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Early results of studies on the levels of depleted uranium excreted by Balkan residents

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Urine samples collected from residents of Bosnia and Herzegovina and Kosovo were analysed to determine their natural and depleted uranium content using MC-ICP-MS. All may have been exposed to depleted uranium released as a consequence of the deployment of armour-piercing rounds by the US Air Force. A ^{236}U tracer was employed to determine chemical recovery. Early results suggest that the levels of natural and depleted uranium excretion by the subjects, which ranged in age from 1 to 71 years, ranged from 2.8 - 58.2 ng d^{-1} and 1.3 - 46.3 ng d^{-1}, respectively. The results suggest accumulated body burdens of depleted uranium ranging from close to zero to 46 μg. All the body burdens predicted are lower than published values for the uranium content of the body (90μg) and health effects are not predicted. Further studies are underway to check the provenance of the results.

KEY WORDS: *Uranium; Urine; Body Burden; Analysis; Bosnia-Herzegovina; Yugoslavia*

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INTRODUCTION

Uranium metabolism and toxicity are reviewed in previous publications (1,2). Uranium enters the body either from the lungs - following the dissolution of inhaled particles or from the gut. Subsequently, the vast majority is excreted within a few days in urine. Of that retained most is present in the skeleton, although significant deposits are also present for a short time in the kidneys. Uranium, like calcium, deposits in bone mineral where it is retained for long periods. Skeletal deposits of uranium are released into the blood stream when bone is resorbed during bone remodelling. Bone remodelling is a slow process in adult humans so uranium deposits persist in the skeleton for decades (Figure 1). In children bone turnover is faster, so that skeletal deposits are lost more quickly. When skeletal deposits of uranium are released into blood then some is re-deposited in bone, but most is excreted. It follows that monitoring the levels of uranium in urine can provide information on both the daily uptakes of uranium and on the size and rate of loss of existing body-burdens of this metal.
Using the uranium retention equations of Hursh and Spoor (3) the retention of uranium in the body at different times after its uptake can be calculated. These suggest that for a long period, spanning 100 days to a few thousand days after uptake about 10% remains in the body. Extrapolations of other data derived from human experiments suggest similar uranium retention during this period, but a much more rapid loss during the first few days post-uptake. In contrast, the predictions of the ICRP biokinetic model for uranium (ICRP, 1994) (4) are consistent with a lower retained fraction of about 2.3% for adults and 5.5% for children (see Figure 1).

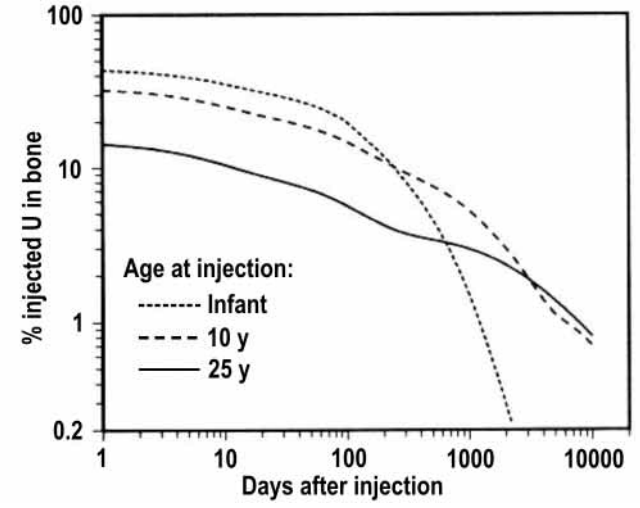


Figure 1. Reproduced from ICRP Publication 69 showing the retained fraction of uranium uptake in the skeleton as a function of time. Note: for long-standing burdens the skeletal deposit represents the vast majority of uranium in the body

