NOTICE

**CERTAIN DATA
CONTAINED IN THIS
DOCUMENT MAY BE
DIFFICULT TO READ
IN MICROFICHE
PRODUCTS.**

GEH-14805

DECLASSIFIED

FILE

IV

Route List

1. ~~Washington~~
2. ~~Washington~~
3. ~~Washington~~
4. ~~Washington~~
5. ~~Washington~~
6. ~~Washington~~
7. ~~Washington~~
8. ~~Washington~~
9. ~~Washington~~

DATE Undated

SUBJECT Summary Report on Measurement and Control of Radioactive Particulate Contamination for Engineering Division, Atomic Energy Commission

To File

FROM T. E. Nelson

COPY No. 10-B

BEFORE READING THIS DOCUMENT, SIGN AND DATE BELOW:

Work	8/17/49
President	10/21/49
F. E. Nelson	5-8-50
CMR	5/22/50
HMP	6/12/50
GEH	
DMC	5/24/51

DO NOT CIRCULATE!
REFERENCE COPY

INVENTORIED
APR 24 1950
DOCUMENT AUDIT AND INVENTORY UNIT

INVENTORIED
MAR 31 1953
DOCUMENT AUDIT AND INVENTORY UNIT

INVENTORIED
JUN 3 1952
DOCUMENT AUDIT AND INVENTORY UNIT

INVENTORIED
SEP 28 1950
DOCUMENT AUDIT AND INVENTORY UNIT

DECLASSIFIED MAR 8 1994

DISTRIBUTION RESTRICTED TO U.S. ONLY

OSTI

GEH-14,805

COPIES OF 1, SERIES MA 24741 Undated

This document consists of 21 pages
No. 10 of 57 Copies, Series B

Plus Title Page

25532-1

Summary Report
on
Measurement and Control of Radioactive Particulate Contamination
for

Engineering Division
Atomic Energy Commission

by

Theodore F. Hatch
Theodore F. Hatch, Consultant
Industrial Hygiene Foundation of America, Inc.
Pittsburgh, Pennsylvania

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

declassified
per ROC 1973

Off Bureau 2/22/82

DECLASSIFIED

MASTER

DECLASSIFIED

Measurement and Control of Radioactive Particulate Matter

on

A.E.C. Operations

Protection of plant personnel against radiation and the suppression of outside contamination by solid, liquid, and gaseous radioactive wastes constitute important phases of every AEC operation. In this connection, airborne radioactive particulate matter has been recognized as a separate problem, requiring special monitoring procedures and control measures. On certain projects particulate matter constitutes the major contaminant; on others, it is secondary in importance to other forms of exposure.

Of particular concern for the past year or more has been the area and off-site contamination by particulate activity escaping with gaseous wastes through process stacks and in the waste ventilating air. The discovery of radioactive particles, some of considerable size, deposited on the ground at varying distances from stacks, lead to more extensive inquiry into the magnitude of particulate emission. Measures taken to reduce the discharge of large specks failed to minimize the escape of microscopic particles and it became necessary to install rather large and costly equipment for air-cleaning to care for this additional problem. An Advisory Stack Gas Working Group was set up to review the gaseous and particulate waste disposal program and, at the recommendations of this Group, the present inquiry was undertaken — to review the "particle" problem, its origin and nature, the analytical procedures employed, and the control measures adopted.

Visits were made to the following sites: Oak Ridge National Laboratory, Hanford Engineering Works, Los Alamos Laboratory, University of California Radiation Laboratory, University of Iowa, Brookhaven National Laboratory, Mounds, and Knolls Laboratories. At each site the purpose of the visit was outlined and the local problems having to do with particulate contamination were discussed. Information pertaining to the nature, characteristics, and magnitude of output of atmospheric particulate wastes was reviewed and the analytical procedures and their limitations were discussed in some detail. Control measures to suppress particulate matter already in service and in the process of design or construction were also reviewed. So far as practicable, actual operations and installations were inspected. Discussions were held mostly with representatives of Technical Divisions, concerned with design of processes and of control measures, and with Health Physics specialists responsible for appraisal of exposure to radiation in the plants and outside.

Broadly, the purpose of this report is to review the characteristics of particulate contamination which set it apart from the other phases of the radiation control program and make it necessary to apply to it different concepts, analytical procedures, and control measures. The problem is treated as one in the appraisal and control of dusts and fumes. The fact that the material is radioactive introduces certain new details, but this does not make the problem basically different from others in the field of industrial dust control.

The report is divided into four parts: (a) discussion of the unique characteristics of particulate contamination, as distinguished from other problems of radiation exposure, and its similarities and differences compared with other industrial dust and fume problems; (b) requirements for proper appraisal of exposure to particulate contamination and for measurement of effectiveness of control; (c) consideration of the basic principles of control of particulate matter and the unusual problems arising here; (d) overall summary and recommendations.

I. RADIOACTIVE PARTICULATE CONTAMINATION

A. ATMOSPHERIC TOLERANCE LEVELS

In the calculation of tolerance levels for radioactive substances it is recognized that certain modifying factors must be considered when the radioactivity is introduced into the body in inhalation of particulate matter. The question is raised as to whether or not a different hazard is involved when a relatively insoluble substance is thus deposited in the lungs to form concentrated sources of radiation in surrounding tissue, compared with the intake of soluble compounds which are quickly distributed in molecular dispersion. In the consideration of this question, it is instructive to convert tolerance levels, which are commonly expressed in terms of activity or mass concentration values in the air, into equivalent numbers of particles. For this purpose and for convenience of reference to published data, the figures given by Morgan (K. Z. Morgan: Tolerance concentrations of radioactive substances. Jour. Phys. and Colloid Chem. 51, July, 1947) are used. Assuming a mass-average particle size of 1μ and particle density of 7, the activity and mass concentration and equivalent numbers of particles (approximate) for various pure substances are as follows:

