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Model of ambient dose equivalent for radium contamination: Dependence on the geometry of the source

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A R T I C L E I N F O

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A B S T R A C T

Industrial activities involving radium sources, such as watchmaking, were still common up until the 1960s. They produced contaminations in building materials and the soil in a large variety of geometries. The potential remediation of such places requires instruments that are properly calibrated as well as adequate procedures. We have developed a model that estimates the rate of ambient dose equivalent $\dot{H}^*(10)$ at 10 cm and 1 m from a source of ^{226}Ra and its progeny in both the soil or the building materials. Our model, described here, uses Monte Carlo (GEANT4) computed yield functions of $\dot{H}^*(10)$ per unit activity induced by point-like sources in different contaminated materials. Fit functions of the yield curve of $\dot{H}^*(10)$ are provided for outdoor contamination. The model can be used for any geometrical activity distribution and we present an example showing the dependency of $\dot{H}^*(10)$ on the diameter and the depth profile of the sources, for both outdoor and indoor contamination.

1. Introduction

The use of radium painting in the watch industry in Switzerland up until the 1960s gave rise to the ^{226}Ra contamination of several external areas and dwellings, especially in the regions around the Jura Mountains. Recently, an extended program of radium decontamination was undertaken in Switzerland under the direction of the Swiss Federal Office of Public Health (Murith et al., 2017). The program involves taking measurements in order to decide if a potential external or internal contaminated area should be remediated. The rate of ambient dose equivalent $\dot{H}^*(10)$ is first measured on 1 m × 1 m grids, at 10 cm and 1 m away from the soil and the building material of the screened areas. In the case where the dose rate at all points of measurement does not exceed 100 nSv/hour the area is considered uncontaminated. In the opposite case, if the dose rate exceeds 100 nSv/hour at one or more positions, additional measurements are taken, involving monitoring of $\dot{H}^*(10)$ on finer grids, as well as collecting and analysing contaminated samples. In general, in order to properly evaluate the level of contamination from all measurements involved in a decontamination procedure, it is essential to have a precise model that quantifies the dependence of $\dot{H}^*(10)$ on the activity and the geometry of the radioactive source. The main goal of this publication was to provide such a model for the case of ^{226}Ra contamination.

Ambient dose equivalent conversion coefficients for mono-energetic gamma and for radionuclides have been computed and published by several authors (Beck et al., 1972; Saito and Jacob, 1995; Clouvas et al., 2000; Lemercier et al., 2008; Saito and Petoussi-Henss, 2014; Gasser et al., 2014; Malins et al., 2015; Bochud et al., 2017). Most of these computations are Monte Carlo (MC) based and use geometrical symmetry and transformation to reduce the variance of the MC simulations. Lemercier et al. (2008), Saito and Petoussi-Henss (2014), and Bochud et al. (2017) have published conversion factors for $\dot{H}^*(10)$ in the case of semi-infinite planar contaminated soils with different relaxation factors of the exponential decrease of the activity in the soil. In these studies, conversion factors are tabulated for mono-energetic gammas and specific nuclides. These factors can be used together with *in situ* gamma spectrometry to estimate $\dot{H}^*(10)$ at 1 m above very large areas of contaminated soil. In our work, we are interested in anthropogenic ^{226}Ra contaminations with limited spatial extension and the hypothesis of semi-infinite contamination is no longer valid. It was therefore necessary to have a model that could compute $\dot{H}^*(10)$ for any kind of activity distribution in contaminated soils or building materials. Gasser et al. (2014) computed the dependence of the kerma rate at 1 m height looking at the depth and diameter of the source but for a homogeneous distribution of the contamination. Malins et al. (2015) computed the dependence of the rate of H*(10) on the diameter, relaxation factors, and height of a cylindrical shape for ^{134}Cs and ^{137}Cs contamination.

In this paper we present a MC based model of ambient dose

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equivalent for ^{226}Ra contamination, that can be used to rapidly compute $\dot{H}^*(10)$ at 10 cm and 1 m from a surface in which any distribution of a specific activity of ^{226}Ra and its progeny may be present. This model is based on the numerical integration of GEANT4-computed yield functions of $\dot{H}^*(10)$ per unit activity of a point-like source of ^{226}Ra and its progeny (Agostinelli et al., 2003; Allison et al., 2006).

2. Method

Our method computes the rate of ambient dose equivalent $\dot{H}^*(10)$ at 10 cm and 1 m away from a stack of flat layers of different materials contaminated with ^{226}Ra and its progeny. More specifically, our method integrates, over the volume of the source, $\dot{H}^*(10)$ yield functions for a point-like source, which are pre-computed with the MC code GEANT4 (Agostinelli et al., 2003; Allison et al., 2006). In this way the time-consuming MC simulations are limited to the computation of the yield curves while the numerical integration over any source shape is done within seconds using a desktop computer.

2.1. Geometry and soil composition

We computed the ^{226}Ra contamination of the soil of an external open field area and the floor and walls of a closed room. For the outdoor contamination, the soil was modelled by a flat 500 × 500 × 3 m^3 earth layer, covered by a 100 m thick air layer to take sky shine into account. For the indoor contamination, we considered a 4 × 4 × 2.5 m^3 room filled with air. The floor was made of a 20 cm thick concrete slab covered by a 1 cm thick wood layer. The walls and the ceiling were made of 20 cm concrete layers covered by a 2 cm inner layer of gypsum. The compositions of the soil, concrete, wood, gypsum, and air considered in the simulations are given in Table 1. The composition of the soil, the concrete, and the gypsum are taken from McConn et al. (2011).