Given the general acceptance of ICRP models and the similarity of

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their predictions to the well-established behaviour of chemically similar elements (e.g., barium) in the body, the ICRP model can be used as a base for predictions of uranium body burden. Overall, it would seem reasonable to assume that, for chronic continuous intakes by adults, an average of 2.3% of uranium uptake is retained and 97.7% excreted in urine. For children the equivalent values would be 5.5% and 94.5%, respectively. If so, it is possible to derive an estimate of uranium body burden if it assumed that the level of daily excretion represents either 97.7 or 94.5% of total uptake. This will be true until body burdens rise to levels where daily losses of uranium from body burden are sufficiently large to produce significant perturbations in the amount of uranium excreted. It follows that an estimate of body burden can be obtained by multiplying the estimated daily uranium retained by the number of days of continuous exposure (Assumption A).
However, it is also possible that uranium in urine represents only that lost from an established body burden with no additional contribution from recent uptake (Assumption B). For body burdens that have been established for 500 to 2000 days the ICRP predicted fractional loss of contemporary body burden approximately equals 0.03% d^{-1} for adults and 0.06% d^{-1} for children. It follows that if this assumption is made body burden may be estimated by dividing the measured output of uranium in urine by the fraction lost each day (i.e., 0.0003 or 0.0006). For the situation in the Balkans it is unlikely that either method will produce an accurate estimate of body burden since it is possible that uptake continues in the presence of an established body burden. Nevertheless, the above methodologies are likely to define the range of possible situations, with the true body burden lying somewhere between the calculated values.
As a result of the deployment of weapons (GAU-8 30 mm rounds fired by US A-10 ground attack aircraft) containing depleted uranium in the Balkans by NATO forces some metal has inevitably deposited in and contaminated the local environment. Subsequently, it has been suggested that civilian populations have become contaminated with depleted uranium either due to the consumption of contaminated food and drink or as a result of the inhalation of condensed uranium fume at the time of weapons deployment. To date, no studies have been undertaken to confirm such human contamination. The objectives of this study were to confirm the presence of and determine the possible extent of civilian contamination within three populations - two resident in Kosovo and one in Bosnia and Herzegovina.

STUDY SUBJECTS

Three populations were examined to determine the levels of depleted uranium excretion by members of the civilian population. The first was an ethnic Serb population possibly exposed to depleted uranium in August 1995 during the deployment of weapons to destroy a munitions factory at Hadžići, which is close to Sarajevo. Over a period of months this population relocated to the town of Bratunac, Republika Srpska in the north of Bosnia and Herzegovina. The second population was resident in the town of Đakovica in Kosovo. This is a population of in-patients at a regional general hospital located adjacent to a destroyed Yugoslavian barracks where NATO has confirmed the use of depleted uranium. The third population was selected from a small group of residents of the town of Klina, Kosovo who were not able to evacuate at the time of NATO attacks on armoured vehicles. This population may or may not have been exposed to locally deployed depleted uranium.
All adult subjects studied were informed volunteers and signed appropriate consent forms. In the case of children parents signed the consent forms. The volunteers were as follows:
- Bratunac, Bosnia and Herzegovina: Four adult males aged between 28 and 60 years.
- Đakovica, Kosovo: Five males aged between 1 and 69 years; four females aged 31 to 71 years.
- Klina, Kosovo: Three males aged between 14 and 67 years, five females aged between 13 and 68 years.
In addition, one member of the BBC crew participated in the study.

METHODS

All chemical methods employed had been previously tested using either fresh urine provided by the investigators or archived ashed urine samples - none of which contained depleted uranium.

Sampling

Each volunteer was provided with a 1L-capacity plastic bottle for urine collection and a data sheet for completion giving personal details of the volunteer. In all cases overnight urine collections were made. Each volunteer collected a timed urine sample, recording the start and finish time of collection on the data sheet. The duration of collection was in most cases about 8 hours, but collections organized by hospital staff in Đakovica were longer and averaged 14 hours.

Treatment of urine and isolation of uranium

All samples collected in the Balkans were transported by air back to the United Kingdom for analysis. Upon receipt, the volume of the samples was measured. They were then transferred to borosilicate glass beakers and evaporated to dryness. The residues were ashed at 500°C for about 10 hours, dissolved in 8M nitric acid, evaporated to dryness and then re-ashed at the same temperature. The resulting white inorganic residues were spiked with a known quantity of a ^{236}U yield-tracer. Subsequently, they were treated in one of two ways - depending

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upon the mass of residue present. Those samples containing small amounts of residue were immediately dissolved in 9M hydrochloric acid for uranium extraction by ion exchange chromatography. Those with a larger amount of residue were dissolved in approximately 1 L of dilute nitric acid and 0.5 g of calcium phosphate and 0.7 g of potassium hydrogen phosphate added. The solution was then neutralised by the addition of excess concentrated ammonia solution to give a final pH of 8.3 - 8.6. At this stage calcium / magnesium / uranium was precipitated as a phosphate. The phosphate was then extracted by centrifugation. The extracted precipitate was converted to chloride and dissolved in 9 M hydrochloric acid. The uranium was extracted using an ion exchange column - Dowex 1X8 200 - that had been pre-washed with 9 M hydrochloric acid. Iron and uranium were then sequentially stripped from this column with 8 M nitric acid. The iron containing solution was discarded and that containing uranium evaporated to dryness in a round-bottom PTFE container and retained for analysis. The chemical recovery of uranium, as determined using the ^{236}U yield-tracer, ranged from 3-86% (mean = 41%). All reagents used were of analytical grade quality. A distilled water sample was processed alongside the urine samples as a process blank.

