

**DEVELOPMENT OF MONTE CARLO METHODS FOR INVESTIGATING
MIGRATION OF RADIONUCLIDES IN CONTAMINATED
ENVIRONMENTS**

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ABSTRACT

This paper presents the results of an ongoing research and development project conducted by Russian institutions in Moscow and Snezhinsk, supported by the International Science and Technology Center (ISTC), in collaboration with the University of Oklahoma. The joint study focuses on developing and applying analytical tools to effectively characterize contaminant transport and assess risks associated with migration of radionuclides and heavy metals in the water column and sediments of large reservoirs or lakes. The analysis focuses on the development and evaluation of theoretical-computational models that describe the distribution of radioactive wastewater within a reservoir and characterize the associated radiation field as well as estimate doses received from radiation exposure. The analysis focuses on the development and evaluation of Monte Carlo-based, theoretical-computational methods that are applied to increase the precision of results and to reduce computing time for estimating the characteristics the radiation field emitted from the contaminated wastewater layer. The calculated migration of radionuclides is used to estimate distributions of radiation doses that could be received by an exposed population based on exposure to radionuclides from specified volumes of discrete aqueous sources. The calculated dose distributions can be used to support near-term and long-term decisions about priorities for environmental remediation and stewardship.

INTRODUCTION

The use of theoretical-computation methods to estimate the transport of contaminants and assess risks to human health can help inform decision-making about remediation and stewardship priorities [1 – 2]. However, because conventional analyses underestimate the uncertainty associated with estimates obtained from observational data, they can be sensitive to the range of parameter values [3]. The use of Monte Carlo methods provides a means for the formal incorporation of uncertainty about parameters into risk assessment [4]. In this paper, we apply Monte Carlo methods to investigate the migration of radionuclides in contaminated environments. Specifically, we focuses on the development and evaluation of theoretical-computational models that describe the distribution of radioactive wastewater within a reservoir and

characterize the associated radiation field as well as estimate doses received from radiation exposure.

Since the 1940s, a large volume of radioactive wastes including high level waste from reprocessing, spent nuclear fuel, transuranic waste (TRU), and mixed wastes has been generated and released in the Russian Federation and other newly independent states of the former Soviet Union. In some cases, because radionuclides and heavy metals in those waste streams have contaminated the water column and sediments of large reservoirs and lakes, simplified turbulence diffusion models that use a distribution function to describe contamination averaged by lake depth have been used to estimate radionuclide migration [5]. In most natural lakes, however, the depth is much less than the width of the lake.

In this paper, we present the results obtained for a homogeneous reservoir in which the rate of mixing inside the reservoir is much greater than the rate of water interchange external to the system. The average contaminant concentration is identical on all horizontal planes of the reservoir, but it can differ vertically (by depth). The precipitation of radioactive sediment that was once suspended in the water column can be both irreversible and convertible. To account for ^{137}Cs penetration in an aqueous environment, we monitored the migration of the contaminated wastewater layer as it dispersed and settled to the bottom of the reservoir and estimated the potential radiation dose as information relevant for the purposes of planning remediation strategies [6]. The distribution of radionuclides between the solid and liquid phase is important when describing the behavior of the radionuclides. For ^{137}Cs , the value of this distribution parameter in fresh water reaches $\sim 10,000$, therefore, a large fraction of radionuclides in the wastewater layer is combined with the suspended sediments [7]. Different turbulence diffusion models that simulate the transport of radionuclides in contaminated environments must be utilized to account for the precipitation of sediments from wastewater.

ADVANTAGE OF A MONTE CARLO METHOD

As noted above, there many problems in which analog methods of simulating physical process are computationally inefficient, and occasionally, yield erroneous results. Calculating the radiation field characteristics over large distances from a contaminated source may result in an increasing estimate of variance. This behavior is defined as a lifetime expansion of particles (rays) and essential deviation of the distribution of the sampling average on the number of histories from a normal distribution [8]. The distribution $f_N(x)$ of sampling average $\overline{x_N}$ on a small number of the histories N occurs as an asymmetric relative expected value Mx , so that the most probabilistic values are $x_N < Mx$. The gradual decrease of $f_N(x)$ at $x < Mx$ maintains the conservation of the mean value, and the reduced distribution more often results in an underestimation. Using Monte Carlo methods to estimate the radiation field characteristics over the irregular geometry of large reservoirs or lakes provides a more efficient technique to incorporate the large outlier values that occasionally influence the distribution and provides a more accurate estimate.

