Localized Instantaneous Dose Rates from Inhaled Particles of $^{239}$Pu

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Abstract—In this work, the authors present instantaneous local dose rates from particles of plutonium-$^{239}$ oxide ($^{239}$PuO) embedded in various regions of the respiratory tract. For comparison, a small number of simulations were performed in a representative region of the respiratory tract with other chemical compounds including pure metallic $^{238}$Pu, $^{239}$PuO$_2$, $^{239}$PuO$_3$, $^{239}$Pu$_2$O$_5$, and $^{239}$Pu(NO$_3$)$_4$. A small number of simulations were also performed with $^{239}$PuO, weapons grade Pu, and Pu from a typical radioisotope thermoelectric generator (RTG) source for the same reason. The self-shielding effect is minor for very small particles but gradually becomes more significant as the particle size increases. For particles that are 0.1 μm and larger (excluding Pu nitrate), the calculated dose rate within several microns of the particle may be sufficient to damage lung cells, but the implications of damage to such a small volume of tissue are unclear. However, it is reasonable to assume that clinical effects will be observed if a large enough volume of tissue is damaged, as might happen when large numbers of particles are inhaled. The instantaneous dose rate around a particle may be predictive of deterministic effects, scar tissue formation, and biokinetics. 

Health Phys. 124(2):75–87; 2023

Key words: biokinetics; hot particles; microdosimetry; plutonium

INTRODUCTION

Inhalation of plutonium is a major concern for internal radiation doses wherever it is handled in significant quantities. Predicting the behavior of inhaled plutonium is essential for predicting the long-term health consequences. The risk of cancer to the respiratory tract depends on the effective dose to the lungs, and the risk is greater when the plutonium is cleared more slowly (ICRP 2019). However, the specific activity of plutonium is high enough that some inhaled particles may influence their own biokinetics via irradiation of surrounding tissues (Poudel et al. 2021a). There is evidence that plutonium may result in the formation of scar tissue in the respiratory tract even at low doses (Poudel et al. 2021b), and plutonium-induced lung fibrosis (PuLF) has also been observed in individuals with doses to the respiratory tract as small as 10 Sv (0.5 Gy) (Newman et al. 2005). Understanding the conditions under which scar tissue and fibrosis occur is of interest for predicting the dose to the respiratory tract tissues.

Inhaled plutonium is deposited as discrete particles and may remain as such long after the inhalation occurs (Romanov et al. 2020). Because the alpha particles from plutonium only travel an extremely short distance through tissue (typically less than 50 μm, as demonstrated below), it follows that the dose to the respiratory tract from inhaled plutonium may be highly heterogeneous. Histopathological studies have confirmed non-uniform distribution of plutonium in the human lung (Guilmette et al. 2002; Hahn et al. 2004; Diel et al. 2007). Furthermore, because of the short range of alpha particles in tissue, the volume of irradiated tissue is strongly dependent on the number of inhaled particles, independent of the inhaled activity and total dose. Stated differently, a given dose to the respiratory tract may be delivered at a very high dose rate from a small number of large particles to a small volume of tissue or at a very low dose rate by a large number of very small particles to a far larger volume. This suggests that the instantaneous dose rate in the immediate vicinity of inhaled particles may be more important than the total absorbed dose for predicting the occurrence of scar tissue and fibrosis.

Past work has attempted to characterize the local doses to the lungs from inhaled plutonium. For the most part, it has relied on autoradiography of excised lung tissues from animals (e.g., Diel 1978; Diel et al. 1984) and humans (Diel et al. 2007) and has used these empirical results to infer the local dose rate to tissues. We are only aware of one instance in which an attempt was made to solve the forward model by explicitly modeling alpha model transport (Caffrey et al. 2017); however, in that case, the volume sections considered (1 cm$^3$) were much too large relative to the alpha particle range to provide detailed results.

In this work, the authors present instantaneous local dose rates from particles of plutonium deposited in various regions of the respiratory tract using Monte Carlo N-Particle (MCNP), and they discuss the possible implications of their findings. These dose rates might be added and combined to predict deterministic effects in a situation in which an aerosol is inhaled, thus providing data for better establishing limits for plutonium exposures to the lungs.

