THE RELATIVE QUANTITY OF AIRBORNE PLUTONIUM DEPOSITED IN
THE RESPIRATORY TRACT AND ON THE SKIN OF RATS*

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Abstract - In assessing the hazard of exposure to airborne plutonium, questions arise as to the relative importance of material deposited within the respiratory tract and on skin. To provide relevant deposition data, shaved rats were exposed to a $^{238}\text{Pu}(\text{NO}_3)_4$ aerosol in a low-speed windtunnel. Half were oriented with noses facing upstream, the other half with noses downstream. Samples of the airstream were taken during exposure to determine the aerosol concentration and its aerodynamic equivalent size distribution. Calculations of the quantity inhaled were made from the aerosol concentration, minute volume and exposure time; skin deposition was estimated from the aerosol deposition velocity, area of exposed skin and exposure time. Immediately after exposure, the rats were sacrificed, skinned and dissected and the plutonium activity on the skin compared to that in lungs, head and intestines. Rats with their noses downstream inhaled only 66% as much as

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the upstream group. In both cases, about 40% of the inhaled material was in the lower respiratory tract, about 40% in the head and 20% in the intestines. Plutonium deposited on the skins of the two groups was almost identical, being 22 to 33% of the respiratory tract value. Measured values agreed with the calculated values within a factor of two.

INTRODUCTION

Questions have arisen as to the relative importance of airborne plutonium deposited within the respiratory tract and on the exposed skin of persons exposed to nuclear plant effluents. The skin deposit has been usually ignored on the grounds that such material would be soon washed off, whereas that deposited in the respiratory tract would have a far longer residence time within the body where virtually all of the radioactive decay energy would be effective in irradiating living tissues. In the case of plutonium compounds deposited on the skin, most of the alpha energy is absorbed in the epidermal layers. However, it has recently been pointed out that this may not be the case (Harvey, 1971). Infants are of particular concern because their epidermal layers may be thinner than those of adults, and they are more likely to have tender skin areas exposed.

In the absence of any data on this subject, we decided to perform a few calculations together with some simple animal experiments.

Theory

The quantity of airborne material inhaled by an exposed individual can be calculated if we know the aerosol concentration (C), the volume of air inhaled by the individual per unit time (MV) and the duration of the exposure (t). Thus,

$$Q_{INH} \text{ (nCi)} = C \text{ (nCi/l)} \times MV \text{ (l/min)} \times t \text{ (min)} \dots\dots\dots(1)$$

This inhaled material will be deposited in one or other of the three regions of the respiratory tract (nasopharyngeal, tracheobronchial and pulmonary) or will be exhaled without deposition, the relative quantities being a function of the aerodynamic equivalent size distribution of the inhaled aerosol and the respiration parameters of the exposed individual (ICRP Task Group --- 1966). For present purposes, the quantity of inhaled material that is in the exhaled air will be ignored since we did not measure it.

The quantity of material deposited on an exposed surface (Q_{surface}) is a function of the nature and orientation of the surface (Clough, 1973), and of the deposition velocity (V) of the aerosol. The deposition velocity is the ratio between rate of transfer of material from air to a unit surface and the air concentration, i.e.:

$$V \text{ (cm sec}^{-1}\text{)} = \frac{F \text{ (nci sec}^{-1} \text{ cm}^{-2}\text{)}}{C \text{ (nC}^{-1} \text{ cm}^{-3}\text{)}} \dots \dots \dots (2)$$

$$\text{where } F = \frac{Q_{\text{surface}} \text{ (nC)}}{t \times 60 \text{ (sec)} \times A \text{ (cm}^2\text{)}} \dots \dots \dots (3)$$

$$\text{therefore, } Q_{\text{surface}} = V \times C \times 60 \text{ } t \times A \text{ (nC)} \dots \dots \dots (4)$$

The aerosol deposition velocity is a function both of particle size and of windspeed, decreasing as the windspeed decreases and passing through a minimum value which also depends upon windspeed as particle size decreases. Tests during the calibration of our windtunnel involving the deposition of a colloidal gold aerosol at various wind velocities onto smooth surfaces oriented in various ways have

shown that V also depends upon the orientation of the surface. The deposition velocity of this aerosol for the top of a horizontal surface was found to be an order-of-magnitude greater than that for the bottom surface or onto vertical surfaces facing upstream or downstream. This applied to polydispersed aerosols having AMADs* in the range 0.7 to 1.0 μm and GSDs in the range 1.55 to 1.81, with wind velocities in the range 0.16 to 20.4 cm/sec. In other words, the aerosols were small enough and wind velocities low enough for the impaction of aerosols not to be significant in relation to deposition by thermal and eddy diffusion or by sedimentation onto horizontal surfaces.