<u>Substance</u>	<u>Atmospheric Tolerance Level, per m³ of air</u>		
	<u>Activity</u> <u>/cc</u>	<u>Mass</u> <u>/μg</u>	<u>Count (1 μ)</u> <u>No. particles</u>
Natural uranium	1.5×10^{-4}	2×10^2	10^8
Enriched uranium	1.4×10^{-4}	3.8	10^6
Graphite	1.2×10^{-1}	2.6×10^{-2}	10^4
Plutonium	1.3×10^{-4}	2×10^{-3}	10^3
Radium	2×10^{-4}	2×10^{-4}	10^2
Polonium	6.4×10^{-4}	1.4×10^{-7}	10^{-1}

These tolerance values are calculated on the assumption that a given fraction of the substance taken into the body is uniformly distributed throughout the controlling body organ and that the resulting concentration per unit weight of that organ produces radiation that is within tolerable limits. Even when the substance is in solution this assumption may not be correct, as Morgan points out. In the case of relatively insoluble material deposited as dust particles in the lungs, the assumption is clearly not valid and may, therefore, introduce an error of some significance in the calculations. Consider, for example, the particulate concen-

tration of plutonium in the lungs. With a daily intake at the tolerance level of 1000 particles/ m^3 , the total number of particles in the lungs under equilibrium conditions would be approximately 10^9 , distributed in more than 10^8 alveoli. Even if it is assumed that there is no more than one particle per alveolus and neglecting the possible concentrating of particles after initial deposition, it is evident that the radioactive substance would be distributed in less than 1% of the total lung volume. With larger particles the situation is more marked; only 100 particles 2μ in diameter are required to fill the daily quota and one 10μ particles will provide an equivalent mass concentration. On the other hand, 10^6 particles per m^3 with a mass-mean size of 0.1μ are required to produce the tolerance dose. This concentration will result in 10^9 particles in the lungs at equilibrium and, consequently, the radiation will be more uniformly distributed. These calculations are not to be taken literally; they are presented, however, to emphasize the physiological difference between particulate concentration and molecular dispersion of radioactive substances taken into the body and the significance of variation in particle size of the particulate material.

The calculated tolerance levels are also based upon the assumption that 25% of the inhaled particles are retained in the lungs. This is probably a minimum retention figure for those particles which are fine enough to penetrate to the alveoli before deposition ($<2\mu$); the possible error is not great, however, since maximum alveolar retention is not more than 60-70%. Relatively coarse particles, greater than $2-3\mu$ in size, on the other hand, have a low probability of penetration to the alveoli before deposition so that, on the average, the assumed alveolar retention of 25% is too high for such particles. One 5μ particle, however, will carry 125 times as much activity as a 1μ particle; this may more than offset its lesser probability of reaching the alveoli before deposition. If it does become deposited in the lungs, the local radiation around it as a source will be of much higher intensity and, therefore, it may be more hazardous than the equivalent deposition in a larger volume of tissue of 125 particles 1μ in size.

It is not known how these conflicting factors add up to determine the variation in toxic effect with differences in size of radioactive particulate substances. It seems clear, nevertheless, that the hazard of particulate activity is sensitively related to particle size and, for an adequate understanding of the problem, it is just as essential to know how particle size varies from one exposure to another or with time at a given location as it is to know the total atmospheric concentration of activity.

The point is emphasized because there is increasing evidence that neglect of this factor has limited progress in the study and appraisal of other industrial dust and fume exposures. In the practical control of silica dust it has been possible to ignore variations in size, perhaps, because of the relatively gross concentrations which are tolerated. On the other hand, it has been a serious omission in the study of lead and more recently of beryllium which involve lower concentrations, although much higher than are considered here. The objective of control on AEC projects is to establish and maintain safe exposures from the outset and not after damage has occurred. Research on the influence of the size factor is clearly indicated but, in the meantime, appraisal of exposures cannot be regarded as adequate without including a study of particle size characteristics. The knowledge of this aspect of the problem is insufficient at the present time.

B. SAMPLING AND MEASUREMENT OF RADIOACTIVE PARTICULATE CONTAMINATION

It is assumed, in the choice of an atmospheric sampling instrument and method of analysis of the collected material, that the results provide a direct measure of the rate of intake and the toxic effect of the contaminant. If there is any variation in the relationship between the amount of contaminant in the air and its physiological effect, the technique of measurement must reveal it. The foregoing discussion of tolerance levels suggests that, in respect to radioactive particulate matter, differences in particle size may greatly alter this relationship. A proper appraisal of exposure must, therefore, provide information on the distribution of the radioactivity in relation to particle size as well as on the total concentration of activity. Methods of sampling and analysis now in common use on AEC operations were reviewed from this point of view and in the following paragraphs, the requirements for additional techniques are considered.

(1) Present methods.

The method of sampling in common use on AEC operations is by filter. It has a number of advantages — high efficiency of collection over the size spectrum, simplicity, ease of handling, and a high volume sampling rate, if needed. Measurement of the collected material is also relatively simple since the activity on the filter is determined by direct counting. It has a fundamental disadvantage, however, in that it does not collect the material in a convenient manner for determination of the particle-size characteristics of the dust. Some attempts have been made to determine the number and size of radioactive particles on the filter by direct radiosautography but not with great success. Individual particles are disclosed by dark spots on the film and the amount of activity per particle is indicated by the relative size of the spots. Differences in specific activity, however, and varying depths of penetration of the particles into the filter, also influence the size of the darkened spots. If the exposure is continued long enough to reveal fine particles there is danger of overshadowing by the greatly enlarged spots produced by large particles and by the general background darkening of the film. At Los Alamos, it has been possible to determine the size of plutonium particles by direct microscopic counting of the number of alpha tracks arising from each point source on the film. Calculations show that particles as small as 0.2μ can be detected with four days exposure. The method is laborious, however, and requires several different periods of exposure to give countable results for particles of different sizes. It is limited, of course, to the study of alpha-radiating materials of known specific activity.

