EUR 4594e

COMMISSION OF THE EUROPEAN COMMUNITIES

AUTORADIOGRAPHY AS A HELP FOR ANALYSING THE DISTRIBUTION OF α-ACTIVE ISOTOPES IN THE HUMAN BODY AFTER AN AIR CONTAMINATION

by

J. P. VAANE, E.M.M. de RAS and Chr. von BRANDENSTEIN

1971

Joint Nuclear Research Centre
Karlsruhe Establishment - Germany
European Institute for Transuranium Elements

LEGAL NOTICE

This report is on sale at the addresses listed on cover page 4

at the price of F.Fr. 6.65 B.Fr. 70.— DM 4.40 It. Lire 750.— Fl. 4.30

When ordering, please quote the EUR number and the title, which are indicated on the cover of each report.

Printed by L. Vanmelle, Ghent
Luxembourg, February 1971

EUR 4594 e

AUTORADIOGRAPHY AS A HELP FOR ANALYSING THE DISTRIBUTION OF α-ACTIVE ISOTOPES IN THE HUMAN BODY AFTER AN AIR CONTAMINATION by J.P. VAANE, E.M.M. de RAS and Chr. von BRANDENSTEIN

Commission of the European Communities
Joint Nuclear Research Centre — Karlsruhe Establishment (Germany)
European Institute for Transuranium Elements
Luxembourg, February — 36 Pages — 9 Figures — B.Fr. 70,—

According to the lung model of the I.C.R.P. the size distribution of the particles of an aerosol is an important parameter for calculating the distribution of radioactive dust in the respiratory tract.
This report gives a review and a short description of autoradiographic methods used to determine the distribution of α-active particles collected by dust filters, and an example of such a calculation after the release of α-activity into the respiratory air.

EUR 4594 e

AUTORADIOGRAPHY AS A HELP FOR ANALYSING THE DISTRIBUTION OF α-ACTIVE ISOTOPES IN THE HUMAN BODY AFTER AN AIR CONTAMINATION by J.P. VAANE, E.M.M. de RAS and Chr. von BRANDENSTEIN

Commission of the European Communities
Joint Nuclear Research Centre — Karlsruhe Establishment (Germany)
European Institute for Transuranium Elements
Luxembourg, February — 36 Pages — 9 Figures — B.Fr. 70,—

According to the lung model of the I.C.R.P. the size distribution of the particles of an aerosol is an important parameter for calculating the distribution of radioactive dust in the respiratory tract.
This report gives a review and a short description of autoradiographic methods used to determine the distribution of α-active particles collected by dust filters, and an example of such a calculation after the release of α-activity into the respiratory air.

EUR 4594 e

AUTORADIOGRAPHY AS A HELP FOR ANALYSING THE DISTRIBUTION OF α-ACTIVE ISOTOPES IN THE HUMAN BODY AFTER AN AIR CONTAMINATION by J.P. VAANE, E.M.M. de RAS and Chr. von BRANDENSTEIN

Commission of the European Communities
Joint Nuclear Research Centre — Karlsruhe Establishment (Germany)
European Institute for Transuranium Elements
Luxembourg, February — 36 Pages — 9 Figures — B.Fr. 70,—

According to the lung model of the I.C.R.P. the size distribution of the particles of an aerosol is an important parameter for calculating the distribution of radioactive dust in the respiratory tract.
This report gives a review and a short description of autoradiographic methods used to determine the distribution of α-active particles collected by dust filters, and an example of such a calculation after the release of α-activity into the respiratory air.

EUR 4594e

COMMISSION OF THE EUROPEAN COMMUNITIES

AUTORADIOGRAPHY AS A HELP FOR ANALYSING THE DISTRIBUTION OF α-ACTIVE ISOTOPES IN THE HUMAN BODY AFTER AN AIR CONTAMINATION

by

J. P. VAANE, E.M.M. de RAS and Chr. von BRANDENSTEIN

1971

Joint Nuclear Research Centre
Karlsruhe Establishment - Germany
European Institute for Transuranium Elements

ABSTRACT

According to the lung model of the I.C.R.P. the size distribution of the particles of an aerosol is an important parameter for calculating the distribution of radioactive dust in the respiratory tract.
This report gives a review and a short description of autoradiographic methods used to determine the distribution of α-active particles collected by dust filters, and an example of such a calculation after the release of α-activity into the respiratory air.

KEYWORDS

RADIOAUTOGRAPHY
DISTRIBUTION
ALPHA PARTICLES
BODY
AIR POLLUTION
LUNGS
AEROSOLS
RESPIRATORY TRACT
DUST
FILTERS

I N D E X

AUTORADIOGRAPHY AS A HELP FOR ANALYSING THE DISTRIBUTION OF α-ACTIVE ISOTOPES IN THE HUMAN BODY AFTER AN AIR CONTAMINATION *)

1. Introduction

The fields for application of autoradiographical methods and their techniques have been described in detail in the work of A.W. Rogers [1] .

One application found for autoradiography in Radiation Protection is the analysis of aerosol filter samples from controlled areas in Nuclear Physics Centres, whereby the radioactivity and size of individual particles of a radioactive aerosol can be measured by autoradiographical methods [2, 3]. In the event of such an air contamination in the respiratory compartments, the radioactive deposits can be estimated with the help of the "Lung Model" developed by the International Committee on Radiological Protection (ICRP) [4] . This model describes the deposition in the respiratory tract as a function of the size distribution of the particles and of their chemical and physical properties.

Dust samples in α-radioactive laboratories are usually taken by drawing air in through filters and particles present in the air are then trapped on or between the fibres. The first measurement of this type of captured radioactivity is usually made electronically. However autoradiography of the filter can be

*) Manuscript received on 21 July 1970

a thousand times as sensitive. The latter method is more time consuming and more complicated but in return permits the size and size distribution as well as activity of the particles to be determined. Often radioactivity can only be detected by autoradiography because of its greater sensitivity.

This report gives a review and short description of autoradiographic methods used to study α-radioactive particles collected by dust filters. A method of calculating the distribution of inhaled radioactive dust, using the I.C.R.P. Lung model is described, for which the size distribution of the aerosol particles investigated by autoradiography is an important parameter.

An example of such a calculation after a release of α-activity into the respiratory air is reported.

2. Description of Autoradiographical Methods

2.1. In general

The presence of radioactive particles on dust filter samples can be investigated with X-ray and nuclear track emulsions. After contact with the radioactive samples and development, the films show blackening in the position in front of the radioactive particles. According to the type of film used the activity of individual particles and the distribution over the filter can be determined from the blackening.

On X-ray films, in general, circular blackenings are formed, whose intensity and size depend upon the radioactivity of the particle and the exposure time. These blackenings are visible without microscope, so the particle distribution on the filter can be easily examined with this method. Under very good conditions it is also possible to estimate the activity of individual particles.

With nuclear track film the size of the silver halide crystals and their concentration in the emulsion are at least an order of ten bigger than with X-ray film, so that every ß and α particle leaves a microscopically visible track when it passes through the emulsion. With autoradiography of α-active filters, which is described in this report, the tracks usually form a star. From the number of tracks in a star the activity of individual particles can be computed.

2.2. X-ray film

The construction of an autoradiograph with X-ray film requires relatively little preparation: as the dust filter sample generally consists of thin weak layers of material such as glass-fibre, it is first mounted on cardboard and for exposure brought into direct contact with the film. Highly contaminated filters are surrounded with a thin Mylarfoil (0.9 $mg.cm^{-2}$) to prevent contamination of the photographic materials. The optimum exposure time is chosen to be adequate for the activity present on the filter, and is determined for a particular filter sample - if necessary, from the results of a previous series of exposures. The times can vary from several

hours to several weeks. If there are radioactive particles present their distribution becomes visible as blackenings after development of the film (Fig.1).

Under certain conditions the activity of the particles can be computed from the spot size.

