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Use of Membrane Filters in Air Sampling

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FIRST efforts to develop a membrane filter, also called a molecular filter membrane, began in Europe over three decades ago. Early investigators^{1,2,3} working with cellulose ester gels for the ultra-filtration of liquids, found difficulty in obtaining a uniform filter material in the dry state. This problem was solved by Goetz⁴ and his co-workers in 1950 at the California Institute of Technology. Since 1951, hydrosol and aerosol membrane filters have been available in this country.

Many authors have described the use of hydrosol filters in the bacteriological examination of water.^{5,6} Time and space required for certain phases of bacteriological analysis have been substantially reduced by using membrane filters, found to give as precise results as are obtained with other techniques.

First and Silverman,⁷ and Goetz⁴ discussed numerous uses and adaptations of the membrane filter for environmental studies in the field of occupational health. In our laboratories we have been using the aerosol-type membrane filter since 1953. This paper describes some of our experiences with the membrane filter and points out certain of its limitations.

Properties of Aerosol Filter

THE AEROSOL-TYPE membrane filter is a dry cellulose ester membrane approximately 150 microns in thickness having 80% to 85% voids. It is composed of tubular pores with smaller diameters at the front or filtering surface than at the back. The pore size of the filter in common use in our laboratory averages approximately 0.8 micron. Air passing through the filter causes a rapid build-up of an electrostatic charge which enables the filter to collect particles in the 0.1 micron range with close to 100% efficiency. High collection efficiencies are

also obtained for smaller aerosols. The membrane filter material has a refractive index of about 1.5 and it can be made almost transparent with an immersion oil of the same refractive index.

FILTER HOLDER: One disadvantage of the membrane filter is its brittleness; it must be firmly supported or it will rupture from the high resistance to air flow. Different supporting materials for the filter are being used including a porous carbon plate, a sintered glass plate⁸ and a wire screen.⁹ We have modified a filter holder originally designed by the Atomic Energy Commission for use with filter paper (Fig. 1). It is made of brass with a threaded collar to hold the filter firmly in place. A 200-mesh, bronze screen held taut by a threaded collar in the holder base supports the filter. A brass ring is placed over the edge of the filter disc and the top of the filter holder is screwed down tight onto the ring. The holder accommodates a standard 1¼ inch filter disc and provides an effective filtering area one inch in diameter. A Schraeder

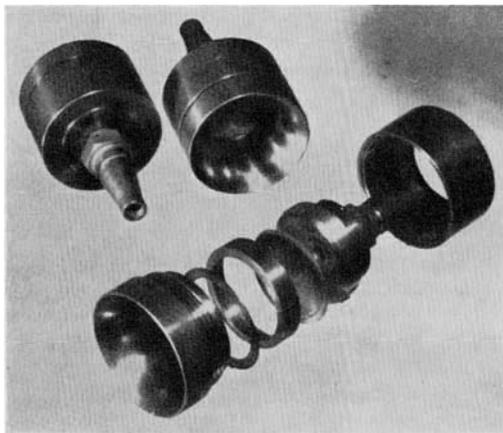


Fig. 1.
A brass holder for the membrane filter.

connector or straight section of brass tube can be used to attach the holder to the source of suction.

SAMPLING CONDITIONS: Sampling rates vary from 0.1 to 0.5 cu ft/min depending on several factors. These include the type and concentration of the contaminant and the type of pump or other source of suction available. We have used the midget impinger pump and other pumps. The midget impinger pump gives a flow of approximately 0.1 cu ft/min at a negative pressure of 12 inches of water, normally used for midget impinger sampling. In our experience, the flow resistances of different filter discs of the same batch are quite uniform, but we have found that resistances vary considerably in different batches. Frequent calibration of the air flow through the filters should be made. If samples are collected for extended periods, increased resistance to air flow must be taken into consideration. In such cases it is best to have some type of metering device or constant flow orifice in the sampling system.

Dust Analysis—Optical Microscope

DIRECT COUNT: After collecting a sample, we place the filters in individual glassine envelopes for transmittal to the laboratory for analysis. Optical dust counting of a sample by the direct method, involves merely adding a drop of immersion oil (refractive index 1.5) to the filter which is placed dust side down on a clean microscope slide, and proceeding with a light-field count. A blank count of an unused filter disc from the same batch of filters must be made. Distribution of an opaque chromite ore dust throughout the filter is shown in Fig. 2. With this relatively good distribution, only a portion of the filter need be used for counting.