MATERIALS AND METHODS

Localized dose calculations were performed using version 6.1 of MCNP (Goorley et al. 2013) with a simplified...
model of an adult human airway. MCNP is a stochastic radiation transport code that simulates the paths of individual particles through a medium specified by the user. In particular, the user specifies the geometry and chemical composition of both the medium and the source(s), as well as the details of the source term (e.g., a radionuclide with a particular decay rate that produces a specific set of emissions). The path of each particle is formed by a random walk through the media, in which each interaction between radiation and an atom/molecule is determined randomly according to known interaction probabilities (e.g., random numbers determine whether an alpha particle will interact with an oxygen molecule, and if so, whether the interaction results in elastic or inelastic scattering, the amount of energy transmitted to the molecule, and the subsequent energy and direction of the scattered particle). After computing a number of particle tracks, the individual random walks are tallied at regions of interest specified by the user to gain information about how the radiation is interacting with the model. Because the path of each particle is stochastic, many simulated particles are generally needed to obtain an acceptable uncertainty at all tally locations.

The model, shown in Fig. 1a and b, was composed of an air pathway, modeled as dry air at sea level with a density of $1.205 \times 10^{-3}$ g cm$^{-3}$, surrounded by a concentric cylinder of tissue, with a density of $1.05$ g cm$^{-3}$, as defined by the ICRP human respiratory tract model (ICRP 1994). The elemental composition and density of each material used in the models was taken from the Compendium of Material Composition Data for Radiation Transport Modeling (McConn et al. 2011).

ICRP Publication 66 (ICRP 1994) was used to determine the size of the air pathway in various regions of the adult male. The thoracic regions of the respiratory tract are divided into the bronchial (BB), bronchiolar (bb), and alveolar-interstitial (AI) regions, each of which has unique radiosensitivity and mechanical transport properties. In turn, these regions are divided into multiple "generations," each of which has its own diameter and length. This study considered a representative sample of generations of each of these regions, each of which was treated as a hollow cylinder. The respiratory generations and diameters used in this study are given in Table 1. Dose rates scaled predictably as a function of respiratory tract diameter (discussed later), so for consistency all results are shown for generation 11 of the AI region.

In each case, a Pu particle of specified chemical composition, isotope, and size was embedded half-way into the tissue, such that one hemisphere was embedded in tissue while the other hemisphere was exposed to the air pathway, as shown in Fig. 1a and b. This configuration was chosen because it simultaneously provides information about alpha penetration into lung tissue, alphas that are parallel with the tissue/airway surface, and alphas that point straight across the airway. However, it is not necessarily a physically realistic configuration for an impacted plutonium particle and should not be construed as such.

The dose rates from a particle were calculated in three regions of interest (ROI). The first region, ROI-1, was in the tissue in which the particle was embedded. The second region, ROI-2, was located at the boundary of the airway and the tissue surrounding the particle. The third region, ROI-3, was the tissue across the airway from the particle. Each ROI is identified in Fig. 1a. The ROI in all the models were modeled identically as 1-μm diameter cylinders split into either 50 or 100 1-μm-thick slices (tallies) depending on the ROI. As a self-check, both a F6 (energy deposition) and *F8 (pulse height) tally were used to calculate the absorbed dose in each tally in all ROI.

For each scenario, dose rates were calculated for 1, 10, and 100 nm particles as well as particles ranging from 1 μm to 100 μm in increments of 10 μm. Only alpha particles were considered for this simulation, as alpha radiation accounts for the large majority of the total decay energy and will always interact local to the particle. The energy of the $^{239}$Pu alpha particles was fixed at 5.101 MeV, as this is the average alpha energy emitted from $^{239}$Pu (Browne and Firestone 1986). The full alpha spectrum of $^{239}$Pu was examined, and it was determined that the difference between the average spectrum and the complete spectrum has no discernable effect on dose rate and introduces an error on the range of not more than 1 μm. The results were also found to be similar to those for the full alpha energy spectrum (scaled by activity) of $^{238}$Pu ($E_{avg} = 5.487$ MeV) (Browne and Firestone 1986). Therefore, to increase the speed at which simulations could be run, average alpha energies were used. For blends of plutonium, the energy of the alpha particles was taken to be the average of the alpha emissions. For weapons grade plutonium, this energy was 5.111 MeV, and for the plutonium blend used in the radioisotope thermoelectric generator, the average alpha energy was 5.517 MeV (Browne and Firestone 1986).

