Convolution Approach to Evaluating Intake Distributions for Inhaled Plutonium Dioxide for the Stochastic Intake Paradigm

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Abstract: For airborne toxic particles, the stochastic intake (SI) paradigm involves relatively low numbers of particles that are presented for inhalation. Each person at risk may inhale a different number of particles, including zero particles. For such exposure scenarios, probabilistic descriptions are required and involve distributions of possible intakes of the toxicant of interest. Methods are presented that evaluate the conditional intake distributions for the inhalation route of exposure to airborne plutonium dioxide (PuO_2) particles. Exposure scenarios of interest relate to Department of Energy workers engaged in decontamination and decommissioning or other operations (e.g., plutonium glovebox work). Only brief exposure scenarios are considered. The conditioning is based on the number of particles inhaled and deposited in the respiratory tract. Conditional radioactivity intake distributions are presented for up to 30 PuO_2 particles that are randomly deposited in the respiratory tract. The methods used involve constructing a single-particle-associated radioactivity intake distribution and convoluting it to obtain radioactivity intake distributions for multiple-particle intakes. The single-particle-associated intake distribution accounts for particle inhalability and particle deposition efficiency based on recommendation of the International Commission on Radiological Protection and is evaluated empirically. Equations and tables are presented that evaluate unconditional intake distributions and the **probability of exceeding the annual limit on intake** (ALI) for a given exposure scenario involving the SI paradigm. In the past, the ALI has been used to control worker intake by inhalation of radionuclides using a deterministic approach in which point estimates of intake based on inhaling many particles are required not to exceed the ALI. The results presented in this paper show how a probabilistic approach can be used to control worker intake of radionuclides for SI scenarios. With this approach, the probability of exceeding the ALI can be required not to exceed a criterion value (e.g., 5%). This 5% value would correspond to a 95% probability of not exceeding the ALI.

Introduction

Currently available radiation dosimetry/health-risk models for inhalation exposure to radionuclides are based on deterministic radiation intake and deterministic radiation doses (local and global). These models are not adequate for brief plutonium (Pu) inhalation exposure scenarios related to Department of Energy (DOE) decontamination/decommissioning (D&D) or plutonium glovebox operations because such exposures can involve the stochastic intake (SI) paradigm. For the SI paradigm, small or moderate numbers of airborne, pure, highly radioactive PuO_2 particles could be inhaled and deposited in the respiratory tract in unpredictable numbers

during brief D&D or other Pu incidents. Probabilistic relationships govern intake via the respiratory tract for the SI paradigm, and intake distributions are more appropriate than point estimates of intake.

An SI-paradigm, Pu glovebox incident occurred on March 16, 2000, at Los Alamos National Laboratory (Los Alamos, 2000)**.** It involved eight workers who inhaled high-specific-activity, alpha-emitting (HSA-αE) $^{238}PuO_2$-contaminated room air (glovebox-failure incident based on current information). Consistent with the SI paradigm, each person apparently inhaled a different amount of $^{238}PuO_2$. Based on nasal swipes, the variability was large. Chelation therapy was recommended for four individuals and one of them appeared to have inhaled much more $^{238}PuO_2$ than the others. The chelation therapy was initiated within 2 hours of the incident after nasal swipes indicated possible levels of intake that would warrant the therapy. However, final estimates of intakes will be at some future time after sufficient bioassay data have been accumulated and evaluated. Researchers at Lovelace Respiratory Research Institute are assisting Los Alamos National Laboratory in characterizing the source term for the incident.

Health-risk estimation is not trivial for the SI paradigm, especially for HSA-αE $^{238}PuO_2$, as different individuals can have very different and uncertain radioactivity intakes for the same exposure duration and same incident. Rather than inappropriate point estimates of intake, dose, and risk, more appropriate probability distributions are needed. A main objective of our research in the U.S. DOE Environmental Management Science Program (EMSP) has been to develop a stochastic dosimetry/risk computer model for evaluating radioactivity intake (by inhalation) distributions for DOE workers who may inhale airborne, alpha-emitting, pure PuO_2 at DOE sites such as Rocky Flats.

