fred Miller	EPA
William Belser	UC Riverside
Thomas Cahill	UC Davis
Walter John	Cal. Dept. of Health Services
Stanley Dawson	Harvard University
Bruce Appel	Cal. Dept. of Health Services
Walter Tyler	UC Davis
Peter Flessel	Cal. Dept. of Health Services

Telephone Numbers
(Area Code 415)

Marriott Inn 548-7920

Oakland Airport 444-4444

San Francisco Airport ..... Call individual airlines

Air and Industrial Hygiene Laboratory ..... 843-7900, ext. 595

Messages may be left with the Marriott Inn Switchboard.

Workshop on

A California Ambient Air Quality Standard for Inhalable Particles

May 10-11, 1979

Marriott Inn Berkeley, CA

Chairman

J. J. Wesolowski

Co-ordinators

E. Jeung
C. Noriye
A. Starr

Sponsored by

California Air Resources Board
Air and Industrial Hygiene Laboratory
California Department of Health Services

SIZE-SELECTIVE SAMPLING TECHNIQUES FOR AIRBORNE PARTICULATE MATTER*

Walter John
Air and Industrial Hygiene Laboratory Section
California Department of Health Services
Berkeley, California 94704

Introduction

My purpose is to address the following question: Are size-selective sampling techniques available and are they adequate to support an inhalable particle standard? Hopefully, my answer will break the circular dialogue between the health effects specialist and the aerosol specialist that Dr. Wesolowski alluded to earlier. It will not be my intent to promote any given sampler but rather to review the current state of the art.

Particle Size Selection Criteria

In discussing the desirable characteristics of a sampler we must first determine what it will be used for, i.e., what is the purpose of the sampling? The primary reason for monitoring of ambient air is to protect the health of the public. Secondary reasons are to preserve visibility and to avoid material damage and contamination from particle fallout. Now the effects of particles on health, visibility and materials all depend on particle size. That is why we are here.

Figure 1 illustrates several points. On the bottom is a graph of a typical ambient air particle size distribution, volume vs. diameter. The nuclei and accumulation modes derive mainly from combustion processes and the coarse particle mode from mechanical processes.[1] There is a minimum at about 2 μm. In the center of Fig. 1, light scattering per unit mass, i.e., visibility reduction is plotted, showing that only the fine particles contribute appreciably.[2]

At the top of Fig. 1 are plotted the lung deposition curves of the Task Force on Lung Dynamics.[3] The "respirable" curve of the ACGIH for insoluble dusts is shown with the 50% cutpoint at 3.5 μm.[4] A cut at 3.5 μm would

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*Prepared for presentation at the Workshop on a California Ambient Air Quality Standard for Inhalable Particles, Berkeley, California, May 10-11, 1979.

also serve to selectively sample those particles which reduce visibility and separate the modes of the ambient air. I am not concerned at this point with the precise location of the cut, whether it should be at 2, 2.5, 3, 3.5 or whatever. We also need an upper size limit in order to complete the definition of the coarse fraction.

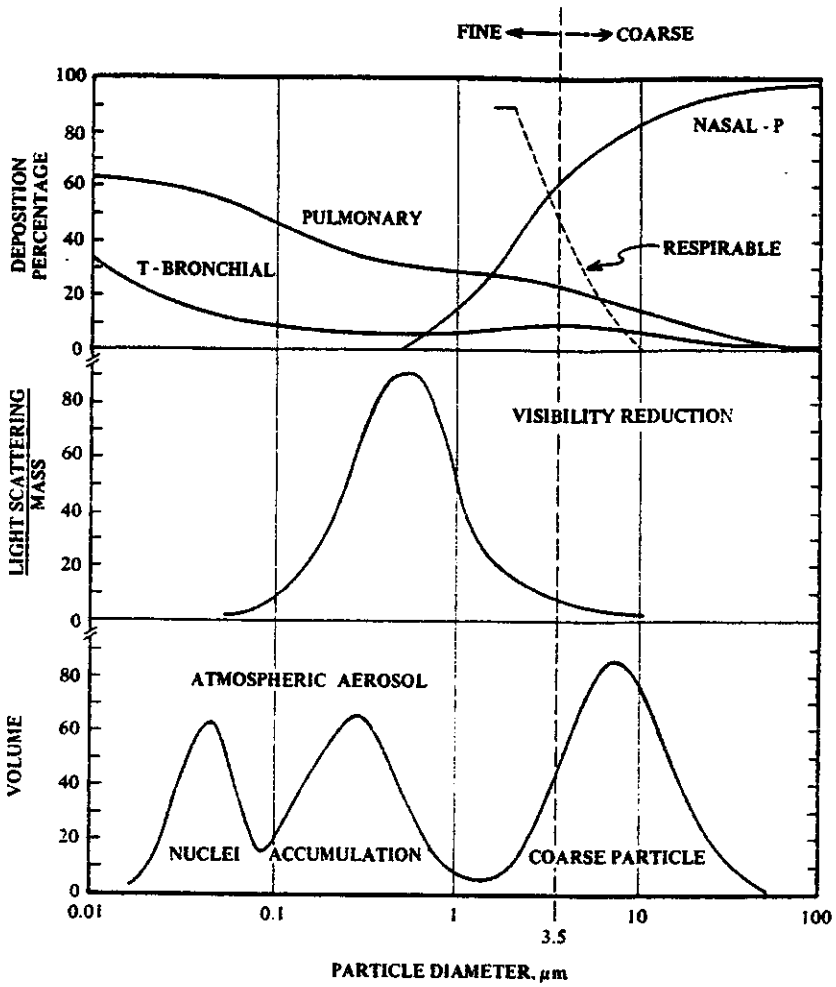


Figure 1. From top to bottom: Lung deposition curves, visibility reduction and ambient aerosol size distribution.

Fig. 2 is a diagram of the human respiratory tract. The EPA is proposing[5] an inhalable particle cut at 15 μm to sample what enters the trachea. The second cut to separate the ciliated and non-ciliated regions is proposed at 2.5 μm. Again, for now we are interested only in the order-of-magnitude of the particle sizes. In Fig. 3 the various mechanisms of particle deposition are shown. For the size range indicated for the inhalable and respirable cuts, mainly inertial forces are involved, namely, sedimentation under gravity and impaction resulting from a particle's inability to follow the changing direction of the gas flow. Since these two mechanisms are responsible for deposition in the lungs, it follows that our sampler should size the particles aerodynamically. This rules out samplers based on optical or electrical properties of the particles. The ambient particles are a complicated mixture of different compositions, shapes, surfaces etc. It is therefore impossible, for example, to convert an optical size to an aerodynamic size.

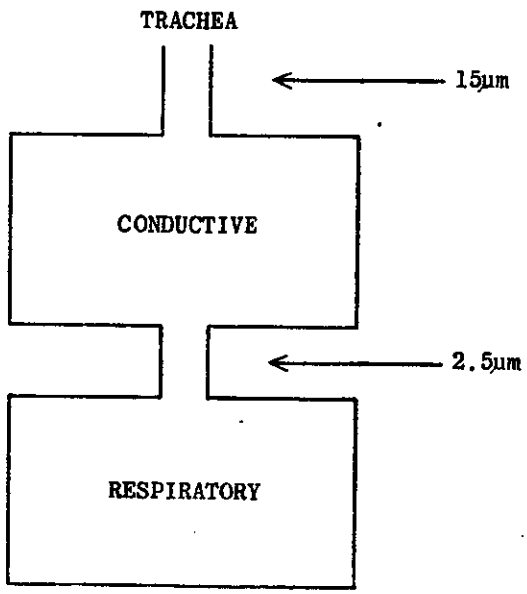


Figure 2. Relation of particle cutpoints to lung regions.

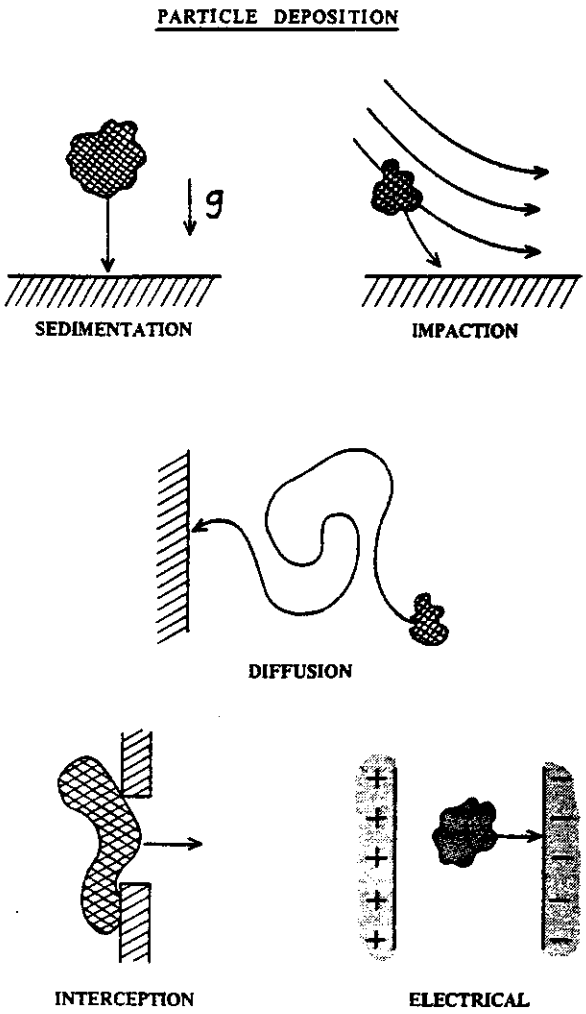


Figure 3. Particle deposition mechanisms.

The Hi-vol

Actually we have been using a size-selective sampler all along. Fig. 4 is the acceptance efficiency of the hi-vol vs. particle size.[6] The 50% cutpoint is in 10 to 15 μm range; however, the tail extends beyond 50 μm. Another difficulty is that the cutoff depends strongly on the wind direction. In Table 1, I summarize the deficiencies of the hi-vol. All of these deficiencies could be eliminated or greatly improved with available technology. I think we should keep this in mind, i.e., a spinoff of going to size-selective sampling can be an improvement over the hi-vol in these respects.

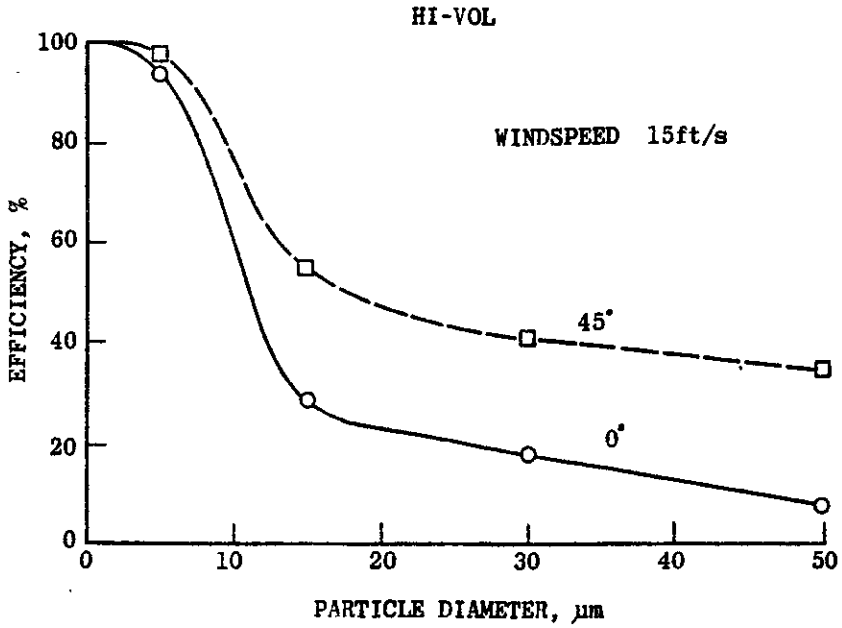


Figure 4. Hi-vol sampling efficiencies measured in a wind tunnel. (Ref. 6)

Table 1. Hi-vol deficiencies

INLET GEOMETRY
EFFICIENCY AFFECTED BY WIND AND FLOWRATE
NON-UNIFORM DEPOSIT
GLASS FIBER FILTER
ARTIFACTUAL SULFATE AND NITRATE
UNSUITABLE FOR X-RAY ANALYSIS

Size-selective Samplers

In Table 2 are summarized some requirements for a size-selective sampler for monitoring (not research). Let's begin by considering the inlet, which in most cases will determine the upper size cutoff. The inlet[7] currently supplied with the virtual dichotomous samplers is shown in Fig. 5. The annular entrance slit assures independence of wind direction. The sizing is a combination of impaction and elutriation, i.e., sedimentation against the upward flow.

