

Differential Decay Analysis of the Alpha Dose of Depleted Uranium and the Neoplastic Risk in the Lungs of Gulf War Veterans

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Abstract

The purpose of this study was to estimate the time-zero lung burden of inhaled depleted uranium (DU) oxide particles in the lungs of Allied Forces' Gulf War Veterans. Eleven British, Canadian, and United States veterans have been quantitatively analyzed for the presence of four DU isotopes in the urine nine years after exposure by thermal ionization mass spectrometry (TIMS). Approximately half of the subjects tested positive for the presence of DU.

We have developed a new non-invasive technique for determining a cumulative minimum radiation dose for alpha-particle radiation from depleted uranium oxide in the respiratory system. Using the minimum biological half-life of DU derived from the Battelle model of simulated interstitial lung fluid, the total inhalational exposure to DU at time-zero was determined. The radiation dose can be calculated by integrating the total number of alpha-particle events from time-zero to ten years after exposure.

A 24-hour urine specimen of a subject containing 0.150 micrograms of DU corresponds to the inhalational exposure of 1.54 mg of DU at time-zero with an alpha-radiation dose of 4.4 millisievert (mSv) during the first year and 22.2 mSv of alpha-radiation to the lungs within ten years. Our values exceed the maximum permissible inhalational dose of uranium and warrant a need for further research of the biological half-life, metabolic pathways, and possible DU induced neoplastic alterations in the lungs.

Introduction

The use of depleted uranium (DU) weapons in the Gulf War and Kosovo conflict pose the problem of mass contamination of military personnel, peacekeepers, aid workers, and the civilian population to varying degrees of depleted uranium exposure. Depleted uranium is a heavy metal having toxicological and radiological properties, being only 43% less radioactive than natural uranium. The decrease in radioactivity is due primarily to the smaller ^{234}U content, not to the change in ^{235}U or ^{238}U concentrations that characterizes depleted uranium. DU has been shown in the recent literature to have adverse health effects including: renal damage¹, neurological deficits², neoplastic transformations³, and mutagenic effects⁴; and environmental effects including ground water and soil contamination⁵ with decreases in functional diversity of the soil bacterial community⁶.

While the industrial uranium compounds have been classified in three dissolution categories, UO_2 compounds correspond to class Y category dominated by the slow dissolution rate and processes of mechanical pulmonary clearance. Burning DU produces very insoluble uranium oxide compounds, UO_2 and U_3O_8 , both of which have a years long lung clearance rate⁷. While insoluble DU compounds are poorly absorbed in the GI system and pose little risk through ingestion, the long-term retention of DU can pose a significant risk in the lungs. Our study attempts to determine the radiation risk to the lung from inhaled DU particles to better understand the hazards from milligram and microgram quantities of inhaled depleted uranium aerosols.

Materials & Methods

Eleven British, Canadian, and United States Gulf War veterans with verified inhalational exposure to dust containing DU during the Operation Desert Storm, 8-9 years prior to sample collection, participated in the study. Each subject signed informed consent and had a 24-hour urine sample analyzed for total concentration and the quantity and ratio of four uranium isotopes: ^{234}U , ^{235}U , ^{236}U , and ^{238}U . The analysis was done by thermal ionization mass spectrometry (TIMS) at the Radiogenic Isotope Facility, Department of Earth Sciences, Memorial University of Newfoundland, Canada.

The analytical methodology involved evaporation, oxidation of organic matter with concentrated nitric acid, purification by ion-exchange chemistry, and mass spectrometry using a Finnigan MAT 262V thermal ionization multi-collector mass spectrometer operating in peak jumping mode using the secondary electron multiplier ion counting detector system to determine the isotopic composition.

Exposure to depleted uranium was indicated by an upward shift in the isotopic ratio of $^{238}\text{U}/^{235}\text{U}$ away from the natural uranium ratio of 137.9. The ratio of 141.0 was taken as the cutoff point for definitive exposure to DU. From the isotopic ratios of natural and depleted uranium and the ratio of the uranium in the sample, the percentage of the uranium that came from a DU source (DU fraction) could be calculated.

Five of the eleven subjects tested positive for DU with a mean $^{238}\text{U}/^{235}\text{U}$ ratio of 191.00 ± 2.90 . For these subjects with definitive DU exposure 8-9 years prior to sample collection, the alpha radiation dose from inhaled uranium was calculated.

Results

The alpha radiation dose from depleted uranium particles in the lungs can be calculated from the DU fraction, DU concentration in the urine (table 1), and the biological half-time of DU particles in the lungs, determined by the Batelle method of simulated interstitial lung fluid model⁸ which was taken as the minimum possible value for the biological half-time.

The rate of DU excretion, dN/dt , is proportional to the amount of DU in the lungs, N (eq1). Solving (eq1) for N gives the traditional decay equation with N_0 as the quantity of DU in the lungs at time-zero (eq2). N_0 can also be found from the current rate of excretion by substituting (eq1) into (eq2) and solving for N_0 (eq3). Integrating the right-hand side of (eq3) with respect to time gives the number of alpha-events over a given time period, N_{α} , where c is the conversion factor between grams of DU to alpha-particle events per day, 25,252 Bq/g (eq4). By substituting (eq3) into (eq4) for N_0 , an equation relating the rate of uranium excretion to the number of alpha-events over a specified time period is obtained (eq5). From this mathematical derivation the number of alpha-events and resulting radiation dose to the lungs from inhaled depleted uranium can be calculated.