Inhaling large amounts of PuO_2 (and daughters) in the nuclear workplace could lead to radiation deterministic effects such as radiation pneumonitis and pulmonary fibrosis (Okladnikova et al., 1994b). Inhaling small amounts in the workplace or from environmental contamination could lead to stochastic effects such as cancer (Tokarskaya et al., 1995, 1997; Khokhryakov et al., 1998). Thus, for years, environmental and workplace exposure to Pu has been a topic of interest to many scientists and clinicians (Diel and Mewhinney, 1983; CDH, 1990; DOE/EPA/CDPHE, 1996; Hickman et al., 1995; Hoover and Newton, 1993; Jones and Zhang, 1994; Krey, 1976; NCRP, 1996, 1999; Okladnikova et al., 1994a, b; Scott et al., 1990; Scott et al., 1993; Scott et al., 1997; Scott and Fencl, 1999; Scott, 2000; U.S. DOE, 1996).

Our research involves the conduct of basic and applied research related to developing improved radiation dosimetry/risk estimates to facilitate environmental management of Pu-contaminated sites. To date, our research has focused on the deterministic intake (DI) and SI paradigms: (1) <u>deterministic inhalation intake</u> of PuO_2-contaminated, resuspended, dust particles (up to millions of particles [Scott, 2000]); and (2) <u>stochastic inhalation intake</u> of relatively small numbers of low-specific-activity, alpha-emitting (LSA-αE) and HSA-αE PuO_2 particles by DOE workers involved in D&D operations or Pu glovebox incidents. Examples of HSA-αE particles are $^{236}PuO_2$, $^{238}PuO_2$, $^{240}PuO_2$, $^{241}AmO_2$, and $^{242}CmO_2$. Examples of LSA-αE particles are airborne oxides $^{239}PuO_2$, $^{242}PuO_2$, and $^{244}PuO_2$ (Scott et al., 1997). Here, we discuss results obtained for the SI-intake paradigm.

With the SI paradigm, some people may have intakes; others may not for the same incident, duration of exposure, and location (e.g., same room). Our research has demonstrated that for the SI-paradigm, point estimates of radioactivity intake and associated dose are highly unreliable, and instead should be replaced by distributions for possible intakes and doses (Scott et al., 1997; Scott and Fencl, 1999). With the distribution framework, each possible dose will

have an associated probability reflecting our degree of belief in that dose. Thus, our probabilistic characterization is similar in interpretation to Bayesian inference.

Particle-Associated Radioactivity Intake Distributions

Single-Particle-Associated Radioactivity Intake Distributions

Here, we summarize characterizing single-particle-associated, radioactivity intake distributions. The distributions help to evaluate radioactivity intake via inhaling multiple particles during D&D accidents or incidents such as the one that occurred at Los Alamos National Laboratory on March 16, 2000, in which a glovebox failed, releasing $^{238}PuO_2$ to room air inhaled by eight workers.

The single-particle-associated radioactivity intake distribution is indicated as $f_1(A)$, where A is the radioactivity intake associated with the single particle deposited in the respiratory tract (or a specific region or subregion) via inhalation. The subscript 1 indicates that the distribution applies to a single particle. The distribution $f_1(A)$ depends on the region of the respiratory tract, the age of the individual, the level of physical activity, and the particle size distribution. $F(d)$, where d is the aerodynamic diameter.

The product $f_1(A)dA$ gives the fraction of single-particle-associated radioactivity intakes with radioactivity in the very small interval (A, $A+dA$). The distribution $f_1(A)$ does not depend on particle presentation, W, but accounts for particle inhalability, $P_I(d)$, and particle deposition probability, $P_{Dep}(d)$ (Scott et al., 1997). In addition, both $P_I(d)$ and $P_{Dep}(d)$ depend on particle density, size, and shape.

In generating $f_1(A)$, particles were sampled from the distribution $F(d)P_I(d)P_{Dep}(d)$ of deposited particles. The diameter d of each selected particle was then converted to the equivalent volume diameter d_{ev}. The diameter d_{ev} is the diameter of a sphere with the same density and mass as the particle of interest. Use of d_{ev} facilitates evaluating particle radioactivity for irregularly shaped particles. The diameter d_{ev} is calculated using mathematical equations presented in ICRP Publication 66 (ICRP, 1994). Particle radioactivity was then evaluated based on d_{ev}.