DEVELOPMENT OF MODIFICATIONS OF A MONTE CARLO METHOD

For many radiation physics problems, we have designed effective non-analog modifications of a Monte Carlo method based on substituting "altered" distributions in place of real physical distributions [9]. The "biased" cross-section Σ' in the probability density function for the free path t is given by Equation 1:

$$p'(t)dt = \Sigma' \exp(-\Sigma' \cdot t)dt, \quad (\text{Eq. 1})$$

Equation 1 minimizes the variance of a random variable $e^{\Sigma \cdot r} / r^2$, proportional to the contribution of the next scattering point to the point detector (*MD*-method).

We introduce a weight factor:

$$W_1 = \frac{\Sigma}{\Sigma'} \cdot \exp[-(\Sigma - \Sigma') \cdot t], \quad (\text{Eq. 2})$$

In Equation 2, Σ is the real cross-section of photon interaction in matter.

The cross-section Σ' is continuously carried out on a particle history sequence. This cross-section is defined by Equation 3:

$$\Sigma' = \Sigma - \frac{k \cdot m}{2} + \sqrt{\left(\Sigma - \frac{k \cdot m}{2}\right)^2 + k}; \quad \Sigma' \leq 4\Sigma, \quad (\text{Eq. 3})$$

where the arguments $k = \frac{2}{R \cdot \sin \theta} \cdot \left(\Sigma + \frac{2}{R \cdot \sin \theta}\right)$ and $m = R \cdot \cos \theta$ that correspond to the geometry of a location of represented by S_n , the scattering point and a pointed detector (Figure 1).

The appropriate contributions of the particles (rays), which scattered near a point detector a great distance from a source, are precisely valued. This simulation results in an optimal choice of assumed trajectories. The algorithm of a *MD*-method can be included by a conventional manner in the common scheme of a flux-at-a-point estimation.

Further development of the computational algorithm was carried out by revision of the flux-at-a-point estimation, utilizing "altered" representation of a collision kernel of Boltzmann transport equation. This modification applies a biasing of the polar scattering angle based on the common concept of a reduced variance of the estimated result [10].

The following expression represents the single-scattered photon radiation:

$$I' = \frac{N_e}{R \cdot \sin \theta} \int_0^{\pi-\theta} E' \cdot \frac{d\sigma}{d\Omega}(E, \theta + \varphi) \cdot \exp\left(-\frac{R}{\sin(\theta + \varphi)} [\Sigma(E) \cdot \sin \varphi + \Sigma(E') \cdot \sin \theta]\right) d\varphi \quad (\text{Eq. 4})$$

Where:

N_e - electron density of a matter;

$\frac{d\sigma}{d\Omega}(\bullet)$ - Klein-Nishina differential cross-section;

$\Sigma(E)$ and $\Sigma(E')$ - the cross-sections for energy before and after a scattering accordingly.

We have included a singularity of $1/\sin\theta$ into a density function. We then received the following distribution of a density scattering function into a solid angle $d\Omega$:

$$p'(\vec{\Omega}) = \frac{1}{\pi^3} \cdot \frac{\pi - \theta}{\sin \theta} \cdot \sin \theta \cdot d\theta \cdot d\varphi, \quad (\text{Eq. 5})$$

along a conditional weight factor

$$W_\Omega = \frac{\pi^3 \cdot \frac{d\sigma}{d\Omega}(\theta_1) \cdot \sin \theta}{\sigma_s \cdot (\pi - \theta)} \quad (\text{Eq. 6})$$

In this case, the sampling of a polar angle of scattering θ is described by the following formula:

$$\theta = \pi \cdot (1 - \sqrt{\alpha}), \quad (\text{Eq. 7})$$

where α is a random number uniformly distributed on the interval (0,1). Using these modifications has considerably increased the capability of a computational algorithm that uses a Monte Carlo method [9].

