

FINAL REPORT OF THE DEPLETED URANIUM  
OVERSIGHT BOARD

SUBMITTED TO

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## FOREWORD

This report presents a summary of the work carried out by the Depleted Uranium Oversight Board during its five-year period of work from 2001-2006. The main report represents the views of the majority of Board members, including all who were appointed as independent scientific experts, and also those who were nominated by the Royal British Legion and the British Red Cross. Four members of the Board, all nominated by the National Gulf Veterans and Families Association, disagreed with some parts of the main report. They were asked by the Chairman to produce a statement, highlighting those aspects of the main report with which they agreed and those with which they disagreed. This minority statement is set out at pages 51-75. Unfortunately, despite constructive discussion, it proved impossible to resolve the differences of opinion that it describes.

The minority statement is presented in a spirit of openness and fairness. The other members of the Board acknowledge that it advances considered opinions which are genuinely held. However, they take no responsibility for the accuracy of its content, and dissociate themselves from its reasoning and conclusions, which they believe to be seriously flawed scientifically.

## SUMMARY

The Depleted Uranium Oversight Board (DUOB) was established in 2001 to oversee a testing programme for British veterans (military and civilian) who wished to know whether they had been significantly exposed to depleted uranium (DU) in the 1990/91 Gulf War or during later military operations in the Balkans.

Following initial pilot work to establish the validity and sensitivity of analytical methods, a test was implemented based on the measurement of uranium isotopes in 24-hour samples of urine. Biokinetic calculations indicated that the assay was sufficiently sensitive to detect past exposures to DU that (according to mainstream medical and scientific thinking) would have material implications for health.

Urine samples were collected through regional clinics based mainly in NHS occupational health departments, and were analysed at one or both of two laboratories. The results were then reported back to the veteran by a company contracted to co-ordinate the day to day running of the programme. To ensure that analytical performance remained satisfactory throughout the programme, the majority of samples were analysed independently in duplicate by the two laboratories. As a further check, a third laboratory prepared urine samples, spiked with known quantities of DU. These were then delivered to the laboratories through the regional clinics and analysed without knowledge of their origin.

The launch of the testing programme in September 2004 was publicised by a press release and advertisement in a national newspaper, and through the internal communications of the armed forces and veterans' organisations. The programme remained open until January 2006, by which time the number of new applications for testing had tailed off virtually to zero. The impending closure of the scheme was also widely publicised.

The results from duplicate analyses and from the "blinded" analysis of spiked samples confirmed that the expected performance of the two analysing laboratories was maintained over the course of the programme.

Altogether, 464 veterans underwent testing in the main programme, including two who were tested twice. Most had served in the armed forces during the Gulf War (219), the Balkans campaign (80) or both (123). None of the veterans tested had detectable exposure to DU. Total 24-hour excretion of uranium exceeded 30 ng in eight samples, the highest value being 497 ng. These higher than average excretions of natural uranium may have reflected unusual dietary or other environmental exposures.

Following their test, 39% of veterans returned a short questionnaire giving their views on the testing programme. Satisfaction with the service provided was generally high.

In addition to their application in the testing programme for veterans, the analytical methods developed under the auspices of the DUOB proved useful in biomonitoring of military personnel with potential exposure to DU during the 2003 invasion of Iraq (Operation Telic).

## Introduction

This report summarises the work of the Depleted Uranium Oversight Board (DUOB) over its five-year history, and describes the findings from the exposure-testing programme that it provided.

The DUOB was established in September 2001, following an announcement by the Minister for Veterans, Dr Lewis Moonie. Its prime purpose was to oversee the development and letting of contracts for a urine testing programme to assess past exposure to depleted uranium (DU) in veterans who wished to know whether they had been significantly exposed in the 1990/91 Gulf War or during later military operations in the former Republic of Yugoslavia. As well as members of the UK armed forces, the programme was to include civilian employees of the UK Ministry of Defence (MOD), civilians working under contract to the UK Ministry of Defence, and civilians employed by other UK Government departments or non-governmental organisations working in support of UK military operations.

The original stimulus to the testing programme had been concerns among Service personnel prompted by media coverage of DU. This had led to an announcement on 9 January 2001 by Mr John Spellar, Minister for the Armed Forces, that a voluntary screening programme would be initiated for veterans. The Surgeon General set up an expert advisory group under the leadership of the Medical Officer-in-Charge of the Institute of Naval Medicine. The group recommended a test for historical exposure to DU based on the measurement of uranium isotopes in urine. After two rounds of public consultation, this recommendation was adopted.

In addition to development and oversight of the testing programme, the remit of the DUOB included:

- consideration of epidemiological studies to establish the distribution and determinants of excretion of uranium isotopes in urine and the relation of historical exposure to DU to possible biological and health effects (assuming that satisfactory methods of testing could be established);
- ensuring promulgation of the findings of the testing and associated research;
- advising on other possible methods of exposure assessment; and
- commenting on the development of biological monitoring tests to be used by MOD in future operations where DU was used.

The Board's full terms of reference can be found at Appendix A.

## Membership

In order to pursue its objectives, the DUOB required expertise in relevant medical and scientific disciplines (radiation medicine, occupational medicine, public health, radiation protection, toxicology, radiobiology, biokinetic modelling, epidemiology, analytical chemistry and mass spectrometry). It was also important to include representation (technical and/or lay) of major stakeholders (MOD, The Royal British Legion, the National Gulf Veterans and Families Association, and non-governmental organisations) as well as independent doctors and scientists. The names and affiliations of DUOB members are set out in Appendix B.

## Summary of activities

The DUOB met 23 times over the course of its work, the first meeting being on 27 September 2001, and the last on 31 October 2006. The minutes of each meeting were published on the DUOB website (<http://www.duob.org.uk/>), and will be archived with this final report.