Approximately half of the simulations were performed with $^{239}$Pu oxide, as this represents a scenario in which the

| Region             | Generation | Diameter (m) |
|--------------------|------------|--------------|
| Bronchial (BB)     | 1          | 1.20 × 10⁻²  |
|                    | 3          | 0.61 × 10⁻²  |
|                    | 8          | 0.20 × 10⁻²  |
| Bronchiolar (bb)   | 11         | 0.1092 × 10⁻²|
|                    | 14         | 0.0603 × 10⁻²|
| Alveolar-Interstitial (AI) | 1 | 0.051 × 10⁻² |
|                    | 9          | 0.039 × 10⁻² |
|                    | 11         | 0.028 × 10⁻² |
particle can be expected to remain in the respiratory tract for a relatively long period of time due to low solubility. Simulations were also performed for $^{238}$PuO, as well as for a variety of different plutonium oxide compounds, a nitrate compound, and a pure metal material. Finally, simulations were performed for chemical and isotopic blends corresponding to weapons-grade plutonium (WGPu) and a typical radioisotope thermoelectric generator (RTG) source. Table 2 summarizes the simulations performed with all the particle sizes specified. All simulations were conducted assuming $^{239}$Pu unless noted otherwise.

Mechanical transport and chemical dissolution of the particle were not modeled. It follows that the calculated dose rates are most likely to be correct at the moment of the initial deposition. Although instantaneous dose rates calculated for $^{238}$Pu are valid, the integrated dose to a particular region, and thus biological effects, may be different because the energy of its decay can cause particles to break up and transport themselves around the airway (Mewhinney and Diel 1983; Park et al. 1997). However, it may be assumed reasonably that the initial dose rates would be approximately 278 (the ratio of specific activities) greater around equivalent-sized $^{238}$Pu particles.

All MCNP simulations conducted were run until the Monte Carlo error was <<1% in the deepest tally to which alpha particles penetrated. Rarely, a single alpha particle would reach 1 μm deeper than all the others. In those cases, the error was controlled in the second deepest tally. This result was then modified outside of MCNP to reflect the correct activity of the hot particle given the size of the particle. In order to improve statistics, two variance reduction techniques were implemented. The first included adding a radial bias onto the particle. This was performed for particle sizes greater than 20 μm as it was found that the alpha particle range in the various plutonium substances was approximately 15–20 μm. The second variance reduction technique

![Fig. 1.](image)

**Table 2.** Summary of simulations performed for this study.

| Material   | Region | Isotopes     | Density (g cc$^{-3}$) | Number of Particles Simulated$^e$ |
|------------|--------|--------------|-----------------------|----------------------------------|
| PuO        | Multiple$^a$ | $^{238}$Pu, $^{239}$Pu | 14.0                  | >5.0E+08                         |
| PuO$_2$    | AI-1   | $^{238}$Pu   | 11.46                 | >5.0E+08                         |
| Pu$_2$O$_3$| AI-1   | $^{238}$Pu, $^{239}$Pu | 10.5                  | >5.0E+08                         |
| Pu Metal   | AI-1   | $^{238}$Pu   | 19.86                 | >5.0E+08                         |
| Pu(NO$_3$)$_4$ | AI-1 | $^{238}$Pu | 2.447                | >5.0E+08                         |
| WGPu       | AI-1   | Mixture$^d$ | 19.84                 | >1.0E+08                         |
| RTG        | AI-1   | Mixture$^d$ | 1.5                   | >1.0E+08                         |

$^a$Includes all of the regions listed in Table 1: BB-1, BB-3, BB8, bb-11, bb-14, AI-1, AI-9, and AI-11.

$^b$Only region AI-1 was modeled for $^{238}$Pu.

$^c$Composition by mass is 93.6% $^{239}$Pu, 6% $^{240}$Pu, 0.2% $^{241}$Pu, 0.14% $^{241}$Am, 0.01% $^{238}$Pu, and 0.03% $^{242}$Pu. Alphas from americium were included in the simulation, but gammas were not simulated. (Material 238, Aged WGPu; McConn et al. 2011).

$^d$Composition by mass is 82.4% $^{238}$Pu, 13.7% $^{239}$Pu, 1.9% $^{240}$Pu, 0.3% $^{241}$Pu, 0.1% $^{242}$Pu, and 1.7% $^{16}$O (Heindel et al. 1995).