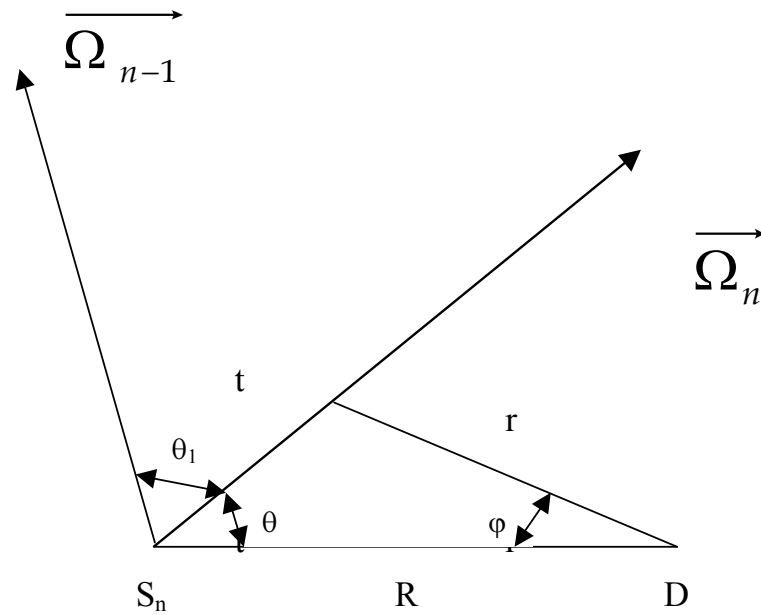


Fig. 1 Geometry of the developed algorithm (S_n - n^{th} scattering point; D - a point detector)

An important property of Equation 5 is the acceleration of a convergence of the results that are comparable with the usual flux-at-a-point estimations. We designed a non-analog simulation process mode of radiation transfer resulting in a finite variance as well. The flux estimation from each collision point in the selected point was carried out on a single-scattered radiation integral (Equation 4).

APPLICATION OF ADJOINT TRANSPORT EQUATION

We took advantage of an adjoint transport equation solution using the Monte Carlo method for problems where the geometry was instituted by a space-distributed source and a point detector. The integral-differential form of an adjoint transport equation is given by Equation 8:

$$-\vec{\Omega} \cdot \nabla \Phi^+(\vec{r}, \vec{\Omega}, E) + \Sigma_t(\vec{r}, E) \cdot \Phi^+(\vec{r}, \vec{\Omega}, E) = \int \Sigma_s(\vec{r}, \vec{\Omega}, E \rightarrow \vec{\Omega}', E') \cdot \Phi^+(\vec{r}, \vec{\Omega}', E') \cdot d\vec{\Omega}' dE' + D(\vec{r}, \vec{\Omega}, E). \quad (\text{Eq. 8})$$

Here $\Phi^+(\vec{r}, \vec{\Omega}, E)$ is an adjoint function or a value function [7]. The solution of an adjoint transport equation is based on the inverse theorem and results from the following expression:

$$J = \int \Phi^+(\vec{r}, \vec{\Omega}, E) \cdot S(\vec{r}, \vec{\Omega}, E) \cdot d\vec{r} d\vec{\Omega} dE \quad (\text{Eq. 9})$$