The presentation, W, is the mean number of particles presented to a human receptor for inhalation over the period of interest, assuming one is dealing with the SI paradigm. The number of particles presented for inhalation is assumed to have a Poisson distribution. The notation $P(n|W)$ has been used to indicate the Poisson probability that exactly n airborne particles of interest are presented to an individual for inhalation during the period of interest. $P(n|W)$ is useful for conducting evaluations over many inhalations and is related to the particle availability, $P_A(d)$, which is associated with a single breath (Scott et al., 1997).

The particle availability, $P_A(d)$, represents the probability that a particle of interest will be contained in a tidal volume of air just before inhaling (Scott et al., 1997). Calculated availability of monodisperse, LSA-αE and HSA-αE PuO_2 aerosols is presented in Figure 1 as a function of particle equivalent-volume diameter, d_{ev}, when the air radioactivity concentration is 1 DAC (derived air concentration) (Scott et al., 1997). Calculations were based on a 1.3-L tidal volume and a particle density of 10 g/cm^3 (Kotrappa et al., 1972; Raabe, 1994) and ICRP Publication 30 DACs (ICRP, 1979).

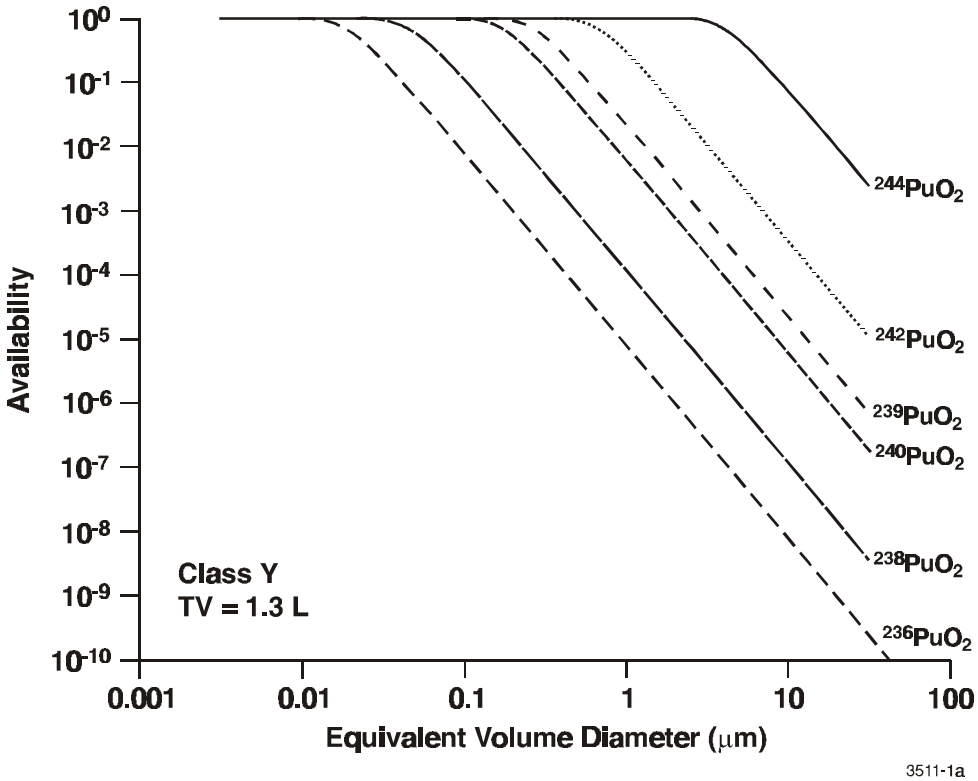

Figure 1. Particle availability, $P_A(d)$, of airborne, monodisperse, PuO_2 particles evaluated at the derived air concentration (Scott et al., 1997).

For the SI paradigm, where, at most, relatively small numbers of airborne particles are inhaled, the particle presentation, W, and associated Poisson probability, $P(n|W)$, are important in characterizing the variability of the unconditional intake or radioactivity (Scott and Fencl, 1999). Here, unconditional intake means that there is no focus on a fixed number of particles being inhaled.