The work of the Board focused mainly on the following topics:

- Review of the relevant science
- Piloting and evaluation of laboratory methods
- Arrangements for admission to the testing programme
- Arrangements for the collection, transport and analysis of samples
- Arrangements for feedback of results to participants
- Information for veterans and their advisors
- Launch, uptake and closure of the testing programme
- Quality assurance and audit of the testing programme
- Statistical analysis of findings from the testing programme
- Archiving of data
- Epidemiological studies of exposure to DU and associations with health outcomes
- Other possible methods of exposure assessment
- Biological monitoring for DU

These areas of activity are described in more detail in the sections that follow.

## Scientific background

The Board was required to develop a test for past exposure to DU based on measurement of uranium isotopes in urine. Natural uranium, as it occurs in the earth's crust, comprises principally the isotope U238, but with lesser quantities of a second, more radioactive isotope, U235. The ratio of U238 to U235 in natural uranium is approximately 137.9. Enriched uranium, which is used as a fuel in nuclear reactors, is derived from natural uranium by a separation process, and has an isotope ratio less than 137.9. Depleted uranium is a by-product of this process (essentially the uranium that remains after the enriched uranium has been extracted), and has an isotope ratio greater than 137.9 (usually between 300 and 500). It is only weakly radioactive, but has useful physical properties. In particular, it is 1.7 times as dense as lead. This has been exploited in engineering applications (e.g. in the construction of counterweights for rudders).

The main potential for exposure to DU in military operations arises from its use in armour-piercing rounds. When a DU round strikes an armoured target, the DU undergoes spontaneous combustion, a substantial fraction being converted to a fine aerosol of largely (> 65%) insoluble uranium oxides. If these particles of relatively insoluble uranium oxides are inhaled, they are ingested by macrophages (cells that scavenge foreign material), some of which remain in the lung or its associated lymph nodes long-term. From here, the uranium is slowly dissolved, and excreted in the urine over a period of many years.

All of us regularly take in small quantities of natural uranium in our diet (for example in drinking water), most of which we excrete in our urine. If a person has been exposed to depleted uranium, either through inhalation of the oxide particles formed when a DU round hit an armoured target, or more rarely because they have been wounded by fragments of a DU round, then some of the uranium that they excrete in their urine will be depleted. In other words, the ratio of U238 to U235 will be higher than the normal value for natural uranium of 137.9.

The theory underpinning the proposed method of urine testing for historical DU exposure is that this perturbation of the isotope ratio should be detectable, and that by measuring the ratio in urine, and also the total daily excretion of uranium, it should be possible to work out how much DU a person is excreting daily. Furthermore, from knowledge of the biokinetics of uranium oxides (i.e. the rates at which they are metabolised and excreted from the body), an estimate can be made of the daily excretion of DU that would be expected at a specified interval after inhalation of a given quantity of DU (Appendix C).

As with any toxic material, the health risks from DU will depend on the route (inhalation, ingestion etc) and extent of exposure. Among the potential adverse effects of inhaling DU, the two that are of most concern are chemical toxicity to the kidney, and an increased risk of cancer (principally lung cancer and perhaps lymphoma) from its radioactivity. Other adverse effects might also occur, but only at higher levels of exposure. Appendix D describes the quantitative relation of radiation dose and kidney concentration of uranium to DU measured in urine, and proposes an upper limit for daily DU excretion by veterans (2 nanograms per day), below which no material impact on health would be expected.

### **Piloting and evaluation of laboratory methods**

Before a testing programme could be initiated, it was necessary to establish that uranium isotopes could be measured in urine with sufficient accuracy and sensitivity to detect exposures with potential implications for health. As a first step, therefore, the Board commissioned a pilot study to assess existing analytical capabilities. Following a tender exercise, five laboratories were each asked to analyse a number of urine samples, 'spiked' with varying concentrations and ratios of uranium isotopes. The spiked samples were prepared by an independent co-ordinating laboratory. The study ended in July 2002 with inconclusive findings. A problem appeared to have occurred from contamination during sample preparation, illustrating the major technical challenge of measuring extremely low levels of an element (uranium) that occurs naturally in the environment and in everyday materials. The study did suggest, however, that three of the analysing laboratories had a better analytical capacity than the others, with measurements that mutually were more consistent.

The DUOB therefore invited these three laboratories to take part in a further pilot study, with a protocol that was designed to minimise the risk of sample contamination. Rather than obtain all samples from a single source, each laboratory produced a set of spiked samples using agreed uranium standards. The spiked samples were then relabelled to disguise their identity, and analysed 'blind' by each of the three laboratories. The second pilot study was completed to the Board's satisfaction in the spring of 2003, and the main findings have since been published in the journal, *Health Physics* (Parrish RR et al, 2006). The study showed that all three laboratories were capable of detecting an elevation of the U238:U235 ratio as low as 144 in urine samples containing less than 5 nanograms per litre of uranium, and that two of the laboratories, which used a more sensitive analytical technique, could perform even better. This degree of sensitivity was more than adequate to detect DU excretion in veterans that would be of practical relevance to health (see Appendix D). The study also indicated that uranium concentrations could normally be measured with an accuracy of +/- 15% or better.

Once the availability of suitable analytical techniques had been established, tenders were invited to provide urine analyses for the main testing programme. A number of laboratories submitted bids, and after a systematic evaluation, contracts were placed with two of the laboratories that had taken part in the second pilot study (Harwell Scientifics and NERC Isotope Geosciences Laboratory (NIGL)). These laboratories used sector field - inductively

coupled plasma - mass spectrometry (SF-ICP-MS) and multiple collector - inductively coupled plasma - mass spectrometry (MC-ICP-MS) respectively. Further details of the analytical methods are given in Appendix E. The MC-ICP-MS method used by NIGL was more sensitive, while Harwell Scientifics had capacity for a higher throughput of samples.