Table 2. Requirements for size-selective sampler

- AERODYNAMIC SIZE SEPARATION
- SAMPLES FOR CHEMICAL ANALYSIS
- MASS DETERMINATION
- SMALL NUMBER OF SIZE CUTS
- DEFINITE UPPER SIZE CUTOFF
- REASONABLE SAMPLING TIME
- EFFICIENCY INDEPENDENT OF WIND
- ENGINEERED FOR RELIABILITY

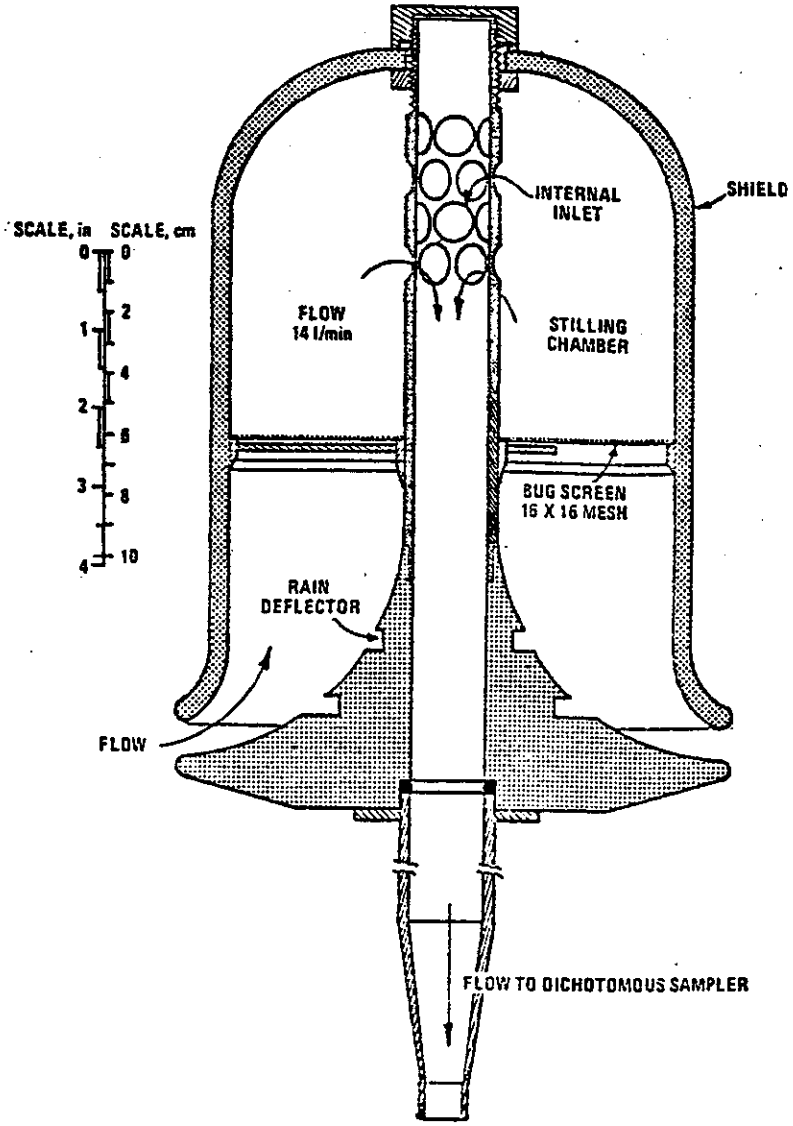


Figure 5. Size-selective inlet for virtual dichotomous sampler. (Ref. 7)

Another type is shown in Figure 6, a design due to A. McFarland.[8] The sizing here is accomplished by a one-stage impaction and the convuluted flow path. This sampler is a "medium vol", 2-4 CFM, for use with a large membrane filter. If a particle standard were adopted with a single inhalable upper size cut, then this kind of sampler would suffice. We could even retrofit the hi-vol, as shown in Figure 7. This is the same type of inlet but with multiple nozzles.[8] This is <u>not</u> my favorite approach since we would still be stuck with glass fiber filters. And anyway, we will need our hi-vols for comparison to any new sampler.

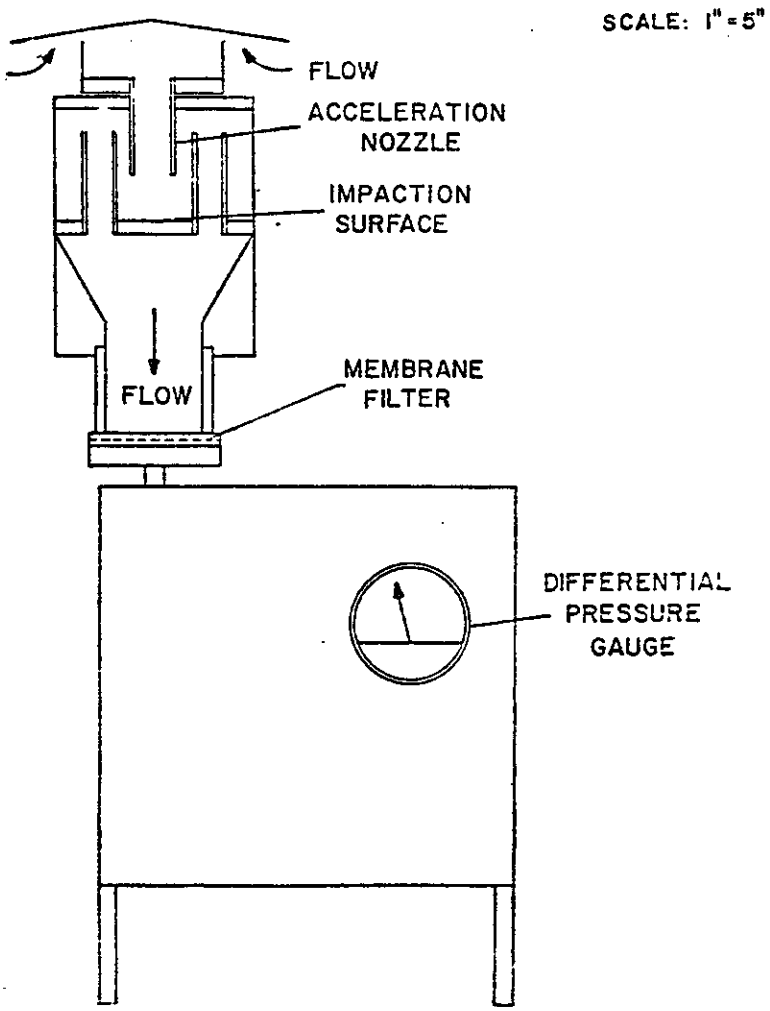


Figure 6. Medium volume sampler with size-selective inlet. (Ref. 8)

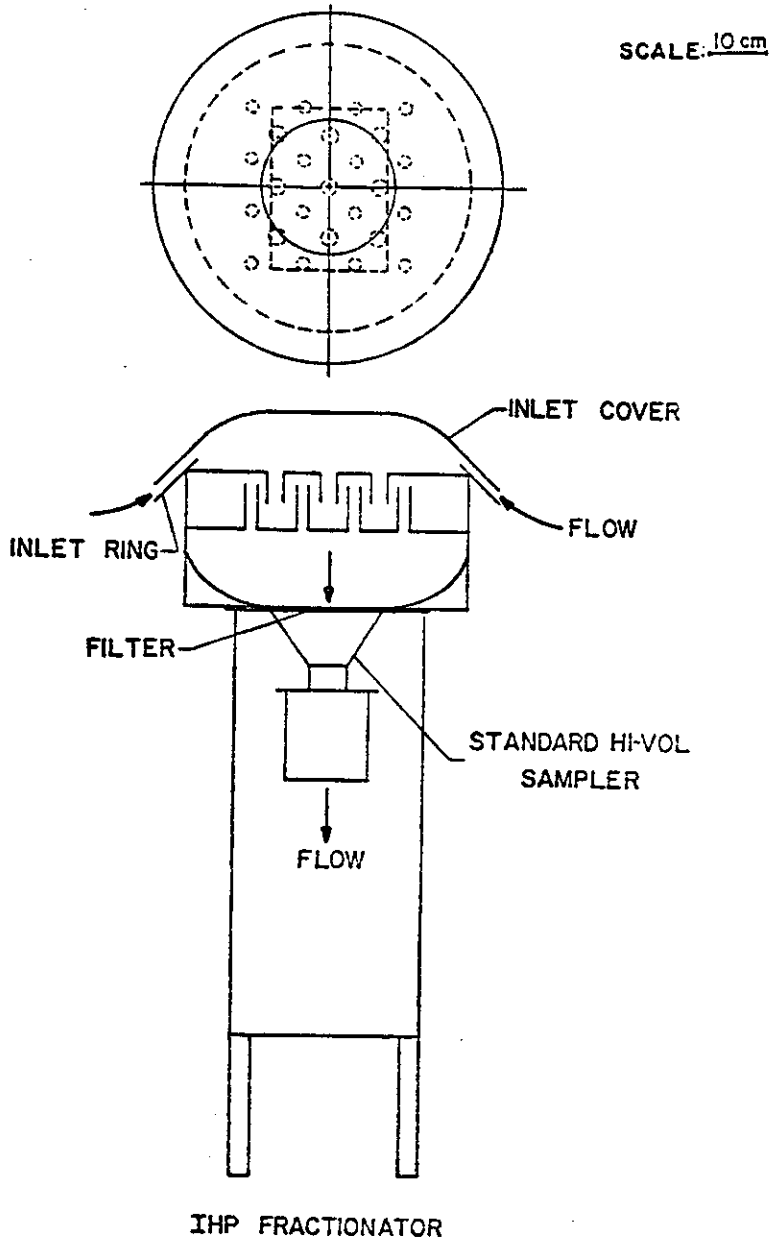


Figure 7. Hi-vol with size-selective inlet. (Ref. 8)

The penetration vs. particle size for these three inlets (Figure 8) have been measured in a wind tunnel.[8] They all have similar curves. The 50% cutpoint is near 15 μm; it falls below 10% by 20 μm and reaches 90% by ∿ 4 μm. Note that the size-selective hi-vol inlet is independent of wind speed. I understand that results for the dichotomous inlet show some effect from wind speed. The inlet problem is the most difficult one that we face. It does not seem possible to sample ideally, i.e., isokinetically in the ambient atmosphere with any reasonable apparatus. However, as you see, currently available inlets are probably acceptable. They are very much better than the "doghouse" roof of the hi-vol. New types of inlets are being designed by at least two people that I know of with the expectation of improved performance.

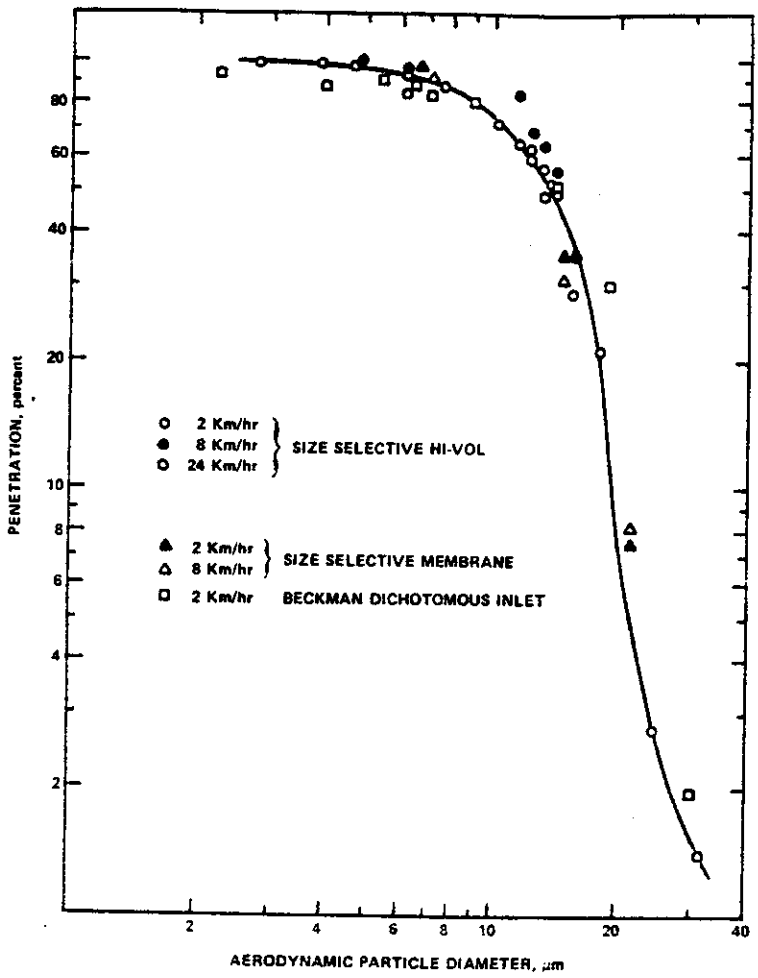


Figure 8. Calibration of inlets with monodisperse particles in a wind tunnel. (Ref. 8)