In addition to its limitations in particle size studies, the filter collector has two other disadvantages. (1) It may capture by adsorption fission-products of near-molecular size which are not properly included in a study of particulate activity. (2) Direct measurement of alpha activity on a filter may be subject to error on account of the variable absorption of radiation within the filter, since with differences in particle size and dust loading there will be a variation in depth of penetration of the particles into the filter. This is particularly true of thick, relatively porous filter media like HV and CNS papers. It is not known how great this error may be. At present a constant correction factor is applied which assumes 30% loss by absorption of alpha radiation. Eisenbud (New York Directed Operation) reported this much loss for relatively coarse uranium dust collected on Whatman 41,

a hard-surface paper. It does not seem reasonable, therefore, to use the same correction for CWS paper and finer particles. Variation in alpha absorption should be considered, for example, in the measurement of performance of an air-cleaning device. Not only is the downstream concentration greatly reduced in such a test (resulting in less surface deposition), but the size of the escaping particles is also reduced. Both changes will increase the depth of penetration into the filter; hence, without a corresponding change in the correction factor, the apparent efficiency will be too high. Recalling that so few particles are required to give significant activity levels, it is evident that there may be considerable variation in the correction factor. A study of the variation in filter absorption with type of filter, dust loading, particle size, is planned in the New York Directed Operations Laboratory.

ARK
note

At Oak Ridge some size separation of the dust collected from the pile-cooling air duct was effected by preceding the CWS filter with an Aerotec cyclone. This device has a high efficiency down to about 5 μ . It serves, therefore, only to remove the gross material from the hygienically significant particles and yields little information of value for the appraisal of hazards. Sampling at Hanford by impingement on fibres of different sizes, according to Langmuir's principle, provided more detailed information on the finer sizes.

(2) Sampling requirements.

The following characteristics are desired in the sampling method selected for analysis of radioactive particulate contamination:

- (a) Particles should be separated into several size fractions in the course of sampling, the size intervals between fractions being selected to give a good picture of distribution over the entire size spectrum.
- (b) The particles should be separated in a way that does not significantly alter their physical state from the original state in the air. That is, aggregations of particles in the air that behave in the respiratory tract effectively as large single particles must not be broken up.
- (c) The sampling method must be limited to the collection of particulate matter and not serve as an adsorber for molecular or near-molecular material.
- (d) The particulate matter must be deposited in a manner to permit accurate measurement of the amount of activity collected in each stage of separation or so that it can be readily transferred into such form. Preferably, it should be possible to determine the number of radioactive particles in each size interval as well as the total activity contributed by each size fraction. In certain situations, it may be necessary to distinguish between liquid droplets and solid particles and between soluble and insoluble particles.
- (e) A sufficiently high sampling rate is required to insure the collection of measurable quantities of material in a period of time properly related to the purpose of the sample. For the determination of average concentrations in the plant or outside atmosphere, a

sampling period of many hours may be employed. To measure the variation in output of contamination during different steps in a process, however, short sampling times related to the duration of each step will be necessary.

To meet these requirements, three devices are proposed for experimental use. Undoubtedly, other schemes will come to mind and it is urged that in the systematic development of particle size studies, the exploration of these and other devices be investigated.

(3) Cascade impactor.

This instrument, first developed for the study of chemical warfare smokes, is the only well-known device designed for the deliberate separation of particles into size fractions during collection.* Four overlapping fractions are obtained by means of successively higher impact velocities against adhesive coated glass slides. Particle deposition is found to obey a systematic dimensionless relationship between impact velocity, particle size and density, expressed as follows:

$$I_n = \frac{12 V D_p^4}{3 \ell}$$

Where

I_n	constant for given efficiency of deposition, N
ρ	particle density
V	impact velocity
D_n	diameter of particle removed with efficiency, N
η	viscosity of air
ℓ	linear dimension of nozzle

In the standard instrument, operating at 17.5 l/m, the highest impact velocity is 34 m/sec. which gives approximately 100% removal of particles 0.6 μ in diameter (density of 7) and 50% for 0.4 μ particles.

By increasing the final nozzle velocity to sonic speed, the 50% particle is reduced to less than 0.2 μ , according to the above relationship. The finest particles, which escape deposition, are captured on a fifth-stage filter. The instrument is calibrated for a given dust by direct microscopic measurement of the particles deposited on each slide and the mass-median diameter determined for each. Results are presented in the form of a cumulative distribution curve. The amount captured in the fifth stage plus one-half of that on the fourth represents the quantity of material smaller than the mass-median size of the fourth stage. The sum of 5th, 4th, and one-half the 3rd stage equals the quantity smaller than the mass-median of the 3rd stage, etc. This procedure assumes that there is no significant overlapping on adjacent slides beyond the median sizes. The cumulative curve, converted to percentage values, may be plotted on log-probability paper for further graphic analysis.

* May, K. R., The Cascade Impactor: An instrument for sampling coarse aerosols. Brit. Jour. of Scientific Instruments, 22, 1945.

The cascade impactor has certain disadvantages for the study of radioactive dusts. The low sampling rate (17.5 l/m) requires a long period of sampling to give measurable results when concentrations are within tolerance levels. Since the dust is deposited in a concentrated narrow band, it is not possible to distinguish individual particles by radioautography because of interference and, possibly, piling up of particles. The adhesive slide coating may interfere with alpha counting. Some dust is deposited on the inner walls of the instrument and it may be difficult to decontaminate. With long continued sampling, some of the deposited dust may be blown off the plates.