Convolution Approach to Multiple-Particle Intakes

The radioactivity intake distribution for inhaling two particles of interest, sampled by inhalation from an airborne particle-size distribution, $F(d)$, is given by $f_2(A)$, where

$$f_2(A) = \int_0^A f_1(x)\, f_1(A-x)\, dx. \tag{1}$$

The radioactivity intake distribution for inhaling three particles of interest, sampled from a given particle size distribution, $F(d)$, is given by $f_3(A)$, where

$$f_3(A) = \int_0^A f_1(x)\, f_2(A-x)\, dx. \tag{2}$$

Similarly, for $n+1$ particles, the radioactivity intake distribution is given by

$$f_{n+1}(A) = \int_0^A f_1(x)\, f_n(A-x)\,dx. \tag{3}$$

Analytical solutions for $f_1(A)$ were not successfully developed. However, an empirical distribution was developed based on particle-size-dependent deposition efficiencies (adjusted for inhalability) associated with the ICRP 66 respiratory tract dosimetry model (ICRP, 1994; Jarvis et al., 1996). The convolutions for multiple-particle intake were carried out via the standard Monte Carlo method using Crystal Ball software (Decisioneering, 1996). For multiple-particle intake, separate single-particle, empirical distributions were used. For example, for two-particle intake, empirical single-particle intake distributions called *One* and *Two* were used. A particle was sampled from distribution *One* and its radioactivity was recorded. A second particle was sampled from distribution *Two* and its radioactivity recorded. Then, both particle radioactivities were added, and the sum was stored. This procedure was repeated 10,000 times with Crystal Ball Monte Carlo software. The software runs inside of Microsoft Excel, which facilitates using Excel macros. This approach yields an empirical intake distribution that varies solely because of sampling, via inhalation, from a distribution $F(d)$.

Figures 2–5 show calculated conditional intake distributions, $f_1(A)$, $f_5(A)$, $f_{10}(A)$, and $f_{30}(A)$, for intake by adults involved in a D&D accident involving brief loss of respirator protection and brief inhalation exposure to $^{238}PuO_2$.

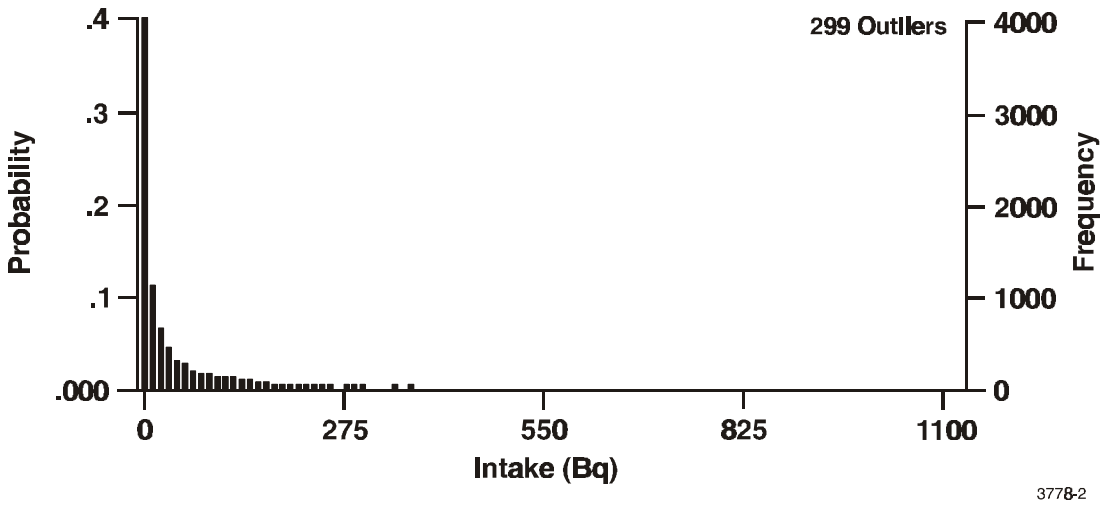

Figure 2. Conditional radioactivity-intake distribution, $f_1(A)$, for a single $^{238}PuO_2$ particle intake by adults engaged in light, work-related exercise (Scott and Fencl, 1999). The distribution was evaluated numerically based on 10,000 Monte Carlo trials, a polydisperse size distribution with an activity median aerodynamic diameter of 5 μm, and a geometric standard deviation of 2.5.