Taking a filter sample with 30 particles of known activity, we found the following relationship between particle activity integrated with respect to exposure time (Z_α) and the spot size D_F (Fig.2):

$$\ln Z_\alpha = 2.9 + 0.2 \ln D_F$$

However it was observed that with the nuclear track film 40 % of the particles investigated on the filter sample were so close together that more than one of them was contributing to one spot. One may accept that this is generally the case and indeed that with more particles on the sample this percentage is increased.

Another source of inaccuracy is the fact that as the exposure time increases, the spot-size tends to a saturation value (Fig.3). For this reason particle sizes will be underestimated for exposure times shorter than those needed for saturation.

R.J.Sherwood and D.C.Stevens [5] have been using a thin layer of α-scintillator placed between the filter sample and the film. The radioactive emission from the sample generates photons in the scintillator and so produces blackenings of the film.

2.3. Nuclear track emulsions

As already mentioned, every α-particle arriving in a nuclear track emulsion produces a track which in the developed film can be made microscopically visible. The lengths of these tracks are a function of the energy of the α-particles on entering the film, though consideration must be given to the great shrinkage of the emulsion during development. The number of tracks produced in unit time is a measure of the activity of the α-active particles. The efficiency (track count per α-decay) depends on various factors which will be analysed in section 2.3.3.

If the isotopic composition of the particles and the efficiency are known then the activity and, if the specific activity is also known, the mass of an individual particle can be calculated (Fig.4). In particular cases it is possible using a stripping film technique (2.3.1.) to make the particles themselves visible (Fig.5).

In autoradiography we distinguish between three types of nuclear track film:

- Stripping film [3]

  Here the emulsion layer lies on a glass plate from which it can be taken off and put over the filter. The filter is made transparent by impregnating it with an Araldite solution, so that the prepared sample can be microscopically examined. In many cases the dust particles themselves as well as the tracks produced in the film are made visible by this method.

- Nuclear track plates

  Here also the emulsion is fitted on a glass plate, but it cannot be taken off as a layer, so the plate has to be placed complete on the filter to be examined. With this method it is not possible to make the particles themselves visible.

- Liquid nuclear track emulsion

  By this method either the filter is dipped in the emulsion or soaked in it. The advantage of this method is that particles which have penetrated too deeply into the filter to be visible by other methods can also be examined and their penetration depths measured.

Next follows a brief description of the three above mentioned techniques.

2.3.1. Strippingfilm method

An analysis with strippingfilm is usually carried out if the distribution of activity has already been determined by the X-ray film method.

The parts of the filter which have been chosen for examination, are cut into pieces of about 1 cm^2. Each piece is then mounted on a warmed, grease-free microscope slide and impregnated with Araldite. To make sure that during the following procedure the film adheres well to the prepared sample, this is first cooled, then dipped in a gelatine solution, and finally dried.

Afterwards an appropriate piece of strippingfilm is cut in a

darkroom and floated on the surface of a water-filled tray. The prepared filter sample is then dipped into the water under the film and lifted, leaving the film lying on the sample, which is then immediately placed in a dark box and left for exposure. The exposure time is estimated beforehand from the activity or, in some cases, by the use of X-ray film.

A nuclear track microscope is used for studying the developed films.

In practice, if at least five tracks appear in a star formation, we conclude that there exists an active particle in the middle of the star. The diameter of the stars, and, on some occasions, the size of the particles are measured with a magnification of 1250 X using an immersion objective and an ocular micrometer. The penetration depth of the particle in the filter can be estimated from the difference in focal distances between the filter surface and the particle.

In many cases when the number of tracks is too large it is impossible to resolve between individual tracks. In such a case the middle of the star is also too black to enable the underlying particle to be recognized. If, however, the developed silver bromide crystals are treated with a solution of potassium ferricyanide and fixing salts in distilled water, the blackening disappears and the particle becomes visible (Fig.5).

2.3.2. Nuclear track plates

The use of nuclear track plates is appreciably simpler than that of stripping film. The active particles themselves cannot however be made microscopically visible. To prepare the sample, the piece of filter is mounted on a flat base and the nuclear track plate laid directly on and in good contact with it. Heavily contaminated filters are covered beforehand with a mylar foil to prevent contamination of the films, which have to be taken off again after the exposure and development process. During exposure the sample is placed in a completely dark box.

B.V.Andersen [6] exposes a series of nuclear track plates to the α-particles on the filter sample being investigated, with exposure times increasing in factors of ten from 12 minutes up to 2000 hours. Particles which produce more than 50 tracks have to be examined with an exposure time shorter by one or more factors of ten as it is practically impossible to count so many tracks owing to the intensive blackening. As with the strippingfilm method, the activity is calculated from the number of tracks and the exposure time. Here the α-particles are not stopped in the filter so much as in the Araldite-prepared filter with the strippingfilm method, the tracks are appreciably longer and the stars are easier to find. A magnification of 125 X is in most cases sufficient.

2.3.3. Calculation of the activity

With the nuclear track methods, using stripping film and nuclear track plates, the activity of the particles (A_α)

can be calculated from the track count (S_α) produced during the exposure time T. The efficiency

$$\eta = \frac{S_\alpha}{A_\alpha \times T}$$

is a function of the following parameters:

- Selfabsorption of α-radiation in the particles,
- Range (R_α) of the α-radiation in filter material and in Araldite,
- Penetration depth E of the particles in the filter.

The self absorption factor S can, according to Sherwood and Stevens [2], be calculated from the following formula:

$$S = \frac{O_\alpha}{A_\alpha} = \frac{3}{2} p \left(1 - \frac{1}{3} p^2\right) \text{ for } p \leq 1$$

where

$$p = \frac{\text{range of } \alpha\text{-radiation in the material of the particle}}{D_p \text{ (Size of the particle)}}$$

O_α = the count rate for α-radiation leaving the surface of the particle.

The absorption of α-radiation in particles with a density of about 10 $gm.cm^{-3}$ and diameter less than 10 μm is practically negligable [2].

If every α-particle entering the emulsion produces a track then for particles with diameter ≤ 10 μm, η is equal to the fractional solid angle, determined from the penetration depth in the filter E, and R_α, so that

$$\eta = \frac{1}{2} \left(1 - \frac{E}{R_\alpha}\right).$$

As both E and R_α depend on many factors, it is not really possible to give a general value for η. The penetration depth E depends upon the density of the filter materials, on the energy of the particles entering the filter and on the size of the particles. For the glass fibre filters we use, and a linear particle velocity of 30 $cm.sec^{-1}$, E varies up to 50 µm.

For the stripping film method the range R_α is determined from the absorption in Araldite and in the fibre material of the filters, and works out at about 30 µm. When using nuclear track plates R_α is merely a function of the absorption in fibre material and lies between 80 and 100 µm.

The efficiency η is therefore not a fixed constant, but depends on the autoradiographical method used.

It is, however, possible to give a mean value for each method, from which the activity of the particles can be accurately calculated to within a factor of two. This means that in respect to the diameter of the identified particle, which is proportional to the cube root of the activity, the maximum error is only 30 %.

For the stripping film method this mean value is found empirically to be $\bar{\eta} = 0.2$; for the nuclear track plate method $\bar{\eta} = 0.4$.

2.3.4. Limit of sensitivity

We define the limit of sensitivity $(A_\alpha)_{min}$ as the least activity of a particle which can be identified on an air

filter sample using the nuclear track film method when exposed for time T. If the existence of a particle is taken to be certain when it produces at least 5 tracks in a star formation, the limit of sensitivity is defined with the following function:

$$(A_\alpha)_{min} = \frac{5}{\eta \times T \times 2.2 \times 10^{12}} \text{ Ci}$$

In fig.6 the limits of sensitivity for nuclear track plates ($\bar{\eta} = 0.4$) and for stripping film ($\bar{\eta} = 0.2$) are shown as a function of the exposure time. The corresponding diameters of $^{239}PuO_2$-, $^{241}AmO_2$- and $^{242}CmO_2$ particles are set out on the ordinate for a number of activity values. It can be seen that in order to identify a $^{239}PuO_2$ particle of 0.1 /um, it must be exposed for one month; that an $^{241}AmO_2$ particle of comparable size can be identified in 13 hours and a $^{242}CmO_2$ particle of 0.1 /um in less than one minute.