Preliminary samples from the pile cooling air at Oak Ridge* gave some new and different information concerning the distribution of activity in relation to particle size than had been found by previous tests. In particular the tests showed that a substantial amount of activity is associated with particles smaller than $1/2 \mu$, contrary to earlier conclusions. The cascade impactor should, therefore, be employed, at least for experimental particle-size studies, on all projects and, to this end, instruments have already been secured at a number of sites. An electric precipitator might be employed in place of the terminal filter to avoid the absorption error in alpha counting and the error from adsorption of sub-particulate fission products.

(4) Multi-stage impinger.

A simple, inexpensive, all-glass instrument for size separation, consisting of a series of four impingers with decreasing nozzle diameters, was tried out at Hanford and Los Alamos with some interesting results. The nozzle diameters were selected to produce the following impinging velocities at a sampling rate of 1 cfm: 37, 75, 150 and 300 m/sec. According to May's relationship for the cascade impactor, given above, this arrangement should give a ratio of 1.7 between the average sizes of particles captured in adjacent stages. Preliminary tests with silica dust gave results which agreed well with this prediction. A terminal filter, electric precipitator or other efficient collector is required to capture the finest particles.

The glass nozzles are easily made to dimension by reaming out the bore with a standard machine drill, four properly sized drills being selected for the purpose. Impingement is on the flat bottom of the flask and the non-critical impinging distance is visually adjusted within approximately 5 mm. The liquid depth of 2-3 cm is also not critical. The principal advantages of the device are cheapness, ease of manufacture, and easier decontamination. The combination of nozzle diameters may be altered for different size spectra, to give the best distribution of material in the several stages; for example, a better series might be 10, 30, 100, and 300 m/sec. to give 2.3 times difference in average size of separation. Using water or alcohol as the collecting fluid, the dust is easily transferred to a counting disk by evaporation, thus avoiding the alpha absorption error of an adhesive coating. Particles may also be spread out for radioautography by evaporating the liquid over a large area. Alpha-track counting for particle size measurement should be simplified when the particles are first separated into size fractions by this means. The multi-stage impinger is shown in the accompanying sketch (Fig. 1) and it is suggested for trial use in particle-size studies.

* By University of Rochester AEC Laboratory

(5) Separation of flocculated particles.

Like the cascade impactor, the multi-stage impinger breaks up aggregations of particles. The size distribution given by such instruments, therefore, may be different from the effective distribution of the airborne material in respect to behavior in the respiratory tract. In order to measure the degree of flocculation in dust clouds, Hemeon devised a simple glass spiral tube (Hatch, T. F. and Hemeon, W. C. L., Effect of particle size in dust exposures. Jour. Ind. Hyg. & Tox., 1948) in which particles and aggregations are deposited by centrifugal force. For 1 cfm sampling rate, an 8 mm ID tube is employed; it is bent into ten turns with a diameter of 35 mm and, in use, is coated internally with a tacky film, such as carbowax. The following calculations indicate the collecting capacity.

1. Centrifugal separation factor:

$$S. F. = \frac{(\text{tube velocity})^2}{\text{radius of turn} \times g} = \frac{(940 \text{ cm/sec})^2}{1.7 \text{ cm} \times 981} = 530$$

2. Minimum size of particle captured:

Since flow is turbulent, assume continuous mixing and exponential settlement; therefore, use 1/5 tube length to determine minimum settling velocity for 99 +% removal:

$$\text{Min. } V_s = \frac{\text{dia. tube}}{\text{Time to travel } 1/5 \text{ tube length}} = \frac{0.8 \text{ cm}}{0.0227 \text{ sec.}} = 35.2 \text{ cm/sec.}$$

$$3. \text{ Equivalent gravitational settling velocity} = \frac{35.2}{530} = 0.067 \text{ cm/sec.}$$

4. Size of particle completely removed (particle density 7)

$$r = \sqrt[3]{1.18 V_g \times 10^{-7}} = 0.9 \mu; \text{ diameter} = 1.8 \mu$$

5. Size of particle with 50% removal = 0.7 μ

Other combinations of tube dimension, diameter and length, and radius of turn may be employed to give finer separation. One arrangement is suggested in Fig. 2. A spiral slot 0.5 cm x 0.8 cm is cut around a 3 cm diameter metal rod and wrapped with a tacky-surface paper, "scotch" tape, or other suitable material, which, in turn, is protected by a removable metal cover. The collecting liner is removed and cut into sections for measurement of the deposited activity in relation to distance downstream. With a sampling rate of 1 cfm, more than 99% of the 1 μ particles will be removed and 0.4 μ particles will be captured with 50% efficiency.

In use, the spiral should be followed by a two-stage impinger operating at 150 and 300 m/sec and a terminal collector (Fig. 1). Since it is the coarse (>1 μ effective size) aggregates, particularly, which need to be removed intact; this will be accomplished by the suggested combination of spiral and impingers.

Results are readily converted into effective size distribution data by graphical analysis, as shown in Fig. 3.

(6) Thermal precipitator.

This instrument is also recommended for trial because of certain basic advantages: collecting efficiency is high over the microscopic and sub-microscopic size range; particles are deposited directly on the collecting surface without impact so that aggregations are preserved; there is no adhesive coating to absorb alpha radiation; the particles may be deposited on the coated screen for direct examination and size measurement in the electron microscope. A serious disadvantage is its extremely low sampling rate of only 7 cc/min.

In the standard instrument with stationary collecting surfaces the particles are deposited in a narrow band with high concentration directly under the hot wire and rapidly decreasing numbers downstream for a short distance. There is some size separation in the direction of air flow. Thus, the deposit is not convenient for particle size study, particularly in the electron microscope, since too many fields must be photographed to give good statistical representation. Also, the particles are too close together for radioautography. There are two modifications which may be employed to overcome these difficulties: the use of oscillating or continuously moving collecting surfaces. An instrument of the first type was developed at the Porton Laboratory. Models have been constructed at the University of Rochester and one is in use at Hanford. The second type, which is described by P. H. Kitto (A new form of thermal precipitator. Presented before Inst. of Mining and Metal., London, April 15, 1948) should be particularly useful for radioautographic studies since the particles are spread out over the length of a standard microscope slide which moves at a constant speed past the hot wire during the sampling period.