$^e$The simulations were terminated when the Monte Carlo error was <<1% in the deepest tally to which alpha particles penetrated. Rarely, a single alpha particle would reach 1 μm deeper than all of the others. In those cases, the error was controlled in the second deepest tally.

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included giving the particles a directional bias toward one of the ROI. These two techniques are showcased in Fig. 2.

RESULTS

The alpha particles from $^{238}$Pu and $^{239}$Pu have a short range in tissue (less than 40 μm) and an even shorter range in plutonium (less than 20 μm). As a result, the energy released is only proportional to the cube of the diameter (i.e., the volume) of the particle for diameters up to about 1 μm. For larger particles, only energy released near the surface of the particle escapes, such that at the limit, the energy that escapes is proportional to the square of the diameter (i.e., the surface area) of the particle. As the particle size increases, the dose rate to a particular region will depend not only on the increased number of alphas that escape the particle but on the fraction of those alphas than can reach the region of interest. It follows that beyond a certain diameter, the dose rate to a particular point will no longer increase at all with particle size. The diameter at which this occurs depends on how far the alphas have to travel, and through what medium, to reach the region of interest. For a point in tissue 25 μm from the surface of the particle, the dose rate limit appears to be achieved at approximately 10 μm (see next paragraph).

In this study, all particles are assumed to be spherical, which minimizes the ratio of surface area to volume. It follows that large non-spherical particles will have relatively higher surface areas and therefore produce higher dose rates.

In ROI-1, the dose primarily comes from particles that traveled in a relatively straight line. As a result, the dose rate scales heavily with particle size between about 1 nm and 1 μm particles. However, above about 10 μm, the volume of material producing alphas that can reach the region of interest is already present. Increasing the particle size further only increases the number of particles that can reach the region of interest after scattering many times, and therefore has a smaller effect on the total dose rate. This can be seen in Fig. 3, which shows dose rates in generation 11 of the AI region. In that figure, order-of-magnitude increases in particle sizes result in very large differences in ROI-1 dose rates up to about 10 μm, beyond which only very small differences are observed. Closer inspection shows that, up to 1 μm, dose rates increase with the cube of the radius (i.e., the volume, which is proportional to activity), indicating that self-absorption is not important. Between 1 μm and 10 μm, the dose rate increases with the square (rather than the cube) of the particle diameter, indicating that only alphas from an outer ‘shell’ of the particle are able to escape, causing it to act more like a plane source. At 10 μm diameter, this outer shell becomes a reasonable approximation of an infinite plane for ROI-1, so that further increases in particle size no longer increase the dose rate.

Unlike the ROI-1 dose region, alpha particles can reach the region across the airway (ROI-3) from a wider range of angles. As a result, increasing the particle size continues to increase the dose rate at least up to 100 μm, although self-absorption continues to be important up to about 10 μm. This can be seen in Fig. 4, which shows dose rates in generation 11 of the AI region.

As might be expected, dose rate in ROI-2 is somewhat of a cross between the ROI-1 dose rate and the ROI-3 dose rate. The difference in dose rates between 100 μm and 10 μm particles can be attributed to alphas traveling a longer distance through air to reach ROI-2 from the portion of the particle that extends into the airway. There is a steep drop in dose rate beyond around 35 μm (the approximate range of plutonium alpha particles in tissue), observed only for particles smaller than 1 μm, suggesting that most of the alphas reaching ROI-2 travel through the tissue. Evidently, that is not the case for larger particles. The ROI-2 dose rates are shown in Fig. 5, which shows dose rates in generation 11 of the AI region.

For a given particle size, the dose rate is inversely correlated with the diameter of the respiratory tract. However, the nature of this correlation (i.e., the shape of the dose rate vs. depth curves) is not affected by the diameter of the respiratory tract. This relation holds true across all the dimensions of the human respiratory tract and for a range of particle sizes. This is shown in Fig. 6.

The chemical form of the plutonium has a significant effect on the dose rate around a particular particle. Less dense
materials contain less plutonium, and therefore, the dose rate will tend to be lower for a given particle size. This is somewhat offset by a corresponding decrease in self-absorption. Fig. 7 shows ROI-1 dose rates around 0.1 μm particles of different chemical forms in generation 11 of the AI region. These results indicate another mechanism (in addition to solubility) by which different chemical forms of inhaled plutonium might have a different tendency to produce deterministic effects in the respiratory tract.