In the direct count of the membrane filter a light dust deposit is necessary. To demonstrate this, consider the following example. If a dust concentration of five million particles per cubic foot is sampled for one minute at 0.1 cu ft/min the filter would collect 500,000 particles. Assuming 0.25 mm² for the ocular grid counting area, the one-inch diameter membrane filter disc contains about 2000 counting fields. This short sample would give an average of about 250 particles per field which is too heavy for

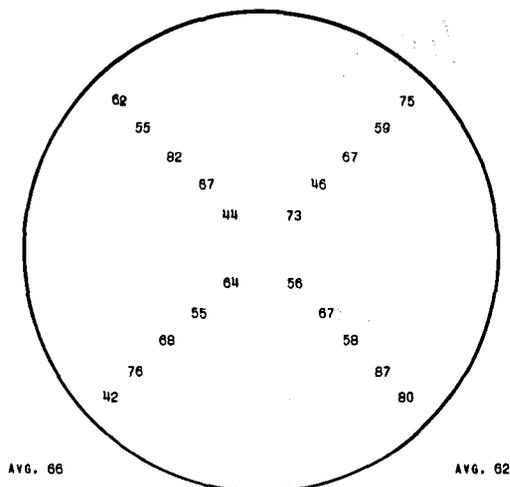


Fig. 2.
Distribution of dust on the membrane filter.

accurate counting. The standard procedure for counting dust using a Dunn cell, adopted by the American Conference of Governmental Industrial Hygienists in 1942, suggests a maximum of 150 particles in a 0.25 mm² field. The same value applies when using the membrane filter. Direct counting of the filter is better adapted to short or grab samples. Because of this limitation it is difficult to compare the impinger count and membrane filter direct count on simultaneous samples.

We have found that small particles of quartz dust with refractive index of 1.54 and diatomite dust with refractive indices of 1.43 and 1.48 will not all be counted because these refractive indices are so close to 1.5 that the dust becomes transparent. Illumination of the top of the filter without oil does not solve the problem since many of the small particles are lodged in the pores of the filter. We have tried staining the immersion oil with an organic dye to bring out the smaller particles but our experience with this technique has not been very encouraging.

DUNN CELL COUNTING: If the refractive index of the dust is too near 1.5, or if the dust deposit is too heavy, the filter can be dissolved in dust free acetone or cellosolve obtained by redistilling the solvents. The count may then be made in a Dunn cell following the usual light-field technique.

Blank counts of filter discs from the same batch are necessary. Comparison counts were made by three observers on samples of opaque dust taken simultaneously with a membrane filter and midget impinger. The direct count values of the membrane filter were made on a short sample taken at the mid-point of the other samples. The results, shown in Table I, indicate a good agreement between the methods of counting as well as among observers.

TABLE I.
DUST COUNTS BY DIFFERENT METHODS AND OBSERVERS IN MILLIONS OF PARTICLES PER CUBIC FOOT OF AIR

Observer	Impinger	Direct Count	Cellosolve
A	4.0	3.3	4.3
B	4.6	3.4	4.2
C	3.1	3.7	3.9

PERMANENT RECORD OF SAMPLE: Another technique developed in our laboratory gives a permanent record of a dust sample. A cell is prepared by fastening a clean membrane filter disc to a microscope slide with a notebook reinforcement ring.¹⁰ An aliquot, 0.01 ml, of a suspension of dust, obtained by dissolving a membrane filter sample in acetone, is placed in the center of the reinforcement ring cell. The acetone evaporates within seconds leaving a transparent film holding the dust in random distribution throughout the cell. This technique apparently had good possibilities but blank counts on unused filters were found to be quite high and variable; so further investigation on this project has been temporarily suspended.

PARTICLE SIZE: The membrane filter is especially adaptable to the determination of particle size by the optical microscope because the particles are collected and sized as they originally appeared in the air. The membrane filter is placed dust side down on a microscope slide and immersion oil is added. The oil immersion (97X) objective can be positioned in the oil. For sizing we use a Porton Graticule in a 15X ocular which yields results comparable with the flar micrometer.

The refractive index of the dust to be sized must be considered as in counting. We found that less difficulty is encountered with this problem when high magnification is used. With different depths of penetration

of the dust into the membrane filter the focus of the microscope must be varied to find and size all the particles. We have had difficulty when sizing a heavy sample with relatively large particles in that the immersion oil tends to cause the particles to migrate to the periphery of the oil drop. In this manner the large particles are usually assembled at one part of the slide and the size determination is biased. To prevent this we are now experimenting with the use of a cellulose base cement* instead of immersion oil. The dust-free cement is applied to the filter, placed dust side up on a microscope slide. A cover slip is then pressed down tightly on the filter to form a permanent slide. Sizing may be done at any time using immersion oil on the cover slip.