To effect the second cut at ∿ 3 μm, we consider the devices listed in Table 3. In Figure 9, the principle of the cascade impactor is illustrated. Although the particle separation is aerodynamic, the impactor has the inherent difficulties of particle bounce and reintrainment. Use of sticky substrates or filter substrates introduces other problems.[9-11] The deposit is also nonuniform. For these reasons I do not consider the cascade impactor to be a suitable sampler for the present purpose. The impactor does have the advantage that we can calculate the 50% cutoff analytically and we can even calculate the cutoff curve by numerical techniques.[12]

Table 3. Size-selective samplers

INERTIAL IMPACTOR
CYCLONE
VIRTUAL IMPACTOR
SPECIAL DEVICES

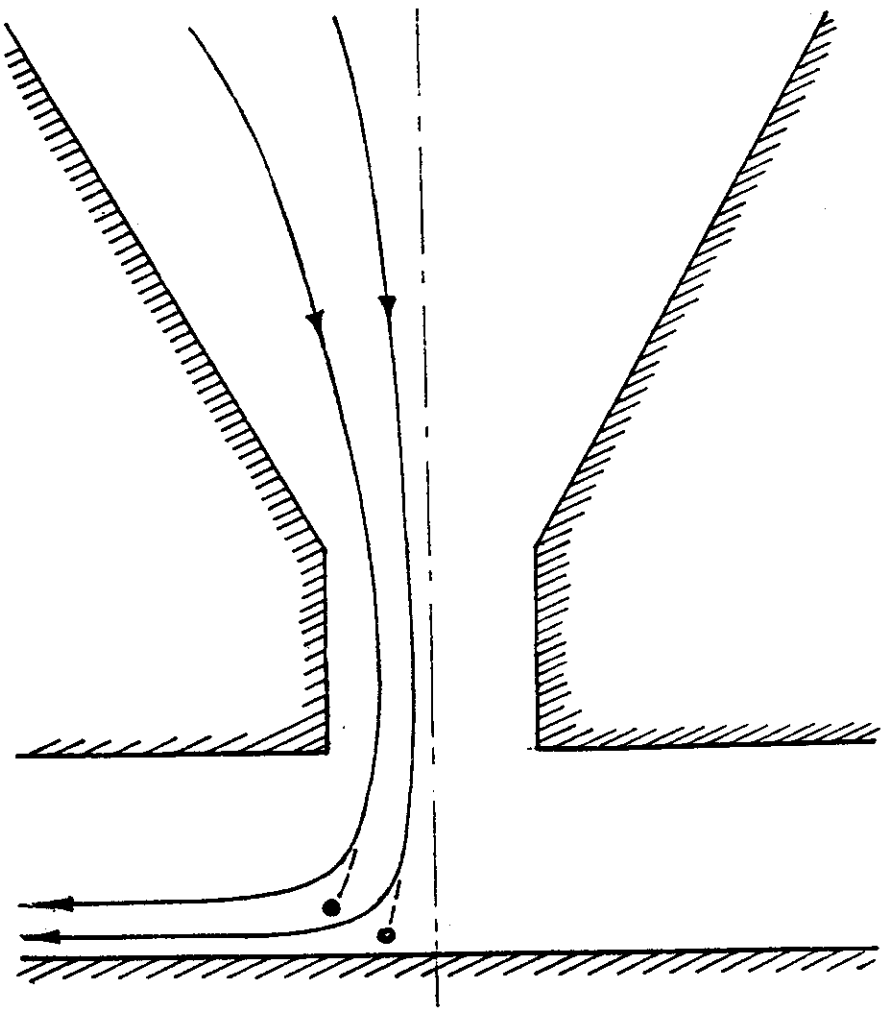


Figure 9. Principle of conventional impactor

The virtual impactor retains these advantages and overcomes the objections. Figure 10 is a schematic drawing. The air is accelerated through a nozzle and then pumped off at 90° through the annular gap between the tubes. The impaction is on the air in the lower tube; this is the origin of the term "virtual". The fine particles will follow the gas flow whereas the coarse particles will enter the lower tube. A small flow, typically 10% of the total, is pumped through the lower tube to sample the coarse particles. In Figure 11 are illustrated the trajectories of two particles, one following the coarse fraction into the lower tube, the other the fine fraction to the side. These trajectories were calculated on a computer by Marple and Chien.[13] The calculations produce the cutoff curve and, in agreement with experiment, show most of the wall loss on the lip of the lower tube. However, the losses cannot be calculated quantitatively. The losses were actually reduced empirically by Bill Loo[14] through proper shaping of the components.

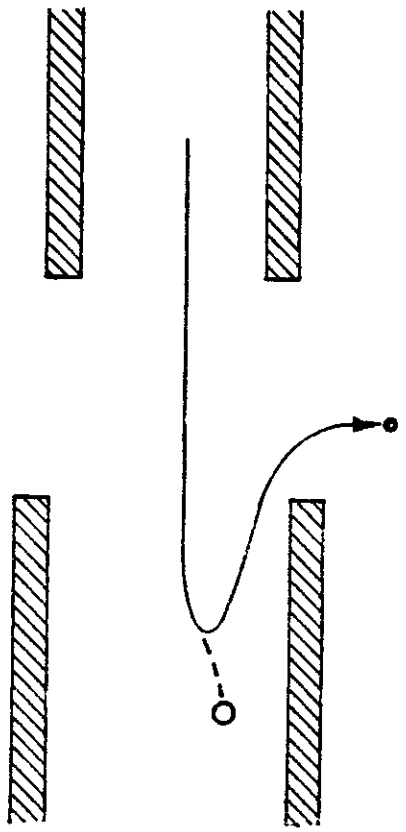


Figure 10. Virtual impactor principle

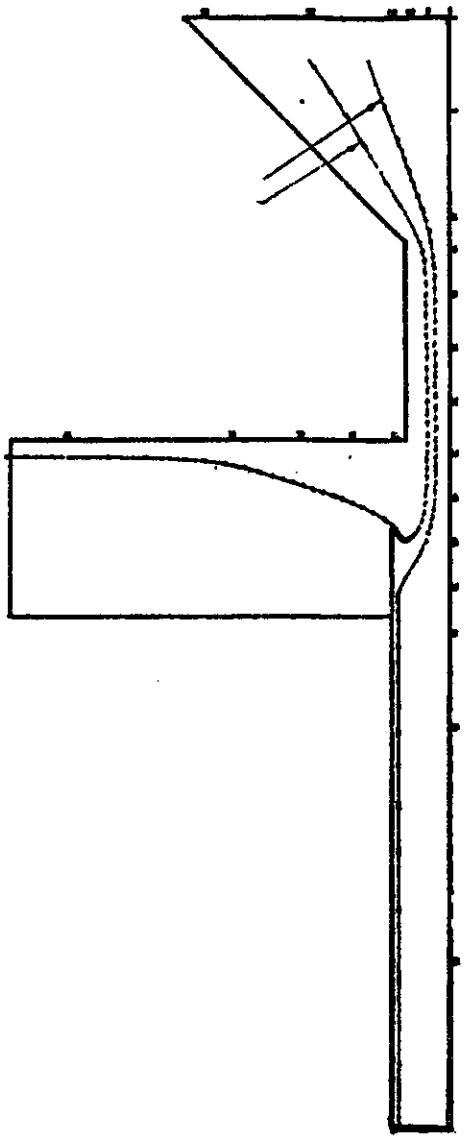


Figure 11. Theoretical particle trajectories in a virtual dichotomous impactor. (Ref. 13)

Figure 12 shows the virtual dichotomous developed by Lawrence Berkeley Laboratory for the EPA,[15] currently available from three manufacturers in a manual version costing about $3000 and an automated version for about $7000. The total flow rate is 1 m^3/hr (16.7 ℓ/min) with 10% going to the coarse particle fraction. This means that the coarse filter contains 10% of the fines and a correction must be made. The two fractions are collected on 37 mm membrane filters. The measured collection efficiency is shown in Figure 13. 50% cutpoint is at 2.5 μm (this can be increased fairly easily with design changes. The flow rate, however, cannot be increased without introducing additional nozzles). The efficiency approaches 10% at small particles as expected and the losses, which peak at the crossover point, are less than about 5%. The previous model of this sampler has been used to collect some 35,000 samples in St. Louis.[16]

Another instrument which samples aerodynamically is the cyclone. An example of the small sampling cyclones is shown in Figure 14. This is actually our own design.[17] The air enters tangentially, forms a vortex, than leaves through the upper tube to an after filter. The cyclone has the advantage of no bounceoff or reintrainment, even under high loadings. The cutpoint can be shifted simply by changing the flow rate. (The virtual impactor operates with low losses only at the design flow rate). The cyclone has the disadvantage that the coarse fraction is deposited in the body, rather than on a filter. Another disadvantage is the lack of an adequate theory of the instrument. This is mainly an inconvenience to the instrument designer. We do have some empirical relations.[18] Figure 15 shows that the cutpoint varies as a power law with flow rate; the exponent is near -1.0. In Figure 16, the cyclone retention vs. normalized particle diameter, $(Dp-D_{50})/D_{50}$ is a universal curve for all flow rates.

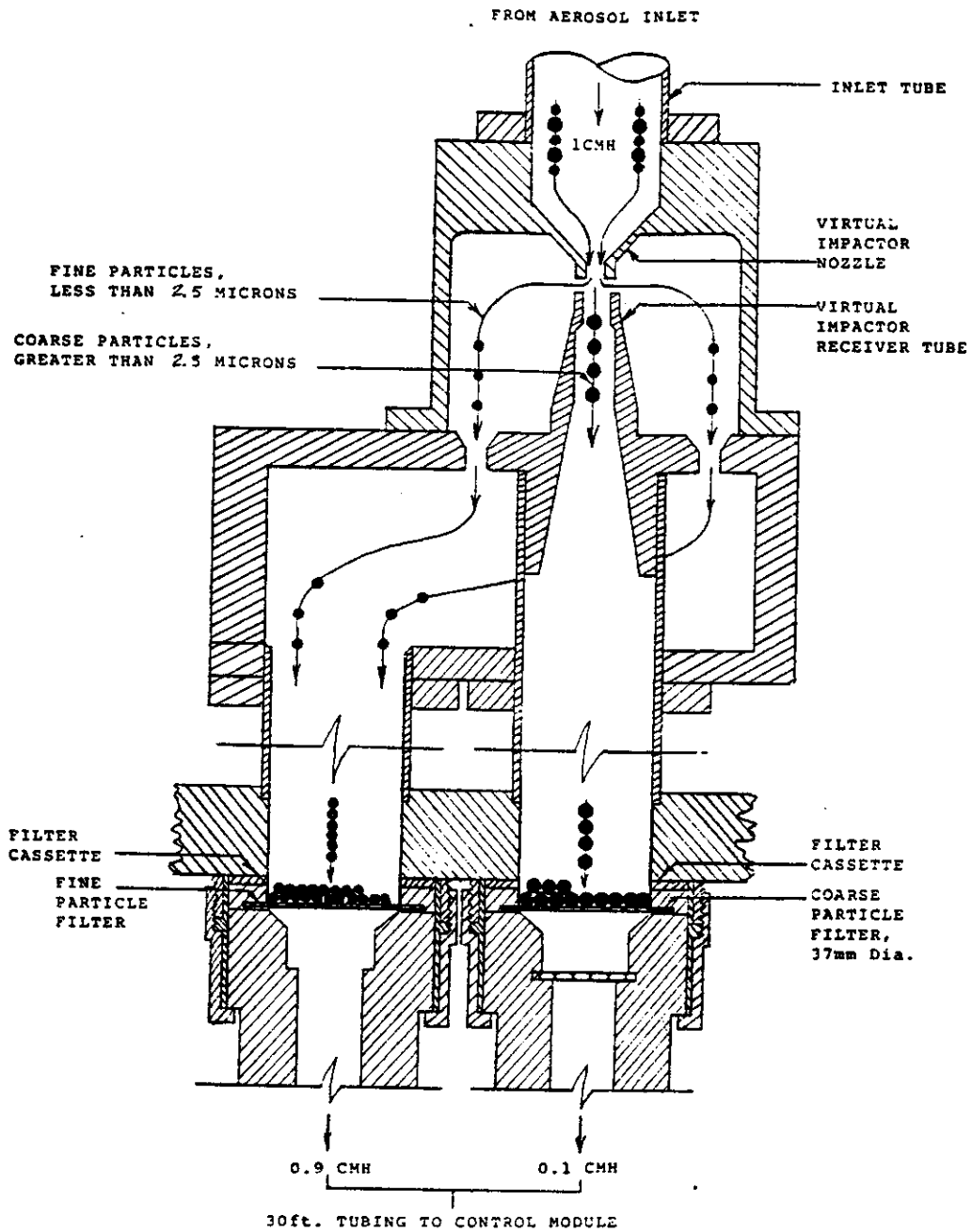
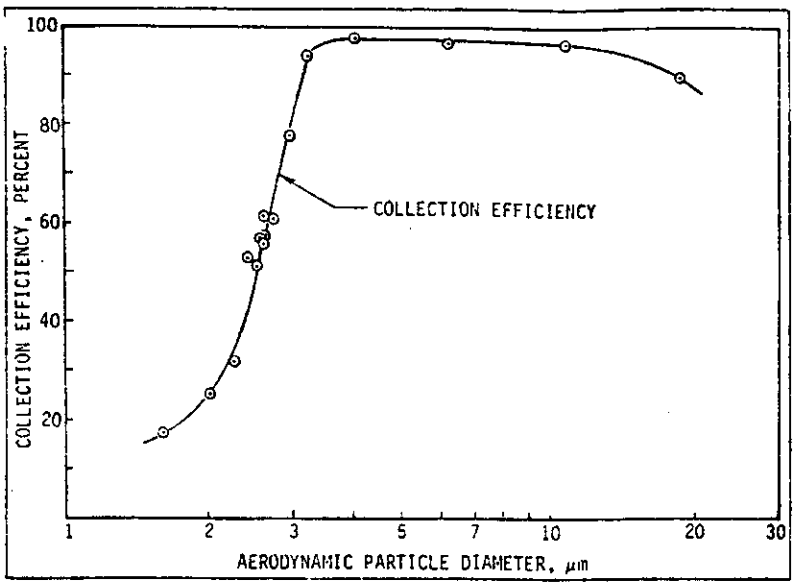
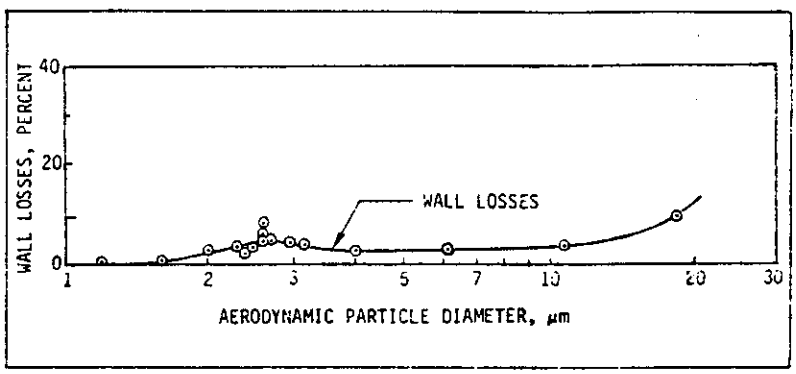


