THE DETECTION AND MEASUREMENT OF PLUTONIUM AIRBORNE CONTAMINATION IN MAJOR PLUTONIUM FACILITIES

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Abstract—A review is presented of air sampling experience in three kinds of plutonium facilities, viz. a laboratory containing lines of glove boxes, a manufacturing facility employing a sealed face to separate operating and maintenance areas, and a workshop used for the cleaning of safety equipment contaminated with plutonium.

The level of airborne contamination as measured on fixed, installed air samplers in these areas is shown to be critically dependent on the type of work being carried out, the position of the sampler and the nature of the ventilation system. Personal air samplers invariably indicate higher levels of airborne contamination than these installed samplers. Over periods of months in the areas under examination, it is generally safe to assume that the integrated exposure measured using personal air samplers will not be more than ten times the exposure measured with the installed samplers. Under incident conditions, however, the installed sampler might underestimate inhalation exposures by several orders of magnitude. The distribution of air sampling data shows a deviation from log normal where incidents occur producing abnormally high airborne contamination.

It is concluded from these observations that there is a requirement in certain plutonium areas to use personal air samplers for the measurement of inhalation exposures. In addition, a personal air sampler with an audible alarm has been developed for the early detection of airborne contamination in operating areas.

Particle size distributions on air samples have been studied, and their significance is discussed in relation to the assessment of the plutonium inhalation hazard.

1. INTRODUCTION

The potential inhalation hazard from plutonium airborne contamination is a major risk on any plutonium manufacturing facility. It is a feature of many plutonium plants, particularly those employing extensive use of glove boxes, that plutonium tends to be released over extremely small time intervals, with long, quiescent periods when no contamination is produced in the working areas. This makes plutonium operational control difficult. Measurement of plutonium intakes by biological methods is a laborious procedure and is post event, also because of the limited sensitivity of these techniques, and the fact that interpretation of results is a lengthy process extending over periods of months, they cannot by themselves provide adequate control of plutonium exposures.

As Dunster has pointed out, [1] although organ doses from long-lived isotopes will only exceed I.C.R.P. recommendations when the maximum permissible organ burden is exceeded, it is sound practice to attempt to limit all intakes to those appropriate to a 13-week period. Because of the intrinsic difficulties in measuring and controlling plutonium intakes, the designer of a plutonium facility must do his utmost to provide absolute containment. This ideal is in practice never achieved and the difficulties of measurement and control of plutonium intakes by inhalation must be faced.

This report considers how detection and measurement techniques for plutonium airborne

contamination have developed with a continuing experience of handling plutonium, and also discusses future developments. Observations and measurements made on facilities at Windscale are also discussed.

2. HISTORICAL DEVELOPMENT

2.1. The earliest type of air sampling programme employed portable air samplers to collect samples over a short duration of sampling (say ½ hr) in all operating areas of plutonium plants and laboratories. Provided that these tests were carried out regularly, it was considered that leakages of activity due to weaknesses in design of operating procedures would be detected. A practical reason for this approach was the lack of air sampling devices which could operate continuously over long periods of time.

It is now well established that under many conditions air contamination levels in operating areas of plutonium plants follow a log normal distribution. With this form of distribution the mode (the value most likely to be observed) may only be one-sixth or less of the mean concentration. It follows, therefore, that "spot sampling" may seriously underestimate the true concentration. Another disadvantage of the short term sample is the lengthy counting period required to give results with reasonable statistical accuracy for an element such as plutonium with its low M.P.C. in air.

2.2. In certain operating areas it was soon realised that a potential airborne hazard did exist and that it might be necessary to limit the time spent by individuals in these areas. To improve the methods of control, more frequent, in fact almost continuous, half-hourly samples were taken with a view to obtaining early information on the levels of airborne contamination. With this information available, breathing apparatus was prescribed if it was estimated that air concentrations were greater than one M.P.C.

This system certainly provided a higher degree of protection for the operators than the earlier approach depending on intermittent air sampling. Also, the accuracy of information about average air contamination levels in an area, derived by counting the filter papers after a time lag sufficient to allow daughter products of radon and thoron to decay, was improved by the introduction of a higher frequency of sampling. A practical disadvantage of this system, however, is the scale of effort that must be mounted (in men and equipment) to assess samples obtained. This is a consequence of the large numbers of samples to assay, and the lengthy counting periods required to produce results of reasonable accuracy because of the small volumes of air sampled. Furthermore, because data accruing from this type of sampling is retrospective, it is not possible to guarantee that releases of plutonium will always be detected early enough to prevent significant personal exposure.

2.3. Because of these defects, the whole philosophy of air sampling in plutonium areas was reviewed. In the first place, equipment was developed to allow continuous sampling in all areas where plutonium was handled. Eventually, within the U.K.A.E.A. a monitoring system was developed, based on continuous sampling through a fixed filter paper which is presented to a scintillation detector with associated counting equipment. Because of the interference from alpha active decay products of radon and thoron, the instrument has a minimum alarm level in the conditions obtaining on the Windscale plants, of about 120 d.p.m./m³ hr.* To provide increased sensitivity, the latest development is a sampler employing a solid state detector and alpha energy discrimination against naturally occurring alpha emitters. By this means an alarm can be given on detection of an exposure of less than 30 d.p.m./m³ hr.

