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AN APPLICATION OF FLUORESCENT PIGMENT TO THE MEASUREMENT OF PARTICLE INHALATION PROBABILITIES

By

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AN APPLICATION OF FLUORESCENT PIGMENT TO THE MEASUREMENT OF PARTICLE INHALATION PROBABILITIES

INTRODUCTION

During the early months of 1954 the frequency with which radioactive particles were found on and in project vehicles caused some concern that workers may be breathing significant numbers of particles while using or servicing these vehicles. It became necessary to establish a control limit for the number and levels of active particles which could be permitted. Two elements entered into the evaluation of the potential hazard and selection of control limits. One was the actual consequence of inhaling a radioactive particle. Insofar as the maximum number of a given activity which could be permitted in the lung little more than opinions of informed individuals were available. The second element was that of the probability of an individual breathing a particle from a vehicle during operating or servicing. Need for precisely defining the consequence from breathing a particle would diminish as the probability of breathing the particle could be shown to be extremely small; and limits could be established with considerable weight given to the probability of an individual breathing a particle.

This work was undertaken to obtain a relatively rapid estimate of the likelihood of a particle being inhaled during certain typical operations involving vehicles. A principal object of the report, however, is to present a sensitive method for tracing the movement of particulate contamination.

SUMMARY

Zinc sulfide powder with a median diameter of about two μ was used to determine the probability of a single radioactive particle being transferred to the air and breathed in several operations involving operation and maintenance of sedans and busses. A known number of particles were

deposited with soil normally present, then the operation giving rise to a potential exposure performed. The number of ZnS particles found on respirators worn or collected on filters used to sample the air during the operation was determined using a microscope and ultraviolet illumination. The probability of breathing a single particle when present was found to range from 6×10^{-7} per hour for driving with minimum air turbulence in the vehicle, to 6×10^{-4} per hour for sweeping the dirt from a bus. Other operations and probabilities are discussed.

GENERAL METHOD

Fluorescent powders have been used to trace the movement of air and to measure the diffusion of fine particles and gases in the atmosphere.[1][2] The particular virtues of a material such as ZnS for this application are its distinctive fluorescence under ultraviolet light and its non-toxicity. Because one particle of ZnS can be detected among hundreds of ordinary dust particles very small concentrations can be determined in a relatively large dust background. The powder can be obtained in a particle size distribution which for many applications provides a satisfactory tracer for the actual particles in question. In the application to be discussed ZnS powder of about two μ median diameter was obtained from the New Jersey Zinc Company, New York. This particle size effectively covers the range of particles known to be carried deep into the lung. The particle density of 4.1 may be somewhat higher than for many of the radioactive particles, say those carried with ammonium nitrate; but is believed to be near enough to the density of the average particle to be a useful substitute.

In the experiments the general approach was to place a known number of ZnS particles in a location known to be susceptible to contamination, such as floorboards, or on tires. The vehicle was then serviced or operated for a period during which air samples were taken from a location near the operator's head, or a respirator was worn by the operator.

After an operation was completed or after a given time interval, the filter used for sampling the air or the respirator filter was examined microscopically under ultraviolet light and the number of particles present determined. After normalizing the sample to 0.5 cfm the fraction found on the filter of those initially present was taken as the probability for inhaling a particle in the given interval.

EXPERIMENTAL EQUIPMENT

A 100X monocular microscope, Gaertner, M-101-A, fitted with a wide field ocular was used for all the particle counting work. A standard type ultraviolet illuminator, Central Scientific Supplies Company, No. 85270, was used. A Whipple disc calibrated with a stage micrometer was used in the eyepiece to delineate a known area of the field. The microscope proved to be easily adapted to use on irregular objects, such as a tire mounted on a sedan. For this type of microscopic examination a cathetometer with the telescope replaced with an adapter to accommodate the microscope support proved very satisfactory. This arrangement is shown in Figure 1.

Air samples were taken with Trico vacuum booster pumps which were operated from a 6-volt battery, and were originally assembled for meteorological studies.(3) For air samples taken while driving a sedan, the vacuum pumps were plugged into the cigarette lighter socket using an adapted trouble-light plug. Membrane-type filters obtained from the Lovell Company, Watertown, Mass., and H-70, 0.018" filter papers manufactured by the Hollingsworth and Vose Company, East Walpole, Mass., were supported in filter holders made from one-half inch pipe unions. A rotameter was used to measure flow rates. Figure 2 is a photograph of this equipment.

American Optical Company respirators R2000 were used in some of the tests. The respirator felt disc furnished was replaced with an H-70 filter of equal diameter. Breathing through the latter filter required somewhat more effort because of the increased resistance.