If we assume a rat to be a right-circular cylinder, we can take the upper half of the curved surface to be horizontally upward, i.e., roughly 40% of the total surface. We can then use two values for the deposition velocity in equation (4), V_1 and 0.4 A for horizontal surfaces, and V_2 and 0.6 A for all other surfaces. The only other unknown in equation (4) is the surface area A of the exposed surface. For the rat, this is assumed to be a function of body weight, i.e.,

$$A (\text{cm}^2) = K W^{2/3} \dots\dots\dots (5)$$

where $K = 10.6$ for rats having a weight W in the range 200 to 300 grams (Biology Data Handbook, 1964).

* AMAD = activity median aerodynamic diameter

GSD = geometric standard deviation

METHODS

The design and calibration of the low-speed windtunnel, built to study the uptake and translocation of plutonium aerosols deposited on plant leaves has been described elsewhere (Craig, in preparation). The windtunnel and its associated glovebox is shown schematically in Fig. 1. The glovebox is 9 feet long and the experimental section of the windtunnel shown with rats in the filing cabinet-type drawer, is approximately 1 square foot in cross-section. This drawer seals the side of the duct, both when withdrawn to allow loading and unloading of experimental material and when closed during an exposure. Wind velocities from 0.15 to about 25 cm/sec (flows of 10 to 1415 ℓ /min) are available, and aerosols are introduced into the airstream just downstream from the air intake filter.

A $^{238}\text{Pu}(\text{NO}_3)_4$ solution, 1.113 mCi/ml in 0.27N nitric acid, was placed into the bowl of a RETEC^{(R)*} nebulizer and dispersed at a pressure of 20 psig. The flow rate in the wind-tunnel was $\sim$ 10 cm/sec (505 ℓ /min). The hair was removed from six 300 g female Wistar rats, using an Oster clipper with No. 40 blades, and they were inserted into individual wire mesh cages which allowed easy passage of the aerosol but prevented the animals from turning around or licking themselves. The rats were then placed in the drawer, three with noses pointing into the wind and three with noses pointing downwind.

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The drawer was closed to insert the rats into the windtunnel; they were exposed to the aerosol for 20 minutes. During this time, two membrane filter samples were drawn both upstream and downstream of the rats, while a cascade impactor sample (Mercer, 1970) was taken to determine the aerodynamic equivalent size distribution of the $^{238}\text{Pu}(\text{NO}_3)_4$ aerosols. All air samples were assessed by counting the 17 keV X-rays of plutonium under a thin crystal feeding into a suitably calibrated single-channel analyzer. The impactor data were analyzed using the method described by Mercer (1964), to obtain the AMAD and GSD values for the aerosol. The assumption of a log-normal distribution was subjected to a Chi-squared "goodness-of-fit" test. The mean upstream aerosol concentration, in general 15 to 20% higher than the downstream value, was calculated and used in all subsequent computations.

Immediately after the 20-minute exposure, rats were removed from their cages and sacrificed by placing them in chloroform. Each rat was skinned and the lungs, the stomach and upper third of the small intestine, and the head were removed. These four parts of the rats were ashed and digested and their plutonium content determined by the liquid scintillation technique of Keough and Powers (1970).

RESULTS

Data used to compute the total activity inhaled by the rats during the exposure and the total activity deposited on their skin are summarized in Table 1. From equation (1), the product of item numbers 1, 2 and 3 yields #4, the calculated inhaled activity:

$$\underline{\underline{Q_{INH} = 2750 \text{ nCi}}}$$

The total activity deposited on the skin is:

$$Q_{\text{surface}} = Q_{\text{horizontal}} + Q_{\text{other}}$$

where, from equation (4)

$$Q_{\text{horiz}} = 60 \text{ t C A X } 0.4 V_1$$

$$\text{and } Q_{\text{other}} = 60 \text{ t C A X } 0.6 V_2$$

$$Q_{\text{surface}} = 60 \text{ t C A } (0.4 V_1 + 0.6 V_2)$$

$$= 4.1 \times 10^5 (0.4 \times 7 \times 10^{-3} + 0.6 \times 8 \times 10^{-4}) \text{ nCi}$$

$$\underline{\underline{\text{i.e. } Q_{\text{surface}} = 1340 \text{ nCi}}}$$

The values for V_1 and V_2 are the means of results obtained in the calibration tests conducted under the same windtunnel conditions, but using a colloidal gold aerosol having almost identical AMAD and GSD values.

