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Plutonium in depleted uranium penetrators

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Depleted Uranium (DU) penetrators used in the recent Balkan conflicts have been found to be contaminated with trace amounts of transuranic materials such as plutonium. This contamination is usually a consequence of DU fabrication being carried out in facilities also using uranium recycled from spent military and civilian nuclear reactor fuel. Specific activities of $^{239+240}$ Plutonium generally in the range 1 to 12 Bq/kg have been found to be present in DU penetrators recovered from the attack sites of the 1999 NATO bombardment of Kosovo. A DU penetrator recovered from a May 1999 attack site at Bratoselce in southern Serbia and analysed by University College Dublin was found to contain 45.4 ± 0.7 Bq/kg of $^{239+240}$ Plutonium. This analysis is described. An account is also given of the general population radiation dose implications arising from both the DU itself and from the presence of plutonium in the penetrators. According to current dosimetric models, in all scenarios considered likely, the dose from the plutonium is estimated to be much smaller than that due to the uranium isotopes present in the penetrators.

KEY WORDS: *Uranium; Plutonium; War; Yugoslavia; Radiometry*

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INTRODUCTION

In the 1990s starting with the Gulf War, again in Bosnia and Herzogvina and most recently in 1999 in Kosovo and Serbia ammunition made from depleted uranium (DU) was used by forces from NATO countries. Although it has not been confirmed it is considered possible that DU ammunition originating from the DU stock of the former Soviet Union or other non-western sources may also have been used on occasions in conflicts in other parts of the world in the 1990s. It is variously estimated that about 30000 rounds of DU armour penetrating ammunition were used in the NATO aerial bombardment of Kosovo in 1999. The majority of these penetrators having missed hard targets remain buried in the ground at depths that make them very difficult to detect and recover. In addition to the main targets that were in Kosovo it is estimated that perhaps another 3000-5000 30-mm-caliber DU rounds were directed at a number of targets in Serbia. Up to the present only a small number of DU penetrators recovered from Kosovo and Serbia has been analysed for their radionuclide content (1). All these analyses are in general good agreement and have confirmed that the penetrators are in fact composed of metallic depleted uranium having the following approximate uranium isotope specific activities (Bq/kg): ^{238}U (12 x 10^6), ^{234}U (1.5 x10^6), ^{235}U (1.5 x10^5) and ^{236}U (5.5 x 10^4). The presence of ^{236}U indicates that recycled uranium from nuclear fuel was probably part of the DU production. In February 2001 it was also confirmed that trace amounts of plutonium were present in some DU penetrators (2). The presence of this plutonium was further confirmation that recycled uranium was probably involved in the production of the DU. With the connotations that exist in the public mind between plutonium and nuclear weapons its presence in the DU penetrators will be perceived by the general public to present an even greater health hazard than that of the DU itself. While such a perception may be grossly incorrect its existence needs to be confronted objectively and addressed. It was therefore considered appropriate to present in this paper information relating to possible radiological health implications of plutonium in DU penetrators used in the 1999 conflict in Yugoslavia. (Other papers in this volume deal in greater detail with the radiological health implications of the uranium isotopes in the penetrators). In order to help quantify the possible doses and risks from the plutonium contamination of the DU penetrators a description is first given in this paper

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of the analysis of a 300 g DU penetrator recovered intact from a May 1999 target site in the southern Serbian community of Bratoselce which is 10 km northeast of Preševo.

ANALYSIS OF DU PENETRATOR

Two grams of DU fragments machined from the penetrator were used for alpha and gamma spectrometry analyses. After the non-destructive gamma analysis was performed, the DU samples went through a variety of strictly controlled chemical treatment stages involving its careful dissolution in acid and other treatments. From the resulting solution two precise 0.5 ml aliquots were taken for a duplicate uranium analysis, and the rest of the solution was reserved for the plutonium analysis.

In the non-destructive gamma analysis fragments from the penetrator were combined in a polyethylene vial and measured by high-resolution gamma spectrometry using an n-type germanium detector with a relative efficiency of 30% and resolution of 1.70 keV (FWHM) at 1.33 MeV. The accumulated gamma spectrum is shown in Figure 1.