Aside from specific activity, the main difference between $^{238}$Pu and $^{239}$Pu is the energy of the alpha particles. The average energy of $^{238}$Pu is approximately 5.5 MeV, compared to approximately 5.1 MeV for $^{239}$Pu (Browne and Firestone 1986). The result is that the range of $^{238}$Pu alpha particles in tissue is approximately 5 μm longer than that for $^{239}$Pu alpha particles. However, because the range of alpha particles in plutonium is not significantly different between $^{238}$Pu and $^{239}$Pu, the relationship between dose rate and particle diameter, scaled for specific activity, is expected to be approximately the same regardless of the isotopic composition of the material. In other words, the dose rate should increase with the cube of the particle diameter up to approximately 1 μm, and the square of the particle diameter up to approximately 10 μm. Fig. 8 shows the ROI-1 dose rates around 0.1 μm particles of $^{239}$PuO$_2$, $^{238}$PuO$_2$, a typical aged weapon’s grade plutonium mixture, and a typical radioisotope thermoelectric generator plutonium mixture (Heindel et al. 1995; McConn et al. 2011) in generation 11 of the AI region.

The dependence of dose rate on particle size and surface area has implications for where the dose is absorbed, as the location at which particles are deposited is strongly dependent on the size of the particle. This is shown in Table 3, which gives fractional depositions as a function of Activity Median Aerodynamic Diameter (AMAD) and Activity Median Thermodynamic Diameter (AMTD) for log-normally dispersed aerosols (Klumpp and Bertelli 2017). For a single particle, the fractional deposition values can be interpreted as the probability of that particle being deposited in a given region. Note that larger particles are much more likely to be deposited in the upper airway (ET1 and ET2, corresponding to nose, mouth, and throat), where the rate of transport into the GI tract is much higher. Large “hot particles” are therefore likely to result in large instantaneous dose rates in the upper airway but may not persist long enough for the dose to accumulate. However, it should be noted that, compared to the AI region, the combined thickness of mucous and epithelial tissues in the ET1 and ET2 regions may be similar to the alpha particle range (ICRP 1994). These tissues may effectively shield the more sensitive basal cells from irradiation with alpha particles, depending on the specific thickness of the protective layers and the depth of impaction of the plutonium particle.

![Fig. 3. ROI-1 dose rates in the lower-AI region (generation 11) from particles with different diameters. Note that particle size has a much larger effect on dose rate below 1 μm compared to the effect above 10 μm.](www.health-physics.com)
Particle size also has a significant impact on the energy spectrum of alphas that escape the particles. For 1 nm and 10 nm particles, there is essentially no self-shielding, and the energy spectrum of escaping alpha particles is similar to that from unattenuated plutonium (Fig. 9). To facilitate comparison of the spectra of different

Fig. 4. Dose rates across the airway (ROI-3) in the lower-AI region (generation 11) from particles with different diameters. Note that, unlike the ROI-1 dose, the effect of particle size is relatively consistent.

Fig. 5. ROI-2 dose rates in the lower-AI region (generation 11) from particles with different diameters. Note the steep drop in dose rate at around 35 μm for smaller particles.
particle sizes, all spectra are normalized so that the modes are equal to 1.

The effects of self-shielding start to become apparent in 0.1- and 1-μm particles, where there are still significant energy “tails” below the main emission peaks. Nonetheless, the energy peaks are still readily apparent (Fig. 10).

For particles which are about 10 μm and larger, self-shielding “smooths out” the energy emission peaks

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**Fig. 6.** Dose rates across the airway from the particle (ROI-3) as a function of the diameter of the respiratory tract from 10 μm and 40 μm particles.

**Fig. 7.** ROI-1 dose rates at 1 μm tissue depth in the Al region (generation 1) from 0.1 μm particles with different compositions. All compounds considered for this plot are 239Pu.
such that they drop off very gradually down to almost 0 keV (Fig. 11).

DISCUSSION

Comparison with past work

Past attempts to model the local dose rate around inhaled plutonium particles have largely relied on inferring local doses from empirical data using analytical expressions for the average energy transfer of alpha particles to media over some distance. For example, an autoradiography study on excised human lung tissues (Diel et al. 2007) characterized the lung dose by the degree of non-uniformity and the relative degree of irradiation to sensitive cells—particularly Clara cells and alveolar Type II cells.