FIGURE 1
MICROSCOPE, SUPPORT, AND LAMP USE
DETERMINE PARTICLES ON TIRE

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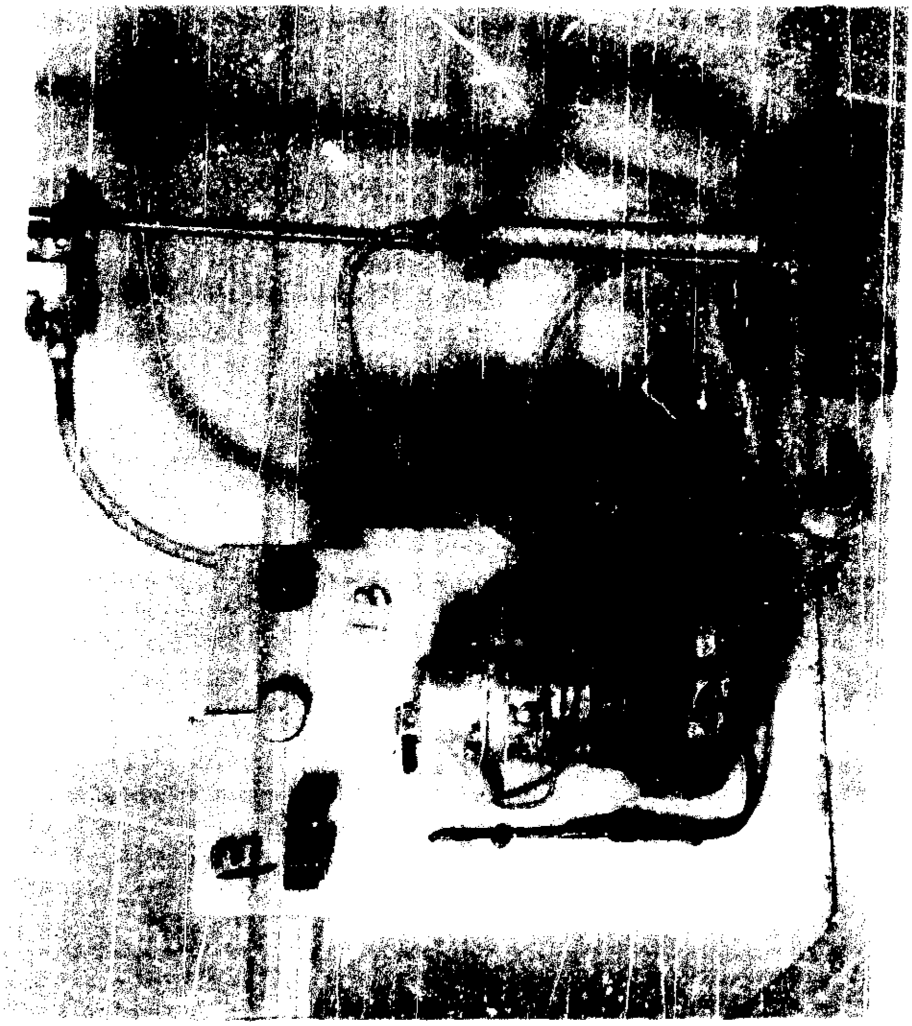

FIGURE 2
AIR PUMP, FILTER SUPPORT, AND FLOWMETER

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MEASUREMENT OF PARTICLES PER UNIT WEIGHT OF ZnS

It was first necessary to determine the number of "observable" particles per gram of ZnS. Since a magnification of only 100X was used, particles below an estimated one μ were not seen. The fluorescence of even very small particles makes detection possible at this magnification. "Numbers of particles of ZnS," when used in the remainder of the report, will refer to those observable with the equipment used. The number of particles per gram was determined by standard techniques. An accurately weighed sample of the material was dispersed in a known volume of water, mixed thoroughly and a portion of the suspension quickly transferred to a Dunn dust counting cell. The liquid was immediately leveled by placing the cover plate over the cell, and the particles in the 1.0 mm depth of liquid allowed to settle for 30 minutes. The slide was illuminated with ultraviolet light and the number of particles per sq/mm determined by counting using 100X. Knowing the volume from which the particles had settled and consequently the aliquot of the original sample, the total number of particles per gram was determined.

Three determinations gave the following number per gram:

a. $1.29 \pm 0.03 \times 10^{10}$

b. $1.40 \pm 0.02 \times 10^{10}$

c. 1.[illegible] ± 0.05 x 10^{10}

Avg. $1.40 \pm 0.09 \times 10^{10}$

The error listed is the standard deviation of the mean of ten fields read in (a) and five fields read in (b) and (c), and gives an index to the reproducibility of this measurement, which was believed to be satisfactory for the intended use of the data.