3. Applications of autoradiographical methods in radiation protection practice

3.1. Introduction

An example of the application of autoradiography is described in the following analysis of an incident when plutonium particles in the form of aerosols were liberated in the respiratory air and that after a routine check it was suspected that one or more of the workers present in that particular laboratory had inhaled plutonium. Various samples (air, nasal

mucous, urine) were analysed autoradiographically and the distribution of inhaled activity in the respiratory tract was calculated. The results of these calculations were compared with the I.C.R.P.-recommended maximum permissible values for radioactivity in the body.

The calculation of the quantity ingested is based on the model of the respiratory tract for internal dosimetry (lung model) [4] recommended by the International Commission on Radiological Protection (I.C.R.P.).

This lung model is an extension of an earlier recommendation of the I.C.R.P. - also called the "simple lung model", in which the distribution of inhaled aerosols was expected to be independent of the particle sizes. In the course of the following discussion "Lung model" is understood to refer to the extended model.

The behaviour of particles after entering the respiratory tract is described in general terms in the lung model, but in the details there are still many uncertainties.

The I.C.R.P. has defined a "standard man" so that standardized physiological and biological figures may be used [7]. It must therefore be realized when interpreting the results of these calculations that deviations from the standard (different kind of respiration, influence of a cold, unknown solubility of the particles in question) exercise a considerable influence on the distribution of the respired aerosol particles.

According to the lung model the size of the particles is a decisive factor in the distribution of aerosols in the body and for this reason autoradiography can make a useful contribution to the calculation of the distribution of ingested radioactivity after air contamination.

In both of the following sections 3.2. and 3.3. we describe firstly the properties of aerosols and then the I.C.R.P. lung model.

3.2. Properties of an aerosol

An aerosol is a system in which solid particles or liquid drops with sizes in the order of /um are suspended in a gaseous medium [8]. For our purposes we are only interested in aerosols produced from solid materials.

The dimensions and shapes of the dust particles depend upon the method of production of the aerosols, in other words, on the type of work and the materials used. The sedimentation rate of the individual particles in the air, that is the speed with which they settle in still air, is a function of these parameters. This applies also to the relative deposition of inhaled particles in the particular parts of the respiratory tracts, though here the process is substantially more complicated than sedimentation in air.

For calculating the sedimentation rate, the diameter of a particle of irregular shape can be defined as the diameter of a sphere having density (ρ_p) and the same sedimentation rate.

Furthermore, according to Stoke's law, the sedimentation rates for particles of equal diameters are inversely proportional to $\sqrt{\rho_p}$. The so-called aerodynamic diameter D_a [8], also called Stokes diameter, of a particle has therefore been defined as the diameter of a sphere of unit density with the same sedimentation rate in air as the particle in question, i.e. $D_a = D_p \sqrt{\rho_p}$.

As an aerosol is composed of particles of different sizes which can be grouped into specific intervals, it is possible to determine the size frequency distribution in each case.

For much of the work with radioactive materials in glove boxes, the frequency distribution of the aerosols produced is log normal, in other words, the logarithm of the sizes form a Gaussian (=normal) distribution. For this type of aerosols the deposition of all particles is, according to the lung model, a function of the so-called activity median aerodynamic diameter (AMAD) of the aerosols [4, p.175]. This AMAD is analogous to the mass-MAD for non-radioactive aerosols in dust research.

The AMAD is defined and determined as follows (table 1):
The diameters D_i (i = 1, ...n) of a sample of n particles representing the aerosol are determined (e.g. by autoradiography) and put in order of increasing value. The activity A_i of each particle is determined. Then for i = 1, ...n, the sum

$$S_i = \sum_{j=1}^{i} Aj$$

of the activity of all the particles with $D = D_i$ is calculated and similarly the percentage P_i of the total activity of all the particles,

$$P_i = \frac{100.S_i}{\sum_{j=1}^{n} Aj} \quad \%$$

The values of P_i set out on logarithm probability paper as a function of the diameter D_i, form a straight line for aerosols with a log-normal distribution (fig.7). The activity median diameter (AMD) is then, by definition, the diameter for $P_i = 50$ %.

Further should be noted that:

$$\text{AMAD} = \text{AMD} \sqrt{\rho_p} \ .$$

The standard deviation of the AMAD is calculated for a log normal probability distribution from the difference of the logarithms of the diameters corresponding to 50 % and 16 % (or alternatively 84 % and 50 %) and is therefore the ratio of these same diameters. This is called the geometric standard deviation (σ_g).

The percentage deposition in the parts of the respiration tract is now according to the lung model a function of the AMAD and σ_g of the aerosols, the respiratory volume and frequency of the person in question, and further dependent on some physical and chemical properties of the particles (e.g. hygroscopic properties and solubility).

3.3. Short description of the lung model of the I.C.R.P.

The I.C.R.P. first gave recommendations for the calculation of the distribution of radioactive dust in the body in 1959 [7, p.33]. In that report a simple scheme for the distribution of an inhaled quantity of radioactive dust in the lungs and gastro-intestinal tract was described and for which only soluble and insoluble materials were differentiated: the size of the inhaled particles was not taken into account in this first model. Later this problem was studied more deeply by a second I.C.R.P. commission (Task Group on Lung Dynamics). The results of these studies were published in 1966 [4] and since then have become known as the "New Lung Model".

The important characteristics of this lung model are the following:

- The respiratory tract is devided into three major compartments, Nasal-Pharynx (N-P), Trachea-Bronchial tract (T-B) and Lungs (L).
- The respiration rate and the volume inhaled and exhaled per minute (Minute volume) are, for normalized circumstances (standard man [4, p.182], doing normal laboratory work) fixed at 15 complete breaths/min and 20 Litres/min. According to the lung model this holds good as a mean working rate. In addition two other figures are given respectively for higher and lower rates.
- The relative deposition of the particles of an aerosol in the particular compartments of the respiratory tracts is

amongst other things a function of their size, the respiration frequency and the minute volume of the person in question. For the deposition of all particles of an aerosol, the activity median aerodynamic diameter of the size distribution and its geometric standard deviation are essential.

The percentage deposition in the naso-pharynx and in the lungs is shown as a function of the AMAD of an aerosol in fig.8 as according to [4]. The figures apply to normal circumstances as previously mentioned. For the deposition in the trachea-bronchial-tract the lung model recommends a fixed figure of 8 %, practically independent of the particle sizes.

The processes whereby particles inhaled into the respiratory tract are distributed to the other organs of the human body are not discussed here. Fig.9 shows the deposition and excretion scheme according to the recommendations of the lung model. From the many publications on this subject we still note [9] and [10].

The following example of the calculation of an inhalation of plutonium dust after an air contamination was worked out according to the previously mentioned figures of the lung model with the help of autoradiographically measured particle sizes. An example of a case of incorporation of ^{242}Cm will be published soon [11].

3.4. Analysis of a plutonium inhalation

In a laboratory in which work was being done with a mixture of

plutonium isotopes (90 % ^{239}Pu) in the form of a metal, plutonium dust escaped into the respiratory air through a fault in its manipulation. The measurement of an air dust filter 3 m away from the location of the laboratory worker in question in the direction of the ventilation, gave a mean α-concentration in the air of 6×10^{-10} Ci.m^{-3}. From this the inhaled quantity of activity was calculated, assuming that this activity was distributed homogeneously in the air. With a standard minute volume of 20 l and a period of respiration of 10 min the inhaled activity was thus found to be 120×10^{-12}Ci.