(7) Routine sampling.

Particle size analysis is time-consuming and it is doubtful if it can be included in the program of routine sampling. Each process should be studied sufficiently by the more refined methods to characterize it in respect to particle size, but for daily, routine sampling, a filter is probably the most practical method. Its limitations in respect to alpha counting must be kept in mind (a hard-surface paper such as Whatman 41 may be preferred over CWS No. 6).

The sparse particle population at tolerance introduces a statistical problem in the routine measurement of exposure to radioactive particulate matter which does not exist in ordinary dust exposures. In the extreme case of polonium, for example, only one particle of $1/\mu$ size is permitted in 10 cubic meters of air. It is obvious that too much dependence cannot be placed upon air samples under this circumstance. If the single particle is missed, a good situation is reported. On the other hand, the capture of a $2/\mu$ particle would put the concentration 8 times over tolerance.

Fixed sampling points are employed in the routine appraisal of exposures. For convenience these are often located some distance away from the breathing zones of the exposed workers and there is some question as to how well they measure actual exposures, especially when the difference between tolerable and excessive concentrations is a matter of only a few particles. Under these circumstances, biological assay by routine urine analysis provides more certain information on the rate of body accumulation of radioactive substances and should be depended upon in preference to air samples.

(8) Atmospheric sampling as an aid in studies of dust sources and design of control measures.

Systematic air sampling is an essential tool in the engineering analysis of dust problems and design of control measures. The requirements for routine surveying of exposures are not as great as are those for collecting the data needed in engineering analysis and design. Consequently, a sampling program designed primarily for survey purposes will not provide the necessary engineering information. Moreover, without proper coordination between the two, interpretation of survey data will be limited. These principles are commonplace in industrial hygiene practice. They have not been as fully developed as need be in the present problem. In the further development of sampling procedures and programs, therefore, it is urged that closer coordination be established between the engineering staff concerned with control and the health physics and industrial hygiene services responsible for appraisal of exposures. An experienced industrial hygienist will be of particular value to both the engineering and health physics groups since his interests overlap both.

II. CONTROL OF RADIOACTIVE PARTICULATE CONTAMINATION

A. CONTROL REQUIREMENTS VERSUS TOLERANCE LEVELS.

With the exception of uranium, the permitted numbers of particles of radioactive materials at tolerance are many times smaller than the tolerable numbers for the more common industrial dusts. In the case of silica, for example, a concentration of 10^9 particles per cubic meter* is permitted. This means that many millions of particles may escape from the source each minute without creating excessive exposure. Hence, in the selection and design of control measures, local exhaust hoods, for example, the continuous escape of a certain amount of the contaminant is deliberately allowed. The tolerable particle concentration for natural uranium (10^8 particles/ m^3) is about like that for lead and here, too, a substantial continuous escape is permitted. Thus, the control of uranium exposures involves no new concepts and the experience and practices in other industries are directly applicable.

The situation with regard to plutonium and polonium is in sharp contrast and finds no parallel in other industries except radium dial painting. With the tolerable level for plutonium set at only 10^3 particles per m^3 , it is evident that the control measures which are applied can make no deliberate allowance for the continuous release of particles. The accidental escape which is expected even with the best control system, allows no margin for anything less. With polonium, perfect control is, indeed, required since tolerance is exceeded with more than one particle in 10 cubic meters of air. The point is well-illustrated in the Mound Laboratory, for example, where the total ventilation rate is approximately 5000 m^3 /min. If main dependence was placed on ventilation for control of exposures, the escape of no more than 500 particles (1/u) could be allowed per minute over the entire establishment with its multitude of points of operation. This means, of course, that ventilation cannot be depended upon for primary control as it is in other industries. Instead, the processes must be completely enclosed and operations carried on by remote control through tight physical barriers. It also requires special facilities to prevent the escape of contamination as materials and equipment are added or removed from the enclosure.

*This concentration is 10 times higher than the figure commonly given, which is based upon low power, light field microscopy, and thus does not include the finer particles which are present

Application of these principles varies on the several projects in relation to the nature and magnitude of the hazard. Properly, the controls at Mound Laboratory are the most extensive since the particulate tolerance is so low. Processes are isolated physically from the operators and access to the process enclosures is through separate service corridors and is limited to a few servicing personnel, who wear extra protective equipment. The ventilating system is designed to serve four separate zones: clean (no active materials handled); low risk (operating areas); high risk (service corridors); and hot (process enclosures). Airflow is modulated by pressure-difference controls which maintain successively lower pressures from the clean to the hot zone, thus insuring airflow always in the direction of the greatest hazard. So-called dry boxes serve the same purpose on other projects. Some difficulty has been experienced in the addition and removal of materials and apparatus even when locks are provided for these operations and despite the inward flow of air. The contaminant does not escape against the air stream; either it is carried out in back-eddies or local displacement of air when the door is opened or it escapes as surface contamination on the object removed. In any event, it emphasizes the need for minimizing, so far as possible, such servicing of dry-boxes. To this end, the practice at Berkley is to plan every new experiment as completely as possible to make certain that all needed equipment and supplies are placed in the dry box in advance. Later additions are made by fastening a container to the dry box before opening from one to the other. At Los Alamos, separately ventilated locks are planned for new dry boxes and processes are being redesigned to minimize the number of occasions for adding or removing materials from the enclosed system.