The convolution equation (Equation 9) adequately estimates the functional using a solution of a direct transport equation with the substitution of an adjoint function, a source function on a flux, or a detector function accordingly. The solution of Equation 8 by the Monte Carlo method means that the architecture of an adjoint random walk of abstract particles known as pseudo-quanta is employed. The basic simulation difficulty of such process is connected to the kernel of Equation 8. This kernel controls the collision of the pseudo-quanta such that they increase energy under the reversed Compton scattering after interaction. Equation 10 solves for the energy of the pseudo-quanta

$$E = \frac{E'}{1 - \frac{E'}{m_0 c^2} (1 - \cos \theta_S)}, \quad (\text{Eq. 10})$$

where E' and E the values of energy before and after a collision accordingly, θ_S is the scattering angle, and $m_0 c^2 = 0.511$ MeV is a rest energy. We suggested the following function for a choice of increasing energy after a collision [12]:

$$f^+(E \rightarrow E') dE = \frac{d\sigma}{dE'}(E \rightarrow E') \frac{dE}{\sigma_S(E)} / \int_{E'}^{E_{\max}(E')} \frac{d\sigma}{dE'}(E \rightarrow E') \cdot \frac{dE}{\sigma_S(E)}, \quad (\text{Eq. 11})$$

where $\sigma_S(E)$ is an integrated microscopical scattering cross-section,

$$E_{\max} = \begin{cases} E_S & \text{- source energy, if } E' \geq \frac{m_0 c^2}{2} \\ \min\left(E_S, \frac{E'}{1 - \frac{2E'}{m_0 c^2}}\right), & \text{otherwise} \end{cases}$$

Using the density function presented in Equation 11 with the condition that allows for the deletion of divergences associated with E_{\max} , for all possible values of scattered energy. Because of physics, the pseudo-quanta energy value cannot exceed the larger source energy value, E_S . Thereby, the pseudo-quanta energy "is locked" inside the existing energy interval. This last condition simultaneously moderates the pseudo-quanta value of energy of each interaction. The simulation increases the number of scatterings per history and illustrates the importance of each history.

The next choice for the value of energy E^* is made by a Neumann method with a weighting factor:

$$W^+ = \frac{\Sigma_S(E^*)}{\Sigma_t(E^*)} \cdot \int_{E'}^{E_m(E')} \frac{d\sigma}{dE'}(E \rightarrow E') \frac{dE}{\sigma_S(E)}. \quad (\text{Eq.12})$$

USAGE OF SOME SIMPLE APPROXIMATIONS

Sometimes it is possible to avoid applying a Monte Carlo method by utilizing semi-analytical methods in calculations that permit reasonable, accurate results for simple geometries. As a rule, such methods are based on integrating a transport equation point kernel. Otherwise, the volume source is represented by a superposition of point isotropic sources [15]. The following paragraphs give examples of volume self-absorbing sources with an equally distributed volume activity q_V .

The self-absorption of an unscattered flux behind a protective layer with a thickness of b for a cylindrical source (R - radius and h - height) is found in Equation 13:

$$\Phi = \frac{q_v}{2\mu_S} Z(R/h, a/h, \mu_S h, b), \quad (\text{Eq. 13})$$

with parameter $\left(\frac{R}{h}\right)$ varying within the limits 0.1÷2.0; parameter $\left(\frac{a}{h}\right)$ varying within the limits from 0 up to 1.0; μ_S as a linear attenuation coefficient in matter of a source $\mu_S = 0,1 \div 10$; and b as a shielding thickness expressed in terms of a

mean free path ($0 \div 10$). The values of function Z for a point on a surface of the cylinder ($a = 0$) without shielding ($b = 0$) are given in Table I [14].

Table I. Values of function $Z(R/h, 0, \mu_s h, 0)$.

R/h	$\mu_s h$							
	0,1	0,3	0,5	1,0	2,0	3,0	5,0	∞
0,1	0,01504	0,04413	0,07198	0,1368	0,2516	0,3437	0,4988	1,0
0,5	0,06418	0,1796	0,2798	0,4770	0,7185	0,8450	0,9509	1,0
1,0	0,1070	0,2875	0,4312	0,6754	0,8932	0,9645	0,9960	1,0
2,0	0,1583	0,4008	0,5706	0,8075	0,9561	0,9885	0,9990	1,0

The flux from an infinite slab with a thickness of h behind shielding b is given in Equation 14:

$$\Phi = \frac{q_v}{2\mu_s} [E_2(b) - E_2(b + \mu_s h)], \quad (\text{Eq.14})$$

in which

$$E_n(x) = x^{n-1} \int_x^{\infty} \frac{e^{-t}}{t^n} dt$$

- an integrated exponential function and the substitution

gives in following expression $E_2(b = 0) = 1$.