A nasal smear taken directly after the incident contained 3×10^{-12} Ci, and a second sample, about 50 minutes after the incident, 0.2×10^{-12}Ci.

An autoradiographical analysis, using X-ray film (fig.1) shows the distribution of Pu-particles on the filter sample. With the stripping film method it was then determined that there were about 750 particles/cm^2 on the filter, with diameters lying between 0.1 /um and 0.9 /um (corresponding aerodynamic diameters 0.35 /um and 3.1 /um).

The size frequency distribution of the particles of these aerosols, calculated according to the scheme in sect. 3.2., is shown in table I and fig.7. It was determined by algebraic analysis that the frequency distribution is log-normal. The AMAD for this distribution is 1.6 /um, with a geometric standard deviation (σ_g) of 1.4; this means that 68 % of the particles have an aerodynamic diameter between $\frac{1.6}{1.4}$ = 1.1 /um and 1.6 x 1.4 = 2.2 /um (real diameter: between 3.8 /um and 7.5 /um).

With the help of the lung model, the quantity of activity deposited in the three parts of the respiratory compartment can now be calculated. For this we are assuming the following figures:

Inhaled activity	120×10^{-12} Ci
AMAD of the particles	1.6 µm
σ_g	1.4

It is further assumed that the laboratory worker in question who worked at a moderate rate during the incident, has the properties of a "standard man".

The quantities deposited in the naso-pharynx and the lung tracts are determined from the graph in fig.8. The results are shown in table II. The quantity deposited in the trachea-bronchial tract is, according to the lung model, 8 % of the inhaled activity, independent of the particle sizes.

The results calculated in this way will be discussed in brief further on and compared with the results of the corresponding measurements.

The activity measured in the nasal smear (3×10^{-12} Ci) amounted to 6 % of the calculated value for the naso-pharynx as shown in table II.

These differences could be explained on the following grounds:
- In general not all the activity is removed by a mucal sample,
- only a nasal smear was taken and not a pharynx sample,
- the removal of activity deposited in the naso-pharynx to the

gastro-intestinal tract has a half time of 4 minutes [4] and in general there are many half times between inhalation and sampling.

According to our calculation, the activity deposited in the lungs is 24×10^{-12} Ci; this amounts to 0.15 % of the ICRP recommended maximum permissible value of 16.000×10^{-12}Ci [7].

Using the lung counter, no plutonium was detected in the lungs. The limit of sensitivity of this counter, which determines the quantity of ^{239}Pu present in the lungs from the outside of the body by measuring the X-rays, is 12.000×10^{-12}Ci. This measurement therefore only proves that the activity present in the lungs is less than the maximum permissible value; the relatively insensitive method does not permit measurement of a small quantity of this order (24×10^{-12}Ci).

We have not payed any attention here to many other samples (blood, urine and faecal tests) which can give more information about the inhaled activity, as this would go beyond the scope of this article.

If however they are used as described above they are a valuable help to Medical Officers and Health physicists for the estimation of the degree of internal contamination after a radiation accident.

We want to thank Dr.Möhrle, head of the Medical Department of the "Gesellschaft für Kernforschung", Karlsruhe, for the urine and faecal analyses which were carried out in his laboratories, and Dr.Semiller, head of the Medical Department of the European Communities, for the discussion of the incident described.

Litterature

[1] A.W.Rogers; Technique of Autoradiography, Elseviers Publ.Cy. (1969)

[2] R.J.Sherwood, D.C.Stevens: Some observations on nature and particle size of airborne plutonium in the radiochemical laboratories, Harwell, A.E.R.E.-R 4672 (1964)

[3] D.C.Stevens: Location and Examination of Alpha Active Airborne Dust Particles Collected on Glass Fibre Filter Paper, A.E.R.E.-M 989 (1962)

[4] Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract: Task Group on Lung Dynamics, Report of Committee II of the International Commission on Radiological Protection, Health Physics 12, 1966, pp.173-207

[5] R.J.Sherwood and D.C.Stevens: A phosphor-film technique to determine the activity of individual particles on air sample filters, Ann.Occup.Hyg. Vol.11, 1968, pp.7-12

[6] B.V.Andersen: Plutonium aerosols particle size distributions in room air, Health Physics, 10, 1964, p.897

[7] Recommendations of the International Commission on Radiological Protection, I.C.R.P. Publ.2, Report of Committee II, Permissible Dose for Internal Radiation, 1959

[8] N.A.Fuchs, C.N.Davies: The Mechanics of Aerosols, Pergamon Press (1964)

[9] E.D.Dyson and S.A.Beach: The movement of inhaled material from the respiratory tract to blood: An analogue investigation of the new lung model,
Health Physics, 15, 1968, p.385

[10] Iral C.Nelson: Theoretical excretion of Plutonium in urine based on the new ICRP lung model, BNWL-SA-1247

[11] J.P.Vaane and E.M.M. de Ras: Analysis of a case of internal contamination with Cm-242 (will be presented at the II Congress of the "International Radiological Protection Association" in Brighton, May 1970)

Table I

Size frequency distribution computed for a sample of plutonium particles on a filter

Aerodynamic particle diameter (/um)	Number of particles	Activity (dpm)	% of total activity	Percentage of total activity attributable to particles of less than stated size
2.7 - 2.8	3	1.200	10.7	100.0
2.2 - 2.3	1	0.200	0.2	89.3
2.1 - 2.2	3	0.570	5.1	89.1
2.0 - 2.1	3	0.470	4.3	84.0
1.9 - 2.0	2	0.280	2.5	79.7
1.8 - 1.9	8	0.936	8.4	77.2
1.7 - 1.8	14	1.383	12.4	68.8
1.6 - 1.7	13	1.030	9.2	56.4
1.5 - 1.6	18	1.114	10.9	47.2
1.4 - 1.5	18	0.925	8.2	36.3
1.3 - 1.4	17	0.728	6.4	28.1
1.2 - 1.3	6	0.222	2.0	21.7
1.1 - 1.2	31	0.889	8.0	19.7
1.0 - 1.1	48	0.820	7.4	11.7
0.9 - 1.0	11	0.132	1.2	4.3
0.8 - 0.9	20	0.188	1.7	3.1
0.7 - 0.8	3	0.021	0.2	1.4
0.6 - 0.7	19	0.106	0.9	1.2
0.5 - 0.6	8	0.030	0.2	0.3
0.4 - 0.5	1	0.002	0.1	0.1
	247	11.246	100.0 %	

Table II

Computed distribution over the respiratory compartments according to the lung model

Location	Reference Fig.8	Percentage of the original respired activity	Deposited activity
Particles in exhaled air	D 2	32	38×10^{-12}
Nasopharynx	D 3	40	48×10^{-12}
Trachea-Bronchial Tract	D 4	8	10×10^{-12}
Pulmonary	D 5	20	24×10^{-12}

