A MODEL FOR BETA SKIN DOSE ESTIMATION DUE TO THE USE OF A NECKLACE WITH URANIUM DEPLETED BULLETS

Paulo Fernando Lavalle Heilbron Filho[1,*], Jesús Salvador Pérez Guerrero[1] and José Francisco de Oliveira Júnior[2]

[1*]Brazilian Nuclear Energy Commission- R. General Severiano 90, 22290-901, RJ-Rio de Janeiro, Brazil

[2]Department of Environmental Sciences, Institute of Forestry, Agricultural Federal University of Rio de Janeiro, Seropédica, 23890-000-RJ-Brazil

*Corresponding author:

Paulo Fernando Lavalle Heilbron Filho
Brazilian Nuclear Energy Commission
R. General Severiano 90, 22290-901, RJ-Rio de Janeiro
Brazil

e-mail: paulo@cnen.gov.br

Abstract

Depleted uranium bullets were use as munitions during the Kuwait – Iraq war and the International Atomic Energy Agency sampling expert's team found fragments in the environment when the war was over. Consequently, there is a possibility that members of the public, especially children, collects DU fragments and use it, for example, to make a necklace. This paper estimates the beta skin dose to a child that uses a necklace made with a depleted uranium bullet. The theoretical model for dose estimation is based on Loevinguer's equation with a correction factor adjusted for the maximum beta energy in the range between 0.1 and 2.5 MeV calculated taking into account the International Atomic Energy Agency expected doses rates in air at one meter distance of a point source of 37 GBq, function of the maximum beta energy. The dose rate estimated by this work due to the child use of a necklace with one depleted uranium bullet of 300 g was in good agreement with other results founded in literature.

Keywords: Beta skin dose, uranium depleted bullet

INTRODUCTION

Depleted Uranium (DU) bullets were used during the Gulf War and there is still a good possibility that children use them to make a necklace. It should be pointed out that the majority of ^{238}U daughters are alpha emitters with a very small range in the air (order of centimeters). In the case of depleted uranium (^{238}U+^{235}U) the radionuclide's of importance due to beta emission are ^{234}Th, ^{234m}Pa and ^{234}Pa for ^{238}U and ^{231}Th for ^{235}U that will be in secular equilibrium respectively with their parents and all beta decay emitters of interested are very low gamma energies emitters (Table 1).

Table 1. ^{238} U and ^{235}U decay daughters of importance for the calculation

Radionuclide	Half-live	Energies (MEV) and emissions probabilities		
		Alpha	Beta	Gamma
^{238} U	4.51x10^{9}y	4.15 (25%)	---------------	
234Th	24.1d		0.063 (3.5%)	0.115 (0.4%)
^{234m}Pa (87%)	1.17m		2.271 (99.84%) 0.074 (0.16%)	0.765 (3.5%) 1.001(0.60%)
^{234} Pa (13%)	6.75h		2.197 (100%)	0,100 (50%) 0.90 (70%)
^{235}U	7.13x10^{8}y	4.38 (100%)	-----------------	
^{231} Th	25.6 h		0.3 (100%)	

The risk of inadvertent exposure to people due to the use or contact with depleted uranium bullets is real and considered an important issue for the International Atomic Energy Agency [IAEA - Assessing Effects of Depleted Uranium:- The IAEA Role (*http://www.iaea.org/newscenter/focus/depleteduranium/iaearole.pdf*).

On this document, the IAEA states that *"Complete depleted uranium ammunition or fragments can still be found at some locations where depleted uranium weapons were used during past wars. Prolonged skin contact with these depleted uranium residues is the only possible exposure pathway that could result in exposures of radiological significance. As long as access to the areas where these fragments exist remains restricted, the likelihood that members of the public could be exposed to these residues is low. The recommendations to the national authorities, in all the cases studied were to collect any depleted uranium ammunition or fragments and any war equipment, which have been in direct contact with these ammunitions and isolate them from the public in appropriate locations until it can be processed as low-level radioactive waste and eventually safely disposed of. Some environmental remedial actions like covering of areas with uncontaminated soils could be convenient at some particular locations, depending on the use of the land." and complemented: "Depleted uranium in munitions is in a concentrated metallic form, and there are understandable concerns about elevated levels in the environment due to spent munitions. There are also worries about people handling intact depleted uranium metal. Contact doses when handling bare DU metal are approximately 2.5 mSv/h, primarily from beta radiation, which is not penetrating and so affects only the skin. Even so, the collection of bare DU munitions needs to be discouraged and, if possible, avoided completely. Doses from depleted uranium are, therefore, real and, in some circumstances, they could be appreciable for military* *personnel. Doses to people in the post-conflict phase are likely to be much lower and should be relatively easy to avoid*."