B. LIMITATIONS OF EXHAUST VENTILATION.

Exhaust hoods with sliding doors are employed in certain instances, some provided with means for regulating the inward air flow so as to keep the velocity constant at 100 fpm. This method of control does not have the same inherent value as a physical enclosure and cannot be regarded as a primary protective measure when concern is with the accidental escape of a few particles. There can be local outward displacement of air, despite the general in-flow, when the arms, for example, are suddenly inserted into the hood or when a piece of equipment is removed. The fact that hoods are most conveniently operated with the sliding doors open will certainly limit the practical protective value of instructions to keep them closed.

C. CONTROL AT THE SOURCE.

Even more basic than the use of enclosures is the design of processes so as to minimize the initial release of particulate matter at its source. The advantage is obvious since every such step reduces the chances of escape of the contaminant and makes it less necessary to depend upon added protective measures. To illustrate: a dry powder dumping operation at Los Alamos was found to be a major source of dust. By means of a locally-enclosing device inside the dry box, the powder was transferred to its storage container with much less contamination of the dry box. The improvement was basic since, without it, the amount of dust released was great enough to overwhelm the protection of the dry box itself and room contamination went up with every dumping. Equally basic is the plan at Brookhaven to surround the uranium slugs in the pile with a positive-pressure atmosphere of helium. This will eliminate the oxidation of the uranium in a leaking casing which was the main difficulty at Oak Ridge in respect to contamination of the pile cooling air. Insofar as dust in the incoming air may contribute to radioactive particulate contamination

after passage through the pile, this, too, will be eliminated at Brookhaven by the installation of GWS filters on the inlet. At Oak Ridge, detailed studies of the several steps in chemical separation of high level materials have shown that the greatest amount of the contaminant is released during one or two steps occupying only a fraction of the total time to complete a run. Moreover, an examination of these processes indicated that, by redesign of certain equipment, considerable reduction can be made in the amount of material released. These experiences are mentioned particularly to emphasize the need for studying every contaminating process from this standpoint, not only to gain greater in-plant control but also to reduce the amount of escaping material which, otherwise, must be removed by air-cleaning equipment before the contaminated air is discharged to the outside. Still more important is the fact that the improvement thus secured is permanent in that it becomes part of the process itself. In the long run, such basic measures are to be preferred over secondary control by air-cleaning.

In the large scale chemical processes at Hanford there is considerable release of particulate contamination from operations beyond the initial dissolving process. By what mechanism are these particles formed and how do they escape from the process vessels into the surrounding cells? There are only two ways for minute particles to be moved, by a conveying air stream or by transfer on a solid surface. Microscopic particles cannot be projected through still air, even when released with high velocity. With these points in mind, the process equipment should be studied with a view to discovering and eliminating every unnecessary mechanism of release and transfer of particulate matter. For example, a centrifuge may act as a fan, drawing air in at one point and discharging it at another. With some relatively minor changes in design, its fan action can be greatly reduced. Such studies, supplemented, perhaps by dry-run operations in the unused Canyon building may very well point to some major reductions in the quantity of radioactive material released from these processes.

D. STACK GAS CONTROL.

To date, major effort has been to design and install air-cleaning equipment for the control of the important sources of particulate contamination. Properly, the equipment has been selected to give the highest efficiency of particle removal, while at the same time, meeting the practical needs imposed by the radioactivity of the collected dust. These include adequate shielding, means for remote installation and removal of filter units, avoiding contamination while handling dirty units, convenience and safety of disposal. To reduce the frequency of changing filter units, a major objective has been to secure the longest possible operating life of the cleaning unit. These special requirements find no parallel in past industrial experience and, as a consequence, it becomes necessary to develop new approaches in design and construction of equipment. The high efficiency of cleaning which is required combined with the extremely low dust loading imposes performance demands upon the equipment beyond anything previously required.

The capacity of air-cleaning installation varies from more than 100,000 cfm down to as little as 5 cfm on individual dry boxes. Several different methods of air-cleaning are in use or under study, including commercial paper and fibreglass filters and electrostatic precipitators, GWS filters, deep fibreglass beds, sand filter. Water scrubbers are employed or under development for removal of liquid droplets and mists, particularly, corrosive substances. Promising results are

reported for wet-type collectors based upon the principle of liquid condensation on the dust particles as nuclei. Such self-cleaning equipment, from which the collected material is constantly removed for disposal, has great practical advantage in that it eliminates the problems of handling contaminated equipment, and presumably, simplifies ultimate disposal of the waste.

Lacking detailed knowledge of the many different processes in use on the several projects, it is not possible to list completely the separate needs for stack gas cleaning. Three major problems are discussed.

(1) Pile cooling air.

The three-stage filtration plant at Oak Ridge has apparently taken care of this source of outside contamination. Absence of slug ruptures since its installation has made it impossible, however, to study performance under maximum load. Also, measurements of activity before and after filtration have given misleading results since they bear no relation to the obviously successful removal of particulate matter in the filters. Insofar as it is proper to judge filter performance by visual inspection and by change in resistance, it can be said that the first layer of FG 50 is giving high performance since there is no appreciable discoloration of the second layer of FG 50 and no significant increase in resistance of the final CNS filter. It is evidently necessary to use more refined sampling techniques to measure performance of this installation and to distinguish between the true particulate matter and the near-molecular decay products which are formed at varying periods of time after the conveying air leaves the reactor. The filter life has proved higher than anticipated. Also the amount of activity deposited on a filter unit has imposed no serious difficulty thus far in respect to direct radiation exposure in handling. Nevertheless, there is need to know more about the relative contribution to the filter load by dust brought in from outside compared with that released within the pile. The relatively crude inlet filters now in use may not remove a high percentage of the finest particles which contribute most to the plugging of the downstream filters. If this is true the build-up of resistance will vary seasonally with the amount of dust in the outside air. The importance of incoming dust as a possible carrier of adsorbed activity also deserves study. The present installation has taken care of the acute situation. The particle problem associated with pile operation should now be investigated in greater detail to determine, in particular, the relative importance of slug rupture and other sources of particulate radioactivity.