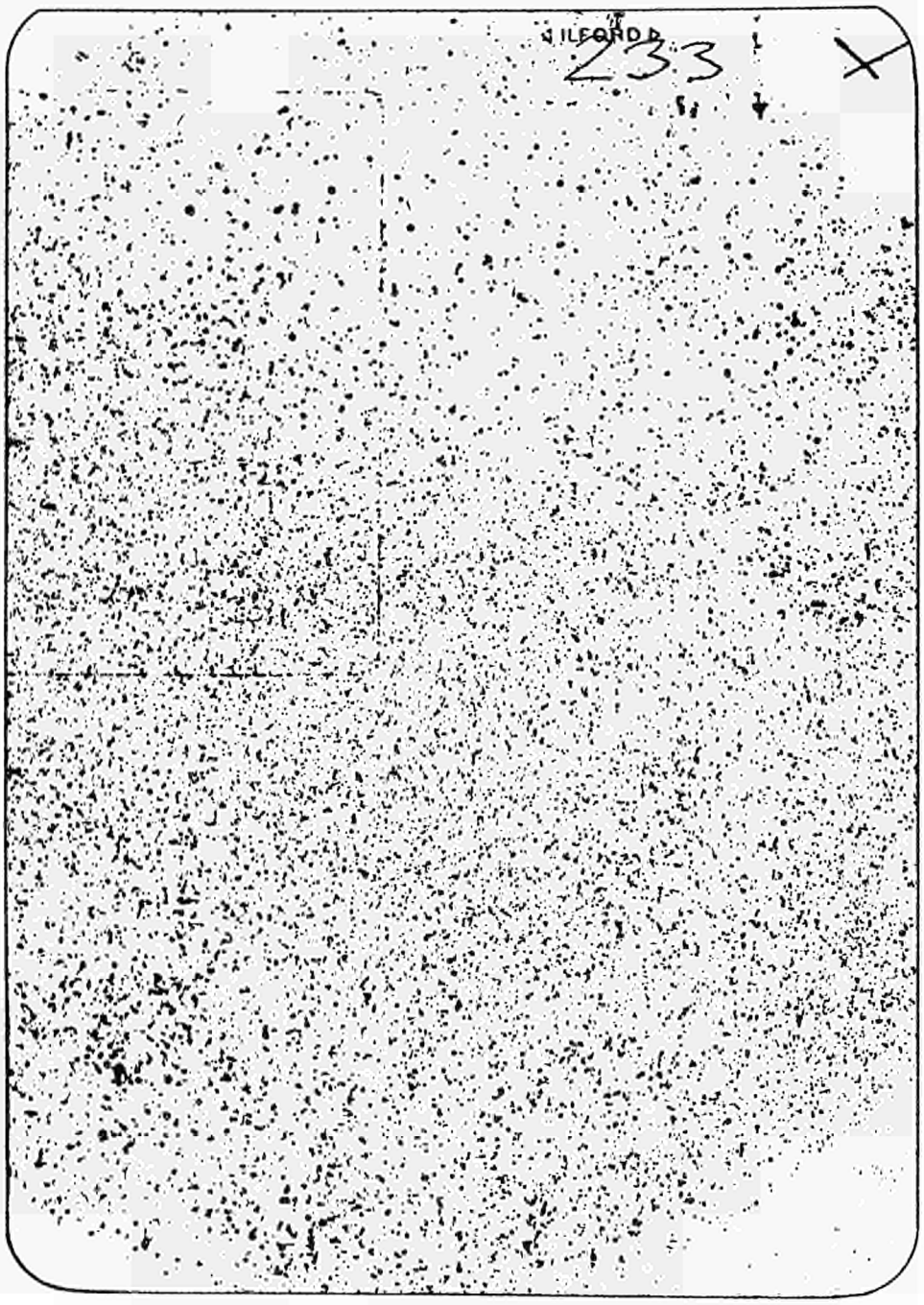


Fig. 1

Autoradiograph of an air filter sample on which there are plutonium particles, using X-ray film. The total α-activity on the filter is 0.01 μCi, the exposure time 7 days.

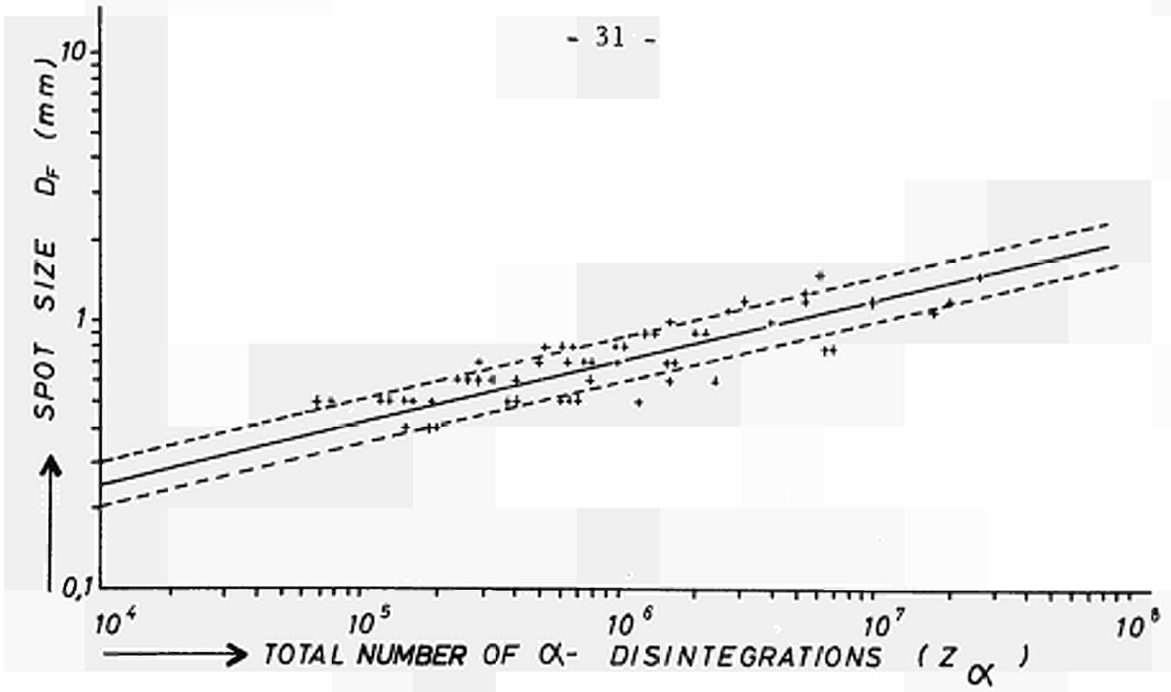


Fig. 2

The relation between spot size from an X-ray autoradiograph and the total count of α-disintegrations during the exposure time.