Particle Size with Electron Microscope

THE MEMBRANE filter with its high collection efficiency for small particles would appear to be an ideal medium for the collection of air samples for electron microscopy. The filter being soluble in certain organic solvents, makes it possible to remove the filter from the collected sample rather than removing the sample from the filter. Several techniques have been suggested for the preparation of membrane filter samples for electron microscopy. First and Silverman⁷ suggested floating the filter face up on the surface of acetone. The filter slowly dissolves until only a thin film remains. This film is supported on a 1/8-inch diameter, 200 mesh screen and is ready for observation in the electron microscope. Although quite simple, this technique is extremely delicate, the timing is critical and the dissolution of the filter must be stopped at the instant before the film completely disintegrates.

Kalmus¹¹ used a substrate of Formvar® to coat the 200 mesh grid. This material is insoluble in acetone while the cellulose esters of the filter are quite soluble. The Formvar coated grid is placed on a piece of filter paper in a petri dish. A small square of membrane filter is placed dust side down on top of the grid. A few drops of acetone are applied to the filter to wet the strip and also the membrane square. More ace-

*DuPont's Duco Cement was used and found satisfactory. Other cements of this type were not tested.

tone is added and the dish is covered. The filter in contact with the saturated paper strip and bathed in a saturated vapor of acetone, is completely dissolved after an eight-hour period. After drying the Formvar film in a vacuum and sterilizing it under ultraviolet light, the specimen is ready for examination in the electron microscope. According to Kalmus it is possible to shadow the specimen with a metal to increase contrast and visibility of the smallest particles. We have found that the particles usually retain a film of gelatinous filter material and the shadows while clear are not usually representative.

We have worked out a method¹² which is similar to that of Kalmus but considerably faster. Ethyl acetate is used as a solvent instead of acetone. The membrane filter sample is placed dust side up on an electron microscope grid coated with Formvar which rests on a coarser 20-mesh stainless steel screen. The screen is placed in the top row of a three-row spot testing plate. Ethyl acetate is added to the depression in the spot plate until the liquid barely wets the membrane filter fragment. The spot plate is then covered with a flat piece of glass. After 10 minutes, the stainless steel screen supporting the electron microscope grid and filter residue is transferred to a clean depression in the spot plate and fresh ethyl acetate added. This washing is repeated after 10 minutes with a final ethyl acetate wash of five minutes. Finally, the stainless steel screens are removed with tweezers and placed on a clean filter paper to absorb the excess ethyl acetate. After drying either in a partly covered petri dish or in an oven at about 85° C, the specimens are ready for observation in the electron microscope. It has been found that if the washing period is insufficient, a background of undissolved filter will remain on the Formvar, while if the wash period is too long, the film will be partially dissolved. The exact timing of the solution period must be ascertained by experiment and may vary from sample to sample. However, it is a relatively simple and quick means of producing acceptable pictures of airborne dust particles. This method has been used to examine particles of cobalt fume, vanadium oxide, granite, uranium oxides, fluorides, diatomite, and other materials.

Chemical Analysis

USE of the membrane filter to collect samples for chemical analysis can simplify the analytical techniques required. The resistance of the filter to strong acids and bases makes it possible to leach the sample with strong reagents without disintegration of the filter. The solution of the sample can be decanted from the filter and a tedious filtration or ashing step eliminated.

Cobalt fume has been collected in our laboratory on the membrane filter and recovered in solution by wetting the filter in a test tube with about 50% HCl and then adding a few drops of HNO₃. The solution was then made up to volume in a volumetric flask with the subsequent washings from the filter. An aliquot of this solution was treated with nitroso-R-salt reagent and the color was read in a spectrophotometer.

Airborne vanadium pentoxide dust was also collected in this way and dissolved from the filter with 10% NaOH. When the solution was decanted from the filter, the vanadium was readily determined colorimetrically.

The resistance of the membrane filter to strong hydrofluoric acid is the basis of a simplified method for the determination of silica in air-borne dusts.¹³ The silica is dissolved from the filter with hydrofluoric acid and the resulting solution analyzed colorimetrically for silica.

X-Ray Diffraction and Spectrographic Analysis

MODERN spectrometer types of x-ray diffraction and spectrographic instruments require a flat sample which the membrane filter provides. In our laboratories the filter is clamped by the edges in a sandwich-type holder. A completely non-destructive determination of the crystalline compounds and major elements present in the sample is possible with one-third the background obtained with samples smeared onto microscope slides. Full-scale recorder response of the diffraction lines of quartz, cristobalite and tridymite can be obtained with less than 1 mg on the filter. Fig. 3 shows the recording of the principal diffraction line of quartz and cristobalite.