2.4. So far, air sampling had been confined to static samplers, i.e. high volume air samplers installed in a fixed position in the working area. As more data became available, however, it was obvious that the levels of airborne activity measured by these samplers was critically dependent on their exact location within a laboratory or other operating area. Large variations in measured airborne concentrations could be

* 120 d.p.m./m³ hr represents the integrated level of air contamination occurring as a result of an air concentration of 120 d.p.m./m³ of plutonium lasting for 1 hr, or 12 d.p.m./m³ for 10 hr, etc.

obtained over a distance of a few feet, the pattern of results being influenced by the type of operations being carried out and the nature of the ventilation system.

These results suggested that the use of static samplers for measuring environmental plutonium airborne contamination could give very misleading results. To investigate this more closely Sherwood and Greenhalgh[2] developed the "personal" air sampler capable of measuring concentrations in the workers' breathing zones. The use of this sampling device on the Windscale Plants during the past two years has confirmed that the static sampler invariably underestimates the airborne concentrations at the point where activity is released to the operating area, which is normally close to the operator who causes the release. The results of these surveys have necessitated a further review of the principles to be applied in the detection and measurement of plutonium air contamination.

3. MEASUREMENTS OF AIRBORNE ACTIVITIES ON WINDSCALE PLANTS

The historical review in the preceding paragraph has been given to present a broad outline of the experiences which have influenced developments leading to present-day practices. However, the techniques employed in this field are still in a state of some flux and further developments are certain to take place. In order to gain a proper appreciation of the "state of the art" now, and likely developments in the future, a survey is given of the data accrued recently from the use of static and personal air samplers.

Three areas were selected for investigation, these are as follows:

A. A facility employing interconnected "free-standing" or "island" glove boxes in an open laboratory, and used for the manufacture of plutonium oxide/uranium oxide ceramic fuel for reactors.

B. A section of a plutonium manufacturing plant, used for the preparation of plutonium metal and oxide via the oxalate precipitation and fluorination route. On this plant the interconnected line of glove boxes in which the process is carried out is sited in a "maintenance area", separated from the operating area by a "sealed face". The maintenance and operating areas are separately ventilated and there are no ambidextrous gloves in the sealed face; movements of plutonium in trays through the line are effected by push–pull rod operation from the operating face. Respiratory protection is always provided for workers in the maintenance area. A block plan of this facility is shown in Fig. 1.

C. A decontamination facility, used for the cleaning of safety equipment (mainly respirators and pressurised suits which have been contaminated with plutonium) and health physics survey instruments.

Areas A and B both handle plutonium on the kilogram scale. In Area C, of course, only microgram quantities are encountered. Thus these three facilities differ considerably from each other in functions, but are each representative of conditions in other areas where measurements have also been made.

3.1. *Static Air Samplers*

Area A. At five positions in the laboratory, at 2 to 3 ft above floor level air is drawn continuously through glass fibre filter papers, at a sampling rate of approximately 5 m^3/hr. The samples are changed at the end of each working period, i.e. at 8-hr intervals. Table 1 shows the mean air contamination levels measured over several months' operations.

In this laboratory the ventilation system is arranged so that clean air is injected into the room through a false roof and is extracted at floor level at a number of positions distributed throughout the laboratory. Contamination released near one particular air sampler should therefore be extracted without seriously affecting the remaining samplers. It would be expected that the relative levels recorded at the sampling positions should reflect the magnitudes of the activity released during operations in their immediate vicinity.

Results on single days in fact showed much wider variations in levels measured at the five