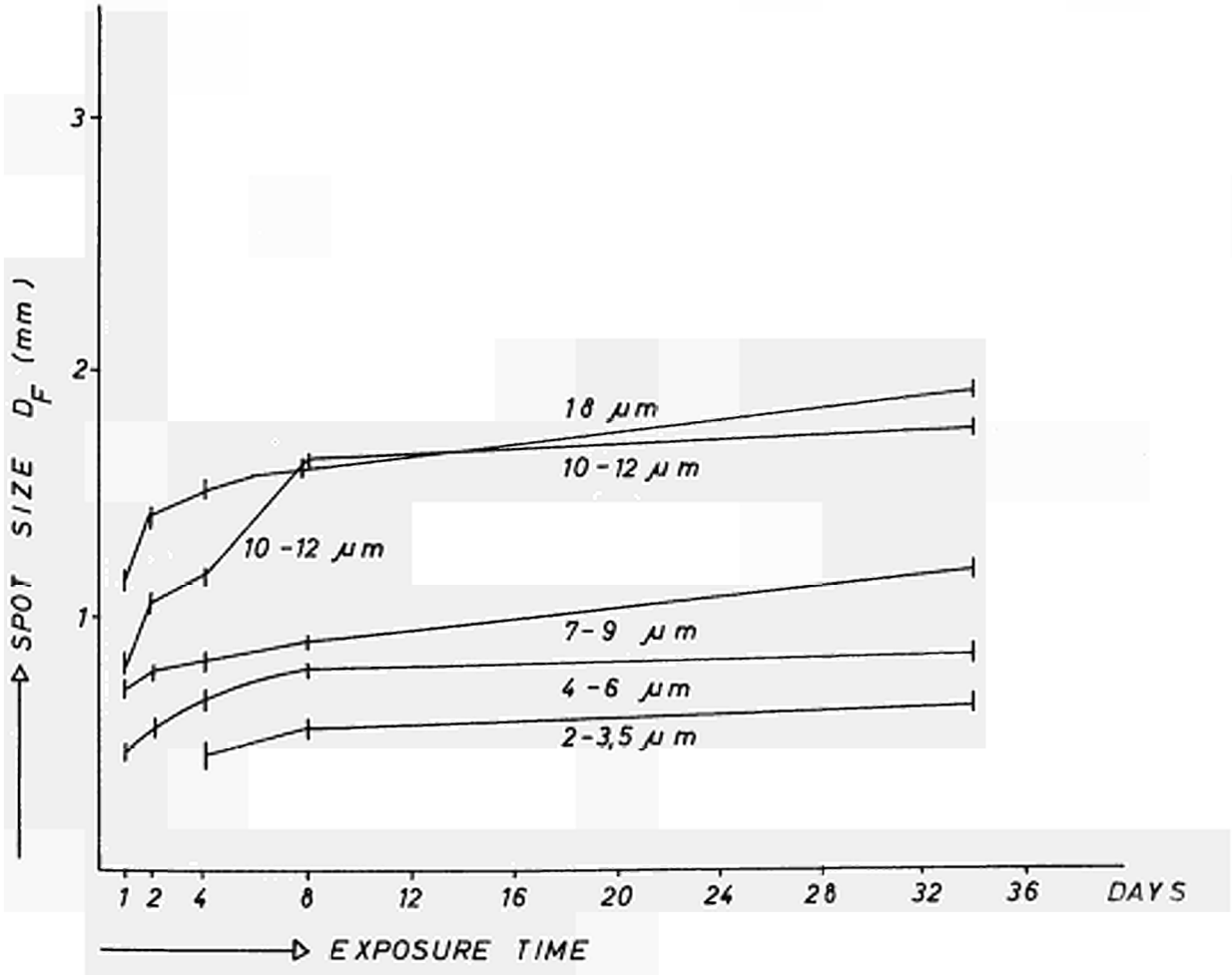


Fig. 3

Spot sizes from an X-ray film as a function of exposure time.

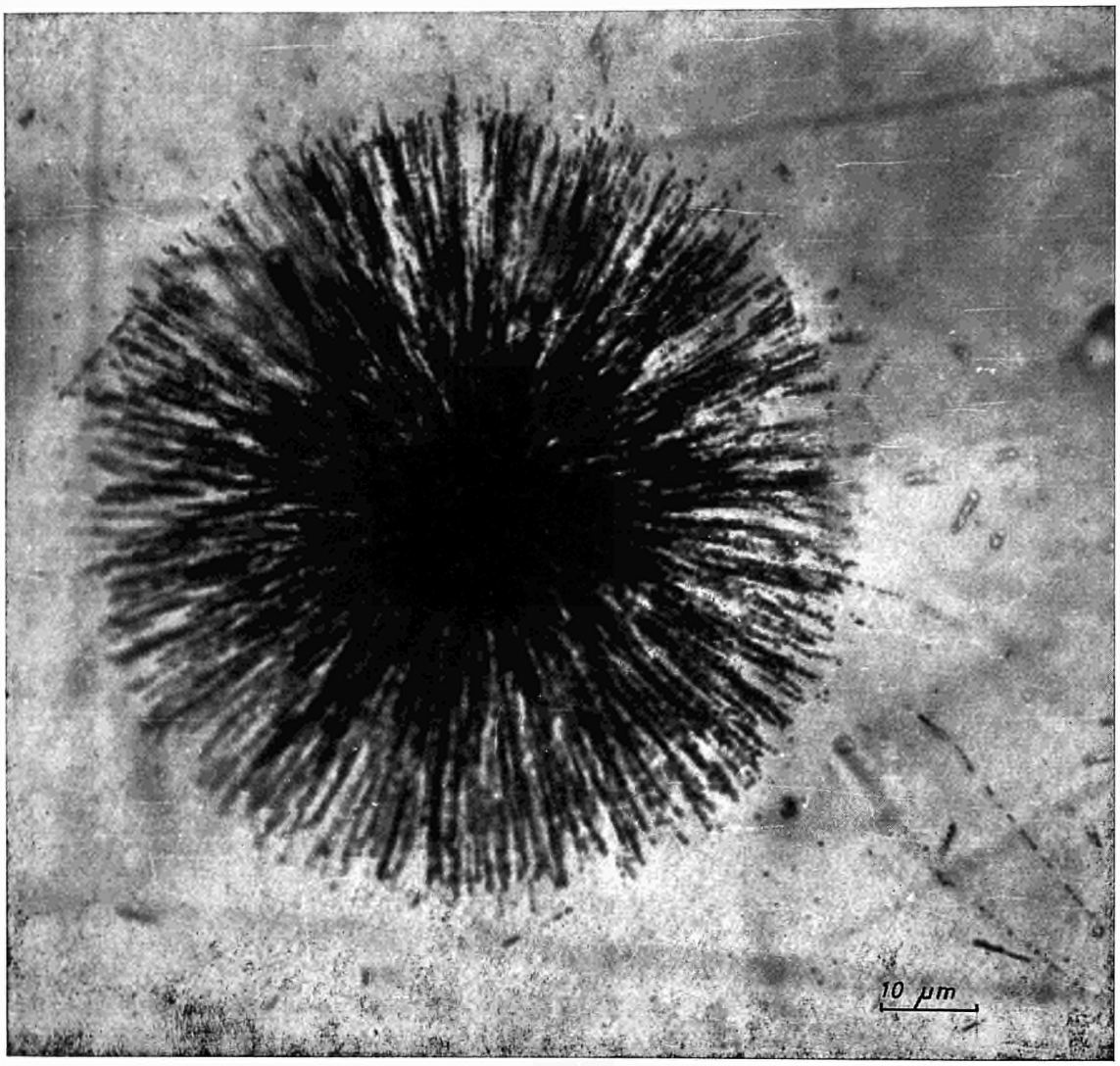


Fig. 4

Illustration of an autoradiograph with a nuclear track film. The star form shows the α-tracks of a plutonium particle (Magn. 660 X).