To avoid the primary difficulty from slug casing rupture at Oak Ridge, Brookhaven plans on surrounding the uranium with helium under pressure. This will prevent oxidation and also serve as a warning when a leak occurs. In addition to this basic improvement, the pile cooling air will be pre-filtered through CNS units and the ducts are being specially treated to minimize corrosion and surface disintegration. Because of these improvements, it is felt that final filtration of the air before entering the stack will not be a primary necessity. The downstream filter, therefore, is intended mainly for short-term emergency protection and woven glass fabric is to be used for this purpose. Since this material does not have the inherent filtering efficiency of a fibreglass mat, it may not meet the needs for continuous air-cleaning should this prove to be necessary despite the basic improvements in pile operation.

(2) Chemical processing of slugs.

A major source of particulate contamination is the dissolving of slug casings and subsequent chemical processing of the uranium. These processes are carried on by remote control within shielded chambers. Gases are actively generated in the dissolver and, to less extent in subsequent processes, and these must be vented to the outside. In addition, the cells become contaminated by material escaping from the process vessels and, as a consequence, the cell ventilating air requires treatment before being discharged to the outside. The rate of generation of process gases is small compared with the rate of cell ventilation. The concentration of activity, however, is greater and there are added complications in that the contaminant is given off in liquid droplets and the gases are corrosive. Stack gas cleaning must, then, be divided into two parts, requiring different kinds of equipment. Dissolver and vessel off-gases must be treated by scrubbers to remove liquid droplets and corrosive substances, followed by filters. Because the cell ventilation rate is great compared with the amount of contaminant, liquid droplets which may be carried in the air stream apparently do not interfere with filter operation and no treatment to remove such particles is required before filtration.

The sand filter at Hanford is apparently giving satisfactory performance in cleaning the cell-ventilating air and tests indicate that a fibreglass bed will perform equally well with the added advantages of lower resistance and (presumably) longer life. Results show, however, that the contribution of contamination from the dissolver after scrubbing, must also be removed to a comparable degree if full benefit is to be realized. Fibreglass beds are to be installed for this purpose.

Tests at Oak Ridge show that high-level chemical operations contribute more units of activity to outside contamination than does the pile. Moreover, the present filters on this process are not highly efficient. Other studies have demonstrated that one or two steps in the whole separation cycle contribute in a few hours a large fraction of the total contamination from the process. Engineering analysis reveals that these major offending steps can be improved in design, thus reducing their contribution of contaminant.

The development of effective means of air cleaning for chemical separation operations is the greatest present need in respect to control of outside particulate contamination. Experience at Hanford and Oak Ridge should be pooled with the studies and plans at Brookhaven and Knolls to develop at the earliest date a complete statement of the problem, and to outline the control requirements and the essentials of design of cleaning equipment. It is particularly urged that detailed studies of the processes be made to determine the characteristics and magnitude of contamination given off by each step and the mechanism of release with a view to reducing the amount of contaminant by improving the design of equipment.

(3) Dry Filtration.

The great majority of air cleaning needs are for the removal of dry particulate matter in the air exhausted from dry boxes and laboratory hoods and in general room ventilating air. Aside from the high efficiency which must be secured against very low dust loadings, no unusual problems are encountered beyond those imposed by the fact that the collected dust is radioactive. For

maximum efficiency, dependence is placed primarily upon CWS No. 6 filter medium. This paper is expensive and not available in large quantity. At present it is not made up in the most convenient form of units for mounting and handling. It contains a substantial amount of non-combustible fibre which is undesirable from the standpoint of disposal after burning.

Undoubtedly, the CWS medium represents the highest present development in filtration and is capable of delivering entirely satisfactory air to discharge stacks, provided, of course, the load brought to the filter is not too high. The practical need, therefore, is for a cheaper and more readily available substitute filter medium of equal efficiency and a practical means of mounting without leakage and for easy replacement by remote control to avoid direct exposure to radiation. The dirty unit must be removed without undue escape of dust into the air and should burn to minimum ash for disposal. Units are required in varying sizes; for a single dry box, the rate of ventilation may be as low as 5 cfm. Laboratory hoods require up to 2000 cfm. On large, central installations the size of the individual unit should represent a balance between convenience of handling and cost.

An objective of the development work by A. D. Little Company is to meet these practical needs. To assist this development a complete list of dry filter requirements should be prepared for each project, including capacity and other engineering specifications. Incidentally, this work requires more information in respect to particle size and the influence of size upon filter performance. Do the conditions indeed require the performance capabilities of the CWS filter? Experience with the pile installation at Oak Ridge suggests that the two-layer fibreglass filter may be sufficient on this particular job. At Los Alamos a soluble filter is desired on certain dry boxes in order to recover the plutonium dust extracted with the ventilating air. Special conditions may also dictate the needs on other projects.

(4) Evaluation of stack emission.

The objective of stack gas cleaning is to reduce the rate of emission of radioactive particulate matter to levels which will insure acceptable concentrations at ground level and prevent the accumulation of deposited materials in excessive amounts. For a given substance, the permitted rate of emission will vary with stack height, local topography and concentration of population and with meteorological conditions. Stack performance requirements may be fixed by other needs as, for example, the safe dilution of radioactive gases. As a matter of public policy, acceptable levels of outside contamination may be set considerably below in-plant tolerance values. This will offset, in part, the dilution factor provided by the stack and make it necessary to reduce the concentration in the stack to a point approaching the in-plant tolerance level. The needs for each area should be determined in this respect and standards of emission established.