Equation 15 gives the flux from a semi-infinite dimensional source ($\mu_s h = \infty$) behind shielding b :

$$\Phi = \frac{q_v}{2\mu_s} E_2(b). \quad (\text{Eq. 15})$$

It is possible to take advantage applicable dose coefficients [13] linking specific activity of water and dose rate for rough estimations of the dose of external radiation potentially received. Presented above, equations 13 through 15 allow the calculation of unscattered radiation characteristics. In reality, we must allow for the contribution of scattered radiation from the source matter. The approximated account of photon-scattered radiation in matter from an infinite source can be estimated using an exponential representation of the buildup factor [11] for a point isotropic source is

$$B = A_1 \cdot \exp(-\alpha_1 \mu d) + (1 - A_1) \cdot \exp(-\alpha_2 \mu d),$$

in which a linear

attenuation coefficient μ is accepted as source material. It is necessary to multiply the corresponding flux on values by buildup factors to compute the total flux for different sources. But, there is a large uncertainty in the choice for distance d .

We used the gamma method with a principle of radiation equal balance to reduce the estimate of scattered radiation for semi-infinite and infinite volume-equal sources given in Equation 16:

$$\dot{D} = 2\pi \cdot \Gamma_{\delta} \cdot q_m / \mu_m , \quad (\text{Eq. 16})$$

in which q_m is specific activity of a source and μ_m is a mass absorption coefficient of energy for photons in a source material. Γ_{δ} is a Kerma constant for energy threshold δ .

RESULTS

In Figure 2, three general migration scenarios for ^{137}Cs , which is contained in a contaminated layer of reservoir water, are illustrated:

- Scenario **a**) is the result of precipitation activity on the reservoir surface. The contaminated layer of water is spreading step-by-step throughout whole volume of the reservoir. There is a self-cleaning of water due to gravitation precipitation, and full activity proceeds in the bottom sediments.
- Scenario **b**) is the precipitation of the radioactive layer at various speeds in such a manner that the low side spreads faster than high side.
- Scenario **c**) is the uniform precipitation with a thickness 10 cm for the radioactive layer.

Figure 3 shows the calculated dose distributions by the Monte Carlo method on the water surface of the cylindrical reservoir. In all calculations, the value of the specific activity was 1.0 becquerel per liter (Bq/L) for a completely filled reservoir. Two processes are considered in Figure 3. The first process is associated with a slow infiltration of contamination on the entire volume of the reservoir. The second process is associated with a self-cleaning of water. As a result of self-cleaning, an upper layer of clean water is created. This layer is shielding against irradiation on the surface of a reservoir.

Figure 4 shows the behavior of the dose buildup factor and the contribution of the scattered radiation from the radioactive water layer. The calculations reveal an important conclusion about solving problems using Equations 13-16. Estimated dose values appear to be half of what is expected when using the more convenient equations when there is a slow precipitation of radioactive sediments. However, in the other case (see Figure 4a) the underestimation can be very large (to two orders). Table II lists the results of the dose estimates obtained as by a Monte Carlo method, and other methods for various cylinders, up to semi-infinite geometry. The results are given both for a non-scattered dose D_0 , and for a total dose D simultaneously with the dose buildup factor B_D . For a semi-infinite source, the values of a non-scattered and a total dose are obtained using Equations 15 and 16, respectively.

Table II. Comparison of the dose results in the units of picoGray (pGy) received by a Monte Carlo method and a numerical method.