### Arrangements for admission to the testing programme

From the outset, it was recognised that some veterans were distrustful of MOD. Therefore, the testing programme was provided by external contractors under the supervision of the DUOB. Nevertheless, before an individual could have a test, it was necessary for MOD to confirm his or her eligibility according to the specifications of the scheme. A system was established whereby veterans who wished to be tested, first contacted the DUOB secretariat at the Veterans Policy Unit, MOD. Once their eligibility had been confirmed, their contact details were passed to an external contractor (Grosvenor Health Ltd), which made arrangements for the collection of a urine sample, and subsequently reported the result back to the veteran.

### Arrangements for the collection, transport and analysis of samples

Originally, the DUOB had hoped to let a contract with a single organisation to collect samples, transport them to the analysing laboratories, receive the test results from the laboratories, and communicate the results back to participants with appropriate interpretation. However, despite informal approaches to several possible contractors, no satisfactory bids were received when a call for tenders was issued. The Board was therefore obliged to issue contracts separately for different components of the work.

#### *Central coordination*

One company (Grosvenor Health Ltd.) was engaged to act as a central coordinator. Its role was: to arrange appointments for participants at a suitable clinic (or in exceptional circumstances a home visit); issue them in advance with a sample bottle (identified by a unique code number) and instructions for collecting a 24-hour urine sample to be brought to the clinic; issue them with a self-administered questionnaire (see below) to be completed and brought to the clinic; receive and retain the completed questionnaires from the clinics; receive and retain the test results from the analysing laboratories; feed back the results of the test to participants with interpretation; provide a telephone helpline to deal with queries about the test from participants or their doctors; and if necessary, refer enquirers to an independent medical consultant for additional advice on the meaning of test results. The company was also required to provide periodic reports to the DUOB on the progress of the testing programme.

#### *Clinics for collection of samples*

To act as collection points for urine samples and questionnaires, contracts were let with six occupational health departments in Bristol, Glasgow, London, Manchester, Sheffield and Stockton-on-Tees. The aim was to minimise the distance that veterans would have to travel in order to deliver samples, both for their own convenience and to reduce the costs for MOD (which reimbursed travel expenses). The clinics received the urine samples and questionnaires; checked that the samples had been collected as intended (e.g. that they covered a full 24 hours); checked that the questionnaires had been satisfactorily completed; forwarded the completed questionnaires to Grosvenor Health Ltd; and arranged for transport of the urine samples to Harwell Scientifics. From the time that they were first received, urine samples were placed in tamper-evident packaging, and a strict chain of custody was maintained at all stages.

### *Questionnaire*

The questionnaire that was sent to participants was drafted by the DUOB. Its primary purpose was to collect information that might be relevant to the interpretation and feedback of an individual's test results (e.g. about the circumstances and timing of potential exposure to depleted, natural and enriched uranium, and about illnesses that could be related to DU exposure). In addition, it asked whether the participant would be willing for his/her data to be used for research purposes. It was recognised that in a voluntary testing programme, participants could in some respects be highly unrepresentative of all who were eligible for testing. For example, it would not be surprising if they had an unusually high prevalence of ill-health. However, if some participants had detectable exposure to DU, analysis of associations with past activities might provide useful information about the major determinants of exposure.

### *Laboratory analysis of samples*

Once a urine sample was received by Harwell Scientifics, its volume and density were measured, one aliquot was removed for analysis of creatinine, and another for measurement of uranium. The remainder of the sample was transported to NIGL in its original container as part of a batch.

The analysis of creatinine was by an accredited laboratory within Harwell Scientifics, using a standard method. Creatinine concentrations were occasionally used as a guide to interpretation where there were doubts about the completeness of a 24-hour urine collection.

On the instructions of the DUOB, some samples were to be analysed for uranium in duplicate by both Harwell Scientifics and NIGL. Others were designated for initial analysis by Harwell Scientifics, with repeat analysis by NIGL only if the measured isotope ratio was 142 or higher (later revised to higher than 140), the estimated daily excretion of uranium was 20 ng or higher, or the findings were in some other way unusual or unsatisfactory. Where a sample was initially to be analysed only by Harwell Scientifics, NIGL stored their portion in case they were subsequently required to test it.

Each laboratory carried out measurements of total uranium concentration and U238:U235 atomic ratio, and also reported whether samples contained detectable U236. U236 is an isotope that is not detectable in natural uranium, but which may occur at low levels in depleted or enriched uranium that is produced by nuclear facilities. Its measurement provided a check against the unlikely possibility that a veteran had been exposed to both depleted and enriched uranium in such a way that the effect of the DU on the U238:U235 ratio in urine was exactly offset by concomitant excretion of enriched uranium. Additionally, the laboratories were asked to give an opinion on whether they thought that samples contained DU, taking into account their measurement of the isotope ratio, whether they found detectable U236, and also any limitations of the analyses for the sample in question (e.g. because the total uranium concentration was unusually low).

### *Piloting of methods for the collection, transport and analysis of samples*

Before the main testing programme was launched, the arrangements for collection, transport and analysis of samples were tested in a pilot group of 32 veterans who had asked to be tested.



## Arrangements for feedback of results to participants

From the measurements reported by the analysing laboratories, a decision was made as to whether a sample showed evidence of DU, and an algorithm was applied to estimate the total 24-hour excretion of uranium.

Samples that were analysed in duplicate by both laboratories were deemed to show no evidence of DU if neither laboratory found an isotope ratio in excess of 142 or reported a suspicion of DU. Samples that were analysed only by Harwell Scientifics were considered negative for DU if the isotope ratio was less than 142 (later amended to 140 or lower) and the laboratory did not report a suspicion that DU was present.

In the calculation of 24-hour uranium excretion, a correction was applied to the urine volume if the collection was incomplete (200 ml added for each missed void). The corrected value was then multiplied by the measured uranium concentration, or if both laboratories had analysed the sample, by the mean of their two measurements.