Consequently, it is important to estimate the beta skin dose to see if it represents a real risk to small children. To do this it´s important to develop an analytical model that takes into consideration the size of the necklace as well as the contribution of the most important beta emitters radionuclides from DU.

SKIN BETA DOSE CALCULATION MODEL

Due to the continuum spectrum of beta particle emission, the dose rates D (Rad.h^{-1}), at a certain distance *x* (cm) of a disk plane source of radius *b* (cm), with a homogeneous activity concentration S_s . (Bq cm^{-2}) (Fig. 1) can be calculated using the famous empirical **LOEVINGER'S (1956)** equations that are strongly dependent of the relation between $c/(v\rho)$ or c/v (when the density ρ of the medium is equal to 1 g cm^{-3}) as well as the radius *b* of the source where c is related to the range of beta particle in air and v (cm^2 g^{-1}) is an empirical absorption coefficient.

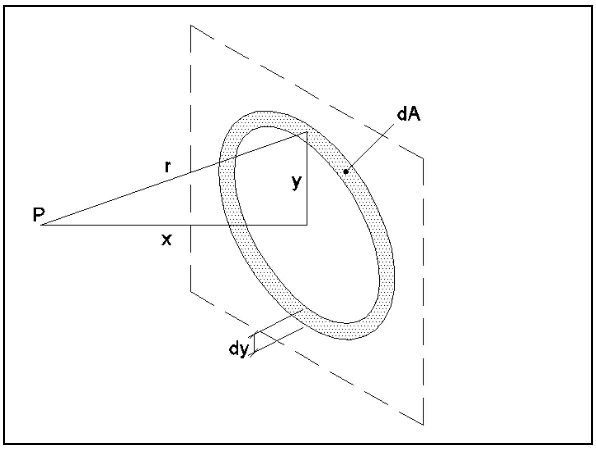

Figure 1. Disk source scheme

Depending on the three boundary conditions (1, 2 and 3 shown on Fig. 2), there are three different empirical equations for the beta skin dose calculation as will be shown.

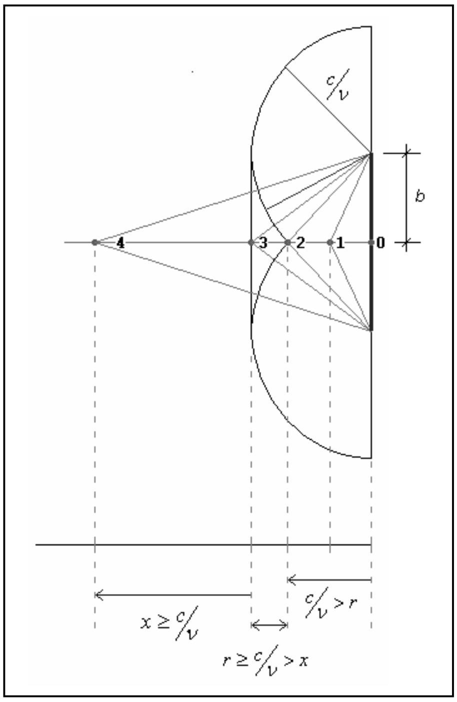

Figure 2. Types of boundary conditions

The dose rate at appoint P (Fig. 1), located on the axis of a finite circular plane source covered by a uniform concentration activity Ss (Bq cm^{-2}) is given by Eq. (1) depending on the relation between $a = (x^2 + b^2)^{1/2}$ and $c/(v\rho)$:

$$D = \frac{2\pi c K S_s}{v^2 \rho^2}[\int_x^a \frac{1}{r} dr - \frac{e}{c}\int_x^a v_e^{-(vr/c)} dr + \frac{e}{c}\int_x^a v_e^{-(vr)} dr] \quad (1)$$