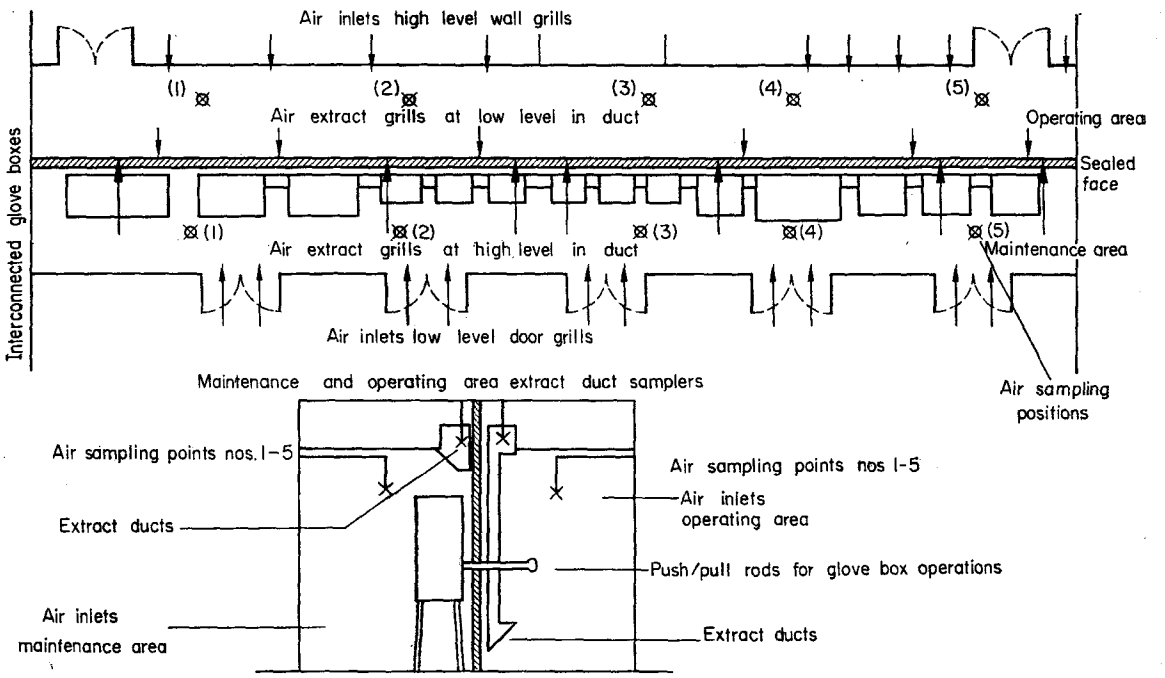

FIG. 1. Layout of Area A showing airflows and locations of air samplers.

sampling positions than is apparent from the mean values shown in Table 1. The ratio of the activities collected by the samplers showing the highest and lowest values over the same working periods varied from 2 to 400, with a mean ratio of 30.

Area B. Air sampling data for both operating and maintenance areas have been examined.

In the maintenance area there are five air sampling locations spaced at intervals of 15 ft down the line at heights of 6 ft 6 in. above floor level. The mean values of air contamination obtained over a period of one year are shown in Table 2. (Samples were collected over 8 or 12 hr periods.)

The situation in this area is that a sampler at one end of the line (position 1) shows much higher levels than the remaining samplers. Those situated in the central positions indicate very similar average air contamination levels, whilst the remaining sampler at the opposite end of the line indicates markedly lower levels than at any other position.

The ventilation system for this area is shown in Fig. 1. Clean air is injected into the line at ground level through grills in the cubicle doors and is discharged to the active extract line in ducts at ceiling level. The air flow across the line is about 1000 linear ft/min, the system being deliberately arranged so that there is a 20%

Table 1. *Mean Air Contamination Levels (d.p.m./m³) Measured at Fixed Positions in the Operating Area of Area A*

Position 1	Position 2	Position 3	Position 4	Position 5
1.29	0.97	0.63	1.68	1.03

Table 2. Mean Air Contamination Levels (d.p.m./m³) Measured at Fixed Positions in the Maintenance Area of Area B

Position 1	Position 2	Position 3	Position 4	Position 5
122	67	76	68	9

higher flow at the centre of the line than at the ends.

With this arrangement, it would be expected that each air sampler would be affected mainly by the air contamination generated in the vicinity of the sampler, although those samplers in the central part of the line would tend to be influenced as well by operations carried out at the extremities of the area. The air sample results confirm this assessment. Position 5 is at the "clean" end of the line where the plutonium metal or oxide is canned and the glove boxes are only contaminated to relatively low levels. Most of the direct handling of dry plutonium as oxide or fluoride is done in the middle of the line and the majority of maintenance work involving breakage of the containment is carried out close to position 1.

In the operating area, clean air is injected through grills almost at ceiling height in the wall at the back of the operator and is extracted at floor level at five separate points into an extract air duct. There are five air sampling locations in the area, the sampling heads being spaced at equal intervals of distance down the line and positioned 6 ft 6 in. above floor level. In addition, continuous samples are taken from the final, common air extract duct from the area. The results of a sampling programme carried out over a period of two months are shown in Table 3 below.

The low levels of air contamination in this area demonstrate the effectiveness of the sealed face concept. The similarity of the measurements at each sampling location, with no large day-to-day variations, probably indicates that the air contamination in this area has its origin in very low levels of surface contamination on surfaces and clothing and that little or no direct transfer of activity occurs between the maintenance and operating areas.

Area C. There are only two static air samplers in this area, one situated centrally and one close to the point where respirator facepieces are monitored after decontamination. Over a sampling period of six months, the mean air contamination levels measured at the two sampling positions were 4.2 and 1 d.p.m./m³ respectively, the sampler immediate to the working position giving the higher result.

The ventilation provided in this room consists of an extract from a central position in the room producing a modest eight air changes/hr. Thus the air is fairly static and variations in air contamination levels throughout the area are the consequence of a gradual dispersion of activity from the point of release. Under these conditions the difference between the value of air contamination measured at the two sampling positions is perhaps somewhat greater than might have been expected.

Comment

The pattern of results illustrated above is one of considerable variations in the levels of air contamination measured in working areas at

Table 3. Mean Air Contamination Levels (d.p.m./m³) Measured at Fixed Positions in the Operating Area of Area B

Extract Duct	Position 1	Position 2	Position 3	Position 4	Position 5
0.2	0.11	0.23	0.2	0.17	0.23

sampling positions a few feet from each other. These variations are caused by local ventilation conditions and the nature of the work being carried out in the vicinity of the samplers.