Figure 12. Virtual dichotomous sampler





13. Collection efficiency and wall loss of a virtual dichotomous sampler. (Ref. 8)

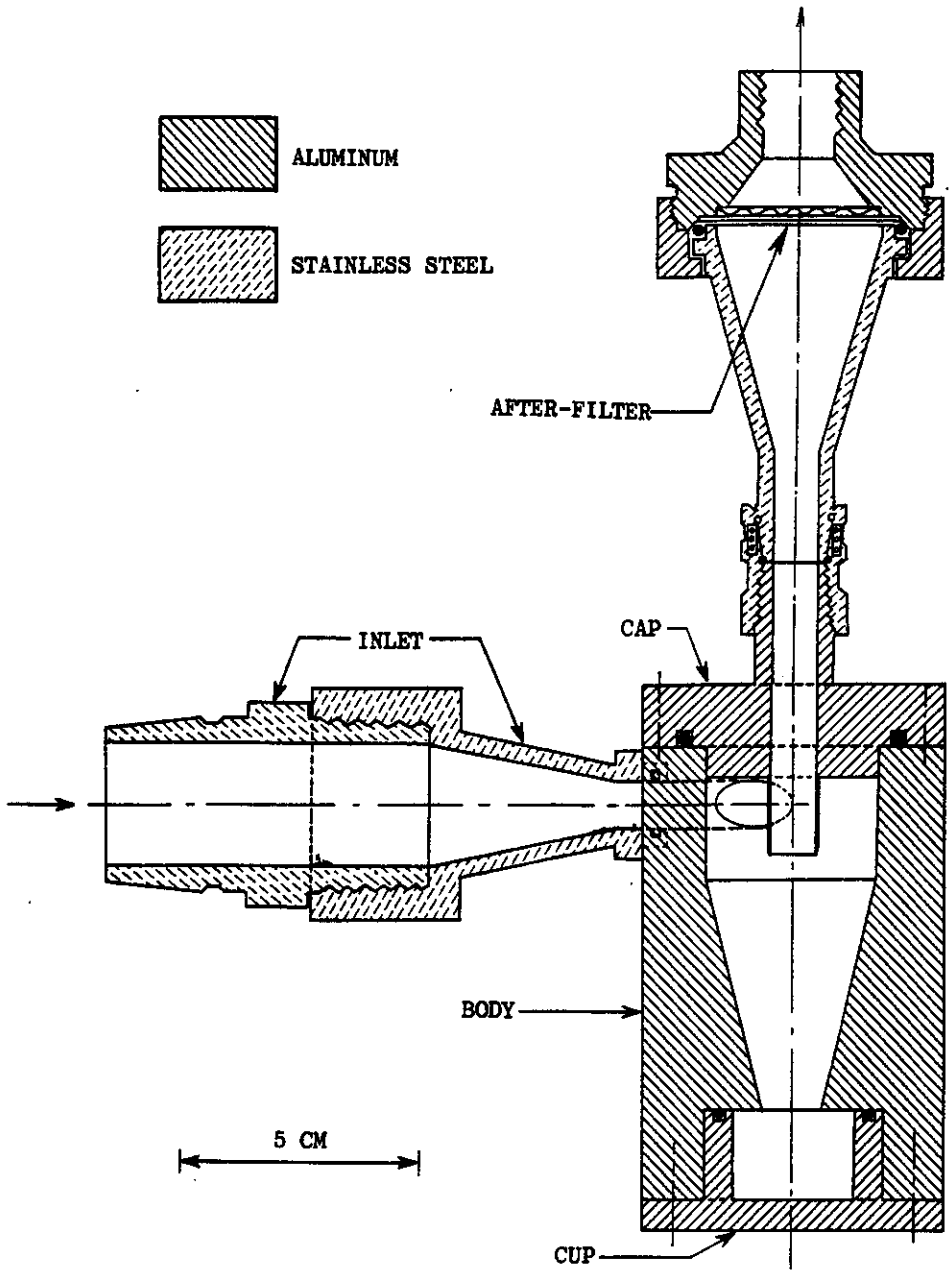


Figure 14. Cyclone sampler. (Ref. 17)

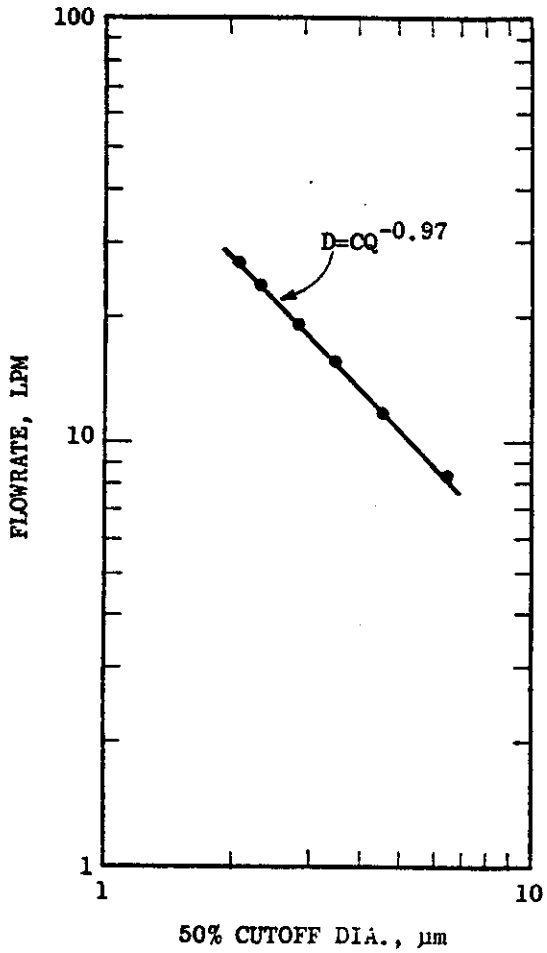


Figure 15. Cutpoint vs. flow rate for the cyclone in Fig. 14.

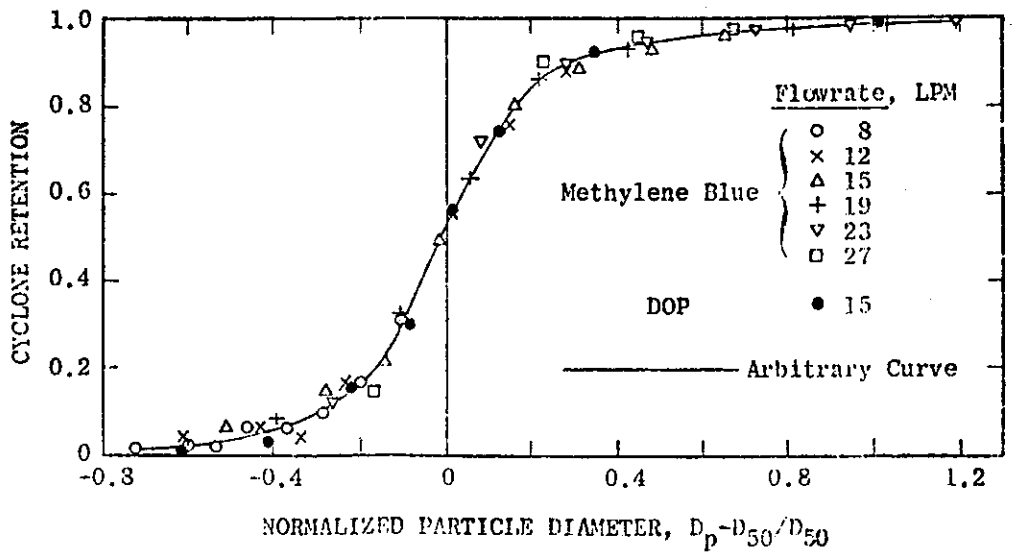


Figure 16. Universal plot of particle retention vs. flow rate for the cyclone in Fig. 14.

So far we have discussed only samplers for ambient air. However, we recognize that an inhalable particle standard will necessitate the sampling of stationary sources with particle size selection. Figure 17 shows a cascaded cyclone system developed by the Southern Research Institute.[19] It is used with the EPA Method 5 sampling train at 1 CFM. Another version, called the SASS train, has been developed for sampling at 4 CFM.[19] Southern is currently developing a cyclone train with the 15 μm inhalable cut.[20]

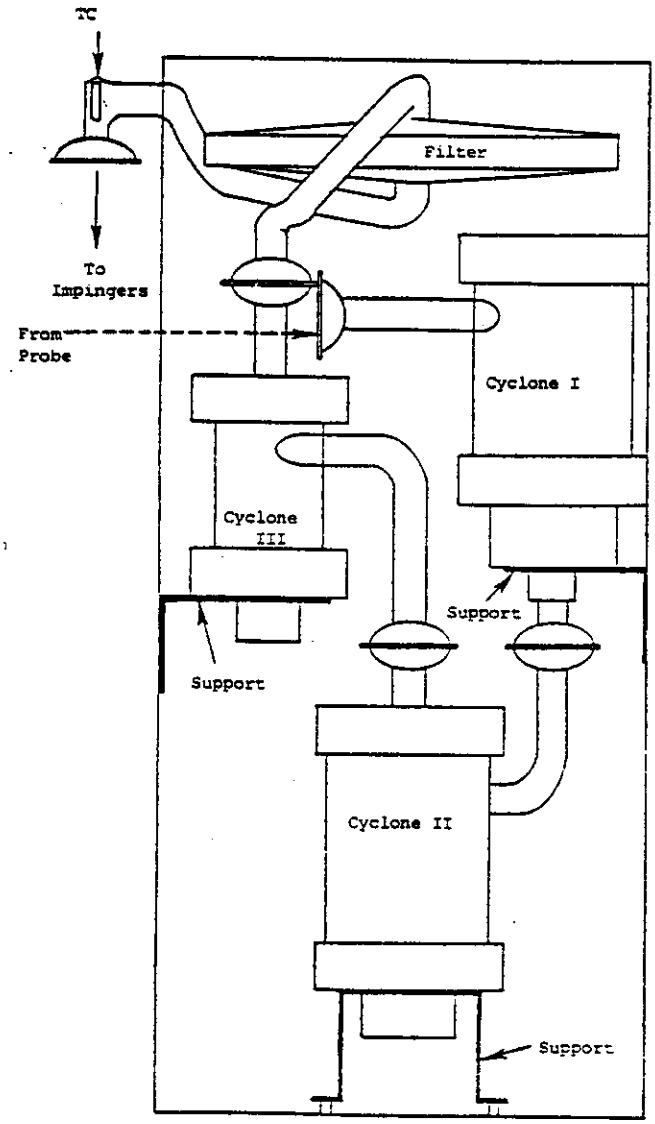


Figure 17. Cyclone train for stack sampling. (Ref. 19)

Other Aspects of Sampling Technique

Besides the size fractionating stages, there are other important aspects of the samplers which should be mentioned. It is important that the flow rate be maintained constant as the filter loads up. Fig. 18 shows that the flow rate vs. pressure drop for a commercial electronic flow controller is constant up to 20 in. of Hg.[21] The virtual dichotomous samplers are equipped with a less expensive mechanical flow controller. Fig. 19 shows that these control well up to 15 in. Hg.[21]

New types of Teflon membrane filters have been developed recently. Table 4 shows efficiencies measured by B. Liu with 0.3 μm DOP particles.[22] Note that 3 μm pore size Teflon shows a small penetration. Our own measurements with ambient particles (Table 5) show the same effect.[23] Thus one can choose say 2 μm pores to minimize the pressure drop while maintaining near 100% efficiency. The Teflon filters show low artifact sulfate and nitrate production. They are suitable for XRF analysis as shown in Fig. 20.