Sizes of cylinder		Numerical integration	Monte Carlo method		
Radius, cm	Height, cm		D ₀ , pGy	D, pGy	B _D
25	50	1.65E-02	1.69E-02	3.00E-02	1.8
50	100	1.77E-02	1.80E-02	3.80E-02	2.1
100	200	1.79E-02	1.80E-02	3.81E-02	2.1
150	300	1.78E-02	1.80E-02	3.80E-02	2.1
200	400	1.77E-02	1.80E-02	3.90E-02	2.2
300	600	1.78E-02	1.82E-02	4.00E-02	2.2
400	800	-	1.83E-02	3.88E-02	2.1
$\infty/2$		-	1.82E-02	4.78E-02	2.6

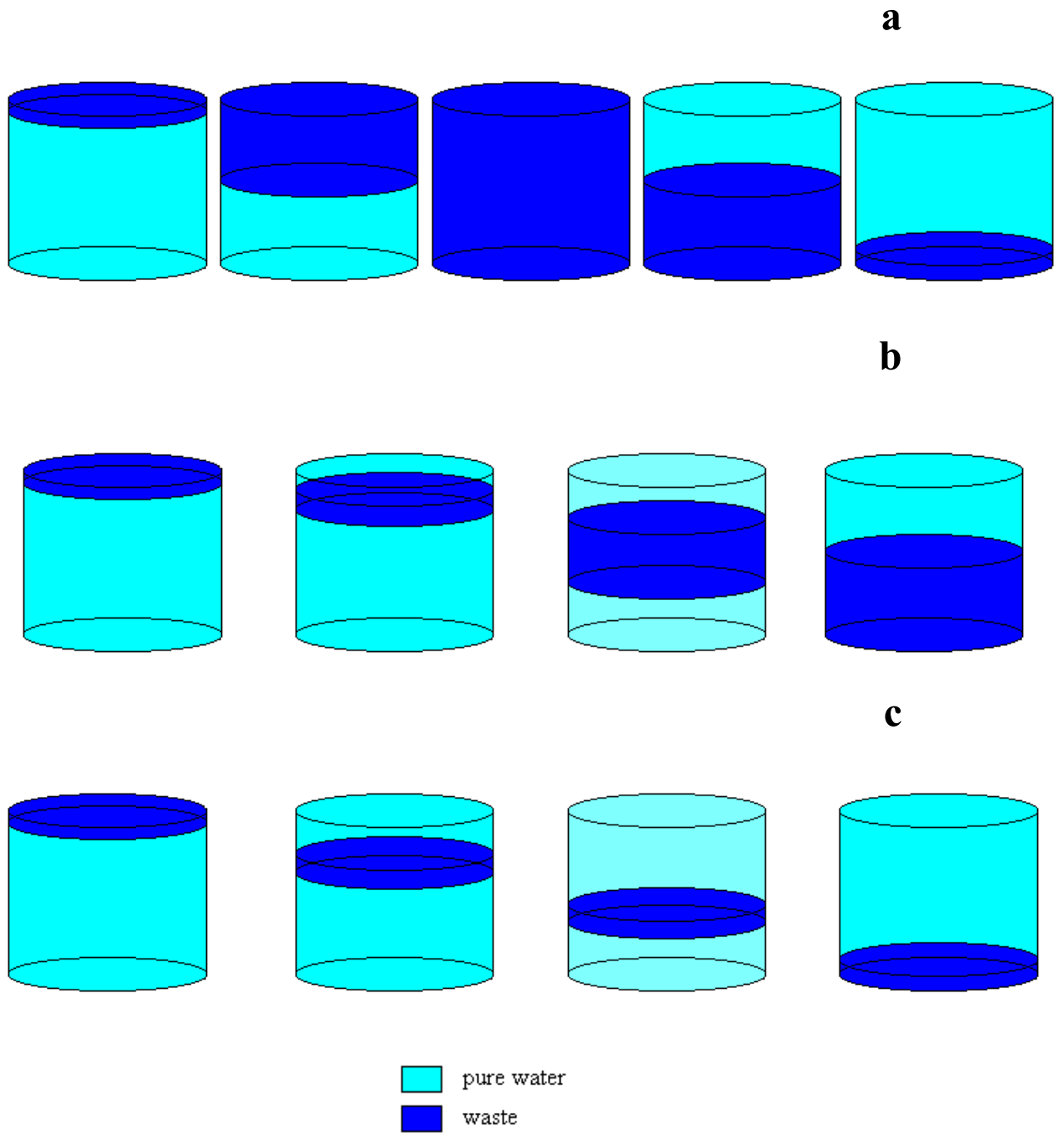


Fig. 2. Various scenarios illustrating the migration of a radioactively contaminated wastewater layer.