Each veteran was told his/her 24-hour uranium excretion, and whether there was detectable exposure to DU. Where he/she had given permission, a copy of the results was also sent to his/her general practitioner.

It was important that the results fed back to participants and their doctors be accompanied by a meaningful interpretation. To this end, the Board agreed a form of words that could be used when there was no detectable exposure to DU and the 24-hour excretion of uranium was less than 20 ng:

“The analysis carried out on your urine sample indicates a total uranium excretion of ..... nanograms per day. This is within the normal range of the general adult population of the UK.

From measurement of uranium isotopes in your urine, there is no indication that you have been exposed to Depleted Uranium (DU).

Although no DU was detectable, this does not rule out a very small exposure while you were in the Gulf or Balkans, too low to be picked up after an interval of 10 or more years.

A small minority of scientists believe that even such very low exposures might carry a significant risk of disease. However, the mainstream view of doctors and scientists in the UK and elsewhere is that they do not. Research on this is continuing.”

As things turned out, the large majority of results were covered by this provision, but in a few cases, special wording was required, and was agreed with the DUOB chairman.

Any queries from veterans about the meaning of their results were addressed first to Grosvenor Health, and if necessary were referred to a consultant physician with appropriate specialist expertise, who was contracted for this task on the recommendation of the DUOB.

## Information for veterans and their advisors

In addition to individual feedback of results and answers to specific queries, more general information about DU and the testing programme was provided in two fact sheets (Appendices F and G). These were drafted up by the DUOB with input from external experts in the communication of health information and a focus group of veterans. They were posted on the DUOB website and available in hard copy on request. A more detailed paper

for veterans and their doctors was also produced, explaining how the results of urine tests reflect past exposure to DU and the potential implications for health (Appendix H).

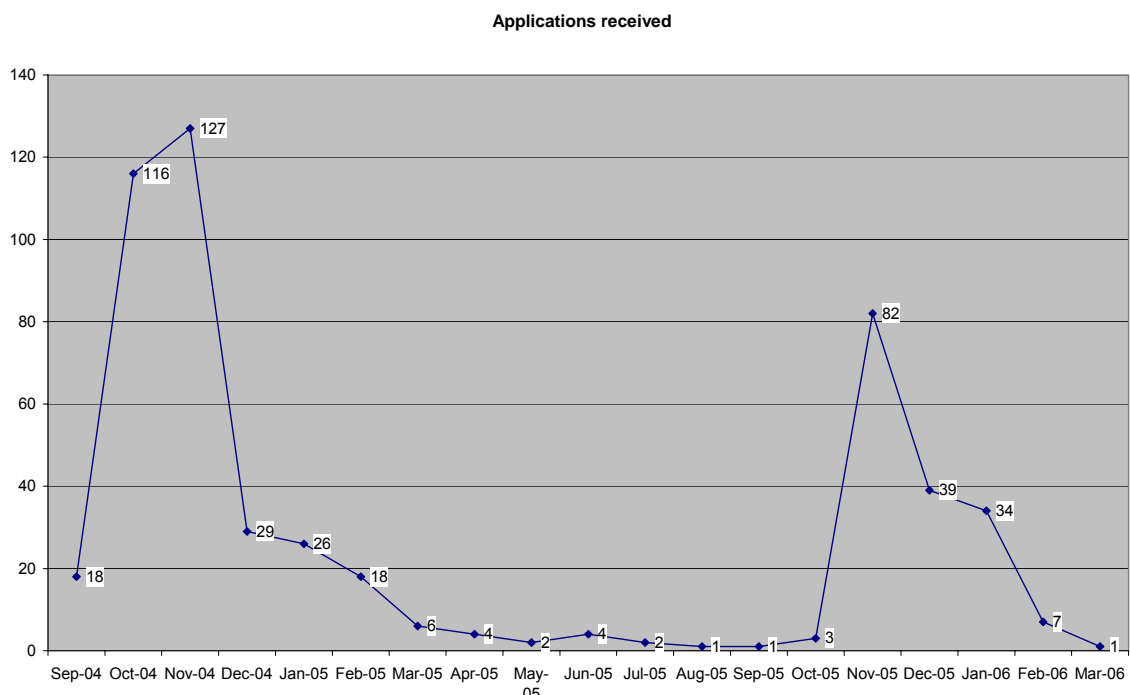
### Launch, uptake and closure of the main testing programme

The main testing programme was launched on 23 September 2004, with a press release and an advertisement in a national newspaper. The press release led to two reports in national newspapers, and also coverage in the specialist press and an interview with Forces Radio. In addition, information about the testing programme was disseminated through the Royal British Legion newsletter, the National Gulf Veterans and Families Association, the internal communication channels of MOD (for veterans who were still serving), the websites of relevant non-governmental organisations, and the DUOB website.

This publicity stimulated an initial surge in enquires and applications to be tested. However, by June 2005, the rate of new applications had tailed off to one or two per month. This relatively low rate persisted, and on 7 November 2005, it was announced that the scheme would close for further applications after 31 January 2006. The announcement of closure was again made through a press release and advertisements (this time in two national newspapers), and through the internal communications of veterans' organisations. It prompted a brief increase in interest, but by January, the rate of new applications had again fallen to low levels.

In total, 518 applicants were eligible for testing, and were referred to Grosvenor Health by the DUOB secretariat. Figure 1 shows the number of applications passed to Grosvenor Health, month by month, over the duration of the programme (two individuals were tested twice – one because of an unusually high excretion of natural uranium excretion on the first test, and the other because of a technical problem at one of the two analysing laboratories). Because there was a short delay while eligibility was confirmed and a small number of late applications were accepted, the last applicant was referred in March 2006.