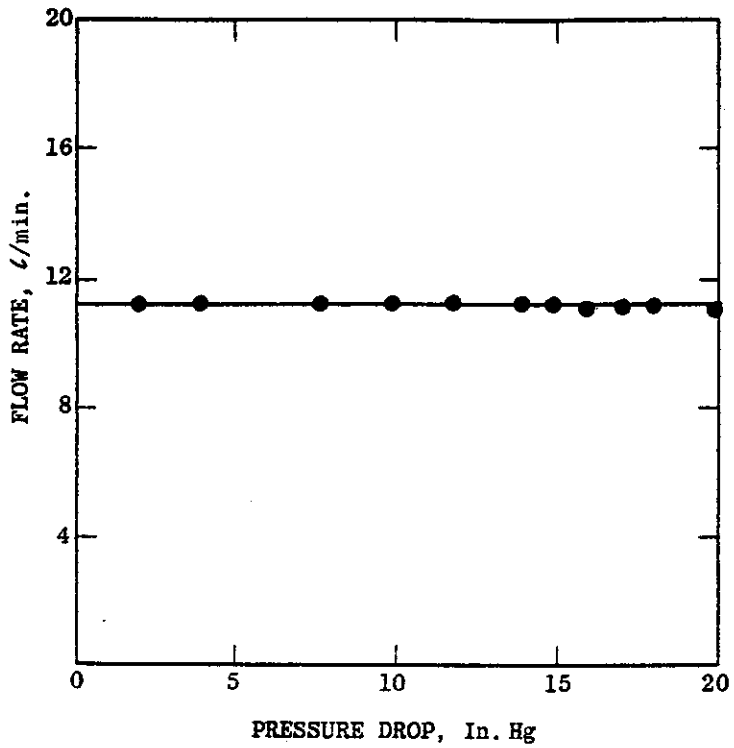


Figure 18. Example of performance of an electronic flow controller. (Ref. 21)

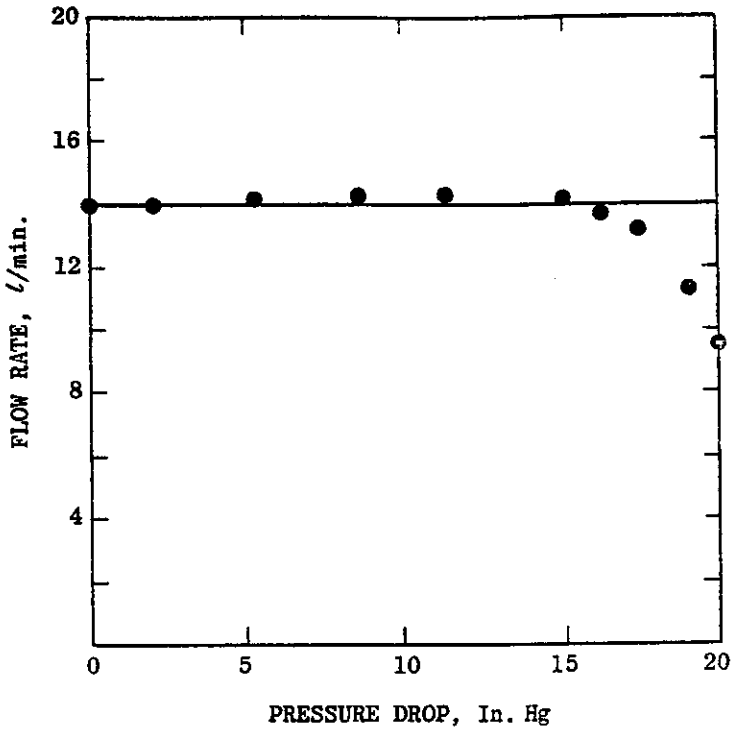


Figure 19. Example of performance of a mechanical flow controller. (Ref. 21)

Table 4. Filtration efficiencies of membrane filters for 0.3 μm DOP particles. (Ref. 22)

MEMBRANE FILTER	VELOCITY cm/s	ΔP cm Hg	MAXIMUM EFFICIENCY
Millipore, cellulose-acetate, 3μm	21	3	99.99%
Fluoropore, teflon			
3μm	24	1	98.1%
1μm	34	3	>99.99%
Ghia, teflon			
3μm	39	1	95.9%
2μm	23	1	>99.98%
Nuclepore, 0.4μm	43	10	89%

Table 5. Filtration efficiencies of Teflon membrane filters for ambient particles. (Ref. 23)

TEFLON MEMBRANE	PRESSURE DROP cm Hg	EFFICIENCY 49cm/s
Ghia 1-3μm	3.6	>99.9%
Ghia 2-4μm	2.1	>99.9%
Ghia 3-5μm	1.0	98.4%
Fluoropore 1μm	-	>99.9%
Fluoropore 3μm	2.1	99.5%

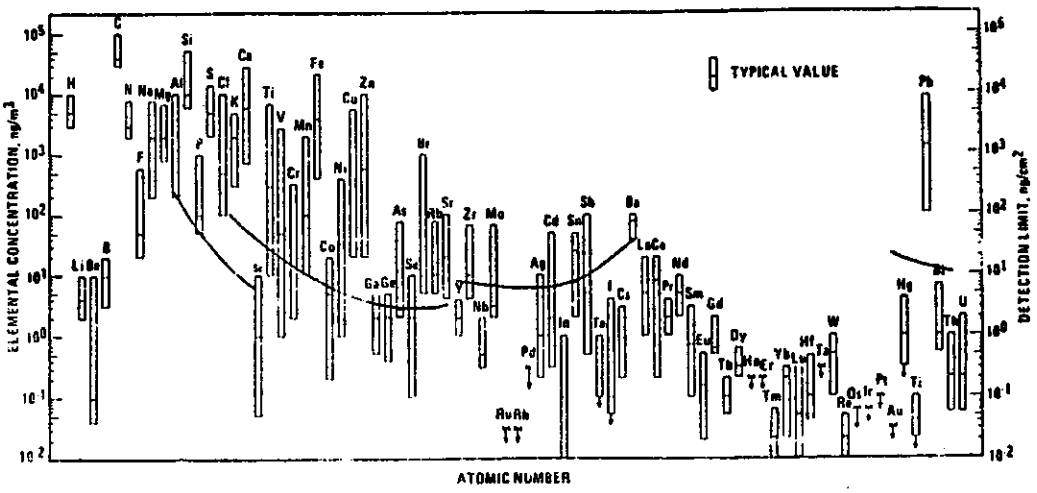


Figure 20. Typical elemental concentrations and detection limits for energy-dispersive x-ray fluorescence analysis. (Ref. 16)

The collected particulate mass can be measured with a beta gage, shown in Fig. 21. The attenuation of the beta particles is directly related to the mass loading. Advantages of this technique include the possibility of measuring the mass without removing the membrane from its mounting frame. The tare weight also does not include the frame. They do have to be tared because 90% of the attenuation is from the filter itself and they are not sufficiently uniform to eliminate this step.

I mention XRF and beta gaging since the smaller sample collected by the dichotomous sampler requires appropriate analysis techniques.

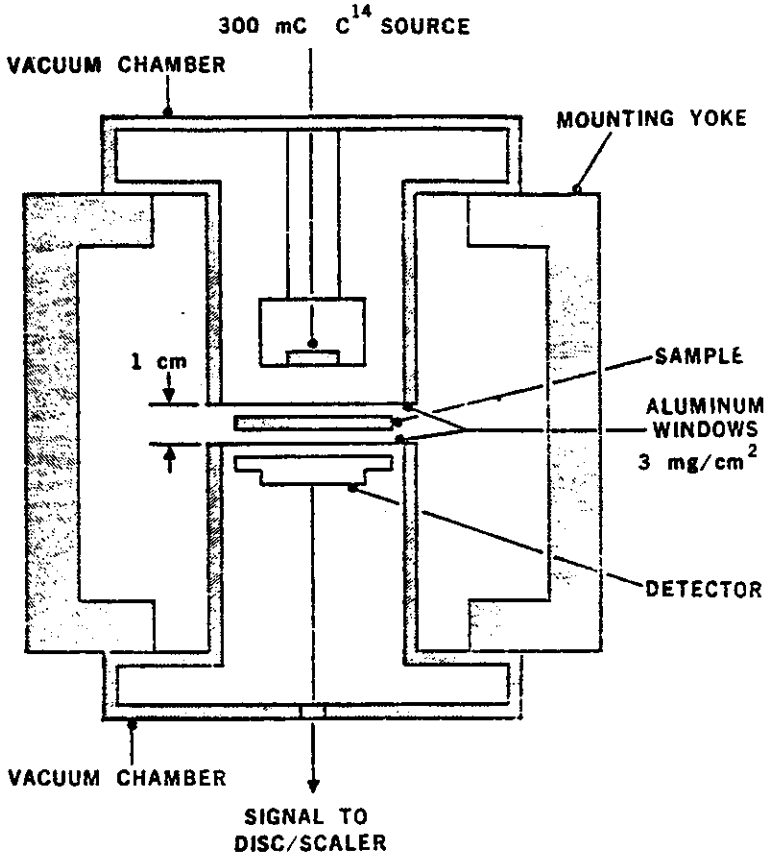


Figure 21. Beta gage for particulate mass measurement. (Ref. 16)

Conclusions

Now for some conclusions. I think it is evident that in the decades since the advent of the hi-vol, substantial advances in sampling technology have occurred. Not only can we design better samplers, but we can validate them rigorously, both in the laboratory and in the field. I can answer the question I began with in the affirmative. Size-selective sampling techniques are available and are adequate to support an inhalable particle standard.

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size considerations for establishing a standard for inhalable particles

Frederick J. Miller, Donald E. Gardner, Judith A. Graham, Robert E. Lee, Jr.
Health Effects Research Laboratory

William E. Wilson
Environmental Sciences Research Laboratory

John D. Bachmann
Office of Air Quality Planning and Standards
Strategies and Air Standards Division
U. S. Environmental Protection Agency

The U. S. Environmental Protection Agency (EPA) is required, under the amended Clean Air Act of 1977, to review the scientific basis for the total suspended particulate (TSP) ambient air quality standard and determine whether a revised particulate standard can be promulgated by December, 1980. It is recommended that research to develop information for a size-specific standard should focus on *inhalable particulate* (IP) matter defined as airborne particles ≤15 μm aerodynamic equivalent diameter. This particle size range relates to that fraction of particulate matter which can primarily deposit in the conducting airways and the gas-exchange areas of the human respiratory system during mouth breathing. It is also recommended that a second particle size cut-point of ≤2.5 μm diameter be incorporated in the air sampling devices, based upon considerations of the chemical composition and the size distribution of airborne particles, and on the predominant penetration of particles ≤2.5 μm diameter into the gas-exchange region of the respiratory tract. Data collected in this size range could be used in conjunction with epidemiological health parameters to refine an inhalable particulate standard in the future.

The need to consider size characteristics and chemical composition in the control of airborne particulate matter has been a matter of continuing concern to EPA. Since the establishment of the current total suspended particulate (TSP) ambient air quality standards in 1971,[1] the Agency has committed a substantial portion of its atmospheric research program to studying the sources, effects, transport, fate, and control technology of fractions of particulate matter thought to be of greatest significance to health and welfare. Much of this effort has focused on fine particles (<2 μm diameter), with an emphasis on sulfate compounds. However, despite the emphasis on these components, most recent reviews have concluded that available scientific information would not yet permit the establishment of meaningful health-based fine particulate or sulfate ambient air quality standards.

Although significant revisions of particulate regulations to control fine particles and/or sulfates might not be possible for several years, the Clean Air Act Amendments of 1977 require EPA to review by 1980 the scientific criteria upon which all of the national ambient air quality standards, including the TSP standards, are based. If, as a result of this review, revisions or modifications to the standards are indicated, the Amendments require that such revised standards be promulgated by December 1980.

In preparation of the mandated evaluation of the scientific criteria for particulate matter, a preliminary analysis was conducted of particulate research and information needs which could realistically be met by the 1980 time frame. This analysis suggested that serious consideration of a possible *respirable* size particle standard was desirable since it could lead to more effective control of those particles most likely to be responsible for adverse health effects. However, such a standard would be practical only if adverse health effects of concern could be related to concentrations of respirable particles and sufficient respirable monitoring capabilities could be established.