Another study (Diel 1978) describes an experiment in which Syrian hamsters inhaled a 0.2 μm monodisperse aerosol of $^{238}$PuO$_2$. The animals were sacrificed several days after exposure, the lungs were excised and radiographed, and dose rates were inferred. In that study, tumor induction was found to be inversely related to the activity of the particle.

The present work serves to complement and validate the past work of Diel and others. In particular, the present

| AMAD (μm) | AMTD (μm) | ET1    | ET2    | BB    | bb    | AI    | Total |
|-----------|-----------|--------|--------|-------|-------|-------|-------|
| 0.020     | 0.01      | 0.1205 | 0.0649 | 0.0302 | 0.1889 | 0.4717 | 0.8763 |
| 0.038     | 0.02      | 0.0728 | 0.0392 | 0.0177 | 0.1261 | 0.4884 | 0.7442 |
| 0.056     | 0.03      | 0.0567 | 0.0305 | 0.0137 | 0.0992 | 0.4160 | 0.6161 |
| 0.090     | 0.05      | 0.0422 | 0.0227 | 0.0102 | 0.0722 | 0.3116 | 0.4590 |
| 0.168     | 0.1       | 0.0383 | 0.0206 | 0.0071 | 0.0457 | 0.2024 | 0.3141 |
| 0.5       | 0.332     | 0.1280 | 0.0689 | 0.0080 | 0.0220 | 0.1214 | 0.3484 |
| 1         | 0.684     | 0.2445 | 0.1317 | 0.0124 | 0.0165 | 0.1066 | 0.5117 |
| 5         | 3.511     | 0.4793 | 0.2581 | 0.0178 | 0.0110 | 0.0532 | 0.8193 |
| 10        | 7.046     | 0.4752 | 0.2559 | 0.0126 | 0.0063 | 0.0237 | 0.7373 |
| 20        | 14.117    | 0.4262 | 0.2295 | 0.0066 | 0.0025 | 0.0072 | 0.6720 |
| 50        | 35.330    | 0.3690 | 0.1987 | 0.0018 | 0.0004 | 0.0008 | 0.5708 |
| 100       | 70.686    | 0.3567 | 0.1920 | 0.0006 | 0.0001 | 0.0001 | 0.5495 |

Fig. 8. ROI-1 dose rates at 1 μm tissue depth in the AI region (generation 1) from 0.1 μm particles with different isotopic and chemical compositions. Alpha dose from $^{241}$Am in aged WG was included (see Table 2).
work does not consider issues of non-uniformity, nor does it explicitly calculate the dose to sensitive cells. However, unlike past works, this study involved a realistic physical model of particle transport, including the effects of self-absorption on the energy spectrum of alpha particles escaping a plutonium particle and how this relates to the range of tissue. This study involved more simulations involving far more particles than anything that has been done in the past and thus provides

Fig. 9. The alpha energy spectra of plutonium nanoparticles displays discrete energy peaks with little or no impact from self-shielding. To facilitate comparison, the spectra are normalized so that the modes are equal to one.

Fig. 10. The alpha energy spectra of smaller plutonium particles displays discrete energy peaks with significant impact from self-shielding. To facilitate comparison, the spectra are normalized so that the modes are equal to one.
more detailed information about the effects of particle size, chemical composition, isotopic composition, and physical configuration in the airway. Notably, one of the studies referenced above (Diel 1978) found that the average effects were similar to those calculated assuming a structureless lung of uniform density. This suggests that the present work can serve to normalize and extend prior results, e.g., in order to infer doses to sensitive cells (Diel et al. 2007).

Implications for deterministic effects

The literature provides limited information about predicting deterministic effects of alpha-emitting particles in the lungs, but it seems reasonable to assume that thresholds would involve a certain volume of tissue receiving a certain dose. Although the volume irradiated by a single plutonium particle would not be sufficient to result in observable deterministic effects, the cumulative volume irradiated by many plutonium particles can cause morbidity and mortality. Therefore, it is of interest to examine what, if any, insights this study may provide into a single particle’s contribution to the induction of deterministic effects.