The “constant” K (Rads/(h.Bq)) is considered a normalizing factor given by Eq. 2, calculated taken into consideration that the rate of energy absorbed over all the sphere is just equal to the rate of the beta energy output by the source (average energy per disintegration) resulting in:

$$K = 4.59 \times 10^{-6} \rho^2 v^3 E_{md} \alpha \quad (2)$$

With,

$\alpha = [3c^2 - e(c^2 - 1)]^{-1}$ and e is the Euler's number (3)

Where, E_{md} is the average beta particle energy in MeV given by one third of the maximum beta energy emitted ($E_{md} / 3$); ρ is the density of the material where the dose will be calculated (g cm^{-3}) v is the mass absorption coefficient for tissue in cm^{2}g^{-1}.

For the skin, the constant c used for the dose calculation can be seen on Table 2 as a function of beta maximum energy.

Table 2. c´s values for skin as a function of beta energy

Value c	Energy range (MeV)
2.0	$0.17 \le E_{max} < 0.50$
1.5	$0.50 \le E_{max} < 1.50$
1.0	$1.50 \le E_{max} < 3.00$

The solution of Eq. (1) is strongly dependent of the boundary conditions situations 1, 2 and 3 shown on Fig. 2 as can be seen from the results shown in Table 3 (x and b can be seen on Fig. 2 and are given in cm).

The mass absorption coefficient for air and skin v (cm^2.g^{-1}) respectively can be seen below:

$$v = \frac{16(2 - factor)}{(E_{max} - 0.036)^{1.4}} \quad (4)$$

$$v = \frac{18.6(2 - factor)}{(E_{max} - 0.036)^{1.37}} \quad (5)$$

Table 3. Dose rate (D) for different type of boundary conditions

First Boundary Condition: $\frac{c}{\nu\rho} > x$
$D = 2{,}89 * 10^{-5}[(Rads/h)/(MeV * Bq/g)] * \nu[cm^2/g] * E_{md}[MeV] * \alpha * S_S[Bq/cm^2] *$ $* \left(c * \left\{ 0.5 * Ln\left(1 + \frac{b^2}{x^2}\right) + e^{\left[1 - \left(\frac{\nu\rho}{c}\right) * \sqrt{\left(x^2 + b^2\right)}\right]} - e^{\left(1 - \frac{\nu\rho * x}{c}\right)} \right\} + e^{(1 - \nu\rho * x)} - e^{\left(1 - \nu\rho * \sqrt{x^2 + b^2}\right)} \right)$ $D = 2.89 \times 10^{-5} \nu\, E_{md} \alpha\, S_s \{c[\frac{1}{2}\ln(1 + \frac{b^2}{x^2}) + e^{1-\omega/c} - e^{1-\nu\rho x/c}] + e^{1-\nu\rho x} - e^{1-\omega}\}$
Second Boundary Condition: $x < \frac{c}{\nu\rho} \leq (b^2 + x^2)^{0.5}$
$D = 2{,}89 * 10^{-5}[(Rads/h)/(MeV * Bq/g)] * \nu[cm^2/g] * E_{md}[MeV] * \alpha * S_S[Bq/cm^2] *$ $* \left(c * \left\{ 1 + Ln\left(\frac{c}{\nu\rho * x}\right) - e^{\left(1 - \frac{\nu\rho * x}{c}\right)} \right\} e^{(1 - \nu\rho * x)} - e^{\left(1 - \nu\rho * \sqrt{x^2 + b^2}\right)} \right)$ $D = 2.89 \times 10^{-5} \nu\, E_{md} \alpha\, S_s \{c[1 + \ln(\frac{c}{\nu\rho x}) - e^{1-\nu\rho x/c}] e^{1-\nu\rho x} - e^{1-\omega}\}$ $\omega = \nu\rho(x^2 + b^2)^{1/2}$
Third Boundary Condition: $x \geq \frac{c}{\nu\rho}$
$D = 2{,}89 * 10^{-5}[(Rads/h)/(MeV * Bq/g)] * \nu[cm^2/g] * E_{md}[MeV] * \alpha * S_S[Bq/cm^2] *$ $\left(e^{(1 - \nu\rho * x)} - e^{\left(1 - \nu\rho * \sqrt{x^2 + b^2}\right)} \right)$ $D = 2.89 \times 10^{-5} \nu\, E_{md} \alpha\, S_s (e^{1-\nu\rho x} - e^{1-\omega})$