Averaged over long periods (of several months' duration) static samplers distributed within a particular laboratory generally produce results which do not differ from each other by more than one order of magnitude and agreement is often much closer. On the other hand, several orders of magnitude difference occasionally appear over single (8 hr) sampling periods under incident conditions. This suggests that air contamination can be highly localised, in which event a static sampler might measure air contamination which is unrelated to the level of air contamination to which the workers are exposed.

3.2. *Personal Air Samplers*

The doubts raised by the above evidence as to the adequacy of static air samplers for measuring environmental air contamination led to the development of a personal air sampler.

The sampler consists essentially of a small diaphragm air pump which samples the atmosphere at about 2 l./min through a 1 in. diameter glass fibre filter paper contained in a sampling head. The sampling orifice is adjusted in size such that the linear velocity of sampling is equivalent to the mean air velocity during breathing.

In the last two years a comprehensive experimental programme, utilising this personal air sampler, has been carried through at Windscale Works. The investigations have been made in a wide variety of working areas and laboratories. However, the results obtained in the three areas chosen to illustrate the significance of static air sampler results are used here because they typify the data obtained in this programme.

Area A

During the same period for which static air sampler data were obtained, twenty operatives wore personal air samplers. Taken over the whole period, the mean exposures incurred by the operatives ranged from 12 to 320 d.p.m./m³ hr, per 8 hr "shift", giving a ratio of about 25 between the exposures recorded for the highest and least exposed individuals. (In this paper, the term "exposure" refers to the integrated level of air contamination as indicated by measurements with personal or static air samplers.) It should be noted, however, that results in the high exposure range were strongly influenced by one or two very high results. For instance, the most highly exposed individual received 80% of his exposure in a single working period, the remaining 20% being spread over 19 similar periods.

The personal air samplers invariably gave results significantly higher than the installed, static air samplers. Using the mean data from the five static samplers, the ratio of the integrated air contamination levels (P.A.S./S.A.S.) measured in units of d.p.m./m³ × hours, varied between 2 and 27 with a mean value of 11.

The ratios for single working periods, however, were spread over a much wider range of values; using the data from the personal air samplers and static air samplers giving the highest results during each working period, the ratio $\frac{\text{P.A.S.}}{\text{S.A.S.}}$ varied from less than 1 to 500, with a mean value of 12.

Another feature of these results is that the ratio $\frac{\text{P.A.S.}}{\text{S.A.S.}}$ shows a tendency to increase as

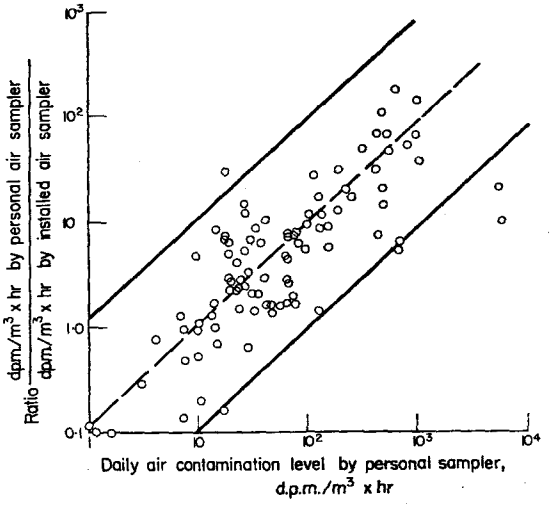

FIG. 2. Daily air contamination level by personal sampler d.p.m./m³ × hours. Highest values of personal and corresponding installed samplers in a plutonium laboratory.

the P.A.S. activity increases and this trend can be seen in Fig. 2. Here there is a wide scatter about a line showing the ratio $\frac{\text{P.A.S.}}{\text{S.A.S.}}$ increasing from 1 to 100, as the personal air sampler results increase from 10 to 1000 d.p.m./m³ × hours. This relationship demonstrates clearly that even static samplers in close proximity to the source of release of plutonium to the operating area are much less sensitive to changes in conditions than personal air samplers. Although not reported here there are similar results for Areas B and C, the only difference lying in the absolute values of the $\frac{\text{P.A.S.}}{\text{S.A.S.}}$ ratio. This ratio is likely to be influenced by a number of factors, including the nature of the work, normal chronic background level of air contamination, proximity of static sampler to the source of release etc.

Area B

Personal air samplers were worn over a two month period by personnel operating the plant from the "clean" side of the sealed face. The mean level of air contamination as measured by these samplers was 0.8 d.p.m./m³ as compared with 0.2 d.p.m./m³ measured on the static samplers, i.e. a $\frac{\text{P.A.S.}}{\text{S.A.S.}}$ mean ratio of 4.

Area C

Three workers engaged in check monitoring of safety equipment and monitoring instruments after a decontamination process, wore respirators over periods of 2 to 3 months.

Analysis of this data is difficult because the workers were not all engaged on this work at the same time. Also the work was of an intermittent nature; often the three workers were only employed for one or two hours each day in the area. It is thus impossible to compare accurately the personal and static air sampler results.