**Figure 1: Applications referred to Grosvenor Health**



## Quality assurance and audit

Although the second pilot study had demonstrated that the contracted laboratories could analyse samples with the required sensitivity and accuracy, it was important to ensure that this level of performance was maintained over the course of the testing programme. In addition, the Board wished to be assured that the programme ran as smoothly as possible, and that veterans were satisfied with the service that they received.

### *Laboratory analyses*

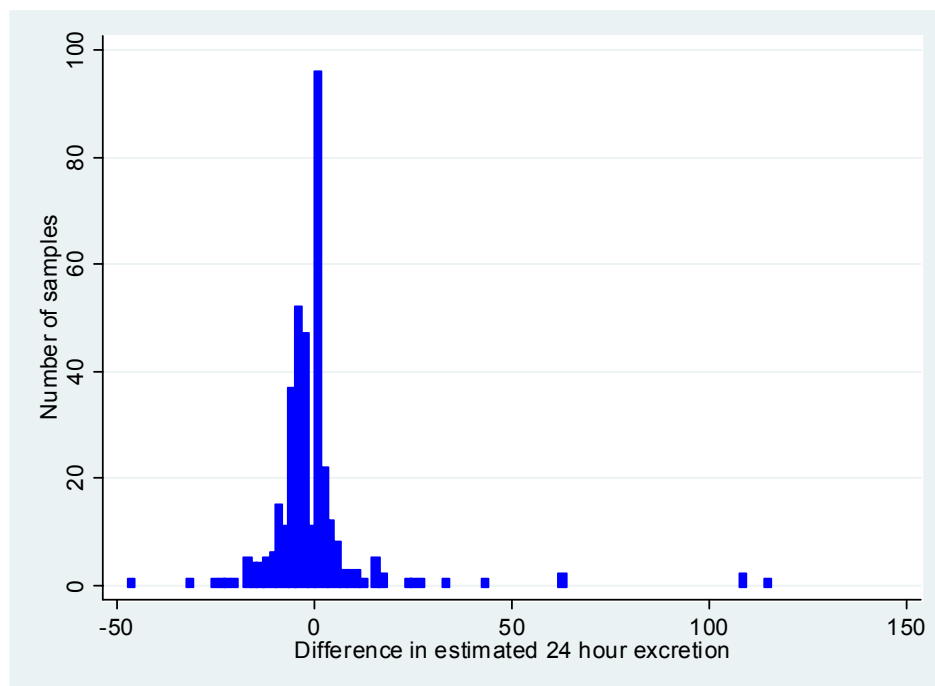
Quality assurance for the laboratory analyses comprised two components. The first 100 samples, and approximately 70% of the remainder were independently analysed in duplicate by the two laboratories, and the results compared.

In addition, the third laboratory that had participated successfully in the second pilot study (Department of Geology, Royal Holloway, University of London) was contracted to produce a series of 50 spiked samples with isotope ratios between 138.0 and 154.2. These were introduced into the system through the clinics in a form mimicking that of normal samples, and were analysed without knowledge that they came from a different source.

The results from these analyses were much in line with what would be expected from the earlier pilot work. None of the duplicate analyses indicated DU, and for only 32 of the 367 samples analysed in duplicate was the discrepancy in uranium concentrations (expressed as a percentage of the mean for the two laboratories) greater than 15% (Figure 2).

### **Figure 2: Inter-laboratory differences in estimated 24-hour urinary excretion of uranium, expressed as a percentage of the mean estimate for the two laboratories**

Analysis was based on 367 samples analysed in duplicate by the two laboratories.



The mean difference in estimated 24-hour excretion of uranium between the two laboratories was 0.070 ng (95% CI -0.087 ng to +0.228 ng).

NIGL, which had the more sensitive assay, reported DU in 40 of the 50 spiked samples. In all of the other ten spiked samples (with U238:U235 isotope ratios ranging from 138.0 to 139.1), U236 was detected, although the U238:U235 ratio was not clearly abnormal.

Harwell Scientifics reported DU in 14 of the 15 spiked samples with U238:U235 isotope ratios greater than 144, and in 12 of the 35 spiked samples with lower ratios.

#### *Operation of the programme and satisfaction of veterans*

Of the 518 applicants referred to Grosvenor Health by the DUOB secretariat, 54 subsequently decided not to proceed with a test. Tests were successfully completed for the remaining 464, including 16 who received home visits and two who were tested twice (see above). The median interval from handover of sample (during clinic appointment or home visit) to report of the test result to the veteran was 157 days (range 49 to 298 days).

All of those tested were sent a short questionnaire about their experience of the programme, and answers were received from 179 (39%). Among the responders:

- 89% felt that the information received prior to their clinic appointment was clear
- 89% felt that the instructions for the production of a sample were clear
- 90% felt that on arrival at the clinic, they were greeted in a friendly and professional manner
- 97% felt that they were seen promptly in relation to their appointment time
- 92% felt that they were treated in a friendly and professional manner
- 88% felt they received satisfactory answers to any questions they asked
- 86% felt that the results and the information that was provided was good
- 20% used the helpline facility, of whom 86% felt the response they received to be good.

#### **Statistical summary of results for veterans**

To maximise privacy, only Grosvenor Health held information that linked test results to named individuals. However, the DUOB was provided with pseudonymised results (identified only by a serial number), which enabled the findings to be summarised statistically.

Of the 464 veterans tested, 453 were men and 11 were women. A total of 422 had been potentially exposed to DU in the armed forces (219 in the Persian Gulf, 80 in the Balkans, and 123 in both theatres), and 46 in other organisations associated with military operations in the Persian Gulf (12), the Balkans (33), or both (1) (a few individuals had potential exposure in both military and non-military deployment). One hundred and seventy eight veterans were still serving in the military at the time of testing.

No excretion of DU was detected in any of the individuals tested. The mean U238:U235 ratio was 137.8 (95% confidence interval 137.7 to 137.9) for the 368 samples analysed by NIGL, and 138.0 (95% confidence interval 137.9 to 138.2) for the 422 samples successfully analysed by Harwell Scientifics (for technical reasons, Harwell Scientifics were unable satisfactorily to analyse the isotope ratio in 44 samples). Neither laboratory clearly detected U236 in any sample.