$$\omega = \nu\rho(x^2 + b^2)^{1/2}$$

On this article the correction factor fc = [2-factor] for ν in the air, as a function of the beta energy, was obtained using equations 5 to 11 of this article combined with the dose rates in air as a function of the beta energy calculated at 1 m from a point source of 37 MBq (Inverse problem - Table 4.) presented by the **INTERNATIONAL ATOMIC ENERGY AGENCY (IAEA, 1987).**

Figure 3 shows the dose rates expected using the correction/calibration factor of Table 4 at 1 meter from a 37 MBq source obtained with the **LOEVINGUER'S** equations in comparison with the **INTERNATIONAL ATOMIC ENERGY RESULTS (IAEA, 1987).** It should be pointed out that **LOEVINGUER** only suggests some values for the factor(E_{md}) to be used in equation 11 such as in the case of Sr-90 (1.17) and Bi-210 (0.77) and not for all beta energy ranges (0.1 to 2.5 MeV) that's why this calibration was necessary.

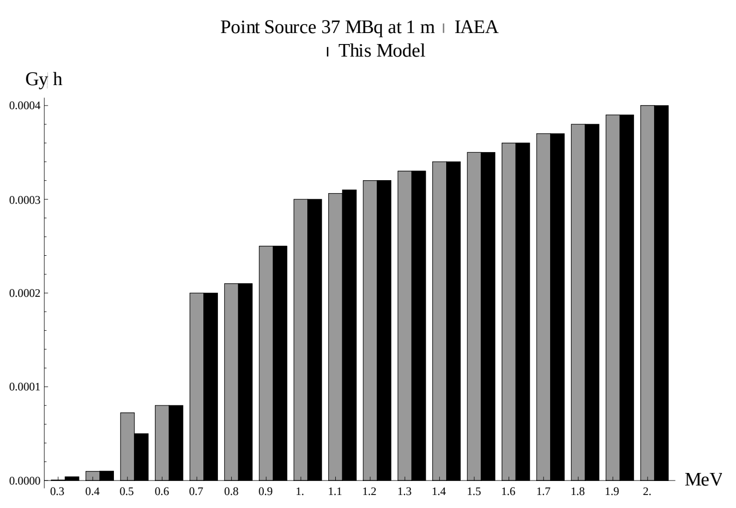

Figure 3. Comparison of the expected dose rate at 1 m from a point source of 37 MBq as a function of the beta energy using this model and the IAEA results.

Table 4. Correction factor as a function of the beta energy

Factor	Beta energy (MeV)
0.01	0.7
0.2	0.7
0.3	0.7
0.4	0.7
0.5	0.400877
0.6	0.63076
0.7	0.552358
0.8	0.637928
0.9	0.71532
1.0	0.78621
1.1	0.856
1.2	0.91929
1.3	0.977288
1.4	1.02951
1.5	1.07598
1.6	1.11676
1.7	1.15195
1.8	1.18175
1.9	1.20637
2.0	1.22606
2.5	1.22606

CONCEPTUAL MODEL FOR THE BULLET AND DATA USED FOR THE CALCULATION

Skin thickness varies not only with the age but also with the part of the body and sex of course. Even epidermis thickness can vary a lot with the factors described above (see Table 10.1 and 10.2 from **ICRP 89 (2002).** As can be seem from ICRP the epidermis thickness for children with ages between 0 and 5 years varies from 23 μm to 380 μm (finger) depending on the part of the body. For adults males with ages between 20 and 60) epidermis thickness varies from 34 μm to 1400 μm (sole) and for females between 18 μm and 1100 μm (sole).

The first challenge question to model beta skin dose is what thickness is reasonable to be considered to calculate the doses on skin (dermis) since it also varies a lot with age, part of the body and sex as shown before.

Usually in literature, 70 microns are considered an average value for the thickness of the dead skin cells (epidermis).