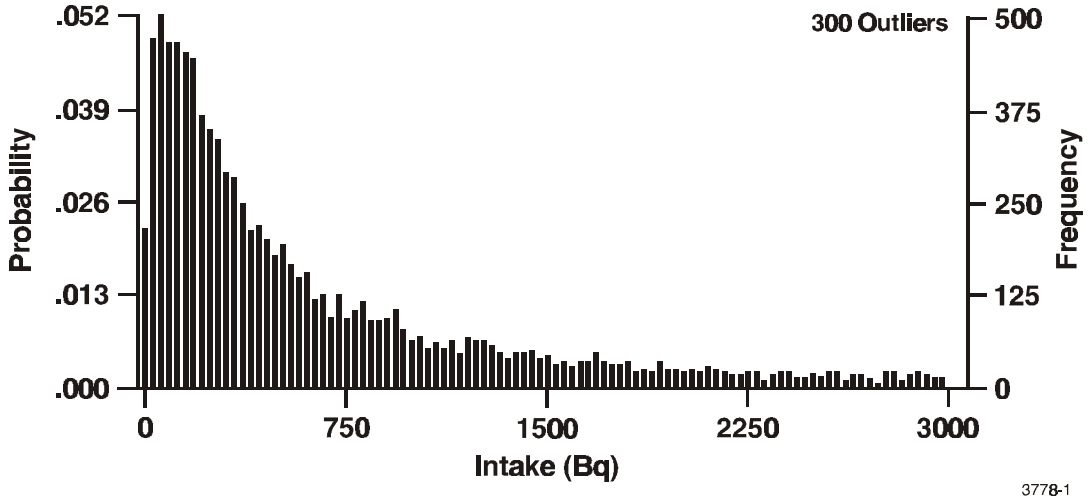

Figure 3. Conditional radioactivity-intake distribution, $f_5(A)$, for a five $^{238}PuO_2$ particle intake by adults engaged in light, work-related exercise (Scott and Fencl, 1999). The distribution was evaluated numerically based on 10,000 Monte Carlo trials, a polydisperse size distribution with an activity median aerodynamic diameter of 5 μm, and a geometric standard deviation of 2.5.

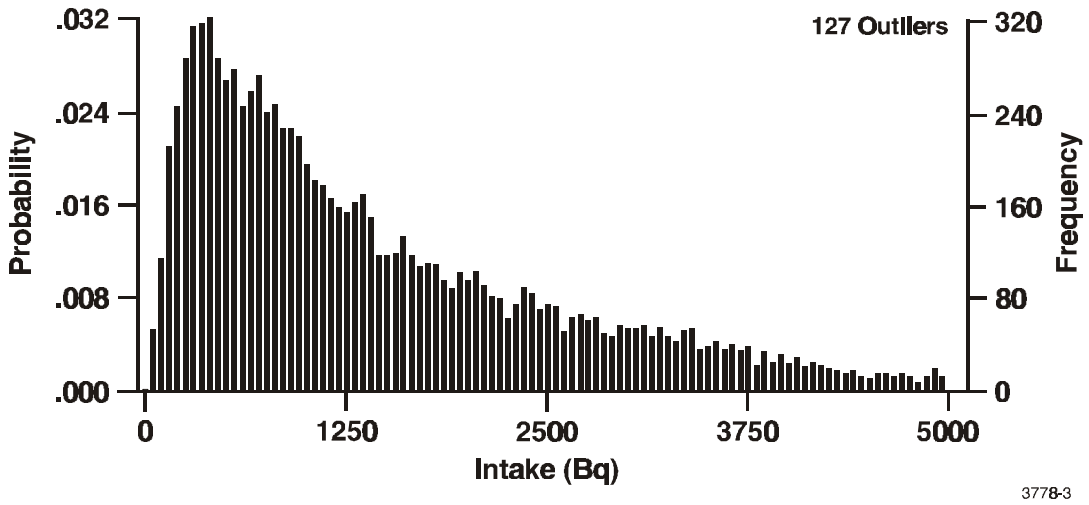

Figure 4. Conditional radioactivity-intake distribution, $f_{10}(A)$, for a 10 $^{238}PuO_2$ particle intake by adults engaged in light, work-related exercise (Scott and Fencl, 1999). The distribution was evaluated numerically based on 10,000 Monte Carlo trials, a polydisperse size distribution with an activity median aerodynamic diameter of 5 μm and a geometric standard deviation of 2.5.