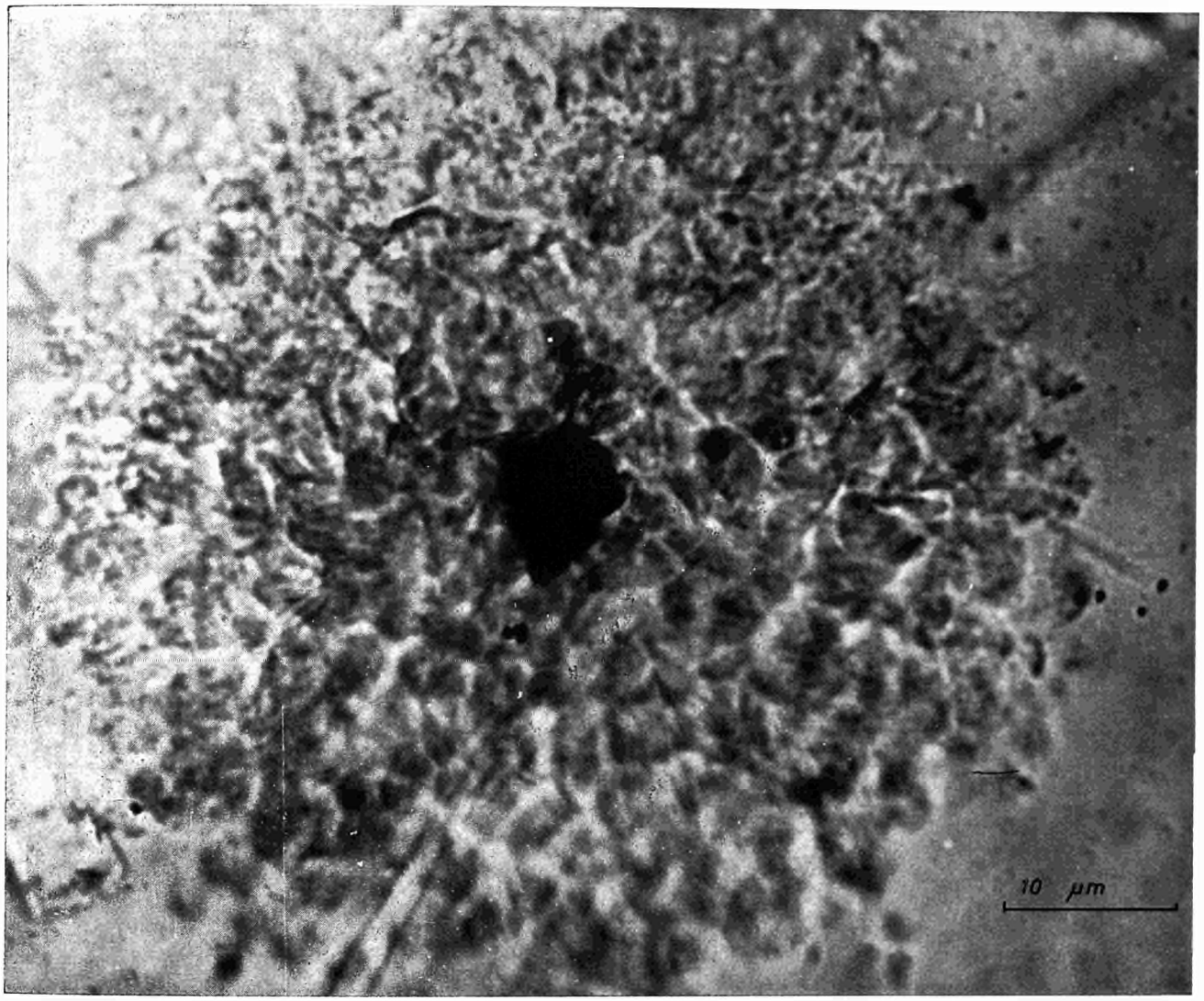


Fig. 5

Illustration of an autoradiograph of a plutonium particle. The α-tracks were chemically dissolved to make the particles visible (Magn. 1250 X).

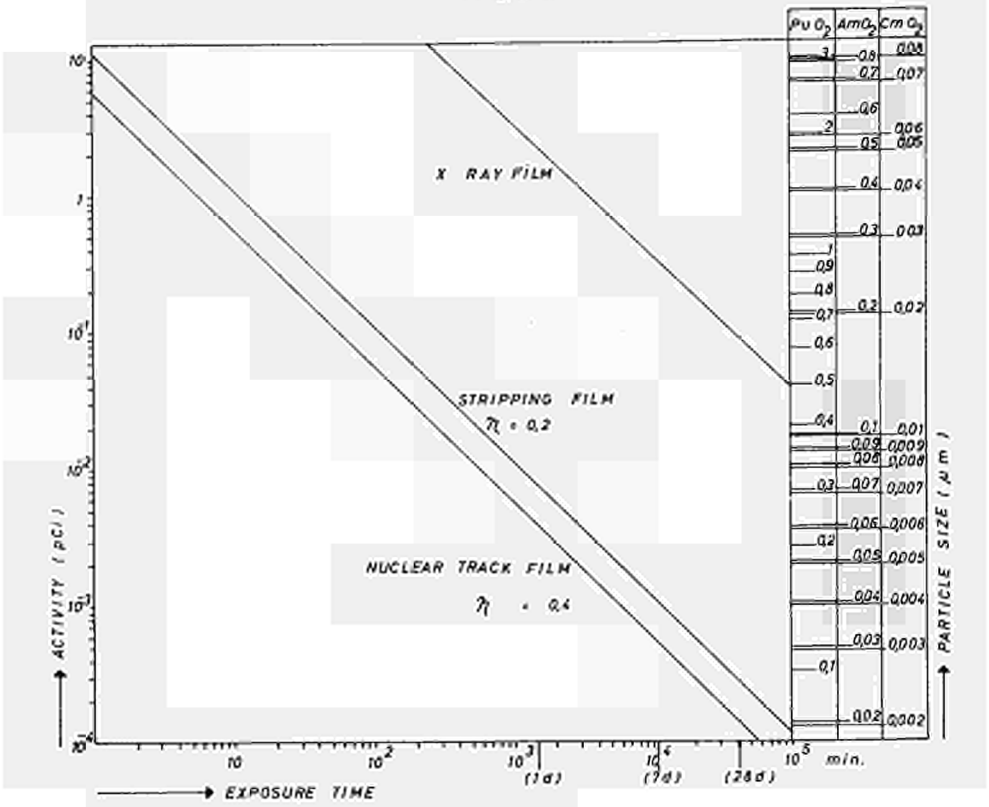


Fig. 6

The limit of sensitivity (activity of a particle which produces fice α-tracks in star formation in a nuclear track film) as a function of exposure time. The activity equivalent diameters for $^{239}PuO_2$, $^{241}AmO_2$ and $^{242}CmO_2$-particles are plotted on the right ordinate.

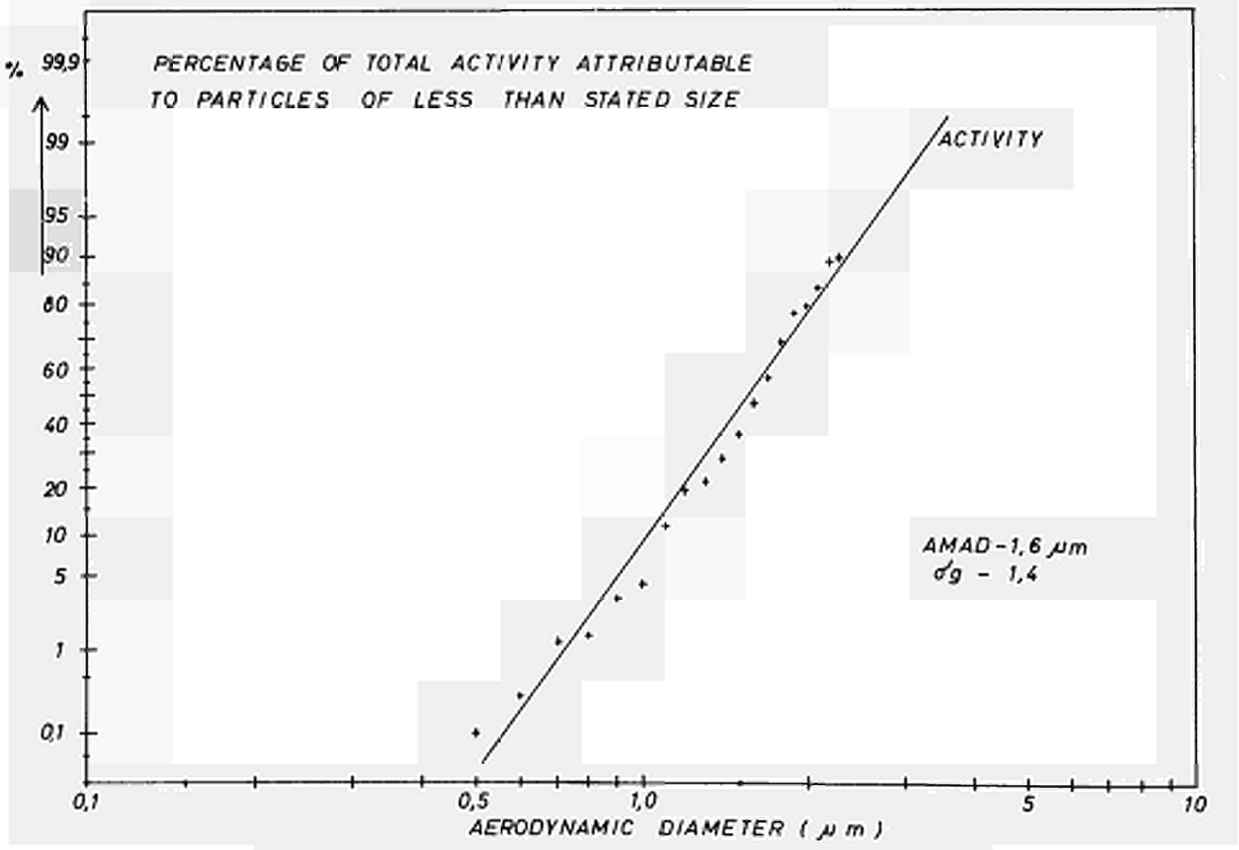


Fig. 7 Illustration of the percentage of total activity attributable to particles of less than stated size.