In fact the skin dose vary very slight between 70 microns (2.8 mSv/h) and 200 microns (2.6 mSv/h) for uranium metallic as can be seen from figure 4 from *http://www.mindat.org/article.php/918/Estimating+the+beta+radiation+dose+rate+from+uranium+minerals*.

So in order to be conservative on skin dose estimation we have to use a thickness higher than 70. A deep skin thickness values between 100 microns and 200 microns, for the authors, sounds a very reasonable values .due to the facts above and were adopted for the calculation of the beta skin dose due to the use of a necklace with deplete uranium.

Regarding the geometry of the beta bullet (cylindrical geometry) to be adopted in the model with should be pointed out that beta particles from DU bullet are self absorbed very easily on the bullet itself because depleted uranium has a very high density of approximately 19.1 g/cm^3 and this limits very much the range of the beta particles on the bullet. The range of the beta particle in metallic uranium is shown on table 5.

Based on the explanation before a good model to estimate skin dose in contact with DU bullets is to considered a very thin disk that logically can be transformed in a surface area but it is also necessary to increase de diameter of the source to take into consideration the total superficial area of the bullet that is in contact and near the skin (due to the cylindrical shape of the bullets-see figure 4).

Table 5. Range and Surface area concentration for beta skin dose calculation

Beta Energy (MeV)	Range on Bullet (cm)	C_{bullet} (Bq cm^{-2})
0.063	0.00028	12.39
2.271	0.033	1457.67
0.074	0.00034	15.34
2.197	0.031	1394.84
0.3	0.0022	98.73

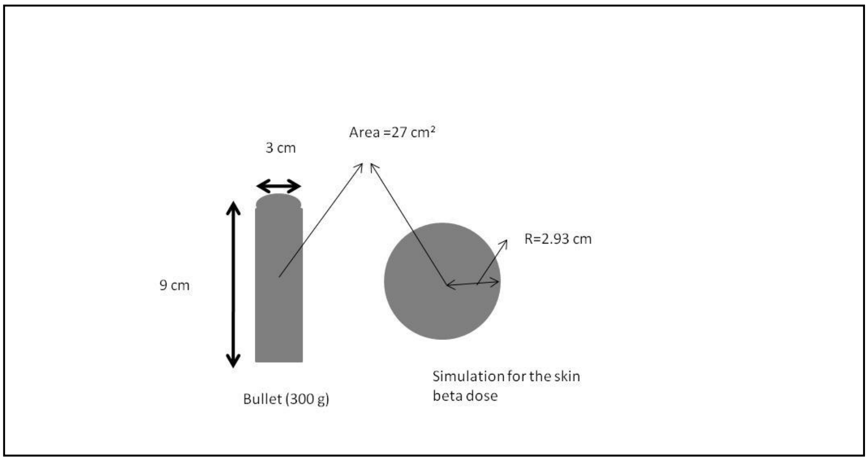

Figure 4. Scheme of conceptual model for beta skin dose calculation of a DU necklace bullet of 30 mm.

Based on the discussions before the beta skin dose estimation due to the use of a bullet in a necklace a conceptual model was developed including the following hypothesis:

- The geometry of the bullet was approximated considering the projected rectangular area of a bullet and a disk source of same area as shown on Figure 4;

- The beta dose was calculated using the formulas shown before for a disk source and a deep of 100 μm and also for 200 μm;
- The range of beta particle (R) in the bullet (equations 6), for each energy (Table 1), in cm was calculated in order to transform volumetric concentration in the bullet Vc (Bq m^{-3}) in surface activity area concentration of the disk C_{bullet} (Bq m^{-2}). A total absorption of 99% was considered for the calculation resulting in the following equations (6 and 7) for the range and surface area concentration:

$$Range = R = \frac{\ln(100)}{(22/E_{\max}^{1.33})\rho mat} \quad (6)$$

$$C_{Bullet} = MA \times W \times (R/D) \quad (7)$$

Where:

MA= Mass activity concentration of the bullet (Bq kg^{-1})

W= Weight of the bullet (kg);

R= Range as defined prior in cm

D= Diameter of the bullet (cm).