A study by Scott and Peterson (2003) reported that a number of factors determine whether inhalation of weapons grade plutonium (primarily 239Pu) would induce morbidity or mortality. The factors that were discussed are the dose, dose rate, solubility, and particle size. The relative impact of each of these factors is uncertain. This study suggests that a particle’s mobility may also play a role. For larger particles, the dose rate is such that the irradiated volume would be killed even if the particle was only present for a very short time. It follows that larger stationary plutonium particles may kill the same amount of tissue for a wide range of plausible solubility values. It is likely that most of the alpha energy emitted by a low-solubility, immobile particle would be wasted on dead tissue, which would act like shielding against healthy tissue. Therefore, mobile particles, which would have constant access to healthy tissue, might have a greater ability to induce deterministic effects than stationary particles. This implies that particles of 238Pu, which are known to break up and transport themselves around the airway due to their decay energy (Mewhinney et al. 1983), may be more harmful per unit of activity than 239Pu. This could partially explain the dog data showing a higher-than-expected occurrence of pulmonary fibrosis at later time periods for lung doses less than 2 Gy from inhaled 238PuO2, which was previously associated with non-radiation-related causes (Muggenburg et al. 1996).

Scar tissue formation

Recent work suggests that the long-term retention of plutonium in the respiratory tract observed in human tissue donor samples may be the result of encapsulation of the plutonium in scar tissues (Poudel et al. 2021). The high local

**Fig. 11.** The alpha energy spectra of larger plutonium particles displays long tails due to self-shielding. To facilitate comparison, the spectra are normalized so that the modes are equal to one.
dose rates around larger particles (e.g., 10–1,000 Gy d\(^{-1}\) around 1 \(\mu\)m particles) could plausibly result in scar tissue formation. If that is the case, smaller particles of plutonium would not become encapsulated in scar tissue and should clear the respiratory tract at a rate consistent with their in vitro solubility. It is interesting to speculate that the physical and mechanical transport of inhaled plutonium may be a function of particle size. This theory could easily be tested through future analyses of donated tissues.

**Plutonium-induced lung fibrosis**

Plutonium-induced lung fibrosis (PuLF) has been observed in humans but has received much less attention and is much less understood than plutonium-induced carcinogenesis. It appears that there is some dose threshold for pulmonary fibrosis, but estimates of this threshold vary by orders of magnitude, ranging from 0.5 Gy (10 Sv) (Newman et al. 2005) to 6 Gy (Scott and Peterson 2003). The largest cohort of plutonium workers diagnosed with PuLF comes from the early days of the Mayak plutonium production facility, where 188 workers were diagnosed with PuLF (Azizova et al. 2020). Of these, 54 cases were of combined etiology (plutonium exposure plus exposure to other toxins or risk factors), and for six workers, PuLF was a primary cause of death. The total radiation dose threshold for lung fibrosis is reported to be 4.0 Gy, although approximately 20% of workers with 1–3 Gy to the lungs from incorporated Pu were diagnosed with PuLF (Azizova et al. 2020). In this cohort, it was found that the absorbed dose to the alveolar-interstitial regions of the respiratory tract was a much better predictor of PuLF than the average absorbed dose to the whole lung. Although the majority of PuLF cases resulted from a mixture of alpha and (uniform) gamma exposures, with the mean effective gamma exposure accounting for 2.1–2.9 Gy, the external gamma dose was found to be almost entirely uncorrelated with fibrosis. It is worth noting that only a small number of the Mayak workers diagnosed with PuLF actually died from it, while nearly half died from some form of cancer (Okladnikova et al. 1994; Romanov et al. 2020; Azizova et al. 2020).

Another important cohort of workers with PuLF comes from a study of plutonium-exposed workers at the Rocky Flats Processing Plant. For this study, 326 plutonium-exposed workers were compared against 194 non-exposed Rocky Flats workers with similar demographic characteristics. These workers received chest x-rays to screen for respiratory tract abnormalities. It was found that workers with cumulative lung doses greater than 10 Sv (0.5 Gy) had a statistically significantly elevated risk of an abnormal chest x-ray. However, only one of these individuals had symptoms of respiratory tract impairment (Newman et al. 2005). Because this study required living individuals to receive chest x-rays, mortality data is not available for this cohort.