Measurement of uranium

Each sample was dissolved in 0.5 mL of 2M nitric acid for uranium and uranium isotope determination by the Micromass IsoProbe, Multi-collector Inductively-Coupled Plasma Mass Spectrometry (MC-ICP-MS). Using this instrument the concentration of ^{235}U, ^{236}U and ^{238}U in the solutions and the ^{235}U:^{238}U ratio were measured. Data were corrected for tail, instrument memory and for mass bias using regularly interspersed analyses of natural U. $^{235}U/^{238}U$ was corrected for ^{235}U and ^{238}U in the ^{236}U tracer: this correction was usually about 1% of the measured ratio, and less than 7% except for the blank. From the ^{235}U:^{238}U ratio the fraction of depleted uranium in the urine was calculated. A measured ratio of 0.007253 (2SE = ± 0.000001) demonstrated the absence of depleted uranium in the sample. Lower ratios demonstrated the presence of depleted uranium in the sample. (The mean ratio measured for two depleted uranium samples, obtained from DU penetrators recovered in Kosovo and Southern Serbia was 0.001996 ± 0.000001.) The lowest ratio measured in urine was 0.004831 ± 0.000001, equivalent to a depleted uranium fraction of 0.51 in the urine. Natural and depleted uranium solutions and blanks were employed to ensure the quality of the analysis. The process blank contained 23pg of uranium, of which ca. 15pg may have been depleted, subject to large uncertainties resulting from correction for 235 and 238 from the ^{236}U tracer. This amounted to 1% or less of the level of uranium in the samples (2.2 - 45.7ng).

Calculation of estimated body burden

The amount of natural and depleted uranium in each urine sample was calculated, as described above, from the total uranium content and the fraction of depleted uranium present. Subsequently, the sample collection period for each subject was used to calculate equivalent 24-hour uranium excretion levels in urine. In the case of depleted uranium the 24-hour excretion levels were used to estimate body burden. Two alternative assumptions were made to produce estimates (see introduction). For the first estimate (Assumption A) it was assumed that the amount of depleted uranium in the urine represented either 97.7 or 94.5% (depending on subject age) of the daily uptake of depleted uranium from the gut or from a slowly dissolving lung deposit. The remaining fraction (2.5 or 5.5%) was assumed to have been retained in the body to build body-burden. An estimate of body burden was, therefore, calculated from the product of the retained fraction and the estimated number of days of exposure. For the second estimate (Assumption B) it was assumed that the estimated 24-hour excretion level of depleted uranium in urine represented that fraction of the established body burden that was lost to excretion each day as a result of bone resorption. In this case, the 24-hour level was divided by 0.0003 (or 0.0006 - the case of a child). This fraction is representative for uranium body burdens that have been established for periods ranging from about 500 to about 2000 days.

RESULTS

Calculated uranium excretion levels and predicted body burdens for volunteers that provided urine for analysis are shown in Table 1.

Table 1. Calculated uranium excretion levels and predicted body burdens for volunteers that provided urine for analysis

Town of Domicile	Subject Gender	Subject Age	NU Excreted ng / day	DU Excreted ng / day	Fraction DU	Body Burden A	Body Burden B
Bratunac	M	56	58.2	30.9	0.35	1.02	31
Bratunac	M	42	36.2	12.2	0.25	0.40	12
Bratunac	M	28	28.3	10.3	0.27	0.34	10
Bratunac	M	60	37.7	8.7	0.19	0.29	9
Djakovica	F	66	20.0	7.6	0.28	0.10	8
Djakovica	F	40	31.6	8.0	0.20	0.11	8
Djakovica	M	69	13.0	5.4	0.29	0.07	5
Djakovica	M	21	9.4	4.5	0.32	0.06	5
Djakovica	M	1	2.8	1.3	0.31	0.01	1
Djakovica	M	6	9.1	4.7	0.34	0.06	5
Djakovica	F	31	13.7	7.9	0.37	0.11	8
Djakovica	M	11	6.9	2.8	0.29	0.04	3
Djakovica	F	71	*	*	0.08	*	*
Klina	F	13	16.6	5.0	0.23	0.07	5
Klina	F	37	32.6	7.8	0.19	0.10	8
Klina	M	15	18.2	4.8	0.21	0.06	5
Klina	F	36	22.1	3.8	0.15	0.05	4
Klina	F	18	31.4	19.1	0.38	0.25	19
Klina	M	46	35.9	36.9	0.51	0.49	37
Klina	F	37	10.5	7.9	0.43	0.10	8
Klina	M	67	53.4	46.3	0.47	0.62	46
Visitor	M	32	34.7	5.9	0.15	0.0	6