ρmat= Density of the medium (g/cm^3)

The radionuclides of importance considered in a uranium depleted bullet is shown on Table 5 and includes the hypothesis of secular equilibrium (^{234}Th, ^{234m}Pa and ^{234}Pa). ^{231}Th and ^{235}U daughters were neglected due to the very low maximum beta energy decay and activity in comparison with the others.

Table 5. Range and Surface area concentration for beta skin dose calculation

Beta Energy (MeV)	Range on Bullet (cm)	C_{bullet} (Bq cm^{-2})
0.063	0.00028	12.39
2.271	0.033	1457.67
0.074	0.00034	15.34
2.197	0.031	1394.84
0.3	0.0022	98.73

The mass concentrations of the radionuclides mentioned above were based on the values shown on Table 6.

Table 6. Bullet data

Radionuclide	Maximum activity concentration in a bullet (Bq/kg) $x10^6$
U^{238}	(12.05 ± 0.55)
Th^{234}	(12.05 ± 0.55) (*)
Pa^{234m}	(12.05 ± 0.55) (*)
Pa^{234}	(12.05 ± 0.55) (*)

The range given in cm calculated using equation 6 for a metallic uranium density material used in the program was also compared with the range formula given by **GRUN(2014)** also in cm, (equation 8), for the same material (metallic uranium $\rho mat = 19.1$ g/cm^3) and the results can be seen on table 7 and shows good agreement between both.

Table 7. Comparison between range equations values

Beta Energy (MeV)	Range in cm This work Equation 6 (cm)	Range in cm Grun Equation 8 (cm)
0.063	2.8 $x10^{-4}$	3.1 $x10^{-4}$
2.271	3.3 $x10^{-2}$	3.7 $x10^{-2}$
0.074	3.4 $x10^{-4}$	3.9 $x10^{-4}$
2.197	3.1 $x10^{-2}$	3.5 $x10^{-2}$
0.3	2.2$x10^{-3}$	2.5$x10^{-3}$

$$Range = R = 0.0825[(1 + 22.4E_{\max}^{2})^{1/2} - 1] / \rho mat \qquad (8)$$

In order to check the results against other absorption coefficient for skin ν (cm^2.g^{-1}) the authors run the program with the values given by **LEICHNER (1994)** (See equation 9) in instead of the values taken from equation 5 but using the same range given by Eq. (6) (see table 8):

$$\nu = 0.474 * (E_{max}/3)^{-2.0} + 5.80 * (E_{max}/3)^{-0.82} \qquad (9)$$

Table 8. Comparison between mass absorption coefficient values

Beta Energy (MeV)	Mass Absorption Coefficient This work Equation 5 cm^2/g	Mass Absorption Coefficient LEICHNER work Equation 9 cm^2/g
0.063	1835.03	1212.62
2.271	7.72	8.12
0.074	1148.97	899.79
2.197	8.08	8.37
0.3	80.73	85.72

RESULTS

A computer program was developed using the "Mathematica" software **(WOLFRAM, 2004)** based on Table 3 and Eqs. (6-7), the values of tables 1 to 6 and resulted in a skin beta dose rate due to the use of a necklace with one 30 mm DU bullet between 3.0 mGy/h (100 μm) and 2.4 mGy/h (200 μm) rate as shown on Table 9.

Table 9. Skin dose rates for a 100 μm and 200 μm (Data from equations 1-7)

Radionuclide	Skin Dose Rate due to a necklace with one bullet 100 μm ($Gy\ h^{-1}$)	Skin Dose Rate due to a necklace with one bullet 200 μm ($Gy\ h^{-1}$)
Th^{234}	$1.3\ x10^{-15}$	$1.3\ x10^{-23}$
Pa^{234m}	$2.9\ x10^{-3}$	$2.4\ x10^{-3}$
Pa^{234}	$7.2\ x10^{-9}$	$5.7\ x10^{-9}$
Th^{231}	$8.4\ x10^{-5}$	$3.3\ x10^{-5}$
TOTAL	$3.0\ x10^{-3}$	$2.4\ x10^{-3}$

Table 10 shows the results obtained for the skin dose rate (100 μm) when using the two different equations for the mass absorption coefficients (Eq. 6) and **LEICHNER (1994)** (Eq. 9).