2.2. GEANT4-computed ambient dose equivalent induced by point-like source

The rate of ambient dose equivalent $\dot{H}^*(10)$ at different distances from the contaminated flat layers of the various materials was computed with the GEANT4 MC code for point-like radioactive sources of ^{226}Ra, ^{214}Bi, ^{214}Pb, and ^{210}Pb placed at different depths within the layers (see Fig. 1). The full chain of processes taking part in the radioactive decay of a source (alpha, beta, electron capture, nuclear de-excitation, and atomic relaxation) was simulated by using the radioactive decay module of GEANT4 based on Evaluated Nuclear Structure Data Files (Tuli, 2001). The resulting secondary flux of gammas and electrons was converted into $\dot{H}^*(10)$ by using fluence to dose conversion coefficients found in the ICRP74 publication (ICRP, 1996). It is worth noting that although in the ICRP74 publication the conversion coefficients for electrons are given for $H'(10, 0^o)$, they can also be used to compute $H^*(10)$ as they were obtained from MC simulations considering parallel beams. For each type of soil and building material, a series of $h_S(z_i,h,r)$ yield functions was obtained from the GEANT4 simulations. It represents $\dot{H}^*(10)$ at distance h from the contaminated layer induced by a 1 Bq source of a radionuclide S placed at a depth z_i within the contaminated material, and in function of the lateral distance r from the position of the source. The yield curves are calculated at h = 10 cm and h = 1 m, and for different discrete depths z_i. In the MC GEANT4 computation the dependence of the yield function on the lateral distance r was obtained by using different radial sectors of disk for the H*(10) tallies. This drastically reduces the variance of the computation compared to using a point-detector tally.

Table 1
Composition of the different materials used in the GEANT4 MC simulations.

Material	Weight Composition (%)	Density (g/cm^3)
air	N 75.53, O 23.18, Ar 1.29	1.2 10^{-3}
soil	O 51.3, Na 0.6, Mg 1.3, Al 6.8, Si 27.1, K 1.4, Ca 5.1, Ti 0.46, Mn 0.7, Fe 5.6	1.52
concrete	H 2.2, C 0.2, O 57.5, Na 1.5, Mg 0.1, Al 2, Si 30.5, K 1, Ca 4.3, Fe 0.7	2.32
gypsum	H 2.3, O 55.8, S 18.6, Ca 23.3	0.84
wood	H 5.2, C 12.4, O 82.4	0.97

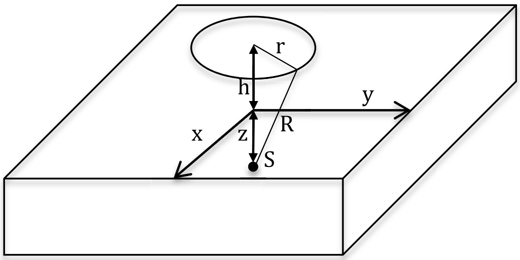

Fig. 1. Coordinates used in the computation of H*(10) yield function for a point-like source of radionuclide S. The source is located at a depth z in the contaminated material. The ambient dose equivalent H*(10) is computed in a plane located at distance h from the external surface of the material, at different lateral distances r from the source. R represents the distance between the source and the position where H*(10) is computed ($R = \sqrt{r^2 + (h+z)^2}$).

2.3. Computed ambient dose equivalent induced by an extended source

The rate of ambient dose equivalent at position (x$_1$,y$_1$,h) induced by an extended radioactive source of a given radioisotope S in a contaminated material is obtained by the following numerical integration

$$\dot{H}^*(10)(x_1, y_1, h) = \int_V A_V(x, y, z) h_s\left(z, h, \sqrt{(x_1-x)^2 + (y_1-y)^2}\right) dx\, dx\, dz \quad (1)$$

where $A_V(x,y,z)$ is the specific activity of the source as a function of its position in the volume, h$_s$ represents $\dot{H}^*(10)$ per unit activity yield function computed with GEANT4, and V represents the volume of the source over which the integral is computed.

2.4. Numerical validation of the model

The numerical validation of our calculations was performed in two steps. First, our GEANT4 computation of $\dot{H}^*(10)$ was validated for the cases of point-like sources in the air with an analytical computation, and with the MicroShield code. The analytical computation of $\dot{H}^*(10)$ consists of convoluting the gamma spectrum of the source with ICRP fluence to dose coefficient (ICRP, 1996). The MicroShield code is a gamma ray shielding and dose assessment program that is used for shielding design (Grove Software, 2013). In the second step, the ambient dose equivalent induced by an extended source as computed with Equation (1) was validated against full GEANT4 simulations for the following two cases.

- $\dot{H}^*(10)$ induced by a homogeneous cylindrical source in a soil
- $\dot{H}^*(10)$ induced in a room by a contaminated wooden floor

2.5. Shape and depth profile of the radioactive source

The radioactive source in the contaminated material was distributed over a cylinder with diameter D and height D_z. The radial profile of the specific activity of the source was considered as constant. The depth profile was described by an exponential function $\exp(-z/\beta)$ with z the depth in the material from the surface and β the relaxation factor both given in g·cm^{-2}. The rates of ambient dose equivalent $\dot{H}^*(10)$ were computed for different values of D, D_z, and β. Full contamination, that