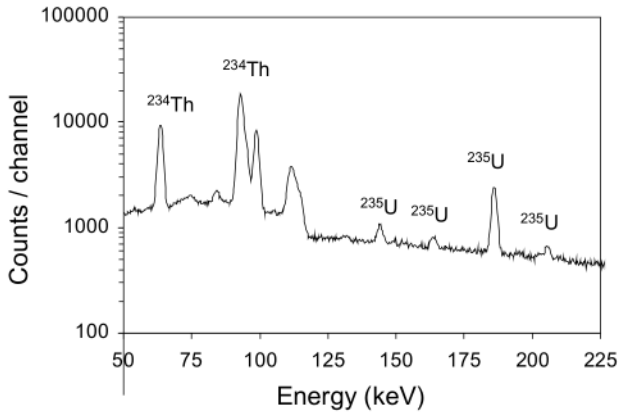

Figure 1. Gamma spectrum of the DU penetrator

The material of the penetrator was confirmed by our analysis to be DU by using a recently proposed convenient and rapid method for discriminating between natural and depleted uranium using gamma spectrometry (3).

URANIUM AND PLUTONIUM ANALYSES

In order to quantify the activities of the uranium and plutonium in the penetrator appropriate amounts of tracer or chemical yield monitor materials were added to a known amount of dissolved sample. In the case of the uranium analyses ^{232}U chemical yield monitor (NIST SRM 4324A) was used and in the case of plutonium analysis the chemical yield monitor was ^{242}Pu (NIST SRM 4334D). A detailed description of the radiochemical methods employed can be found in León Vintró and Mitchell (4).

At the end of the radiochemical processing samples of the resulting solution were transferred to an electrolytic cell and plated onto stainless steel discs. The alpha activities of the electroplated discs were measured using a multiple alpha spectrometer detector system, specially designed for low-level analysis using *Canberra*, passivated (ion-) implanted planar silicon (PIPS) detectors.

Uranium analysis: The alpha spectrum of the purified uranium fraction contains a number of well-resolved peaks including that of the ^{232}U tracer, as shown in Figure 2.

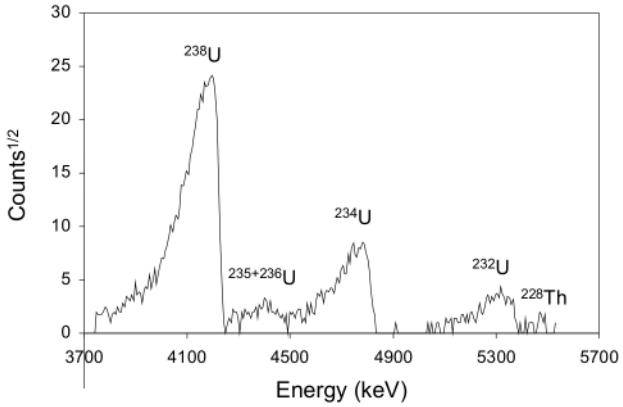

Figure 2. Pulse height spectrum of uranium from the DU penetrator

The results for all uranium isotopes are in good agreement with those determined by other researchers (Table 1).

Table 1. Uranium specific activities (Bq kg^{-1}) in DU penetrators (1,2)

Sample	Analysis Lab	^{238}U	^{234}U	^{235}U	^{236}U
UCD-01	UCD	12.20×10^6	1.43×10^6	1.66×10^5	7.40×10^4
UCD-02	UCD	12.10×10^6	1.44×10^6	1.90×10^5	7.40×10^4
ZA/R-00-505-01	AC Spiez	12.37×10^6	1.16×10^6	1.60×10^5	6.10×10^4
ZA/R-00-505-02	AC Spiez	12.37×10^6	1.39×10^6	1.39×10^5	6.19×10^4
ZA/R-00-500-16	AC Spiez	12.37×10^6	1.62×10^6	1.62×10^5	6.10×10^4
Kokouce	STUK	12.70×10^6	1.55×10^6	1.55×10^5	5.72×10^4

The presence of ^{236}U in the penetrator indicates likely contact of the DU during its production with recycled nuclear fuel.