COMPARISON OF FILTER MEDIA

Some tests were made to determine the relative merit of H-70 paper and the membrane filters for collecting and for counting the number of particles present. An accurately weighed amount of ZnS was diluted with a known weight of talc. The two materials were mixed by turning the container end over end about 20 times a minute, for 24 hours. Five milli-gram portions of the mixture were weighed into small gelatin capsules. The filter to be tested was placed in a support and about 0.5 cfm flow maintained. A gelatin capsule containing the diluted ZnS was pierced with a very small hole near the bottom of the smaller half of the capsule which was then inserted snugly into the end of a Tygon tube, forming a tight plug except for the very small pin hole in the capsule. The upper half of the capsule was withdrawn, the capsule held close to the filter support inlet, and by very careful air pressure applied to the end of the tube, the talc and ZnS were gently introduced in small quantities to the air stream flowing through the filter. Only minor amounts adhered to the inlet walls, and the emptied capsules were reweighed to determine the actual weight of ZnS and talc transferred.

The filters were carefully removed and the number of ZnS particles counted under ultraviolet illumination using 100X. The number thus determined was compared with the number present calculated from the particles per gram and the weight of ZnS known to be delivered to the filter. Since only 5 mg was taken to avoid excessive shielding by the inert material, a weighing error of about ± 5 percent was introduced, and though spreading on the filters was reasonably uniform, undoubtedly some error was introduced from this source, and from shielding by the diluent talc.

Table I presents the data obtained:

TABLE I

RELATIVE MERIT OF MEMBRANE AND H-70 PAPER FOR COLLECTION AND COUNTING ZnS PARTICLES

Determination	Paper	Particles on filter as Determined by wt. of ZnS	Particles on filter by counting
1	Membrane	4.5×10^4	3.9×10^4
2	Membrane	2.8×10^4	2.5×10^4
3	Membrane	4.0×10^4	3.9×10^4
4	Membrane	5.7×10^4	4.4×10^4
5	H-70	2.3×10^4	2.8×10^4
6	H-70	3.2×10^4	2.8×10^4

The data show that on the average 87 per cent of the calculated number of particles present were found when using the membrane filter, which, considering some of the uncertainties mentioned appeared satisfactory for the precision required. A satisfactory collection and reading efficiency was shown for the H-70 paper, but because of the ease with which the membrane filters were counted, these were used in preference to the H-70. In some experiments both were used to get additional comparative data. (Since completing this work it has come to the author's attention that black membrane filters can be obtained, which would be particularly useful in this application because of the contrast which would result.)

TRANSFER FROM FLOORBOARDS AND SUBSEQUENT INHALATION DURING DRIVING

A one gram portion of ZnS was mixed with the dirt normally found below the brake pedals of a sedan. The sedan was driven for approximately one hour during which an air sample was collected adjacent to the operator's head at approximately nose level. The number of particles found on the filter was determined using the 100X microscope and ultraviolet lamp. A test was made during minimum air flow through the car, that is, all windows and vents were closed. Other tests were made with the hood ventilator open and the side windows open resulting in much turbulence in the front seat region. The fraction of the total number of particles which had been collected by the filter was determined. Table II gives a summary of these probabilities.

TABLE II

PROBABILITY OF INHALING ONE PARTICLE INITIALLY PRESENT ON THE FLOOR AREA OF A SEDAN WHILE DRIVING FOR ONE HOUR

With no ventilation in vehicle	6×10^{-7}
With high air flow through vehicle	(a) 1.3×10^{-6}
" " " " " "	(b) 2.4×10^{-5}
" " " " " "	(c) 2.3×10^{-6}

Run (c) was made with the same vehicle used for run (b) the day before and contained the original particles. The higher probability obtained in (b) is believed to result from particles airborne during the placement of the particulate material, and collected early in the hour's run. Since the degree of turbulence could have been quite different in the runs, it might be expected that rather large differences in probability would be found. The data are useful in giving an order of magnitude of probability which was adequate for the application.