Therefore, the obvious first step in consideration of a possible respirable particles standard was to define just what particle size range should be considered as respirable. A number of definitions of *respirable particles* have appeared in the literature, usually with specific application to insoluble particles. However, as discussed below, most of these definitions are related to the protection of specific, occupationally-exposed groups and have not taken into account Clean Air Act requirements for establishing ambient air quality standards. In particular, the Clean Air Act requires that ambient air quality standards protect the health of the most

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sensitive or susceptible groups of the population from adverse effects associated with the regulated pollutant. Because of its multiple definitions, the term *respirable particulate matter* is not useful here. Rather, as will be presented, the term *inhalable particulate* matter should be used to define that fraction of particles penetrating into the lower respiratory system.

This paper presents the rationale used to define an upper size limit for inhalable particles capable of having effects on the lower respiratory system. This size limit can be used in developing a research data base—from both a monitoring and a health effects viewpoint—to allow the Administrator of EPA to make appropriate judgments concerning whether or not the TSP standard needs to be revised.

Particle Deposition in the Respiratory Tract

In man, the respiratory tract can be divided into three main areas in which experimental and theoretical research has been conducted on the deposition of inhalable particles, as shown schematically in Figure 1. These areas are the upper respiratory tract, extending from the anterior nares to the larynx, the conducting airways, and the gas-exchange areas. Beginning with posterior portions of the nares, the nasal turbinates, the trachea, the bronchial airways are ciliated and lined by mucus arising from glands and/or secretory cells. The nonciliated portions of the lung, which are lined by surfactant, are comprised of the respiratory bronchioles and the alveolar regions and represent the gas-exchange areas.

Others have defined respirable particles as that portion of inhalable particles which is deposited in the nonciliated portions of the lungs.[2-4] However, these definitions were specifically restricted to those particles which produce chronic diseases of alveolar region tissues, e.g. silicosis and coal workers' pneumoconiosis. For airborne particles which may have toxic effects in the airways of the head and/or tracheobronchial region, other definitions of respirable particles are needed. Due to the paucity of human and animal data on the influence of particle size on observed health effects, it would be desirable that terminology be adopted which is specific to anatomical features and is devoid of implications of possible adverse health effects. It is clear that the toxicity of particles is closely tied to their chemical composition. As such, different types of particles deposited in the same area in the lung may have vastly different health consequences.

Practically all models of respiratory tract deposition have employed a normalizing technique based upon *aerodynamic equivalent diameter*, which is defined as the diameter of a unit density sphere that has the same terminal settling velocity as the given particle. Unless specified otherwise, all references to particle size will relate to aerodynamic equivalent diameter.

There are five mechanisms by which particle deposition can occur within the respiratory tract. These mechanisms involve interception, electrostatic precipitation, impaction, sedimentation, and diffusion; the latter three are the most important of these mechanisms.[5,6] Inertial impaction of inhalable particles is the principal mechanism for large particle deposition in the respiratory tract, acting on particles ranging from a few micrometers to greater than 100 μm in diameter. Sedimentation is one of the main mechanisms of deposition of inhalable particles having diameters of 0.5 to 2.0 μm, whereas diffusional deposition is important for particles less than 0.5 μm in diameter. The relative predominance of these mechanisms, with respect to deposition in the head, the conducting airways, and the gas-exchange areas has been studied.[7-10]

The deposition of particles within specific regions can be influenced by changes in respiratory flow rate, respiratory frequency, and tidal volume. Thus, the activity level of the individual and the route of breathing can significantly alter regional, as well as total, respiratory tract deposition of inhalable particles. Deposition in the conducting airways can be altered by physiological or pathological factors. Lippmann, *et al.*[10] have shown that deposition in the conducting airways is greatly enhanced for asthmatic and bronchitis patients and is higher than normal in cigarette smokers who inhaled 1–5 μm particles.

Using the equation developed by the Task Group on Lung Dynamics[11] for the probability of deposition of particles within the head during nose breathing, the probability is essentially one that particles $\geq$13.2 μm are retained in the head during normal respiration (i.e. nasal breathing with an inspiratory peak flow rate of 15 l/min).

Figure 2 shows that deposition of monodisperse aerosols in the head during inhalation via the nose is essentially 100% for particles $\geq$10 μm with average inspiratory flow rates on the order of 30 l/min, i.e. flow rates corresponding to moderate exercise.[6] However, during mouth breathing the nasal passages are bypassed, increasing the fraction of particles of a given size entering the trachea. For example, Figure 3 indicates that overall head deposition of 10 μm particles is only 65% when breathing by mouth at an average inspiratory flow rate of 30 l/min.[6] Thus, this method of breathing provides increased deposition in the conducting airways and gas-ex-

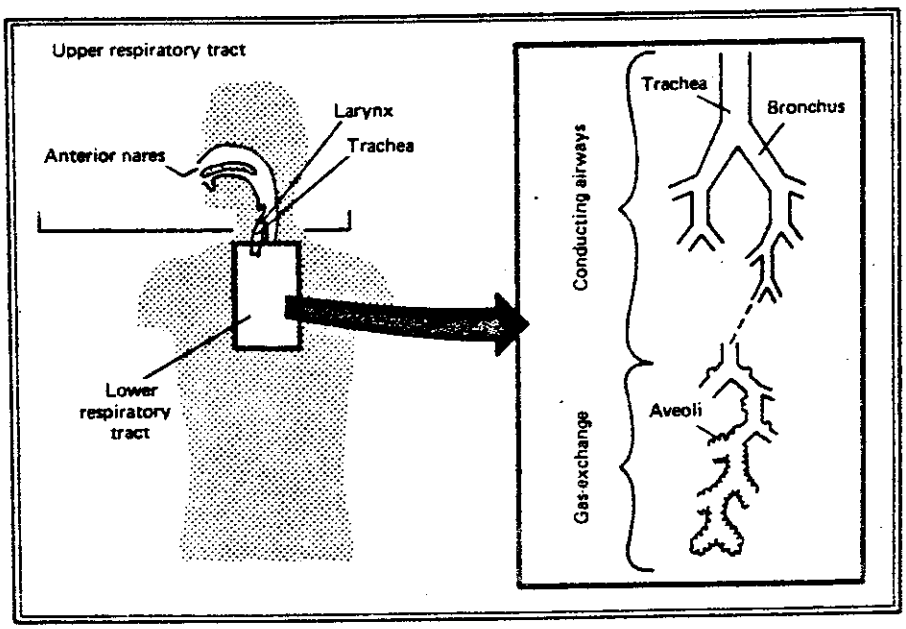

Figure 1. Diagrammatic representation of the human upper and lower respiratory tract.

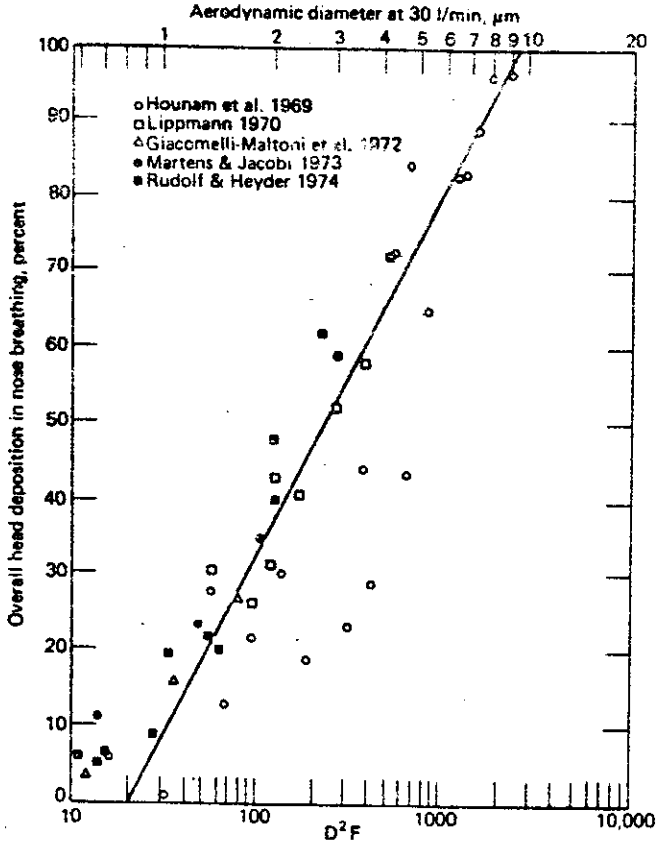

Figure 2. Deposition of monodisperse aerosols in the head during inhalation via the nose versus D^2F, where D is the aerodynamic equivalent diameter (μm) and F is the average inspiratory flow (l/min).[12-16] The inspiratory flows in the individual studies of this composite range from 5 to 60 l/min. The heavy solid line is the International Commission on Radiological Protection Task Group deposition model.[6]

change areas. Furthermore, the data of Figure 3 indicate that, even under these inhalation conditions, only a small percentage (<10%) of particles >15 μm would penetrate to the trachea. Various studies indicate that the deposition site within the nasal airways may be a crucial factor in determining the likelihood of possible adverse health effects because mucociliary transport may be rapid in some areas and relatively slow in others.[12,17] All available data demonstrate that direct health effects from inhalable particles >15 μm are primarily restricted to the upper respiratory tract.* Thus 15 μm would be a reasonable particle size cut-point to include in the design of a sampler which would differentiate particles deposited in the upper vs. lower respiratory tract.

Conducting airway deposition includes deposition both by impaction in the larger airways and by sedimentation in the smaller airways. While it would be desirable to have a sampler that had a cut-point that could delineate a particle size range that would primarily be associated with deposition in the conducting airways, the tremendous variability among individuals in deposition in this region prevents such a refinement. Palmes and Lippmann[18] identified a *characteristic airway* parameter which relates to the average size of an individual's bronchial tubes. Deposition curves for isopleths of this parameter[6] illustrate the variation in conducting airway deposition associated with particles in the 2–10 μm range. For example, in nonsmoking healthy males, deposition of 5 μm particles varies between 33 and 77%, depending upon an individual's characteristic airway dimension value. On the other hand, conducting airway deposition of 5 μm particles ranges from 30 to 95% for cigarette smokers. The range of conducting airway deposition values is less variable for particles <2 μm or >10 μm, due in large part to the fact that conducting airway deposition is slight for particles <2 μm and is nearly complete for particles >10 μm. Therefore, there is no *standard* conducting airway deposition curve, and hence, there appears to be no clear basis for establishing a particle size range which is exclusively restricted to the conducting airways.

For mouth breathing at 30 l/min average inspiratory flow rate, a maximum gas-exchange area deposition of approximately 50% is associated with a particle size of ~3 μm in nonsmoking healthy males (Figure 4). When the route of breathing is nasal, a maximum deposition of about 25% occurs with 2.5 μm particles, with a nearly constant deposition of 20% for all particles between 0.1 and 4 μm. Deposition patterns in the gas-exchange areas of the lung are not well defined for cigarette smokers and for individuals with chronic lung disease. At first glance it would appear that a cut-point anywhere between 2 and 3 μm would reflect particle deposition primarily associated with the gas-exchange areas of the lung, since deposition in the head is slight (5–10%) for particles in the 2–3 μm size range (Figure 3). However, conducting airway deposition is much more variable for 3 μm particles (11–40% deposition) than for 2 μm particles (5–22% deposition).[6]

Aerosol Chemistry Considerations

Aerosol chemistry information also supports the desirability of a cut-point in the vicinity of 2 μm. The distinction between *fine particles* (<2 μm) and *coarse particles* (>2 μm) is a fundamental one, as shown in Figure 5. There is now an overwhelming amount of evidence that not only are two modes usually observed in the mass or volume distribution of well-mixed urban and rural aerosols, but that the fine and coarse modes are normally quite different in chemical composition. Of particular importance is the general division of acidic

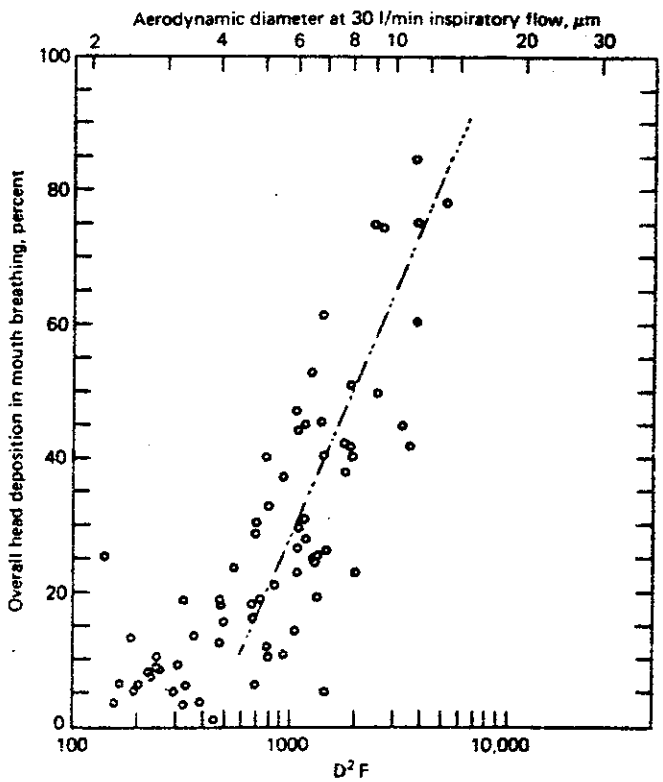

Figure 3. Deposition of monodisperse ferric oxide aerosol in the heads of nonsmoking healthy males during mouthpiece inhalations as a function of D^2F where D is the aerodynamic equivalent diameter (μm) and F is the average inspiratory flow in liters/min. An eye-fit line describes the median deposition between 10 and 80%.[6] The fitted line has been extrapolated to 15 μm.