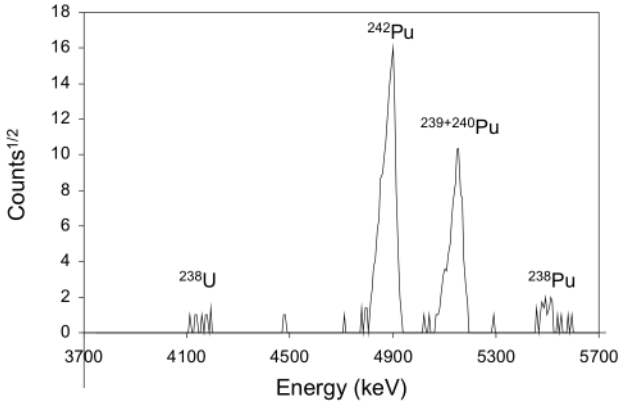

Figure 3. Pulse height spectrum of plutonium from the DU penetrator following separation by ion exchange and solvent extraction

Plutonium analysis: The pulse height spectrum for plutonium, following radiochemical separation by ion exchange and solvent

extraction with TIOA - xylene, is shown in Figure 3.

The specific mass activity of $^{239+240}$Pu in the DU penetrator analysed is given in Table 2, together with $^{239+240}$Pu specific mass activities reported by other laboratories for other DU penetrators used in the Balkans.

Table 2. Plutonium specific activities in DU penetrators (1,2)

	Laboratory	$^{239+240}$Pu (Bq kg^{-1})
UCD-01	UCD	45.4 ± 0.7
ZA/R-00-505-01	AC Spiez	<0.8
ZA/R-00-505-02	AC Spiez	3
ZA/R-00-500-16	AC Spiez	1
Kokouce	STUK	<0.8
Ceja Mountain	SSI	12

Although the specific mass activity of $^{239+240}$Pu in the DU penetrator analysed at University College Dublin (UCD) at about 45 Bq/kg is higher than that reported by other laboratories, it is in good accord with values quoted for mean $^{239+240}$Pu concentrations in DU armour used in tanks, at 85 Bq kg^{-1} (5). The ^{238}Pu/$^{239+240}$Pu activity ratio as determined from the alpha spectra, at 0.039±0.008, is typical of low burn-up plutonium. The presence of plutonium indicates that the DU employed to manufacture the penetrator was mildly contaminated with plutonium, at some point in time, most likely during the enrichment process.

HUMAN EXPOSURE IMPLICATIONS

Uranium: The overwhelming majority of the DU penetrators used by NATO forces in Yugoslavia in 1999 still remain buried in the ground and did not strike targets of sufficient hardness, such as armour, to cause them to be aerosolised. It remains to be confirmed but it is likely therefore that only a very small number of persons may have inhaled or ingested DU in aerosol or small particle form during and in the immediate aftermath of hostilities. International attention up to now has focussed primarily on the possible health effects from DU on servicemen or other personnel from abroad stationed in the Balkans with the effects on the local population receiving minor attention. For the local population in target areas the most likely long term future exposure to DU in the Yugoslav environment will therefore be by skin contact when penetrators are picked up or by ingestion of drinking water or perhaps food contaminated by the DU which has dissolved and migrated in the environment. In the case of small children ingestion of soil contaminated by DU is another possible pathway. In the case of handling the penetrators the radiation dose to skin in contact with DU metal is about 2.5 mSv /hour but radiation damage deterministic effects, such as erythema or radiation burns, should not occur at this skin dose rate for short term contact with the DU. However, for prolonged (i.e. some weeks) direct skin contact with DU there will be a non-trivial increase above the baseline risk of skin cancer both fatal and non-fatal (6). For DU, which is inhaled or ingested, its partitioning within the body is governed by many factors such as its chemical state and biological parameters. While uranium deposits on bone surfaces and may remain there for many years in most cases nearly 90% of the intake will be excreted in the urine within a few weeks. A detailed discussion of uranium metabolism and both its chemical- and radio-toxicities is outside the scope of this paper (see other papers in this volume). Recent reviews of the extensive literature dealing with these topics suggest that DU health effects to communities living in target areas will in most cases be negligible with DU intake at any conceivable level unlikely to have an appreciable potential for chemical or radiological carcinosis (7). In this context it is appropriate to point out that the two largest human studies to date on the health effects of uranium were cohort studies, mainly carried out some decades ago, of adult (mainly male) uranium workers in the US and in the UK (7). By the exacting standards of present day radiation and chemical epidemiology these studies had numerous deficiencies which in some degree calls into question their transferability and usefulness in assessing the long term health effects of DU exposure in a general population, which includes children and persons with existing health problems (6). It is therefore considered prudent and equitable to the general populations in the target areas that as a pre-requisite to actual health studies a properly designed and executed assessment of their exposure to DU should be carried out. In a pilot study earlier this year for the first time DU was found to be present and was measured in the urine of a small number of persons from Bosnia and Kosovo (8). Estimates of DU burdens as high as 288 μg were made. If, to a first approximation, it is assumed that DU body burdens are log-normally distributed in an exposed general population then a small percentage of the population may have DU body burdens much higher than this. This suggests that DU concentrations in the urine of the general population in target areas should be measured systematically and also the dissolution and migration rates of DU from buried penetrators into local drinking water supplies should be investigated. These and other related proposed investigations are to be encouraged by the appropriate Yugoslav authorities. In view of the multidisciplinary nature of such work and the need for access to specialised analytical equipment collaboration on an international basis in this work would seem to be scientifically desirable.