However, a number of deductions are possible from the data available. Even though these workers were employed on identical work, the mean air contamination levels as recorded by their personal air samplers varied by factors of two or three. In each case the personal air samplers indicated higher levels of air contamination than the static samplers; the mean ratio of personal to static air sample activities expressed in d.p.m./m³ hr, was 5. The use of personal air samplers in fact demonstrated that conditions in this area were not satisfactory and led to the introduction of improved local ventilation.

Discussion

The most important fact which emerges from these measurements is that personal air samplers invariably indicate higher levels of air contamination than static air samplers. The explanation for this would appear to be that air contamination in operating areas is nearly always associated with the actions of individual workers, as opposed to unexpected malfunctioning of plant or equipment. Because of the nature of the work carried out on plutonium facilities, particularly during glove box operations, this means that the contamination will initially be generated close to the worker, before being dispersed by the ventilation system. Hence, the personal air sampler, with its sampling head in close proximity to the operator's breathing zone should collect samples which are representative of the air contamination to which the worker is exposed.

The inference to be drawn is that in some areas the measurement of environmental air contamination by static air samplers is an unreliable technique. In Area A, for example, about 50% of personal air sampler results at the 1000 d.p.m./m³ × hours level coincided with static air sampler results not significantly different from the normal background level of air contamination in the laboratory. Most of these exposures would have remained undetected if personal air samplers had not been worn, since there were no obvious causes for the exposures and the incidents did not reveal themselves by producing abnormal surface or personal contamination.

On the other hand, there is some evidence from this area that for exposures of the order of 5000 d.p.m./m³ × hours on personal air samplers there are likely to be significant increases in static air sampler results. Also, in Area B operating area the results obtained with static air samplers, suitably corrected by a factor

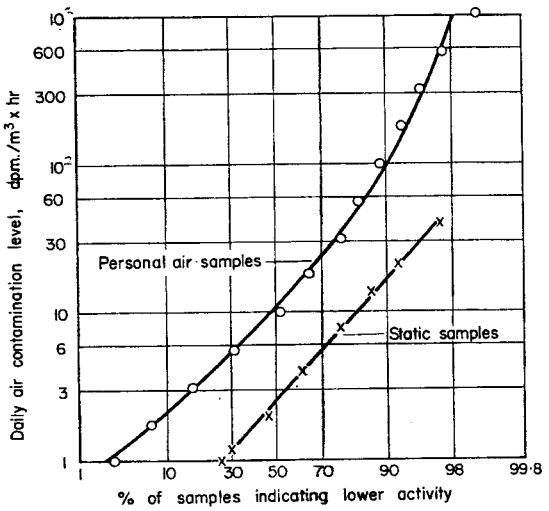

FIG. 3. Air sample measurements in a plutonium laboratory.

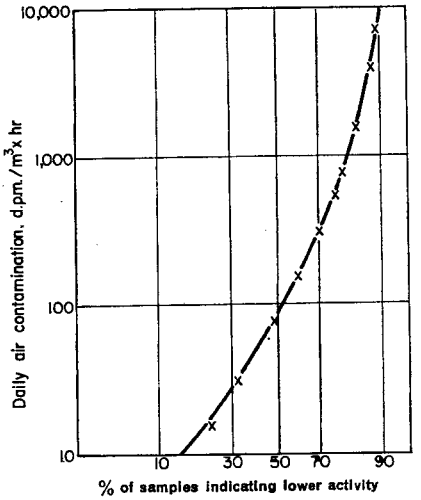

FIG. 5. Static air sample measurements in a maintenance area of a plutonium manufacturing plant.

of about 4 give reasonably reliable estimates of the exposure of workers in this area. The results obtained from the routine use of personal air samplers in areas such as this would add little information to that obtained from static samplers. The personal air sampler results for this area supplement the evidence from static air samplers in justifying the use of a sealed face, on major plutonium facilities, to isolate the operating area from the main plant.

Both the static and personal air sample data are plotted on log probability paper in Figs. 3 to 6 inclusive. In Area B operating area and in Area C the observations lie on a straight line. demonstrating a true log normal distribution, The personal air samples from Area A, and the static air sample results for the maintenance area of Area B, however, show a deviation from a log normal distribution caused by a larger number of high values than would be expected from a log normal distribution of data.

It is suggested that where the spread of air-

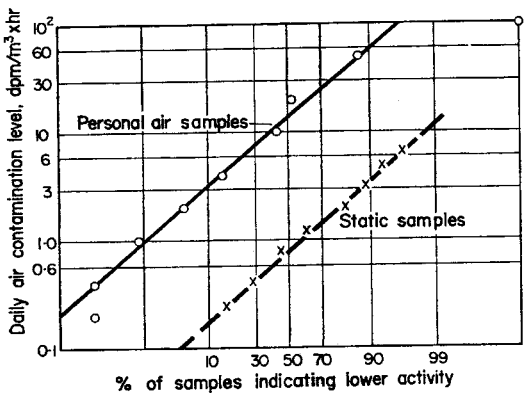

FIG. 4. Air sample measurements in a plutonium manufacturing plant.