* It should be noted, however, that a small number of large aeroallergen particles of the order of 25 μm aerodynamic diameter have been found in the deep lung parenchyma, and thus, the possibility exists that a direct contact mechanism may be operative in the genesis of pollenic asthma. See F. B. Michel, J. P. Martz, L. Gort and P. Cour, "Penetration of inhaled pollen into the respiratory tract," *Am. Rev. of Resp. Dis.* 115:569 (1977).

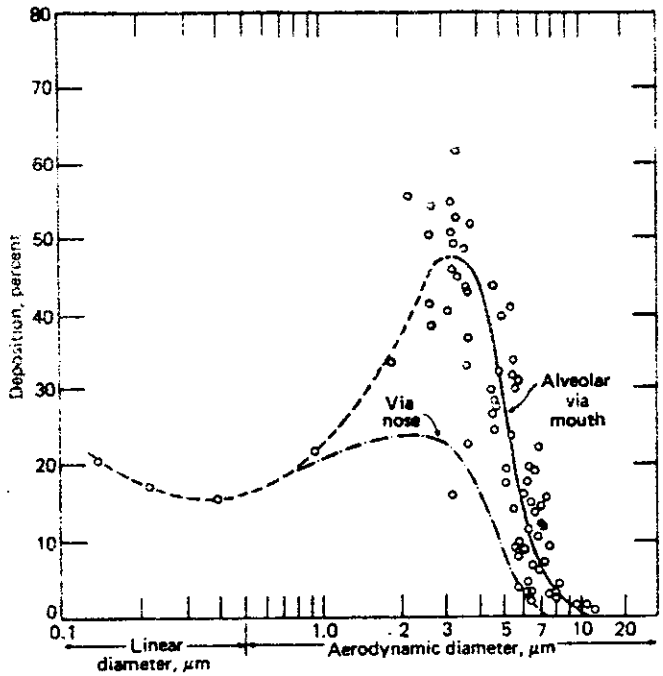

Figure 4. A comparison of the deposition in the alveolar region by mouth and nose breathing as a function of diameter. Lippmann and Albert utilized the data of various investigators in forming these eye fit curves.[6]

material into the fine fraction and of basic material into the coarse fraction. Measurements of acidity (pH) require that the sample be dissolved in water. If the fine and coarse fractions are collected in the same sample, the acid particles in the fine fraction will dissolve and be neutralized by coarse basic particles.

The fine and coarse particle modes in general: originate separately, are transformed separately, are removed from the atmosphere by different mechanisms, require different control techniques, have different chemical composition, and have different optical properties. Therefore, the distinction between fine and coarse particles is of fundamental importance to any discussion of the physics, chemistry, measurement, or air quality standards of aerosols. Fine and coarse particles differ substantially in their deposition patterns in the respiratory tract. Furthermore, other differences, described below, make it desirable to collect and analyze them separately.

The physical separation of the fine and coarse modes occurs because condensation/coagulation processes produce fine particles, while mechanical processes produce mostly coarse particles.[19] This is shown in an idealized schematic in Figure 5. Individual sources of primary aerosols may produce fine or coarse aerosols; some chemical species in the coarse mode may have a tail extending into the fine mode. Secondary aerosols, formed in the atmosphere from primary gaseous emissions, will normally be fine. The nuclei mode is observed only near combustion or other high temperature sources. These particles rapidly coagulate into the accumulation mode. The dynamics

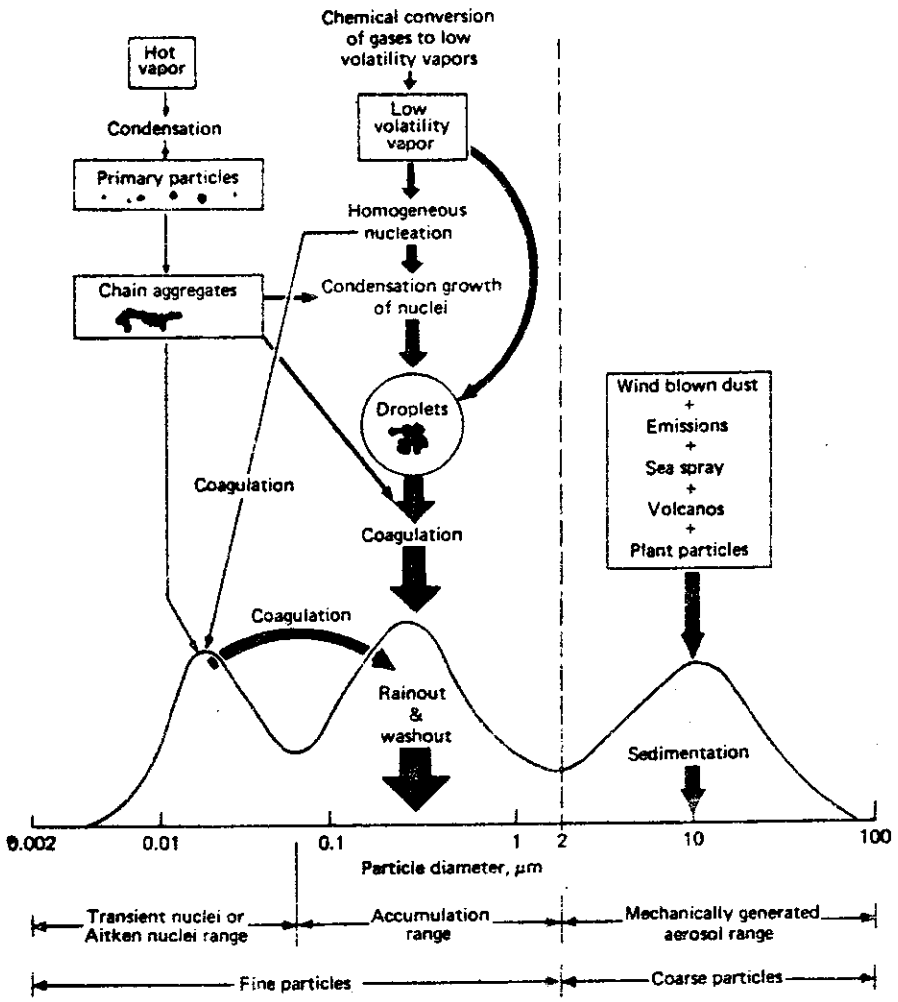

Figure 5. Idealized schematic of an atmospheric aerosol surface area/mass distribution showing principal modes, main sources of mass for each mode, and the principal processes involved in inserting mass in each mode and the principal removal mechanisms. (from K. T. Whitby and B. Cantrell, "Atmospheric Aerosols—Characteristics and Measurements," in *International Conference on Environmental Sensing and Assessment*, Session 29, Fine Particles, Las Vegas, Nevada, September 14–17, 1975, Institute of Electrical and Electronics Engineers, New York, 1976, with permission).[19]

of particle growth normally prevent accumulation mode particles from growing larger than about 1 μm.

The existence of a bimodal distribution with fine and coarse modes has been clearly demonstrated by cascade impactor studies[20-27] which yield mass-size distributions and by number distribution studies[28] which may be converted into volume distribution. These size distribution studies suggest 1–3 μm as the most appropriate range for a cut-point for fine and coarse aerosols. However, practical considerations of reducing plugging of impactor orifices indicate that 2.5 μm is a more appropriate cut-point, especially for particle size fractionating devices such as the dichotomous sampler.[29,30]

Impactor studies in which chemical composition has been determined as a function of particle size also demonstrate the division into fine and coarse modes and show the difference in chemical composition of the two modes. Except for a few trace elements, the chemical species are either primarily coarse, primarily fine, or bimodal.[20-27,31,32] On the basis of such studies, it is possible to divide the major chemical species observed in atmospheric aerosols into several groups shown in Table I.

Table I. Classification of major chemical species associated with atmospheric aerosols.[20-27,31,32]

Normally Fine	Normally Coarse	Normally Bimodal	Variable
$SO_4^=$, C (soot), Organic (condensed vapors), Pb, NH_4^+, As, Se, H^+	Fe, Ca, Ti, Mg, K, $PO_4^=$, Si, Al, organic (pollen, spores, plant parts)	NO_3^-, Cl^-	Zn, Cu, Ni, Mn, Sn, Cd, V, Sb

The major components of the fine fraction of the atmospheric aerosol are sulfate, ammonium, nitrate ions, lead, carbon-containing material including soot and condensed organic matter. In urban areas the fine fraction, as a percent of total suspended particulate matter, varies from 15–25% in Denver[22] to 40–60% in the Los Angeles area[33] and New York-New Jersey urban areas.[21] The percent of the fine particle fraction which is secondary varies from 60–80% in these urban areas (percentages based on short-term intensive studies). Also, several studies have shown that potentially toxic carcinogenic species, such as polynuclear aromatic compounds, As, Se, Cd, Zn, which can exist as vapors, are more concentrated in the fine particle fraction.[33-36]

The coarse fraction consists mainly of crustal material, such as Fe, Ca and Si. The major sources are wind erosion products, primary emissions, sea spray and volcanic eruptions.

Particle Size Recommendations for Ambient Air Measurements of Inhalable Particulate Matter

A complete characterization of ambient aerosol distributions through the use of multi-stage impactors or other such sampling devices in a national network would probably provide the best data for evaluating the health effects associated with inhaled particles. However, such a sampling network may not be cost effective from a monitoring and control implementation viewpoint. The cut-points established for a sampling device must maximize the potential for meaningful health monitoring data while not ignoring logical cut-points suggested by aerosol chemistry information.

Cut-points of ≤2.5 μm and ≤15 μm are recommended to be included in samplers for a particulate sampling network. A 15 μm cut-point provides a particle size above which tracheobronchial deposition is slight, even if individuals are exercising moderately. Also, an upper cut-point of 15 μm would eliminate collection of airborne allergens in the inhalable particulate fractions, a feature that would be desirable. There is, however, a need to continue air monitoring of larger size particles >15 μm to maintain continuity with previous health studies and other historical air quality data.

While the ≤2.5 μm fraction underestimates particles penetrating to gas exchange areas, the extent of the underestimation will not usually be great. Only a small fraction of the total ambient aerosol mass is between 2 and 3.5 μm, this latter particle size being the cut-point used by the American Conference of Governmental Industrial Hygienists and the Department of Energy in particulate sampling. Also, the minimum mass concentration based on aerodynamic particle size is usually closer to 2.5 μm than it is to 2 μm. Data collected in the ≤2.5 μm size range could be used in conjunction with epidemiological studies to refine, if necessary, a particulate standard in 1985.

Summary

It is recommended that the term *respirable* particulate matter be avoided because of its vagueness and that the term *inhalable* particulate matter be used for particles ≤15 μm aerodynamic equivalent diameter. The suggested cut-points of ≤15 μm and ≤2.5 μm aerodynamic equivalent diameter provide guidelines for future human and animal research to obtain a data base for evaluating health effects of inhalable particulate matter.

Disclaimer

This report has been reviewed by the Health Effects Research Laboratory, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

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Dr. Miller is a senior biostatistician in the Statistics and Data Management Office, Health Effects Research Laboratory, Dr. Gardner is the Chief of the Inhalation Toxicology Branch, Health Effects Research Laboratory, Ms. Graham is a research microbiologist with the Inhalation Toxicology Branch, Health Effects Research Laboratory, and Dr. Lee is the Deputy Director of the Health Effects Research Laboratory, U.S. Environmental Protection Agency. Dr. Wilson is Scientific Director, Regional Field Studies Office, Environmental Sciences Research Laboratory and Mr. Bachmann is an environmental engineer in the Strategies and Air Standards Division, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.

Health Effects of Airborne Particles

Rodney R. Beard, MD MPH

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The discussion of this topic must of necessity be as broad and varied as the composition and characteristics of all the particles which occur in the atmosphere.

This colloquium is aimed to review facts and ideas that can guide the California Air Resources Board to decisions about an air quality standard for particulate air pollutants, without specification as to their chemical or other identity.