Plutonium: There are traces of ^{239}Pu in the environment which occur naturally due to the capture by ^{238}U in uranium ores of neutrons from spontaneous fission and alpha-neutron reactions. The mass concentrations of plutonium which are formed naturally are of the order of 1 part in 10^{11} which is exceeding low (9). The plu-

tonium of most interest from the radiological health perspective is anthropogenic and is produced as ^{239}Pu in a nuclear reactor as a result of neutron capture by uranium (^{238}U).

^{239}Pu has a radioactive half-life of 24360 years and emits alpha particles of energies between 5.115 (73.2%) and 5.51 (10.6%) MeV (the MeV is a unit of energy used to describe the energy of the radiation emitted from radioactive substances). Plutonium concentrations found in DU ammunition are commonly quoted as being $^{239+240}Pu$ because it is technically difficult using alpha spectrometry to distinguish between the alpha particles from ^{239}Pu and those from another isotope ^{240}Pu .

The most important pathway into the body for plutonium is by inhalation. On the other hand for ingested plutonium only about 1 part in 10^4 passes through the gut wall into the bloodstream with the rest being excreted. As wide variations in the ingested plutonium uptake by animals have been reported considerable fluctuations should also be anticipated in man. Inhaled plutonium activity deposited in the lung may, depending on factors such as its solubility, transfer into other body tissue and will concentrate mainly in the liver, kidneys and in particular in the skeleton. In the case of plutonium in the skeleton in common with other multivalent metals the activity will be concentrated in the bone surfaces as distinct from the internal volume of the bone. Plutonium seems to bond with the phosphate groups found in organic compounds on bone surfaces. Of particular note in this regard is the fact that internal bone surfaces surround bone marrow. Therefore alpha particle emitting radionuclides such as plutonium deposited on internal bone surfaces may irradiate bone marrow. The possibility that this could cause leukaemia in children and young adults has been the focus of much research in recent decades. In particular many studies have taken place in the UK to determine if, *inter alia*, plutonium in the environment as a result of activities at the British Nuclear Fuels Limited (BNFL) reprocessing plant at Sellafield in Cumbria (UK) could be a contributor to the incidence of leukaemia in the region. Due to the confounding effects of other environmental and socio-economic factors it has yet to be scientifically established if the leukaemia incidence in Cumbria is enhanced by the presence of radionuclides such as plutonium discharged to the environment by the operations at Sellafield (10). Nevertheless it is considered prudent by most radiological protection agencies to avoid contamination of the environment by plutonium.