TRANSFER AND INHALATION DURING SWEEPING FLOORBOARDS

An operation performed quite frequently is that of broom sweeping the front floor area of a vehicle. Since the air is stirred up vigorously it might be expected that exposure risk would be high. Two filters were used to draw air samples and a respirator worn by an individual who spent about four minutes sweeping the front floorboards of a car on which had been distributed one gram of ZnS. Resulting probabilities of inhaling a single particle during an hour of sweeping were determined as described and found to be:

From particle count on respirator filter . . . . $6.4 \pm 0.4 \times 10^{-4}$
From particle count on H-70 filter sample . . . $3.3 \pm 0.2 \times 10^{-4}$
From particle count on membrane filter sample . . . $6.2 \pm 0.3 \times 10^{-4}$

Satisfactory agreement was obtained using the three media for collecting when it is recognized that a nominal breathing rate of 0.5 cfm was assumed and to which the filter sampler flow rates were normalized.

TRANSFER AND INHALATION DURING SWEEPING A BUS

Five similar experiments were performed in which 41-passenger busses were swept by the bus operator in the usual manner. One gram of ZnS was mixed with dirt and distributed on the floor throughout the bus. Air samples were collected during the subsequent sweeping operation, which took about five minutes. The number of particles per filter was determined and normalized to a flow of 0.50 cfm for one hour's sweeping time.

The probability of breathing a single particle per hour of sweeping was determined in these five experiments to be $4.8 \pm 0.05 \times 10^{-4}$. The error indicated is the standard deviation of the mean for the five determinations.

TRANSFER AND INHALATION DURING A TIRE REPLACEMENT

Two experiments were performed to obtain an estimate of the probability of inhaling a given radioactive particle present on a tire during a tire replacement. Since rather energetic handling and manipulation are required in changing a tire and since the mechanic is always in close contact with the tire, it was believed that a fairly high inhalation probability might exist.

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Zinc sulfide was thoroughly diluted with talcum in the ratio of one part of sulfide to five of talcum. This mixture was applied to the front tire of a sedan by uniformly sprinkling the powder over a strip of paper placed in the path of the front wheel, then driving back and forth over the paper thus prepared. The vehicle was then driven immediately forward into a building whose floor was covered with clean paper. Using the microscope and light arrangement shown in Figure 1, the number of ZnS particles was determined by counting in representative known areas of the tread. The total area was determined by planimetering an imprint of the tread over a known fraction of the circumference. It was recognized that particles may lodge in the grooves as well as on the surface of the tread. Using the contact tread surface only in determining the number of particles present would result in a higher than the actual probability of exposure, since fewer particles would be counted on the tire than were actually present. After the number of particles present had been determined, a tire mechanic changed the tire. This operation consisted of raising the car with a jack, removing the wheel, dismounting the tire from the rim, deflating the tube, removing the tube, examining the casing. The reverse procedure was then followed which left the tire and wheel mounted on the vehicle. The mechanic wore a respirator fitted with an H-70 filter, and 0.5 cfm air samples were taken from a location adjacent to the mechanic's head during this operation which required about 20 minutes. The number of particles collected on the respirator and air sampler filters was determined as already described, and the probability of a single particle being inhaled calculated. Results are reported in Table III.

[redacted]

TABLE III

PROBABILITY OF INHALING A SINGLE PARTICLE DURING A TIRE CHANGE

	Test 1*	Test 2**
Total particles on tire	1.7×10^9	1.0×10^9
Particles collected on respirator filter	6.0×10^4	2.0×10^4
Particles collected on H-70 filter	4.0×10^4	3.2×10^3
Particles collected on membrane filter	-	6×10^3
Probability of inhaling a single particle if present	3×10^{-5}	2×10^{-5}

* Tire worn nearly smooth.
** Tire was virtually new.

These experiments were more subject to uncertainty because of inability to determine precisely the number of particles present on the tire. They do permit, however, a reasonably good estimate of the probability of breathing a particle transferred to the air during a tire replacement.

After changing the tire the vehicle was driven for some 10-15 miles and the tire again examined to determine the number of particles remaining. In the first case only 0.0004 of the original number remained; in the other case 0.008 of the original number remained, indicating that the particles were very lightly held. In the actual case of radioactive particulate contamination it is highly probable that a vehicle is driven much further than 15 miles between the time of contamination and tire servicing, hence the probabilities quoted in Table III are very likely higher than in the actual case, if it can be assumed that the remaining particles are more difficult to remove in handling the tire. It was determined that 30-50% of the initial ZnS particles could be removed by pressing Scotch tape against the tire tread, showing the ease with which this powder was removed.

[redacted]

CONCLUSIONS

When the frequency and location of contamination and the number of exposure hours per man are known, it is possible for the operations described to estimate the probability of an individual breathing a radioactive particle. Resulting probabilities for the particular situation faced in early 1954 are reported in reference,[4] and were found useful in setting control limits for vehicle contamination.

The general method should find rather wide application to problems of this kind when it becomes necessary to trace the movement and fate of particulates.

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L. C. Schwendiman

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