Total daily excretion of uranium was generally less than 30 nanograms, but eight samples showed higher values (range 38 to 497 nanograms).

## Archiving of data

Because of the possibility that results from the testing programme might subsequently be challenged, or might be cited in relation to a claim for compensation, the DUOB made provision for the safe archiving of data.

As part of its routine company policy, Grosvenor Health undertook to retain all questionnaires and laboratory reports for five years. The Board asked that before the end of this period, MOD should consult with veterans' organisations before deciding how the records should be archived in the longer term.

As a further safeguard, some veterans' representatives requested that, where permission had been granted by the participant, copies of his/her questionnaire and laboratory reports should be lodged with an independent third party. A firm of solicitors, nominated by the National Gulf Veterans and Families Association, was asked to take on this task, but they declined. Eventually, however, it was agreed that the Royal British Legion would undertake the role.

It was agreed that where a sample showed no detectable DU at either laboratory and a 24-hour excretion of uranium less than 20 ng, laboratories could destroy any residual urine three months after the findings had been transmitted to the person tested (assuming no queries were raised in the meantime). Permission was given for the destruction of all other samples three months after all testing had been completed and all results communicated to participants.

## Epidemiological studies of exposure to DU and associations with health outcomes

### *Urinary excretion of uranium in the general population*

When the DUOB began its work, it was unclear what the findings from tests in veterans would reveal. The Board was concerned that interpretation of apparently elevated uranium excretion or detectable DU might be difficult without knowledge of the distribution and determinants of uranium excretion in people who had not served in the military. The Institute of Occupational Medicine (IOM) was therefore commissioned to investigate this question.

Their research was planned in two phases. First, a study would be carried out in a sample of hospital patients to compare findings from spot urine samples (using uranium:creatinine ratios as an index of excretion rate) with those from a 24-hour sample provided by the same person. If this indicated that spot samples (which are much cheaper and easier to collect) provided an adequate proxy for 24-hour collections, the second phase would then look at the levels and determinants of excretion using spot samples from larger numbers of people selected to be representative of the general population in different geographical locations.

Phase 1 of the research was successfully completed, and demonstrated that spot samples would indeed serve as a reasonable proxy for 24-hour collections (Jones et al, 2005). However, by the time this became known, it was apparent that any detection of DU in veterans would be rare, and that their excretion of uranium was generally low. Therefore, the second phase of the research was no longer considered necessary.

### *Epidemiological studies of DU exposure and potential health effects*

The DUOB was conscious that if past exposure to DU could be detected in a sufficient proportion of veterans, then there could be scope for epidemiological research looking at the

relation of measured exposure to health. Possible study designs that were discussed included case-control investigations nested within cohorts for whom relevant health outcomes had already been ascertained, and cross-sectional surveys looking at the association of exposure with cytogenetic abnormalities. The Board therefore maintained close liaison with the Medical Research Council, with a view to facilitating the commissioning of such research at an early opportunity if it looked feasible. However, once it became clear that any detectable exposure would be extremely rare, this idea was abandoned.

### Other possible methods of exposure assessment

In addition to the measurement of uranium isotopes in urine, the DUOB discussed two other possible approaches to the retrospective assessment of exposure.

The first was through the measurement of uranium isotopes in lung tissue obtained at autopsy. A major obstacle would be the logistics of obtaining consent for autopsy in a timely fashion, and then ensuring that the sample was collected and stored in a way that would be suitable for subsequent analysis. This would be compounded by the rarity of deaths in veterans over the next decade because of their relatively young average age. Thus, the Board did not recommend any proactive attempt to secure lung samples for analysis. It was recognised, however, that if such samples did become available from even just one or two veterans, and they were analysed for uranium isotopes by valid methods, the findings might provide a useful test of the validity of data obtained from urine tests.

The other approach discussed by the Board was the measurement of chromosome aberrations in lymphocytes. The majority view was that this assay would not be specific for exposure to DU. For example, chromosome aberrations might arise from other sources of exposure to ionising radiation such as X-ray examinations. Thus it would not provide an index of exposure as reliable as that which could be obtained from measurement of uranium isotopes in urine. However, it was agreed that if evidence of past exposure to DU could be demonstrated in a sufficient proportion of veterans, then there could be scope for an epidemiological study looking at the association between measured exposure and the frequency of chromosome aberrations. As things turned out, no exposure was detectable in the veterans tested, so this was not taken forward.

### Biological monitoring for DU

In accord with its terms of reference, the DUOB provided comment in 2002 on a new draft policy on biological monitoring for military personnel deployed in operations in which DU munitions are used. In response to these comments, a number of revisions were made. The policy was subsequently put into effect the following year, when DU weapons were used in the invasion of Iraq (Operation Telic). Implementation was assisted by the understanding of measurement methods for uranium in urine that had built up as a consequence of the pilot work overseen by the DUOB.

As well as commenting on biological monitoring for military personnel from Operation Telic, the DUOB also provided input to a scientific study of DU exposure in soldiers who served in Operation Telic. This study, which was led by Professor Simon Wessely at King's College London, used the methods for measuring uranium isotopes in urine that had been developed and tested under the auspices of the DUOB. None of the soldiers tested had detectable excretion of DU.

## Overview

In planning and executing its programme of work, the DUOB faced several challenges.

Techniques for measuring traces of uranium isotopes in aqueous media (principally for geochemical research) were already well established, but their validity and sensitivity when applied to urine samples was uncertain. Pilot work was therefore required before the main testing programme could begin. Although it took some time, this preliminary investigation was important in ensuring that the test provided to veterans would be sufficiently reliable to detect clinically important exposures to DU in the Gulf War or Balkans campaigns. Comparison of duplicate analyses and results for spiked samples that were analysed “blind” confirmed that laboratory performance was maintained throughout the duration of the testing programme.