DOSES AND RISKS

One can consider the health implications of the radioactive heavy metals uranium and plutonium from both radiological and chemically toxic perspectives . Uranium is nephrotoxic but even its toxic effects on the kidneys at most appear to cause minor and reversible renal damage. The plutonium concentration of about 44 Bq/kg of DU which we have measured in a 300 gram DU penetrator recovered from south Serbia means that the total mass of plutonium present in such a penetrator is about 5.5×10^{-9} grams equivalent to about 0.019 ppb (ppb = parts per billion = 10^{-9}). While this is comparable to the level of plutonium naturally produced and present in uranium ores the removal of natural plutonium by chemical processing of the ore means that the plutonium in the penetrators almost completely originates from reactors. Due to its minute mass concentration even if it was possible for all the plutonium from a single DU penetrator to be ingested or inhaled its chemical toxic effects are incalculable and in any case would not be possible to detect. In a similar manner the radiological risk posed by the plutonium present in the DU will be exceedingly low. In this case however it is possible to calculate the radiation dose from the plutonium and to compare it to both the doses from the DU itself and also with the natural radiation doses that the population always receives. To make a worst case estimate we will consider the radiation doses arising from inhaled DU particles. This is because the radiation doses from both uranium and plutonium arising from inhalation are much greater than those if the same mass of material is ingested. Based on current models of the ICRP (International Commission on Radiological Protection) we estimated the radiation doses that would arise from the inhalation of 1 mg of insoluble DU particles containing the plutonium at a specific activity of 45 Bq/kg (11,12). For the DU itself the dose was estimated to be about 0.15 mSv and that from the plutonium to be about 0.7 nSv (nSv = 10^{-9} Sievert). For the DU the ALI (Annual Limit of Intake), based on the public annual dose limit of 1 mSv, would be about 5 mg. At the uranium ALI the plutonium contribution to dose would be only about 5 nSv which is by any standards an exceedingly low dose. If the DU contaminated with plutonium were ingested rather than inhaled the doses would be even smaller. It should be noted that the average annual radiation dose from natural radiation (i.e. radon, external gamma, cosmic radiation) in Yugoslavia is probably about 2.5 mSv while in parts of Kosovo due to uranium mineralisation much higher natural radiation levels are known to exist (13). On the basis of these calculations there is no likely scenario of human intake in which the radiation doses and associated risks from the plutonium in these DU penetrators could even approach a level that would be considered a matter of health concern for radiological reasons by such bodies as the ICRP or for reasons of chemical toxicity by the WHO. This is not however the case for the chemical toxic effects of the uranium isotopes in the DU. On the basis of the chemical toxicity of uranium the WHO has derived a provisional guideline for drinking-water quality of 0.002 mg/l

which for a 500 l/year consumer corresponds to an ALI of 1 mg (14). It should however be pointed out that this is provisional and is substantially more stringent (due to still existing uncertainties in our knowledge of uranium chemical toxicity) than the corresponding guidelines of other agencies. Depending on the isotopic mix in the DU based on a 1 mSv/year public limit the radiological ALI for ingested uranium is about 1 g and the radiologically based DDWC (Derived Drinking Water Concentration) is about 2 mg /l. (Even in this case the WHO has the more stringent guideline of 0.28 mg/l.) These observations further emphasise the greater importance of chemical toxicity relative to radiological toxicity in the case of uranium. It also strengthens the recommendations, already given above in this paper, that a careful study of the dissolution and migration of DU into the drinking water supplies in target areas coupled to urine analyses of the general population should be given priorities by the relevant national and international agencies.

CONCLUSION

Plutonium has been detected in trace amounts in DU penetrators recovered from 1999 attack sites in Kosovo and Serbia. In this paper the level of plutonium measured in one such penetrator was about 44 Bq/kg of DU. The associated dose from the plutonium for an inhaled intake of 1 mg is estimated to be about 0.7 nanoSievert (10^{-9} Sv) . Even allowing for the uncertainties in such an estimate this is an exceedingly low radiation dose and is a minute fraction of natural radiation doses. The presence of plutonium in the DU penetrators does however raise a number of ethical questions. For example its presence would seem to be in conflict with the basic radiation protection principle of justification as the plutonium serves no useful purpose as part of the penetrator. The fact that the DU penetrators used so far would appear to have plutonium contamination at a very low level of almost no radiological health significance does not mean high levels of plutonium may not appear in DU from other sources used in future conflicts. In contrast to plutonium it would appear that studies should be carried out both into the dissolution and migration of the DU into drinking water supplies in target areas and also that the DU content of the urine of the local population should be assessed.

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