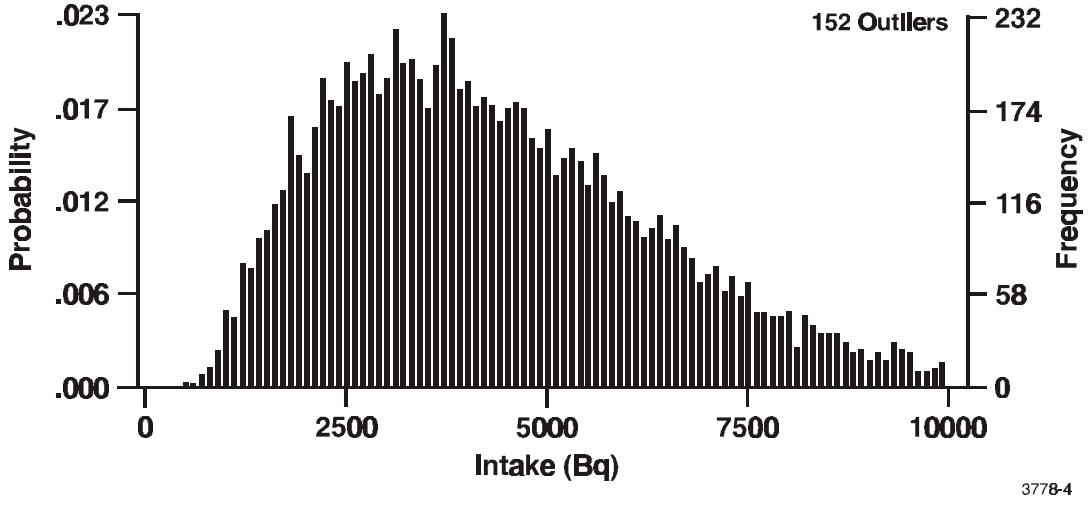

Figure 5. Conditional radioactivity-intake distribution, $f_{30}(A)$, for a 30 $^{238}PuO_2$ particle intake by adults engaged in light, work-related exercise (Scott and Fencl, 1999). The distribution was evaluated numerically based on 10,000 Monte Carlo trials, a polydisperse size distribution with an activity median aerodynamic diameter of 5 μm and a geometric standard deviation of 2.5.

Tables that summarize conditional intake distributions for adults engaged in light, work-related exercise have been constructed based on results obtained for $^{238}PuO_2$ using the

convolution approach summarized above. See Tables 1 and 2 below which were reproduced from Scott and Fencl (1999).

Table 1. Percentile level of the conditional $^{238}PuO_2$ intake distribution associated with fixed amounts of radioactivity intake (Scott and Fencl, 1999)[a]

Intake (Bq)[b]	Percent Level by Number of Particles Deposited in the Respiratory Tract of Adults[a]									
	Number of Particles									
	1	2	3	4	5	6	7	8	9	10
100	75.9[c]	53.1[d]	36.5	22.3	13.5	7.49	3.98	2.08	1.08	0.54
200	84.4	67.6	53.5	39.2	28.9	19.4	13.3	8.60	5.77	3.76
300	88.2	75.0	63.3	50.6	41.1	30.3	23.1	16.4	12.4	9.08
400	90.6	80.0	70.3	58.8	50.2	39.7	31.4	24.3	19.3	15.4
500	92.4	83.2	75.0	64.9	57.0	47.1	38.8	31.4	25.9	21.4
600	93.6	86.0	78.9	69.6	62.7	53.0	45.7	38.0	32.1	27.0
700	94.6	87.8	82.0	73.5	67.0	58.5	51.3	43.6	37.7	32.0
800	95.3	89.3	84.0	76.7	70.4	62.6	55.9	48.8	42.8	37.1
900	95.9	90.7	85.8	79.5	73.9	66.5	59.8	53.0	47.4	41.8
1000	96.5	91.5	87.2	81.6	76.9	69.6	63.2	57.0	51.5	46.3

[a]Based on 10,000 trials for each set (column) of percent-level values. Results conditional on $P(\Omega) = 1$. Respiratory tract parameters based on male adults engaged in light, work-related exercise. Multiply column 1 by $S_r/280$ to obtain corresponding Bq for other PuO_2 particles.
[b]Intake amount in Bq.
[c]This number means that 75.9% of the intake via single-particle deposition is ≤ 100 Bq.
[d]This number means that 53.1% of the intake via two-particle deposition is ≤ 100 Bq.