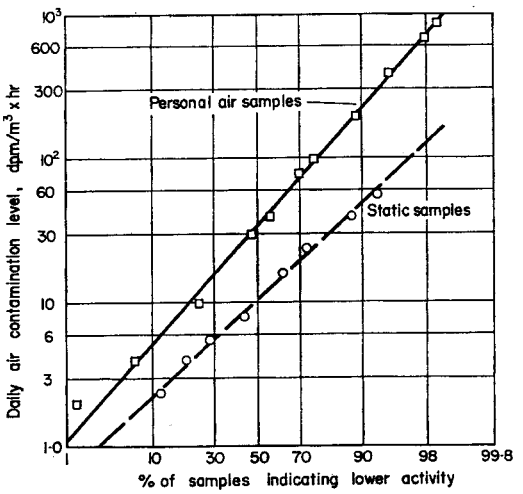

FIG. 6. Air sample measurements in a decontamination centre.

borne contamination is due only to abrasion of activity from contaminated surfaces, the observations are more likely to follow a log normal distribution. Such are the conditions in Area B operating area and Area C. In the other two areas, however, the chronic air contamination pattern will be modified by infrequent incidents due to accident or mal-operation, in which more significant quantities of activity will become airborne. Most incidents of this nature are caused by glove failures. The deviation of the data from log normal will be dependent on the frequency and magnitude of the "incidents" in each area.

4. PARTICLE SIZE ANALYSIS

4.1. Measurements of total air contamination in the breathing zone are not necessarily an accurate indication of the hazard to individuals. This is dependent on a number of other factors, notably particle size and solubility in tissue fluids of the material inhaled. Investigations made to ascertain the size spectra of plutonium aerosols in operating areas at Windscale are described below.

4.2. *Method*

The activity size distributions of plutonium particles collected on filter samples were determined using the autoradiographic technique described by Carter.[3] In this method the alpha particle energy is converted to visible light by means of a zinc sulphide screen and the light output is recorded on a photographic plate. The zinc sulphide is spread over vinyl backed sellotape and placed in contact with the sample. A photographic plate is placed under pressure in contact with the screen and, after a suitable exposure time, developed and fixed. Plutonium particles on the sample produce black spots on the plate, the diameter of a given spot being indicative of the associated particle activity. Individual particles of known activity are used to calibrate the system. By using an enlarger to view the plate, the minimum detectable activity size for an exposure of 10^4 min is about 0.01 d.p.m. The size spectrum of particles in a given area was determined by assuming that all particles assessed autoradiographically were of pure plutonium dioxide of density 11·4 g/ml.

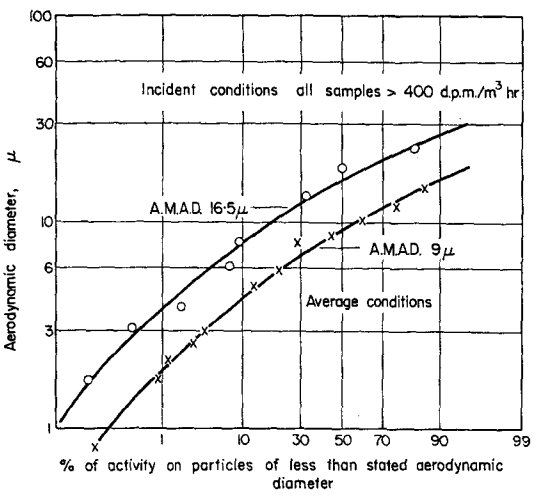

FIG. 7. Plutonium particle activity size measurements in a laboratory area.

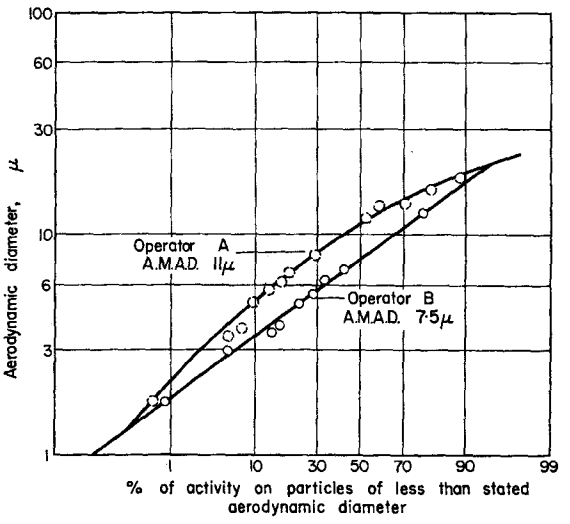

FIG. 8. Plutonium particle activity size measurements in a decontamination centre. Operator A—Decontamination of respirators. Operator B—Monitoring of respirators.

4.3. Personal air samples from Areas A and B have been analysed autoradiographically. Samples were taken in Area A during the manufacture of 25% PuO_2/75% UO_2 pellets. All personal air samples taken over a prescribed period were analysed and the results for samples indicating an integrated air contamination in

excess of 400 d.p.m./m³ × hours were analysed as a separate group.

In the decontamination centre (Area C) the personal air samples associated with the operators responsible for (a) the decontamination of respirator facepieces and (b) the monitoring of facepieces after decontamination, were analysed as two separate groups. The duties of both these operators were carried out in the same room.