A second major challenge was the uncertain demand for the test. Until the programme was launched, it was unclear how many veterans would request a test from the 50,000 or more who were eligible. It was important that requests for testing be dealt with as quickly as possible. On the other hand, the Board wished to avoid wasting taxpayers’ money by contracting in advance for substantially more analytical capacity than was required. It was helpful, therefore, that many contractors were prepared to charge according to the volume of work performed rather than for a total sum fixed in advance. Conveniently, the number of tests eventually requested was close to the laboratory capacity that the Board had originally commissioned, but had it turned out to be much higher, there would have been a need to recruit additional laboratories, which might have been difficult.

Apart from the initial pilot study, where there appeared to be a problem with contamination of spiked samples, all phases of the project ran closely to plan. The median interval from handover of sample to report of results (157 days) was longer than the Board would have liked, but was an acceptable compromise given the need for laboratory analyses to be performed in batches, and for the findings to be cross-checked before they were fed back to veterans. Once the large initial surge of tests had been dealt with, there was less pressure on the laboratories, and turnaround times were reduced.

Feedback from the questionnaire survey of veterans indicated generally high levels of satisfaction with the service provided, and the Board is grateful to all of the contractors who participated in the programme for their contribution to this outcome.

None of the individuals tested showed evidence of significant exposure to DU, and for both laboratories, the mean isotope ratio across all samples from veterans was almost identical to that for natural uranium. The Board is thus confident in concluding that, within the sample of veterans tested, there was no exposure to DU in the Gulf War or more recently, at a level which, according to mainstream medical and scientific thinking, would have any material impact on health. In their potential for exposure to DU, the veterans who participated in the programme may not have been representative of all those eligible for testing. Nevertheless, this finding suggests that clinically significant exposure of British military personnel to DU in the Gulf War and Balkans, if it occurred at all, was relatively uncommon.

Eight of the urine samples tested indicated 24-hour excretion of natural uranium in excess of 30 ng, presumably reflecting higher than average dietary or other environmental exposures. In general, however, uranium excretion was relatively low.

All of the tests provided to veterans were based on 24-hour urine samples. However, the study carried out for the DUOB by the Institute of Occupational Medicine and the earlier piloting of laboratory methods together suggest that if a similar programme were required in the future, then it could probably be based on spot samples of urine without undue loss of

accuracy. The option to analyse a 24-hour sample could still be available if required for individuals with suspicious or uncertain results on initial testing.

The DUOB was pleased that its work proved useful not only as a service to veterans who wished to know about their past exposure to DU, but also in the design of the biomonitoring programme that was successfully implemented during Operation Telic. This biomonitoring, together with the recent survey of DU exposure in personnel from Operation Telic by King's College, will further enhance our understanding of the potential for clinically important exposures when DU is used in military operations.

## Endnote

Set out at the end of this report is a minority statement supported by four Board members. The main arguments that it presents were discussed by the Board at its meetings, and failed to persuade the other members of the Board. In particular, the majority group reject the claim that the urine test was unreliable because excretion of DU may have been masked by concomitant excretion of enriched uranium. Their main reasons for rejecting this view are:

- a) In the opinion of the analysing laboratories, after final analysis, none of the individual urine samples showed an isotope ratio indicative of enriched uranium.
- b) No individual urine sample showed detectable U236, as would be expected if a combination of depleted and enriched uranium were present. We know of no DU that has been shown not to contain U236.
- c) The mean isotope ratio for all veterans tested was extremely close to the ratio for natural uranium. If exposure to DU were being masked by the presence of enriched uranium, the levels of enriched uranium would have to have been such as to offset almost exactly any DU that was present. There is no scientific reason to expect an exact offset of this sort, and its occurrence by chance seems highly improbable.

The majority group acknowledge that there are scientific uncertainties in the biokinetic models for excretion of DU, and these are described in Appendices C and D. The extent of these uncertainties was taken into account in the conclusions that were drawn about implications for health of measured urinary excretion of DU. The existence of alternative, minority scientific views was acknowledged in the wording that was used to report results back to individual veterans (Page 8). This wording was agreed by all members of the Board.



## Acknowledgements

The DUOB thanks the many individuals and organisations that contributed to the success of the DU testing programme – in particular:

NERC Isotope Geosciences Laboratory  
Harwell Scientifics  
Department of Geology, Royal Holloway, University of London  
NEQAS – School of Biomedical & Life Sciences, University of Surrey  
Grosvenor Health Ltd.  
Avon Partnership Occupational Health Service  
Glasgow Occupational Health  
Guy's and St. Thomas' Hospital Trust  
Medigold Health Consultancy Ltd.  
Sheffield Occupational Health Service  
University Hospital of North Tees  
Professor Anthony Seaton  
The Institute of Occupational Medicine  
The College of Health and those veterans who participated in the information focus groups  
The Radiation Protection Division, Health Protection Agency  
The Royal British Legion  
The National Gulf Veterans and Families Association  
The British Red Cross  
The Ministry of Defence, in particular the DUOB secretariat and the staff of the Veterans Policy Unit

## References

Parrish RR, Thirlwall MF, Pickford C et al. Determination of the  $^{238}\text{U}/^{235}\text{U}$ ,  $^{236}\text{U}/^{238}\text{U}$  and uranium concentration in urine using SF-ICP-MS and MC-ICP-MS: an interlaboratory comparison. Health Physics 2006;90:127-38.