Table 10. Skin dose rates for a 100 μm skin thickness for different absorption coefficients data

Radionuclide	Skin Dose Rate due to a necklace with one bullet 100 μm Mass absorption coefficient from authors ($Gy\ h^{-1}$)	Skin Dose Rate due to a necklace with one bullet 100 μm Mass absorption coefficient from Leichner ($Gy\ h^{-1}$)
Th^{234}	$1.3\ x10^{-15}$	$4.3\ x10^{-13}$
Pa^{234m}	$2.9\ x10^{-3}$	$3.0\ x10^{-3}$
Pa^{234}	$7.2\ x10^{-9}$	$7.3\ x10^{-9}$
Th^{231}	$8.4\ x10^{-5}$	$8.3\ x10^{-5}$
TOTAL	$3.0\ x10^{-3}$	$3.1\ x10^{-3}$

CONCLUSIONS

The skin dose rates obtained in this work (3.0 mGy/h for 100 μm skin deep and 2.4 mGy/h for 200 μm skin deep) for the use of a necklace made with just one DU bullet, with the characteristics shown on Table 5 and dimensions of figure. 4, is in good agreement with

BLEISE ET AL. (2003) estimation (2.0 mSv/h) and the **HPA HOMEPAGE** (2.5 mSv/h).

Table 10 shows that a there is no significant difference between the two results ($3.0x10^{-3}$ and $3.1\ x10^{-3}$ Gy/h).

Taking into account the **ICRP60's (1991)** tissue weighting factor for skin of 0.01 results in an effective dose rate between 30 µSv/h and 24 µSv/h or between 263 µSv/y 210 µSv/y considering a full time exposure of 8760 hours/year that is not negligible in terms of radiological protection considering just one bullet.

It should be pointed out that this work did not considered the possibility of use of more than one bullet of 30mm in the necklace, the risk of inhalation by children of dust of depleted uranium due to leaching or destruction and ressuspension of bullet materials.

According to **BLEISE (2003)** the effective dose from inhalation of 1 mg uranium depleted to 0.2% is in the order of 0.12 mSv and considering the 1 mSv annual standard for the public it is equivalent to 8.3 mg (one bullet has approximately 300 g but not in a dust form). On the other hand for DU the ingestion risk is the order of 0.71 mSv/g. It was also not intention of this article to consider the possibility of ingestion of DU by children due to hands contamination, contamination of soil (crops) and water by DU ammunition.

Finally yet importantly, the program can be easily adjusted for the calculation of the beta dose rate due to the use of a necklace with other DU bullet size and numbers.

REFERENCES

BLEISE A, DANESI PR., BURKART W. Properties, use and health effects of depleted uranium (DU): a general overview. Journal of Environmental Radioactivity 64:93–112; 2003.

GRUN R., Beta dose attenuation in thin layers-Department of geology-McMaster University – Ontario- Canada https://www.aber.ac.uk/temp-ancient-tl/issue4_1/grun_atl_4%281%29_1-8.pdf /Accessed 07 May 2014.

HPA- HEALTH PROTECTION AGENCY - Available at: http://www.hpa.org.uk/Topics/Radiation/.../DepletedUranium/. Accessed 19 September 2011.

IAEA -International Atomic Energy Agency. Explanatory Material for the IAEA Regulations for the Safe Transport of Radioactive Material. 1985 Edition. 2. ed. Viena, IAEA (Safety Series, n°. 7); 1987.

ICRP PUBLICATION -60 - International Commission on Radiation Protection. Recommendations of the International Commission on Radiation Protection Oxford:Pergamon Press; ICRP Publication 60, Ann ICRP21 (1/3); 1991.

ICRP PUBLICATION 89- Basic Anatomical and Physiological Data for Use in Radiological Protection Reference Values-Ann. ICRP 32 (3-4), 2002.

LOEVINGER R, JAPHA EM, BROWNELL G L., in Radiation Dosimetry ed G L Hine and G L Brownell. New York: Academic Press; 1956.

LEICHNER, PETER K. -A Unified Approach to Photon and Beta Particle Dosimetry Department of Radiology, University of Nebraska Medical Center, Omaha, Nebraska- The Journal of Nuclear Medicine V.35 No. 10 October 1994.

WOLFRAM S. THE MATHEMATICA 5.1, Cambridge, UK, Wolfram Media; 2004.