Table 2. Percentiles (in Bq) for the $^{238}PuO_2$ conditional intake distribution for adults (Scott and Fencl, 1999)[a]

Level %	Percentile (Bq) by Number of Particles Inhaled and Deposited[b]									
	Number of Particles									
	1	2	3	4	5	6	7	8	9	10
2.5	0.19[c]	2.46[d]	8.26	19.7	32.1	53.9	79.1	108	136	173
5.0	0.43	4.47	13.5	30.1	48.4	80.3	112	152	187	226
50.0	18.9	86.7	174	295	397	547	677	825	961	1097
95.0	746	1532	1911	2374	2588	2866	3156	3454	3619	3858
97.5	1244	2239	2523	2889	3150	3351	3729	3993	4252	4449
Other Statistics:										
Mean (Bq)	143	306	436	598	718	887	1040	1194	1332	1473
CV	2.55	1.82	1.49	1.27	1.15	1.03	0.97	0.91	0.85	0.81
Skewness	4.62	3.19	2.69	2.27	2.07	1.80	1.81	1.65	1.49	1.38
Kurtosis	28.2	14.6	11.4	9.55	7.93	6.60	7.23	6.23	5.53	5.14

[a]Based on 10,000 Monte Carlo trials per data set. Evaluated based on respiratory tract parameters for adult males engaged in light,work-related exercise with $P(\Omega) = 1$; CV = coefficient of variability; skewness = coefficient of skewness; kurtosis = coefficient of kurtosis.
[b]Multiply results by $S_r/280$ to obtain corresponding results for other PuO_2 particles.
[c]This number means that 2.5% of the intake from single-particle deposition ≤ 0.19 Bq.
[d]This number means that 2.5% of the intake from two-particle deposition ≤ 2.46 Bq.

Scaling for Different Pu Isotopes Using S_r

Each possible value for the possible intake of radioactivity has an associated probability reflecting the degree of belief in the value. Male and female differences were judged as negligible so that calculations based on respiratory tract parameters for adult males are presumed to apply (to a first approximation) to adult females. Evaluations are based on a polydisperse size distribution with an activity median aerodynamic diameter of 5 μm and a geometric standard deviation of 2.5, as recommended in ICRP publication 66 (ICRP, 1994) for nuclear workers. The results clearly indicate that considerable variability in intake by nuclear workers would be expected when small numbers of the HSA-αE $^{238}PuO_2$ particles are inhaled. Here, the variability in intake is solely related to particle polydispersity (i.e., varying sizes). The results for $^{238}PuO_2$ can easily be scaled to $^{239}PuO_2$, $^{240}PuO_2$, or $^{242}PuO_2$, based on particle relative specific activity, S_r (Table 3). The relative specific activity has been evaluated relative to $^{239}PuO_2$.

Particle radioactivity A (*in Bq*) is evaluated using (Scott et. al., 1997)

$$A \text{ in } Bq = rS_r d_{ev}^3/845. \tag{4}$$

The parameter, r, is the particle density in g/cm^3, and d_{ev} again is the equivalent volume diameter.

Table 4 gives information related to means (for 1 to 10 PuO_2 particle intakes) of conditional intake distributions for the adult population considered. Information is also provided on the coefficient of variation, coefficient of skewness, and coefficient of kurtosis for the conditional distributions.

Table 3. Relative specific activities for PuO_2 aerosols (Scott et al., 1997)

Type of Particle	Specific Activity Class[a]	Relative Specific Activity S_r (dimensionless)
$^{242}PuO_2$	LSA-αE	8.3×10^{-2}
$^{239}PuO_2$	LSA-αE	1.0[b]
$^{240}PuO_2$	HSA-αE	3.7
$^{238}PuO_2$	HSA-αE	2.8×10^{2}

[a]LSA-αE = low-specific-activity, alpha-emitting source; HSA-αE = high-specific-activity, alpha-emitting source.
[b]Corresponding absolute specific activity = 2.26×10^{3} MB/g.