Sensitivity to a few micrograms of the major elements of a sample is possible by the x-ray spectrographic technique when the sample is evenly distributed over the

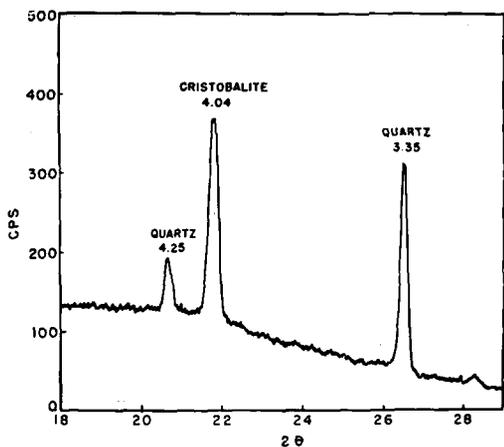


Fig. 3. Intensity of principal x-ray diffraction lines of quartz and cristobalite given by one milligram of each on a membrane filter.

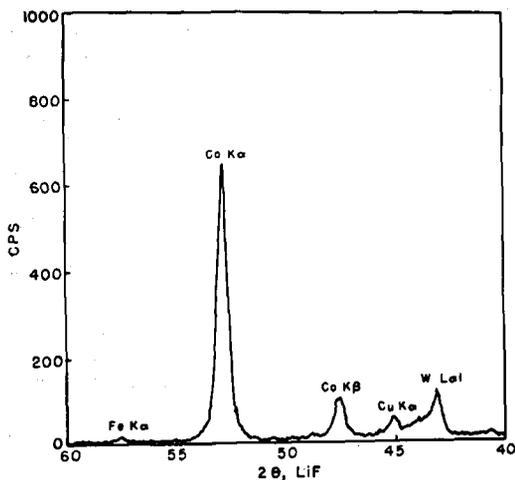


Fig. 4. Intensity of principal fluorescent wavelengths of cobalt given by 180 micrograms on a membrane filter.

surface of a membrane filter. A portion of the x-ray fluorescence spectrum of a sample collected on a membrane filter is shown in Fig. 4.

Size Analysis by Photometric-Sedimentation

BURKE¹⁴ has shown the utility of the membrane filter in sampling dust for particle size analysis by the sedimentation method. Size analysis can be carried below 0.5 micron with an improved instrument designed

in our laboratory.¹⁵ The membrane filter is dissolved in cellosolve and the size analysis obtained by following the sedimentation of the resulting suspension with a recording photometer.

Reflectance Photometry

PARTICULATE matter deposits more uniformly on the membrane filter than on filter paper. Since most of the particles are at the surface and because of higher initial reflectance, the membrane filter gives a higher sensitivity than filter paper, whether the concentration of the contaminant is determined by visual comparison with a series of standard stains (Fig. 5) or by reflectance photometry. Fig. 6 shows the difference in reflectance readings of filter paper and membrane filter samples of vanadium pentoxide dust. The membrane filter curve shows much less scatter of points and consequently is the more reliable.

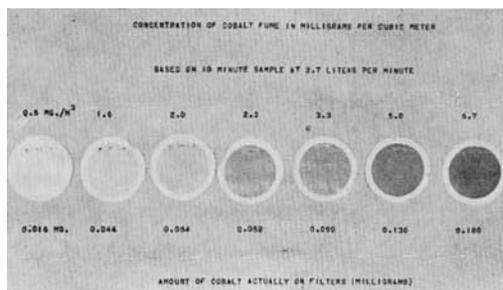


Fig. 5. Visual comparison of cobalt fume samples.

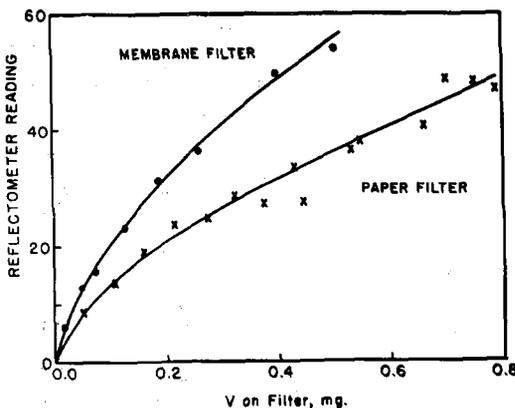


Fig. 6. Reflectance readings of vanadium pentoxide dust on filter paper and vanadium membrane filter.

One detracting feature of the membrane filter when used in this way for dust determination is its high efficiency for smoke and fume particles. A small amount of smoke may prevent accurate interpretation of the dust stain.