The weighted average alpha-particle energy for the four isotopes was found to be 4.27 MeV per alpha-particle (table 2). The total number of alpha-events over a time period from (eq5) multiplied by the average alpha-particle energy gives the total energy imparted to the lungs. This is averaged over 2 kg of lung tissue to convert to units of Grays.

The weighting factors meant to modify absorbed dose to reflect no-threshold effects as a function an internal radiation event in the body are equal to 20 for a quality factor as a function of linear energy transfer. The dose in Gray multiplied by the value of the relative biological effect of alpha particles on the tissue gives units of milliSieverts.

Table 3 shows the gram-quantity of DU present in a 24-hour urine sample, the number of alpha-particles this amount of DU gives off per day, and the total number of alpha-particles and the resulting milliSievert dose over the first year and the first decade.



Equations

$$\text{Eq 1: } \frac{\partial N}{\partial t} = -\lambda N \quad \text{where } \lambda = \frac{\ln(2)}{T}$$

$$\text{Eq 2: } N = N_0 e^{(-\lambda t)} \quad \text{Eq 3: } N_0 = \frac{\left(\frac{\partial N}{\partial t}\right) e^{(\lambda t)}}{\lambda}$$

$$\text{Eq 4: } N_{\alpha} = c N_0 \int_0^{t_2} e^{(-\lambda t)} dt = \frac{N_0 \left(e^{(-\lambda t_1)} - e^{(-\lambda t_2)} \right)}{\lambda} \quad 0 \leq t_1 \leq t_2$$

$$\text{Eq 5: } N_{\alpha} = \frac{c \left(\frac{\partial N}{\partial t}\right) e^{(\lambda t)} \left(e^{(-\lambda t_1)} - e^{(-\lambda t_2)} \right)}{\lambda^2} \quad 0 \leq t_1 \leq t_2$$

Table 2

Radioactive Decay of Uranium Isotopes in Natural and Depleted Uranium				
Isotope	Natural Uranium (Bq/g)	DU (Bq/g)	Radiological Half-life (Years)	Alpha-Particle Energy (MeV)
^{234}U	12,342	1,815	2.45×10^5	4.76
^{235}U	568	159	7.04×10^8	4.39
^{236}U	0	71	2.34×10^7	4.48
^{238}U	12,342	12,406	4.47×10^9	4.19
Totals	25,252	14,451		4.27

Table 3

Radiation Dose to the Lungs from Depleted Uranium				
Subject	Excreted DU/24hrs (mg)	Time-Zero Lung Burden of DU (g)	Over the First Year (alphas) (mSv)	Over the First Decade (alphas) (mSv)
1	1.10×10^{-7}	1.13×10^{-3}	4.70×10^5 0.00	2.38×10^6 0.02
2	4.18×10^{-6}	4.29×10^{-2}	1.79×10^7 0.12	9.07×10^7 0.62
3	1.50×10^{-4}	1.54	6.41×10^8 4.39	3.25×10^9 22.22
4	3.55×10^{-5}	3.64×10^{-1}	1.52×10^8 1.04	7.70×10^8 5.26
5	4.92×10^{-6}	5.05×10^{-2}	2.11×10^7 0.14	1.07×10^8 0.73
Average	3.89×10^{-5}	3.99×10^{-1}	1.66×10^8 1.14	7.10×10^8 5.77
Std Dev	6.36×10^{-5}	6.52×10^{-1}	2.72×10^8 1.86	1.42×10^9 9.43
Std Error	2.84×10^{-5}	2.92×10^{-1}	1.22×10^8 0.83	4.34×10^8 4.22

Conclusion

Recent studies of the carcinogenic and mutagenic risk of depleted uranium (DU) suggest that long-term exposure to DU could be a contributing factor in the development of neoplastic disease in humans⁹. The linear extrapolation of high dose data to low dose lung cancer risk from alpha particles is currently being reevaluated for underestimating the exposure risk¹⁰. Internal contamination with DU particles has been described as a contributing factor in malignant alterations in different experimental models¹¹. The risk of inhalation of DU aerosols has been studied in the veterans of the Balkan conflict, exposed to the impact of Tomahawk missiles, by the simulated Gaussian plume model¹². Long term internal contamination with DU particles has contributed to the modification of established policies of DU contamination¹³ particularly in reference to the exposure assessment of the military, humanitarian aid workers personnel, and civilian population in the area of deployment of DU weapons¹⁴.

The interpretation of uranium urinary excretion measurements should be used in conjunction with the measurements of air samples in the scenarios of DU aerosol release. Our study provides a model of calculating the alpha radiation dose to the lung, from the urinary concentration, biological half-life, and DU fraction, derived from the isotopic ratio.

Table 1

Isotopic Data for Individual Samples					
Subject	% ^{238}U	% ^{235}U	$^{238}\text{U}/^{235}\text{U}$	% DU	U/24hrs (mg)
1	99.3154	0.6758	146.96	8.52	1.21×10^{-6}
2	99.4280	0.5663	175.58	29.64	1.41×10^{-5}
3	99.5603	0.4304	231.34	55.86	2.68×10^{-4}
4	99.4876	0.4945	200.77	43.28	8.20×10^{-5}
5	99.4862	0.4966	200.34	43.08	1.14×10^{-5}

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