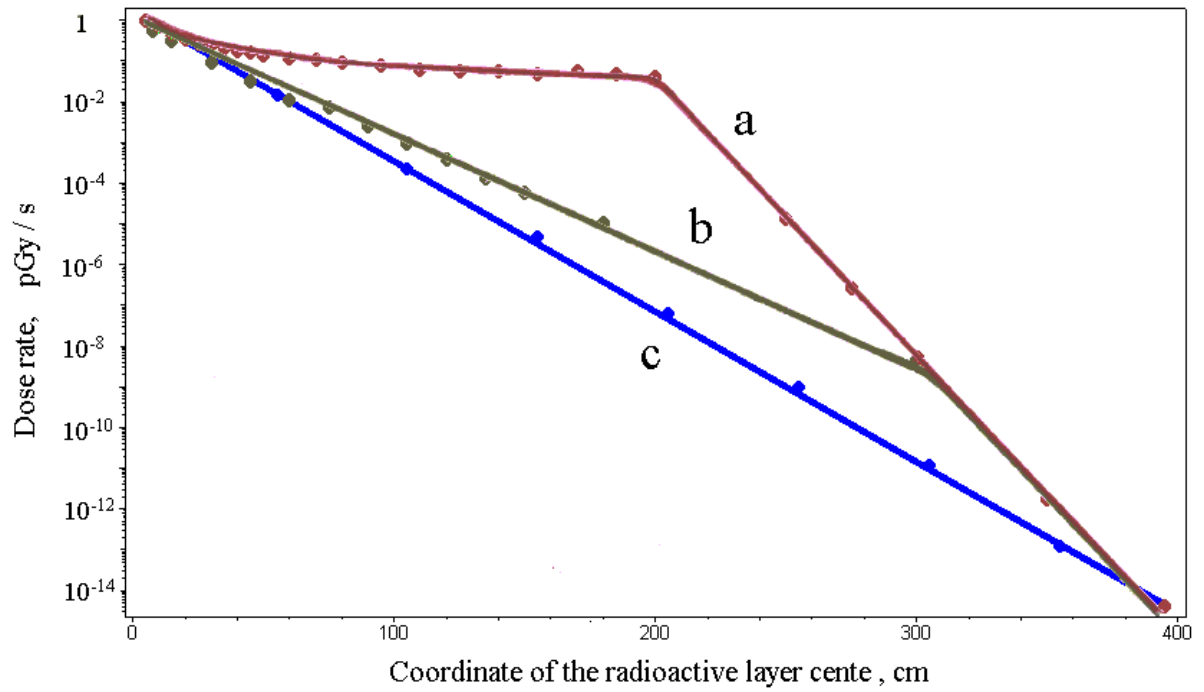


Fig. 3. Dependence of the calculated dose rates based upon the depth of the center of the radioactive wastewater layer relative to the surface of a reservoir with a 200 cm radius and 400 cm height (a, b, and c correspond to scenarios a, b, and c, respectively).