Metallic Constituents

HIGHLY sensitive spectrographic analyses of samples collected on membrane filters should be preceded by a determination of the metallic constituents present in the filter material. The tacit assumption that these filters are mineral free is one which is untenable on the basis of spectrographic analysis of three types of membrane filters.

We have analyzed, spectrographically, filters of the following types: (1) type AA, white, plain; (2) type AA, black, plain; and (3) type AA, white, grid (ink ruled). Sets of five or 10 filters were wet ashed with concentrated nitric acid which had been redistilled in an all-glass still. The ash was dissolved in 0.5 ml of redistilled hydrochloric acid and double distilled water. The solutions were analyzed by a spectrographic procedure described elsewhere.¹⁶ Semi-quantitative estimations of the constituents were made by a visual comparison of sample and standard spectra in a spectrum comparator except where otherwise indicated. These results are presented in Table II. It is apparent that certain filter constit-

uents occur in concentrations which should be evaluated before using a given type of filter for atmospheric sampling and subsequently for spectrographic analysis. Sampling at the threshold limit level for lead (0.15 mg. Pb/m³) at a rate of 0.1 cu ft/min for 10 to 30 minutes would result in the collection of four to 12 micrograms of this element. These amounts would be difficult to determine in a matrix whose lead blank is as high as those reported for the grid and the black filters. For the same reason, the grid type of filter does not appear suitable for collecting chromate dust at concentrations in the threshold limit region (0.1 mg CrO₃/m³).

For general sampling of toxic dusts and fumes, the plain white filter appears to be most desirable from the standpoint of a minimum of heavy metal contamination.

Summary

THE AEROSOL type membrane filter with its high collection efficiency for very small particles has proven to be very useful in environmental studies of particulates. Use of the membrane filter for the collection and quantification of dust has the added advantage of simplicity over some of the methods used in the past. Light dust deposits may be counted directly on the filter whereas heavy deposits require the solution of the filter in a suitable solvent followed by standard dust counting procedures. Particle size of dust collected by this medium has been studied by the optical microscope, the electron microscope and by photometric sedimentation. With the optical microscope a limitation of the filter with respect to the refractive index of the dust must be recognized.

An accurate and rapid method for the transfer of dust from the membrane filter to an electron microscope grid allows one to take full advantage of the high collection efficiency of the membrane filter for very small particles. Removal of microquantities of toxic dusts and fumes from the filters by simple procedures has aided chemical analysis of cobalt fume, vanadium pentoxide and free silica. Blank determinations of metal constituents present in the filter may be necessary in view of spectrographic analysis of three types of aerosol filters. Studies of dust concentration by reflectance pho-

TABLE II.
SEMI-QUANTITATIVE SPECTROGRAPHIC ANALYSES
OF MEMBRANE FILTERS
(Expressed as Micrograms per Filter)

Filter type Diameter, mm	AA, White, Plain	AA, White, Grid	AA, Black, Plain
	32	47	29
Element			
Al	0.4	1.3	0.3
Ba	n.d. ††	1.4	n.d.
Bi	0.002	n.d.	n.d.
Ca	60.	0.8	1.0
Cd	0.09	n.d.	n.d.
Co	0.02*	n.d.	n.d.
Cr	0.0 †	38. ‡	0.4 †
Cu	0.6	1.1	2.0
Fe	0.1	0.7	24.
Mg	1.2	0.4	0.6
Mn	0.1	0.5	0.3
Mo	n.d.	n.d.	0.4
Pb	2.3	112.	10.
Sn	0.02	0.08	0.1
Sr	n.d.	trace	n.d.

* Quantitative Spectrographic Method

† Diphenylcarbazide Method

†† None Detected

tometry demonstrate the advantage of the membrane filters over filter paper in this application.

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Symposia on Industrial Health Problems

A FULL-DAY session on major industrial health problems will be held on November 14, 1957, during the Annual Meeting of the American Public Health Association. The symposia will be co-sponsored by the Occupational Health Section of the American Public Health Association, the Industrial Medical Association, and the American Industrial Hygiene Association. Attention will be focused on the extent of knowledge on several outstanding industrial health problems, their importance in magnitude and scope, and what needs to be done to promote further progress in their control. Recognized authorities will participate in symposia on (1) Problems Resulting from the Use of Habituating Drugs in Industry, and (2) Status of the Pneumoconioses. Other programs of interest to occupational health personnel, such as air pollution, radiation protection, and impact of health services on workers and their families, will also be included at the Annual Meeting of the American Public Health Association, to be held in Cleveland, November 11-15, 1957. The complete program for the Annual Meeting will be found in the September issue of the Journal of the American Public Health Association.



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