The results of these analyses can be seen graphically in Figs. 7 and 8. Median particle diameters have been converted to equivalent aerodynamic diameters by using a multiplying factor $\rho^{\frac{1}{2}}$ assuming a density of 11·4 g/ml.

4.4. *Discussion*

The particle size spectrum in both areas closely approximates to log normal. There is, however, some variation in the activity median aerodynamic diameter (A.M.A.D.) for each distribution. Analysis of the most active personal air samples in Area A indicates an A.M.A.D. of 16–17 μ. The A.M.A.D. based on analysis of all the personal air samples is 9 μ, thus indicating that high air contamination is generally associated with the presence of larger particles, rather than a general increase in the numbers of those normally present. However, it should be noted that incident conditions are infrequent, and contributed only 7% of the total air contamination over the period of interest.

In Area C the A.M.A.D. for the activity on samples associated with the operator carrying out the decontamination of respirator facepieces is about 11 μ and, not unnaturally, somewhat greater than the A.M.A.D. which is associated with the duties of monitoring decontaminated facepieces (7·5 μ).

The interpretation of these results in terms of relative hazard requires a knowledge of the deposition and translocation of particulate matter in the respiratory tract. The currently adopted maximum permissible concentration for insoluble plutonium in air (4×10^{-11} μc/cc or 88 d.p.m./m³) is based on an assumed 12½% retention of inhaled activity in the lung and a biological half life of one year. A task group appointed by the International Commission on Radiological Protection (I.C.R.P.) has recently proposed a new lung model defining the deposition and translocation of particulate matter in the respiratory tract.[4] The model assumes a log-normal distribution of particle diameters and a knowledge of the A.M.A.D. and the nature of the activity is all that is required to define the M.P.C. in air for a particular distribution. For average conditions in Area A for example (A.M.A.D. : 9 μ) the derived M.P.C. for plutonium dioxide in air is about 250 d.p.m./m³. Under "incident" conditions (A.M.A.D. : 16–17 μ) the M.P.C. rises to about 350 d.p.m./³. In Area C the M.P.C.'s associated with the decontamination and monitoring of respirator facepieces are respectively 270 and 220 d.p.m./m³.

Some caution is required, however, before applying the data in this manner. As has been remarked earlier, the technique of particle size analysis used in these investigations only measures the diameter of the radioactive component of the sample. Inert dust adhering to active particles is not detected, nor is uranium because of its low specific activity. Therefore unless pure plutonium oxide is collected, particle sizes are likely to be underestimated.

On the other hand, particle activities of less than 0·01 disintegrations/min (equivalent to pure PuO_2 particles of 0·25 μ diameter, or 0·85 μ aerodynamic diameter) were not detected, mainly because of the limited exposure time of the sample papers (1 week). This will introduce an error leading to an overestimate of the A.M.A.D.'s. The error is unlikely to be serious, because of the large numbers of small particles required to make any significant difference to the A.M.A.D.'s. However, particle size investigations are now in hand, using longer exposure periods to reduce significantly the threshold detection level.

5. CONCLUSIONS

For each type of facility considered in this paper it has been demonstrated that static air samplers invariably underestimate air contamination levels in the vicinity of plutonium workers, as measured by personal air samplers. This is particularly true under "incident" conditions; our experience shows that individual exposures of at least 1000 d.p.m./m³ × hours might easily escape detection by static air

samplers sited in the operating area. There is limited evidence which suggests a much higher probability of detecting exposures greater than 5000 d.p.m./m³ hr. Static samplers will give more representative results the nearer they are sited to possible releases. Unfortunately, it is often impossible to guarantee optimum siting of these samplers because of the numbers of individuals at work and their movements around the plant. In these circumstances, a routine air sampling programme involving the use of static air samplers in the working area is of doubtful value.

On all plutonium plants there is a requirement for a monitoring system to give immediate warning of abnormal air contamination. It has been shown that static air samplers sited in the general working area cannot be relied on to fulfil this function. This requirement can best be met at the present time by sampling the air in the ducts which extract air from operating and maintenance areas. The use of a fixed air sampler for this purpose is illustrated in Fig. 9.

Since an instrument of this type samples on a continuous basis, all the air extracted from the area being monitored, it can also be used to establish trends in air contamination conditions in the area.

To establish an accurate picture of the plutonium air contamination levels it is necessary to use personal air samplers. Particle size analysis of a selection of the filter papers used in the personal air samplers can be carried out to determine how these measurements should be modified to give a true picture of the inhalation hazard. A study of particle sizes on two plutonium facilities at Windscale, has indicated that measurements of total air contamination in these areas using personal air samplers are likely to overestimate the inhalation hazard.