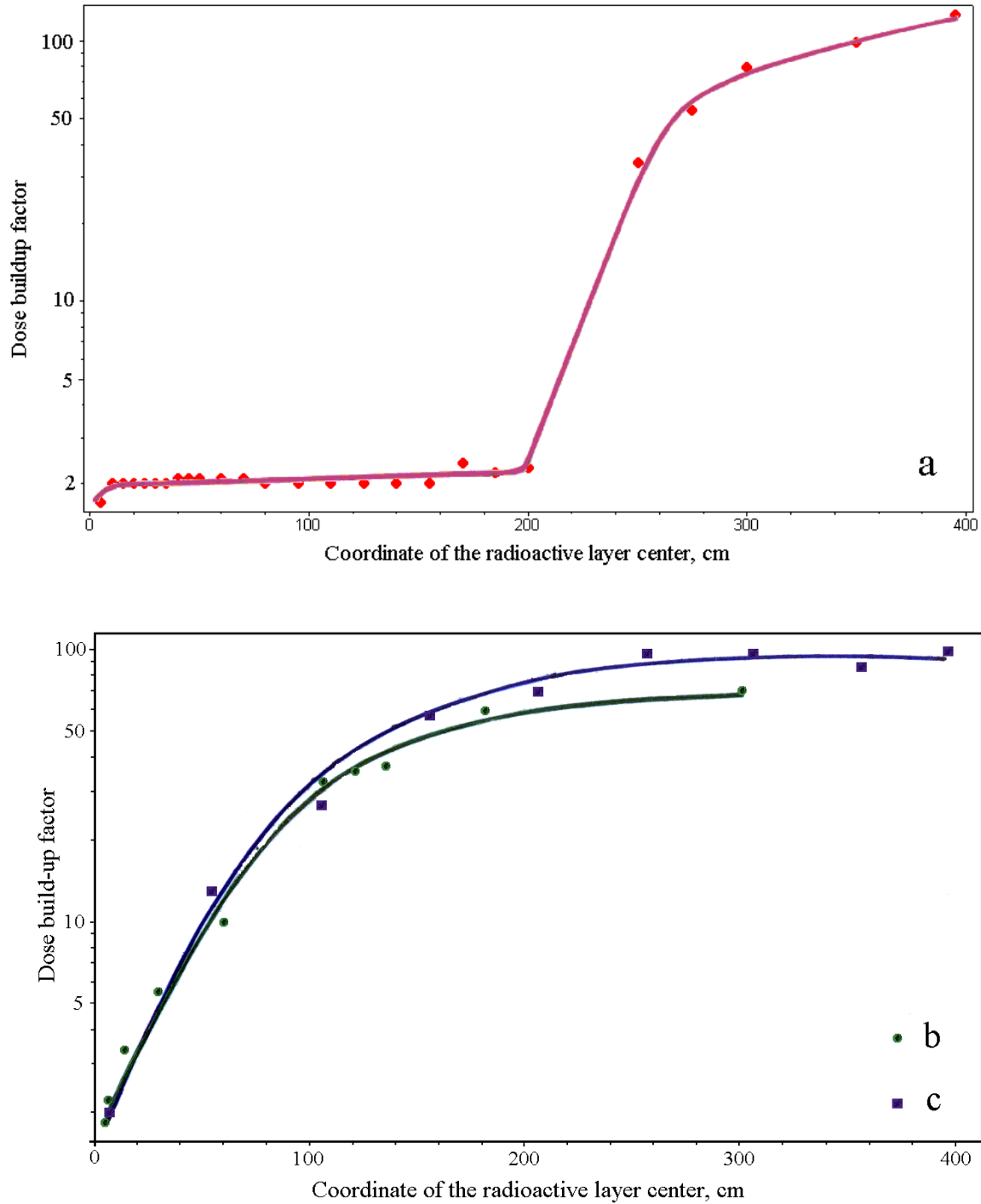


Fig. 4. Relationship of the calculated dose buildup factor and the depth of the center of the radioactive wastewater layer relative to the surface of a reservoir with a radius of 200 cm and a height of 400 cm. (a, b, and c correspond to scenarios a, b, and c, respectively)

CONCLUSION

Using simple methods for calculating radiation field characteristics in large reservoirs or lakes can result large errors in characterizing contaminant migration and potential exposure. This study demonstrates the advantages of using the Monte Carlo method to calculate radiation field characteristics and estimate the impact on the potential radiation doses received that result from exposure to contaminated wastewater. Non-analog simulation of photon radiation using an adjoint Monte Carlo method allows efficient calculations of transport for a three-dimensional problem. The calculated characteristics provide information about the migration of a radioactive water volume that can inform decisions about possible remediation and stewardship activities.

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