The animal data are summarized in Table 2, only the means for each of the two groups being given. The lung value includes material deposited in both the pulmonary and the tracheobronchial regions, except for the material that was cleared by the ciliated epithelium

and swallowed prior to sacrifice. Similarly, the head contains all the material initially deposited in the naso-pharynx that was not swallowed. The sum of the activity in the lung, the head and the intestine gives us the total initial deposition.

DISCUSSION

From Table 2, it is apparent that there is little difference between the activity deposited on the skins of the two groups. In fact, there was remarkably little scatter in the individual values ($SE = 16.2\%$). However, the total initial deposition of $^{238}\text{Pu}(\text{NO}_3)_4$ in the rats facing downstream was only 66% of that in the rats facing the other way. This is due in part to a decrease in concentration of the aerosol further downstream.

The calculated inhaled activity Q_{INH} is 20% lower than the total initial deposition measured for the rats facing upstream. In the light of uncertainties in the minute volume of the rats, this is reasonable agreement. It seems clear that very little of the inhaled aerosol is being exhaled again and most of what might normally be exhaled in the human or the dog is probably deposited in the head, i.e., roughly 40% of the total initial deposition.

The measured quantity of material deposited on the skin of these rats is about half the quantity Q_{surface} calculated on the basis of the stated assumptions. The most obvious shortcomings

are: some rats were above others and may have offered some shielding of the lower ones; the choice of a single deposition velocity for a polydispersed aerosol; and the assumption that the rat can be treated as 40% upward-facing horizontal surface and 60% other surfaces upon which deposition occurs by diffusional processes only. The relative importance of deposition on these other surfaces decreases as the particle size increases. Under the circumstances, and given the fact that shaved rat skin is very far from being a smooth surface, agreement between measured and calculated surface deposition is fairly good.

Data presented in Table 2 show that skin deposition is 22 to 33% of the total initial respiratory tract deposition for this aerosol. Since the deposition velocity onto horizontal surfaces increases from 4×10^{-3} cm/sec to 3×10^{-1} cm/sec as particle diameter for unit density spheres increases from 1 μm to 10 μm (Clough, 1973), and also increases as the wind velocity increases, the relative importance of the material deposited on the skin will vary with particle size and the wind velocity. However, it seems unlikely that the skin deposition would ever greatly exceed the total initial deposition in the respiratory tract, since larger particles will settle out close to the point of emission. The distribution of inhaled material in various regions of the respiratory tract will, of course, also change as the particle size changes.

The good agreement between calculated and experimentally determined values in this study suggest that deposition on the skin and in the respiratory tract can be reasonably estimated for varying conditions.

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Table 1. Values used to compute activity inhaled by rats and activity deposited on their skin for a $^{238}\text{Pu}(\text{NO}_3)_4$ aerosol

#	Item	Quantity
1.	Mean Aerosol Concentration	740 nCi/l (= 7.4×10^{-1} nCi cm^{-3})
2.	Assumed minute volume of rats	0.185 l/min
3.	Exposure time	20 min (= 1.8×10^3 sec)
4.	Calculated inhaled activity	<u>2750 nCi</u>
5.	Size Distribution of Aerosol	
	a. AMAD	1.03 μm
	b. GSD	1.81
	c. $\Sigma\chi^2$ for d.f. = 5	6.05*
6.	Deposition velocity	
	a. Onto horizontal surface (V_1)	7×10^{-3} cm/sec
	b. Onto vertical and other surfaces (V_2)	8×10^{-4} cm/sec
7.	Mean Body Wt (g)	286
8.	Mean Skin Area (cm^2)	460
9.	Percentage of surface of rats	
	a. Horizontal facing upwards	40%
	b. Other	60%
10.	Calculated activity on skin of rats*	<u>1340 nCi</u>

* -If $\Sigma\chi^2 < 9$, the aerosols are normally distributed with respect to the logarithm of particle diameters.

Table 2. Mean $^{238}\text{Pu}(\text{NO}_3)_4$ activity found in organs of rats exposed in windtunnel

	<u>Item</u>	<u>Rats Facing Upstream**</u>		<u>Rats Facing Downstream**</u>	
		<u>Activity nCi</u>	<u>% of Total*</u>	<u>Activity nCi</u>	<u>% of Total*</u>
1	Lung	1280	39	845	39
2	Head	1420	43	815	38
3	Stomach and Intestine	570	18	500	23
4	Total 1+2+3	3270	100	2160	100
5	Skin	705	22	715	33
6	Skin as % of Lung	55		85	

** Mean of 3 animals each.

* Total internal deposition.

TRANSVERSE SECTION OF GLOVE BOX AND AEROSOL EXPOSURE WIND TUNNEL