Table 4. Calculated conditional PuO_2 intake means (Bq) for adults as a function of the number of particles inhaled and deposited in the respiratory tract (Scott and Fencl, 1999)[a]

Particles Inhaled[b]	$^{244}PuO_2$[c] (Bq)	$^{242}PuO_2$ (Bq)	$^{239}PuO_2$ (Bq)	$^{240}PuO_2$ (Bq)	$^{238}PuO_2$ (Bq)	$^{236}PuO_2$[c] (Bq)	CV[d]	Skewness[d]	Kurtosis[d]
1	0.00015	0.0327	0.511	1.89	143	4341	2.55	4.62	28.2
2	0.00032	0.0699	1.09	4.04	306	9289	1.82	3.19	14.6
3	0.00045	0.1	1.56	5.76	436	13236	1.49	2.69	11.4
4	0.00062	0.136	2.13	7.89	597	18123	1.27	2.27	9.55
5	0.00074	0.164	2.56	9.49	718	21796	1.15	2.07	7.93
6	0.00092	0.203	3.17	11.7	887	26927	1.03	1.80	6.60
7	0.00108	0.238	3.71	13.74	1040	31571	0.97	1.81	7.23
8	0.00124	0.273	4.26	15.78	1194	36246	0.91	1.65	6.23
9	0.00138	0.304	4.76	17.6	1332	40436	0.85	1.49	5.53
10	0.00153	0.337	5.261	19.5	1473	44719	0.81	1.38	5.14

[a]Respiratory tract parameters based on results of adult males engaged in light exercise were obtained by multiplying results obtained for $^{238}PuO_2$ by $S_r/280$. Results are conditional on $P(\Omega) = 1$.
[b]Particles inhaled and deposited in the respiratory tract.
[c]Not reported as being present at Rocky Flats but included for completeness.
[d]CV = coefficient of variability; skewness = coefficient of skewness; kurtosis = coefficient of kurtosis.

Unconditional Intake Distributions

The unconditional intake distribution is represented as *F(A)*, where

$$F(A) = P(1|W)f_1(A) + P(2|W)f_2(A) + P(3|W)f_3(A) + \ldots \quad (5)$$

The probability (Prob) of exceeding a given level of intake A* is therefore given by

$$\Pr ob(A > A^*) = \int_{A^*}^{\infty} F(A)dA. \quad (6)$$

From Equations 5 and 6, it follows that

$$\Pr ob(A > A^*) = \sum_{i=1}^{\infty} P(i \mid \Omega) \int_{A^*}^{\infty} f_i(A)dA. \quad (7)$$

Tables such as Table 1 can be used for evaluating the integral on the right-hand side of Equation 7. Equation 7 was used to evaluate the probability of exceeding the annual limit on intake (ALI) for a hypothetical PuO_2 intake scenario (Scott and Fencl, 1999). However, the equation was not presented in the cited reference.

Setting A*=ALI allows use of the ALI concept for the SI paradigm. Unlike its use for the DI paradigm, where point estimates of intake are required not to exceed the ALI, Equation 7 yields a probability. The probability could be set to a criterion value (e.g. 0.05 or 5%). With such a criterion, the probability of not exceeding the ALI would be 95%.

For multiple radionuclides, the situation is more complicated but could also be addressed using a probabilistic approach related to the sum of ratios concept used for the DI paradigm (RSALOP, 1999). Based on the intake distributions for individual radionuclides, distributions of sums of ratios could be generated. The distribution could be required to not exceed the value of 1 by a given criterion value (e.g., 5%). This would correspond to a 95% probability that the sum of ratios does not exceed the value of 1.

Conclusions

The convolution approach to evaluating intake via inhalation of airborne PuO_2 during a D&D or other PuO_2 incident was presented. The approach allows evaluation of the variability of intake of radioactivity due to sampling by inhalation of a relatively small number of PuO_2 particles (via inhalation) from a polydisperse size distribution. Considerable variability in intake was implicated for the SI paradigm. This result is consistent with real-world observations following a $^{238}PuO_2$ inhalation incident that occurred at Los Alamos on March 16, 2000, which involved eight workers. Different intakes were implied for each of the workers based on preliminary results from nasal swipes even though the workers were in the same room and were at risk for about the same amount of time. Such variability will impact on variability in organ radiation doses and associated health risks for the eight workers. The convolution approach presented will facilitate evaluating variability in intake of radioactivity, organ radiation dose, and health risks associated with inhaling PuO_2 for the SI paradigm. A probabilistic approach to

controlling worker exposure based on the ALI was introduced and recommendations for dealing with multiple radionuclides were presented.

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