If the assessed concentrations of "respirable" activity are below the maximum permissible

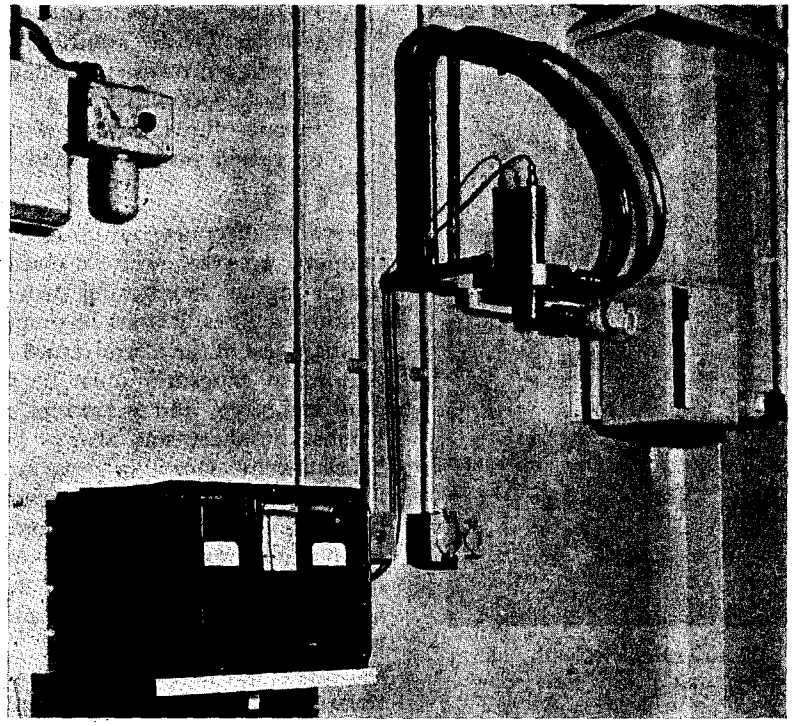

FIG. 9. Extract duct air samples.

concentration, it may be possible to dispense with the use of personal air samplers unless the duct samplers suggest a deterioration in working conditions. The use of personal air samplers is recommended during the commissioning of new plant as an aid to detecting weaknesses in design or operation which lead to the release of plutonium to the working area. To enhance their usefulness in this respect, it would be advantageous if an audible alarm system could be incorporated in their design. Such a device, employing a semi-conductor detector

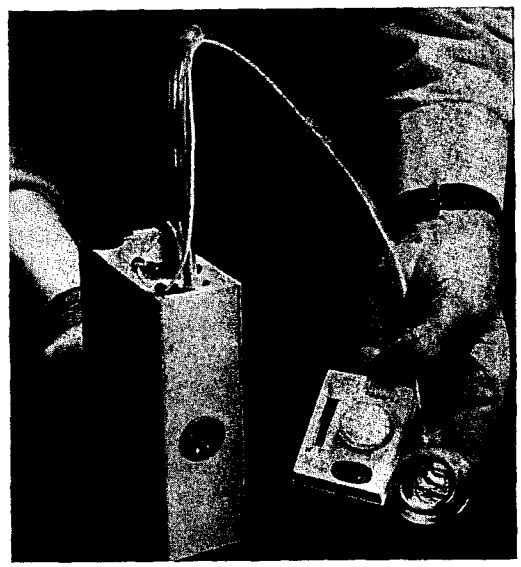

FIG. 10. Personal air sampler, with alarm unit.

mounted in front of the filter paper feeding a transistorised amplifier circuit activating a whistle alarm, is being developed at Windscale (see Fig. 10). This prototype instrument has been designed to alarm at an exposure greater than 400 d.p.m./m^3 hr.

Summarising, therefore, the evidence presented in this paper suggests the following basic principles for an air sampling programme on plutonium facilities:

1. Continuous sampling in extract ducts for the purpose of triggering alarm systems in the event of abnormally high air contamination, and for *detecting* trends in air contamination levels in working areas.
2. Personal air samplers to be used for *measurement* of total air contamination in the breathing zone, in areas where static air sampling has been shown to give unreliable or ambiguous results. They may also be used in the collection of samples for particle size analysis and other operational investigations.
3. Portable, or preferably "personal", air samplers with audible alarm level to investigate transient releases of activity—such devices will indicate the time of the release, the location of release and the operation which caused the release to occur.

Finally, of course, although an effective air sampling programme is an essential part of the health physics monitoring programme on plutonium facilities, it must be emphasised that the ultimate test of the effectiveness of the control of plutonium exposures must be made by biological sampling, and personnel monitoring. At Windscale, there has always been a comprehensive urine sampling programme backed up by faecal sampling where significant intakes have been suspected, and in common with other radiological protection groups we are developing techniques, using our whole body monitoring facility, for measurement of plutonium in the lungs.

Despite the discrepancies that have been reported in this paper, between static and personal samplers, this biological monitoring has demonstrated that the design of our plutonium facilities and operational procedures have been effective in controlling plutonium intakes to acceptable levels. The continuous development of improved air sampling techniques will reduce still further the possibility of individual plutonium exposures.

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REFERENCES

1. H. J. DUNSTER. Some problems in the application of I.C.R.P. maximum permissible levels for occupational exposure. *Health Physics* **12,** No. 1 (1966).
2. R. J. SHERWOOD and D. M. S. GREENHALGH. A personal air sampler. *Am. Occup. Hyg.* **2,** 127–132 (1960).
3. R. F. CARTER. An autoradiographic method for particle size estimation of plutonium dioxide. A.W.R.E. Report, unpublished.
4. Task group on lung dynamics, Deposition and retention models for internal dosimetry of the human respiratory tract. *Health Physics* **12,** No. 2 (1966).