**Plutonium “hot particles”**

The National Council on Radiation Protection and Measurements (NCRP) has published limits for hot particle exposure to the lungs (NCRP 1999). It is thought that the carcinogenic effects from a hot particle are less than that from a uniform exposure (Coggle et al. 1986; Lang et al. 1995). However, the effects of highly localized doses such as from alpha-emitting “hot particles” are not known. This is evident in a recent study (Caffrey et al. 2017), which published dose rates for alpha-, beta- and gamma-emitting hot particles in the lungs of rabbits. However, while the results were meaningful for beta- and gamma-emitters, the resolution of the study was not sufficient to capture the local dose distribution from alpha emitters.

In fact, there are a variety of different definitions of “hot particle,” and it is not clear that any of them are appropriate for alpha-emitters such as plutonium. However, the results of this study point to some possible definitions that might be useful in various contexts or for addressing certain questions.

First, because of self-absorption, dose rates to surrounding tissues plateau for particles larger than about 10 \(\mu\)m. Therefore, if any Pu particle is to be considered “hot,” all particles larger than 10 \(\mu\)m must be considered “hot.”

A more practical definition for “plutonium hot particle” might be related to the reliable occurrence of cell killing in the immediate vicinity of the particle. The threshold for cell killing occurs when the dose rate is such that all of the cells within the irradiated (by alphas) volume are likely to be killed. The authors do not know what this limit is, but it is difficult to imagine any cells receiving 100–1,000 Gy d\(^{-1}\), as occurs around 1 \(\mu\)m Pu particles, could survive. By this measure, a 1 \(\mu\)m Pu particle would certainly be considered “hot.”

Finally, the term hot particle implies a particle that is significantly radioactive. When the number of alpha particles escaping the particle is so low that the effects on surrounding cells are stochastic, it should not be considered a hot particle. This may also be considered the upper limit for when microdosimetry is useful. Once again, the authors do not claim to know exactly where this limit is. However, a particle that releases less than 1 alpha particle per day [e.g., a sphere of 239PuO smaller than approximately 0.1 \(\mu\)m (100 nm)] would likely not qualify as a hot particle under this definition.

All of these definitions depend on the dose rate and alpha fluence around a Pu particle. These depend on size, shape, specific activity, and self-shielding. Therefore, it would not be possible using any of the definitions above to define “plutonium hot particle” purely in terms of size.

**Plutonium microdosimetry**

In general, microdosimetry refers to a collection of techniques that are needed to characterize biological effects of discrete particles when the activity of the particle is so low that the local effects around any single particle must be treated as stochastic (Hofmann et al. 2020). Because of the
relatively high techniques of microdosimetry are likely to be very small. The activities of a 0.1 μm particle to a 1 μm particle of 239PuO2 are 1.45 decays per day and 1,450 decays per day, respectively. Therefore, for 239Pu in the chemical form, the threshold for microdosimetry might be assumed reasonably to lie within that size range. Such small particles have a very high effective surface area and in most cases are likely to dissolve into the blood plasma very rapidly. Some particles may also be small enough to pass through vascular walls without dissolution. These particles are likely too mobile and/or transient for meaningful analysis with microdosimetry. Therefore, the size window for Pu particles that could be meaningfully analyzed with microdosimetry may be quite narrow or may be limited to low-density particles.

**CONCLUSION**

Dose rates to various regions of the respiratory tract were calculated for single particles of plutonium oxide embedded in the airway. The ROI-1 dose rate increases significantly with particle size up to about 10 μm. The ROI-2 dose rate and the ROI-3 (across the airway) dose rate increases with particle size without an observed upper limit. The self-shielding effect is minor for very small particles but gradually becomes more significant as the particle size increases. The dose rate from these larger particles, which are expected to be deposited primarily in the nose, mouth, and throat, will be highly dependent upon the surface area and thus shape of the particle.

For particles that are 0.1 μm and larger (excluding Pu-nitrate), the calculated dose rate within several microns of the particle may be sufficient to damage lung cells, but the implications of damage to such a small volume of tissue are unclear. However, it is reasonable to assume that clinical effects will be observed if a large enough volume of tissue is damaged, as might happen when large numbers of particles are inhaled. This work might therefore be useful in predicting whether an individual might be at risk for deterministic effects and, therefore, be a candidate for chelation or lavage. Because of the non-linearity of the dose rate with particle size, it makes sense to attempt to estimate particle size following an inhalation event. If this is not feasible to do retrospectively, an alternative might be to determine in advance the range of particle sizes that are produced by a given activity or process. If the particles are larger than 50 μm, it also might make sense to try to determine the shapes of the particles being produced.

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