There are a number of toxic materials which may be encountered as airborne dusts, fumes, fogs, or nuclei. Most of these arise from specific sources, and are subject to emission controls. Their levels of toxicity are reasonably well established, except as they may promote carcinogenesis. These include arsenic, beryllium, cadmium, chromium, fluorides, manganese, nickel, silica and vanadium. They do not contribute much to the total number of particles in the air. A second category of inhalable toxic agents includes asbestos, lead, pollens, and polynuclear aromatic hydrocarbons. These are generated from many small sources, and consequently they can not be controlled as easily as those in the first category. They have to be considered individually because of their unique characteristics. Their toxicity is also fairly well understood.

What remains is still a very heterogeneous mixture,

ranging from dusts from quarries and sandblast operations to acid aerosols formed by chemical reactions in the atmosphere. Sulfates, nitrates, ammonia salts, sodium chloride, clay, organic materials, and many other species are included. It is probable that some of these will be found to be so toxic that they should be controlled individually.

The earliest and most persistent efforts for the control of air pollution have been aimed at the control of suspended particles in the form of smoke. Ill health caused by smoke inhalation was invoked from the outset as the justification for controls on burning. However, it was generally believed that some reactive chemical must also be associated, and pungent, irritating sulfur dioxide was readily accepted for this role. Consequently, particulate pollution is always discussed together with sulfur dioxide emissions. As Ferris says in his excellent 1978 review of the health effects of the controlled air pollutants, "These two pollutants will be discussed together because, in general, they come from a common source - combustion of fossil fuels; and sulfur oxides exist as a gas, sulfur dioxide, and as a particle, sulfate." (1)

The most conspicuous examples of illness and death caused by air pollution are clearly related to exposures to excesses of smoke and airborne sulfur compounds, and in the

most dramatic instances, with fog. The Meuse Valley, Donora, and London established this for acute episodes. The decrease of chronic bronchitis in London after constraints were put on the use of coal as domestic fuel supports the view that chronic lung disease has similar causes.

A key question is, Do chemically non-reactive particles cause lung disease or interfere with lung function, if exposures are long-continued?

It appears reasonable to believe that a heavy deposit of foreign material, even if inert, could clog air passages, occlude alveoli, or impede the flow of lymph (or tissue fluid) and thus impair respiratory function. Also, the loading up of the mechanisms for the removal of foreign particles from the lung might impair the protective reaction to infection, and interference with blood flow might lead to tissue degeneration or to effects upon the heart.

The first factor which might affect this is the penetration and deposition of particles in the respiratory tract. A number of studies have been done in animals, and extensive speculation and calculations have led to the development of widely-quoted models to describe the deposition of dust in the lungs of man. However, few measurements of regional deposition in humans have been attempted, and the National Research Council Committee on

the Medical and Biological Effects of Environmental Pollution recently published the conclusion that "None of the previously proposed models provide reliable estimates of aerosol deposition in healthy normal adults, because their predictions for total and alveolar deposition efficiencies differ from the best experimental data." (2) This is obviously of urgent importance for our understanding of the effects of inhaled particles and the prediction of safe levels for community air. This topic will be developed by other participants in the Workshop.

The second factor to consider is the fate of the deposited particles. In healthy persons, those which are caught in the nose, throat, trachea, bronchi and bronchioles will almost all be carried to the throat by the mucociliary transport system, and will be swallowed, within the space of a day. There are large differences among individuals, and even within one person, from time to time. In people with respiratory disease, several complicating factors may appear: the mucus layer may be thick, narrowing the airway and thus modifying the airflow rate and thereby the deposition of particles; the composition, the volume, and the flow rate of the mucus may be altered, increasing or decreasing the rate of clearance (2). Camner et al (3) report that inhalation of 11 micrometer carbon particles at a concentration of 50 grams per liter led to more rapid clearance of previously deposited 6 micrometer teflon

particles in 6 of 8 healthy subjects.

Nau et al (4) exposed several animal species to inhalation of very small particlesof carbon black, 0.025 and 0.035 micrometers in diameter, and found no effect other than accumulation of the dust in the lungs.

In a study using a polydispersed aerosol of calcium carbonate, with particles from 0.2 to 5 micrometers in diameter, Norris and Bishop (5) elicited a variety of disturbances of pulmonary functions in both healthy human subjects and in patients with lung diseases, mainly characterized by uneven ventilation and impaired gas exchange.

Albert, Lippmann, et al (6) studied the deposition and clearance of radioactive tagged iron particles in human subjects. The particle size was closely controlled, and in different experiments ranged from about 2 micrometers to about 6 micrometers. Clearance time for 90% of the particles from the bronchi was extremely variable among different subjects, ranging from 2 to 20 hours in nonsmokers. The intrasubject variation on repeated tests was less marked, about 30 %. Smaller particles, deposited more distally, were cleared less rapidly than large particles. Cigaret smokers showed more rapid clearance than nonsmokers, and

smokers showed longer clearance times after abstaining from cigarets.

The National Research Council (NRC) Committee (2) suggests that acute dust exposures increase mucus production and mucociliary transport; continuation of the exposure leads to hypertrophy of bronchial mucus glands and hyperplasia of goblet cells; eventually, mucociliary transport becomes inadequate for removal of the excess secretions; chronic cough, accumulation of secretions, and increased susceptibilty to inhaled particles, noxious gases, and pathogenic organisms then follows. This progression was originally proposed by Albert, Lippmann, et al as an explanation for the course of smoker's bronchitis. It has not been shown that inert dusts cause hypertrophy of the mucus glands.

Particles that reach the alveoli can be removed by macrophages, large ameboid cells which can ingest them. The macrophages then migrate to the ciliated portion of the bronchial tree and thence to the throat. It is uncertain whether they move through the interstices of the lung tissue or on the surface, in a moving film of secretions.

Particles may also penetrate the alveolar walls and enter the interstitial spaces, remaining there or being transported by the flowing lymph or tissue fluid toward the

central structures of the chest. Collections of lymph nodes, filter-like structures through which lymph flows, are located along the courses of the bronchi. These nodes show increasing deposits of foreign particles with advancing age, and persons who have been heavily exposed to particulate pollution by occupation or smoking have very dense accumulations. Urban dwellers usually show more than country people.

Some particles make their way into the blood capillaries and are carried to other parts of the body.

The NRC review (2) summarizes studies on the kinetics of alveolar clearance. It appears that there may be two phases, one with a half time measurable in weeks, and another with a half time of months or years. The length of the second phase appears to be related to the solubility of the particles. These generalizations are based on animal experiments, done mainly in dogs.

Two laboratory studies of alveolar clearance in humans have been reported. Albert and Arnett (7) found two-phase clearance of radioactive particles in two subjects. The first phase lasted about a month. A third subject with a chronic cough, a heavy cigaret smoker, showed a single-phase clearance pattern.

Morrow et al (8) found only a single alveolar clearance phase in four subjects who inhaled manganese dioxide particles. The half time ranged from 62 to 68 days. A second set of experiments gave similar results for manganese dioxide and for ferric oxide (9).

Susceptibility to lung infection was studied by Vintinner and Baetjer (10). They exposed rats to coal dust and smoke, and found no alteration of reponse to inoculations of Type I pneumococci.

The effects of inert particles on pulmonary functions, such as flow resistance, compliance, forced vital capacity, timed vital capacity and maximal expiratory flow rate have had little effective study.

Amdur and Underhill (11) found no increase in pulmonary flow resistance after one-hour inhalations of carbon, manganese dioxide, open hearth dust, ferric oxide, manganous chloride, ferrous sulfate or sodium orthovanadate particles in guinea pigs. The particles were under 0.5 micrometer in diameter.

DuBois and Dautrebande (12) reported constriction of airways and increased resistance to flow in isolated guinea pig lungs and in human subjects after large doses of various inert dusts, but the studies were directed to other

interests and the report leaves much uncertainty about details of dust exposures and related results.

We turn now to consideration of the combination of inert aerosols with gaseous pollutants. As was said before, sulfur dioxide has had the most attention. However, the earliest report on this subject is credited to Dautrebande(13), who, in 1939, reported that concentrations of mustard gas which had little effect in rats became lethal when combined with a sodium chloride aerosol. When such an effect of two agents acting together to produce an effect greater than the sum of their individual effects is observed, the term "synergism" is applied.

LaBelle et al (14) suggested that a highly soluble irritant gas that would ordinarily be absorbed in the upper respiratory tract, such as formaldehyde, could be carried to the lungs by small particles on which the gas could be adsorbed, and thereby cause more severe injury. Conversely, adsorption on small particles would diminish the toxicity of the sparingly soluble gases, such as nitrogen dioxide. His experiments supported this. Dalhamn and Reid (15) found that carbon particles enhanced the toxicity of ammonia. Boren (16) reported protection of mice from the pulmonary edema expected from exposure to a high concentration of nitrogen dioxide if they were simultaneously exposed to a carbon aerosol. After repeated exposures to the mixture,

the animals developed focal pulmonary lesions.

Amdur devised methods for the measurement of airflow resistance and lung elasticity, or pulmonary compliance, in guinea pigs, and made extensive use of these techniques. She showed synergism among several substances in combination with sulfur dioxide (17). The effect was to increase airflow resistance and to decrease lung elasticity. In one study, she compared several combinations and concluded that synergism occurred only if the particles were soluble in water, and also that the effect was related to the solubility of sulfur dioxide in the salt solution used to generate the aerosol. It has been suggested that guinea pigs are uniquely susceptible to constriction of the airways caused by inhalation of irritants.

Amdur and Corn (18) demonstrated the inverse relation between particle size and toxic effect. They used zinc ammonium sulfate in a mass concentration of about 1 milligram per cubic meter. Four polydisperse aerosols with particle sizes averaging 0.3 to 1.4 micrometers diameter were used. There was a dramatic inverse relationship between particle size and the effect on airway resistance. This may have been related either to the greater penetration of the smaller particles or to the greater ratio of surface area to volume. This greater effect of smaller particles was also shown for sulfuric acid (19).

It should be noted that studies of these and other highly soluble aerosols are complicated by their hygroscopic nature. The particles will grow rapidly in size where the humidity is high, and this factor must be carefully controlled in the operation of experimental exposure chambers.

Amdur and Corn (18) and Amdur and Underhill (11) compared the effect of several sulfate aerosols. They found the following order of potency: sulfuric acid, zinc ammonium sulfate, ferric sulphate, zinc sulfate, ammonium sulfate, copper sulfate, ferrous sulfate, and manganese sulfate. The latter two had negligible effects.

Burton, Corn et al (20), Frank, Amdur, and Whittenberger (21), and Small and Luchsinger (22) have failed to elicit synergism between sulfur dioxide and sodium chloride aerosol in man, although such synergism was reported by Nakamura (23) and by Toyama (24,25).

Ultimately, effects in humans are the markers which we seek to guide our judgments about the importance of air pollution in keeping our health. Studies in animals show which organs are likely to be injured, and what kinds of injury to look for. They often give us measures of comparative toxicity,. But the only conclusive observations must come from studies of mankind. Because of the strict

limitations on experiments in human subjects, we must get much of our knowledge from "natural experiments" in which groups of people have been exposed to pollutants because of ignorance or lack of caution or negligence. Such epidemiological studies often provide convincing evidence of concentrations of pollutants which can be dangerous. More often, important parts of the desired evidence are lacking, and the conclusions can only be tentative. In recent years, as we have learned more about the method, epidemiological studies have become more persuasive and less subject to differences of interpretation.

The NRC report (2) discusses the epidemiology of health effects of airborne particles at length. It concludes with a table which summarizes the available data as follows: (Table 8.3).

Table 8-3. Health effects and dose-response relationships for particulates and sulfur dioxide

Averaging time for pollution measurements	Place	Particles (mg/m^3)	SO_2 (mg/m^3)	Effect	Reference
24-hour	London	2.00	1.04	Mortality	102
		0.75	0.71	Mortality	446
		0.50	0.50	Exacerbation of bronchitis	450
	New York City	6 COHS[a]	0.50	Mortality	297
		3 COHS	0.70	Morbidity	296
	Chicago	Not Stated	0.70	Exacerbations of bronchitis	125
	New York City	0.145 (+?)	0.286	Increased prevalence of respiratory symptoms	156a
	Birmingham, Ala.	0.18–0.22	0.026	Increased prevalence of respiratory symptoms	156a
	New York City	2.5 COHS	0.52	Mortality	277
Weekly mean	London	0.20	0.40	Increased prevalence or incidence of respiratory illnesses	39
Six winter months	Britain	0.20	0.20	Bronchitis, sickness, absence from work	547
Annual	Britain	0.07	0.09	Lower respiratory infection in children	202
		0.10	0.10	Bronchitis prevalence	432
		0.10	0.12	Respiratory symptoms and lung function in children	497, 498
	Buffalo	0.08	0.45[b]	Mortality	823, 824
	Berlin, N.H.	0.18	0.73[c]	Decreased lung function	229

[a] Coefficient-of-haze units.
[b] mg $SO_3/cm^2/30$ days.
[c] mg $SO_3/100\ cm^2/day$.

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