The extensive area and off-site survey program at Hanford provides valuable information on the ultimate disposition of particulate matter. At Brookhaven a large-scale research program designed to correlate stack performance with meteorological measurements is in progress. On other projects, also, outside surveys have been carried on in varying degrees. Such work is time-consuming and expensive. It is necessary, therefore, to design the survey program for maximum usefulness while, at the same time, avoiding unnecessary or relatively valueless tests. The work to date should be reviewed to determine the most useful future program. For example, the Hanford experience probably represents the most extensive survey of stack

performance that has ever been made over so large an area. To what extent is it possible to generalize from these findings in respect to stack performance? Are there gaps in the data which limit their usefulness beyond describing local conditions of contamination?

Characteristics of stack performance, standards of permissible stack emission, area survey procedures, and minimum requirements in respect to number and location of samples should be standardized, so far as possible, after reviewing the work done to date.

III. SUMMARY OF PARTICLE PROBLEM

1. Present atmospheric tolerance levels for radioactive dusts and fumes may not be based upon sufficient correction for the varying effects of particle size of the particulate matter. The possible physiological difference between particulate concentrations of radioactive substances in the lungs and molecular dispersion in body fluids requires further study.
2. There is, at present, insufficient knowledge of the particle size and other physical characteristics of the radioactive particulate matter for adequate description of the conditions of exposure. Particle size measurements are as important as determination of total atmospheric concentration in the basic study of conditions of exposure and appraisal of control measures.
3. The method of air sampling in current use is not well adapted for the study of particle size and other physical properties of the contaminant. More refined methods are suggested for study and adaptation to the work.
4. There is need for closer association between the Health Instruments and Technical Division on the various projects to insure fuller utilization of atmospheric concentration data in the analysis of dust problems and design of control measures.
5. Owing to the small numbers of particles permitted at tolerance, the requirements for dust control on A.E.C. projects differ from those of the common dusty industries in this basic respect: control measures can make no allowance for the continuous release of any amount of contaminant, since with the small numbers of particles permitted at tolerance, the inevitable accidental escape of a few particles allows no margin for anything less than 100% efficiency in the design of the control device. This means that exhaust ventilation, the common method of control in industry, cannot be regarded here as a primary control measure (except in uranium processing plants where the tolerable dust concentrations approximate those of other, more common dusts).
6. The first step in a long-range control program is to examine every process to determine the mechanism of generation and release of particulate contamination with a view to redesign, where possible, to reduce the contaminant at its source. Not enough has been done so far in this regard.
7. Second to reduction at its source, is the use of physical enclosures to prevent the escape of particulate matter into the atmosphere of the work room. An essential part of this method of control is the arrangement of operations to reduce to a minimum the need for moving materials or equipment in or out of the enclosures. The inherent advantages of this method of control

are recognized and are generally employed on well-defined production operations.

8. An exhaust hood, even with means for regulating the airflow to insure constant inward air velocity, does not have the inherent protective value of a physical enclosure. Without constant supervision, doors will remain open simply because that is the most convenient way to use hoods. The use of hoods for control is generally limited to low-risk operations.
9. In the establishment of air-cleaning requirements for stack gases, the first need is to determine the maximum permissible rate of stack emission to keep within the tolerance levels which are established for outside contamination. The minimum dilution rate should be determined or estimated for every source in relation to local topography, as a basis for calculating the permissible emission rate from the stack. In this connection, the area survey data at Hanford and Oak Ridge and the meteorological studies at Brookhaven should be utilized fully to establish minimum requirements for area survey programs.
10. There are two major needs in respect to stack gas cleaning:
 - (a) The standardization of design requirements for handling off-gases from the dissolver and other process vessels employed in large-scale chemical separation.
 - (b) The development of an adequate substitute for the CWS filter medium and practical means for mounting filter units without undue exposure of workers and for safe and simple disposal of the collected material. The special needs for dry filters should be summarized for each project, to aid in the filter development program.

IV. RECOMMENDATIONS

1. Particle size studies: It is recommended that systematic studies of the particle size characteristics of airborne radioactive particulate matter be undertaken on all projects, to parallel the present program of measurement of total activity.
2. Coordination of atmospheric sampling with engineering control: The closest association is recommended between the Technical and Health Physics Divisions in the planning of tests, selection of sampling means, methods of analysis, time of sampling in relation to operations and interpretation of results. The industrial hygienist, if available, should be utilized fully in this joint work.
3. Control of particulate contamination at its source: Every major source of particulate contamination should be studied in detail to determine the nature, magnitude and physical characteristics of the contaminant, the manner in which it is released. Control measures should be applied whenever possible, to reduce the amount of contaminant at its source.
4. Cleaning off-gases: Experience and plans on the several projects for the control of off-gases from chemical separation processes should be reviewed and summarized with a view to a common solution of this problem.

5. Dry filter requirements: It is recommended that the dry filter requirements for each plant be summarized in terms of capacity, dust or activity loading, particle size, and other physical properties, and including details of use, mounting, and disposal, and other information needed to aid in the filter development program.
6. Standards of stack emission: A review and summary of the sampling and analytical procedures, meteorological analysis, and findings of the several site programs now in progress is recommended with a view to establishing standards of permissible stack emission in relation to stack performance, local topography, and meteorological conditions and to establish minimum requirements for routine area survey programs.

TWO-STAGE IMPINGER
SUGGESTED SIZE ANALYSIS ASSEMBLY

FIG. 1

INDUSTRIAL HYGIENE FOUNDATION
1400 FIFTH AVE. PITTSBURGH, PA.

BY TFM

DATE

CLEARANCE FOR LINER
(SCOTCH TAPE)

SPIRAL SLOT 0.5x0.8 cm.
(15 TURNS)

2 mm

3 cm dia.

2 cm

SPACING LEGS

FIG. 2
SPIRAL ASSEMBLY

INDUSTRIAL HYGIENE FOUNDATION
4400 FIFTH AVE., PITTSBURGH, PA.

BY TFH

DATE

SECRET

INDUSTRIAL HYGIENE FOUNDATION
4470 FIFTH AVE. PITTSBURGH, PA.

DATE

FILMED

4/19/94

END