* no result (^{236}U tracer not added)

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Continuing studies

The results presented below are provisional and further studies are being undertaken to confirm them. The ^{235}U:^{238}U ratios measured are precise and accurate, but the possibility of accidental sample contamination by depleted uranium, while considered unlikely, has not been finally excluded. Studies are underway to examine the provenance of the results obtained. These include the analysis of repeat samples to be collected in Kosovo and the processing of "unexposed" control urine collected in the UK. Given the production timeline imposed upon the study by its sponsor, BBC Scotland, it was not possible to make these checks prior to the release of the data into the public domain.

Urinary levels of depleted uranium

Table 1 shows the indicated levels of natural and depleted uranium excretion, by the subjects. It can be seen that, for the volunteers from Bratunac, the estimated daily excretion levels ranged from 8.7 - 30.9 ng d^{-1} (mean = 15.5 ng d^{-1}) and the equivalent mean estimate body burdens ranged from 0.29 - 31 μg. For the hospital patients in Djakovica the levels were lower. Estimated 24-hour excretion levels of depleted uranium ranged from 1.3 (in the case of a 1 year-old infant) - 8.0 ng d^{-1} (mean = 5.8 ng d^{-1} (excluding the result for the infant)) and the equivalent best estimate of body burdens ranged from 0.1 - 0.11 μg. For the residents of Klina, estimated daily excretion levels were very variable and ranged from 3.8 - 46.3 ng d^{-1} (mean = 16.5 ng d^{-1}). However, in the case of Klina three volunteers, one a 67-year old male, one a 46-year old male and the other a 18-year old female excreted much more depleted uranium than the others measured. The urine sample provided by the single BBC team member analysed indicated a daily, depleted uranium excretion of 5.9 ng (since this person had had no known previous exposure to depleted uranium and, therefore, no known pre-existing body burden to mobilise). This level was typical of the levels excreted by most of the other Kosovan residents.

Table 1 shows that, as expected, the body burdens calculated using each assumption were quite different and were greatest when it was assumed that urinary depleted uranium resulted from the loss of uranium from an established body burden.

CONCLUSION

The preliminary results described above suggest that all of the subjects assayed showed some level of depleted uranium excretion. Since not all were likely to have been sufficiently close to the impact sites of the deployed depleted uranium to have sustained significant intakes this finding, if confirmed, most likely indicates that the metal is now present in the food chain and / or drinking water. This conclusion is supported both by the finding that depleted uranium was excreted by a subject born after the conflict and by the visiting BBC staff member. Within both of the Kosovan groups studied daily, depleted uranium intakes from these sources would seem to be about 6 or 7ng (cf. a mean of 24.9ng d^{-1} for natural uranium). Where excretion levels are much higher they may indicate additional environmental intakes or the presence of established body burdens resulting from specific exposures. The size of these body burdens is unknown, but should not exceed the maximum predicted burden of 46μg, for a resident of Klina. Given the lower amount of depleted uranium deployed in Bosnia and Herzegovina than in Kosovo - 3T rather than 10T and the remoteness of Bratunac from the site of depleted uranium deployment in Sarajevo - the results produced for the subjects from Bratunac are consistent with their claims that they were exposed to depleted uranium released during the bombing of the Hadžići munitions factory. If so, then these exposures were most likely by inhalation and it is possible that body burdens will continue to grow as depleted uranium deposits in the lungs continue to be mobilised. However, at 1660 days since exposure most of the lung deposit will have cleared and the potential for further systemic depleted uranium uptakes will be limited. Overall, all of the estimated body burdens are much less than the reported body burden of natural uranium in man (90μg (5)). It follows that the radiation dose from uranium to the skeleton of the volunteers is likely to be radiologically insignificant and will be dominated by that dose received from their, more radioactive, body burdens of natural uranium. In turn these doses will be much smaller than those received from other alpha-emitting radionuclides that are more common in the body - including radon-220 and its daughters.

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