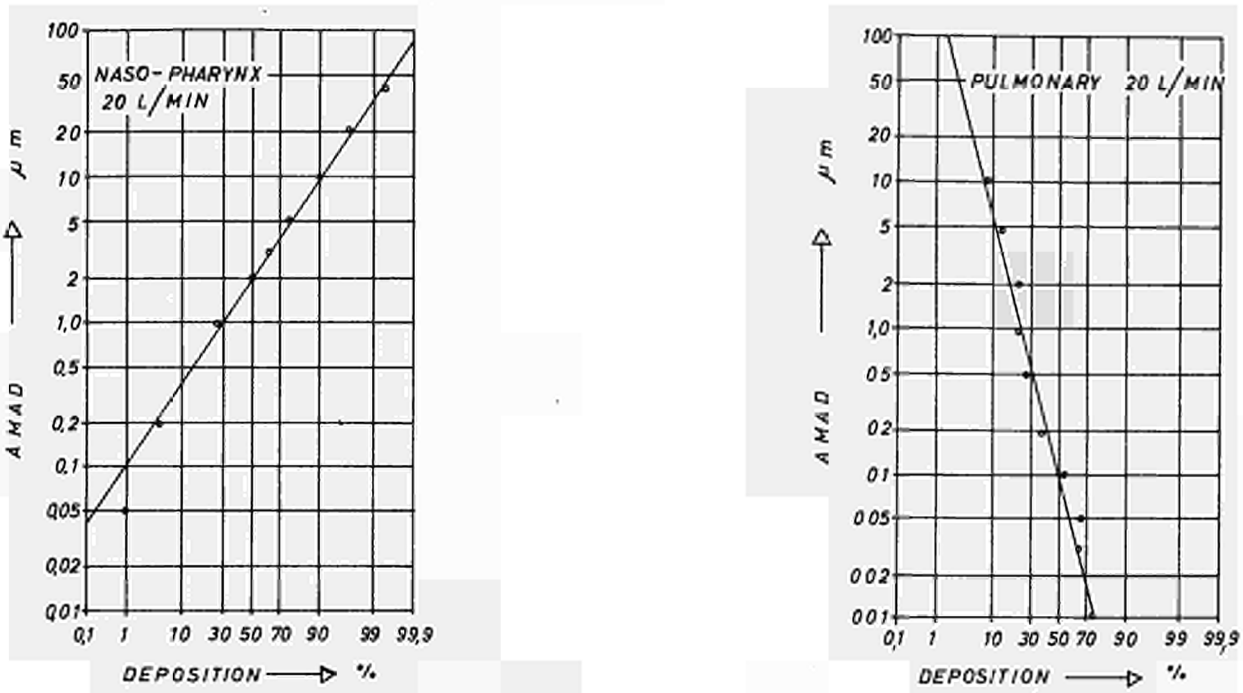


Fig. 8

Dust deposition in lungs and naso-pharynx according to the lung model for a moderate working rate. The various size distributions for the inhaled particles are shown in AMAD.

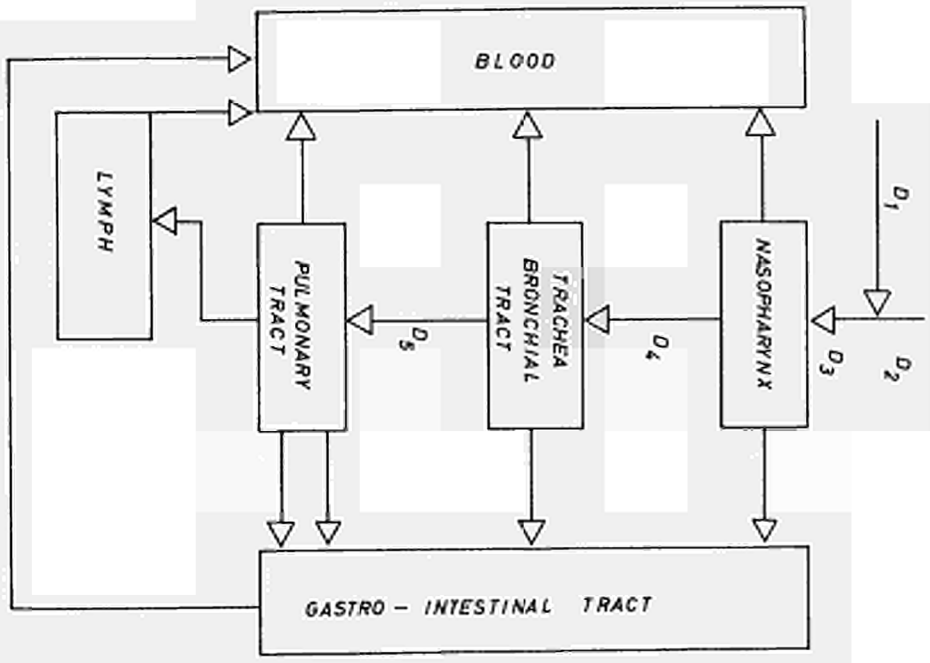
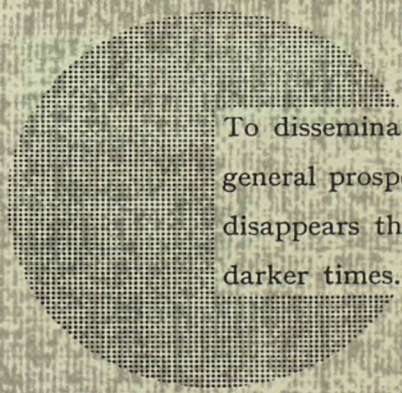


Fig. 9

Distribution scheme of an inhaled amount D_1 in the human body, according to the ICRP lung model.



To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

SALES OFFICES

All reports published by the Commission of the European Communities are on sale at the offices listed below, at the prices given on the back of the front cover. When ordering, specify clearly the EUR number and the title of the report which are shown on the front cover.

OFFICE FOR OFFICIAL PUBLICATIONS OF THE EUROPEAN COMMUNITIES
P.O. Box 1003 - Luxembourg/station
37, rue Glesener, Luxembourg (Compte chèque postal Nº 191-90)

BELGIQUE — BELGIË
MONITEUR BELGE
Rue de Louvain, 40-42 - B-1000 Bruxelles
BELGISCH STAATSBLAD
Leuvenseweg 40-42 - B-1000 Brussel

DEUTSCHLAND
VERLAG BUNDESANZEIGER
Postfach 108 006 - D-5 Köln 1

FRANCE
SERVICE DE VENTE EN FRANCE
DES PUBLICATIONS DES
COMMUNAUTÉS EUROPÉENNES
rue Desaix, 26 - F-75 Paris 15e

ITALIA
LIBRERIA DELLO STATO
Piazza G. Verdi, 10 - I-00198 Roma

LUXEMBOURG
OFFICE DES
PUBLICATIONS OFFICIELLES DES
COMMUNAUTÉS EUROPÉENNES
Case Postale 1003 - Luxembourg/gare
37, rue Glesener - Luxembourg

NEDERLAND
STAATSDRUKKERIJ
en UITGEVERIJBEDRIJF
Christoffel Plantijnstraat - Den Haag

UNITED KINGDOM
H. M. STATIONERY OFFICE
P.O. Box 569 - London S.E.1

Commission of the
European Communities
D.G. XIII - C.I.D.
29, rue Aldringer
Luxembourg