Jones AD, Miller BG, Walker S et al. Depleted uranium (DU) normative value pilot study: levels of uranium in urine samples from the general population. IOM Research Report TM/05/03, July 2005. [[http://www.iom-world.org/pubs/IOM\\_TM0503.pdf](http://www.iom-world.org/pubs/IOM_TM0503.pdf)]

## APPENDIX A

### TERMS OF REFERENCE

1. The purpose of the Depleted Uranium Oversight Board is to:
  - a. Oversee and co-ordinate the process of letting the contracts, and undertaking testing, for uranium isotopes in urine to assess historical exposure to DU.
  - b. Act as a Project Board, to direct, endorse and oversee the work of the MOD Project Manager who will:
    1. Develop a draft Statement of Requirement for a DU sampling protocol, a chain of custody for samples and a quality control protocol for endorsement by the Board.
    2. Invite proposals for testing.
    3. Prepare an assessment of proposals received.
    4. Manage a pilot study to demonstrate the performance, precision, accuracy and validity of the method, including the techniques for collecting, splitting, storing, transporting and analysing samples.
  - c. If satisfactory methods of testing can be established, agree proposals for one or more epidemiological studies using those methods, to determine the distribution and determinants of excretion of uranium isotopes in urine, and to explore the relation of historical exposure to DU to possible biological and health effects.
  - d. If satisfactory methods of testing can be established, agree arrangements for testing additional individuals who are not part of the epidemiological studies. These will include the arrangements for the involvement and briefing of GPs, and procedures for any accompanying medical assessment.
  - e. Monitor progress of the testing, including auditing and quality assurance of the data.
  - f. Ensure that the findings of the testing and research are appropriately promulgated.
  - g. Report to the Under-Secretary of State for Defence and Minister for Veterans' Affairs, on progress issues and concerns.
2. The Oversight Board will explore and advise on other possible methods of historical exposure assessment.
3. The Oversight Board will be invited to comment on:
  - a. The development of biological monitoring tests to be used by MOD for future operations where DU is used.
  - b. Proposed epidemiological studies to examine possible ill-health effects of service in the Balkans.
  - c. Possible arrangements for a Veterans' Assessment Centre

## APPENDIX B

### MEMBERSHIP OF THE DUOB

Period of service

Surgeon Commodore Nick Baldock  
Central Air & Admiralty Medical Board

Mr Ray Bristow  
National Gulf Veterans and Families Association

2002

Mr Ron Brown  
Dstl Radiological Protection Services

Dr Chris Busby  
Low Level Radiation Campaign

Dr Peter van Calsteren  
Open University

Until 2003

Professor David Coggon (Chairman)  
MRC Epidemiology Resource Centre  
University of Southampton

Mr Ivor Connolly  
National Gulf Veterans and Families Association

Maj Gen (Retd.) R P Craig  
The Royal British Legion

Since 2003

Professor Nick Day  
IPH

Until 2003

Dr George Etherington

Since 2003

Radiation Protection Division  
Health Protection Agency

Miss Frances Fry  
Division Head  
National Radiological Protection Board

Until 2003

Professor Ian Gilmore  
President, Royal College of Physicians

Mr Jim Glennon  
National Gulf Veterans and Families Association

2003 – 2004

Sir Muir Gray  
UK National Screening Committee  
NHS Executive  
Institute of Health Sciences

Until 2005

Miss Beverley Green *2002 – 2003*  
Head of Benevolent Department  
The Royal British Legion

Dr Derek Hall *Since 2004*  
National Gulf Veterans and Families Association

Dr Gideon Henderson *Since 2003*  
Department of Earth Sciences  
Oxford University

Professor Malcolm Hooper  
Department of Health Sciences  
University of Sunderland

Dr Len Levy *Since 2002*  
Head of Toxicology and Risk Assessment  
MRC Institute for Environment and Health

Dr David Lewis  
Laboratory Technical Manager  
Institute of Naval Medicine

Dr J Gordon Paterson  
Chief Medical Advisor  
British Red Cross

Mr Shaun Rusling *Until 2002*  
National Gulf Veterans and Families Association

Dr Margaret Spittle *Since 2002*  
Consultant Oncologist  
Middlesex Hospital

Professor Brian G Spratt  
Department of Infectious Disease Epidemiology  
Imperial College

### Observers

Mrs Karen Davies  
Health and Safety Executive

Mr Alan Duncan  
Hodge Jones & Allen Solicitors

Mr Neville Higham  
Ionising Radiation Policy Unit  
Health and Safety Executive

Dr Steven Laitnor  
NSC

Dr Hilary Walker  
Department of Health

## THE SOLUBILITY OF INHALED DU AND ITS INFLUENCE ON URINE EXCRETION

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National Radiological Protection Board, Chilton, Didcot, Oxon, OX11 0RQ  
(September 2004)

### 1. Introduction

This note briefly discusses the process of absorption of inhaled material from the respiratory tract, and describes how it can be modelled. It then presents some of the available information on absorption of depleted uranium, and describes the effect of variability in absorption on urinary excretion. It is in no way intended to be a comprehensive account; for this, reference should be made to Annexes of the report of the Royal Society Working Group on the Health Hazards of Depleted Uranium Munitions (RSWGDU) (Royal Society, 2001). Annex A give an account of the current ICRP models used to assess intakes of uranium. In addition, Annexes G and H of the RSWGDU report give summaries of the available information on the absorption characteristics (ie "lung solubility") of particulate DU resulting from penetrator impact and combustion in fires respectively.

### 2. Absorption from the respiratory tract

Inhaled material is cleared from the respiratory tract by three mechanisms. In regions of the respiratory tract other than the nose, clearance results from a combination of movement of particles towards the gastro-intestinal tract and lymph nodes (*particle transport*), and movement of material from the respiratory tract into the blood and then to body fluids (*absorption*). Material deposited in the nose is cleared by *nose blowing*, particle transport and absorption.

It is generally assumed that :

- all clearance rates are independent of age and sex;
- particle transport rates are the same for all materials;
- absorption into blood, which is material specific, occurs at the same rate in all regions except the front of the nose, where none occurs.

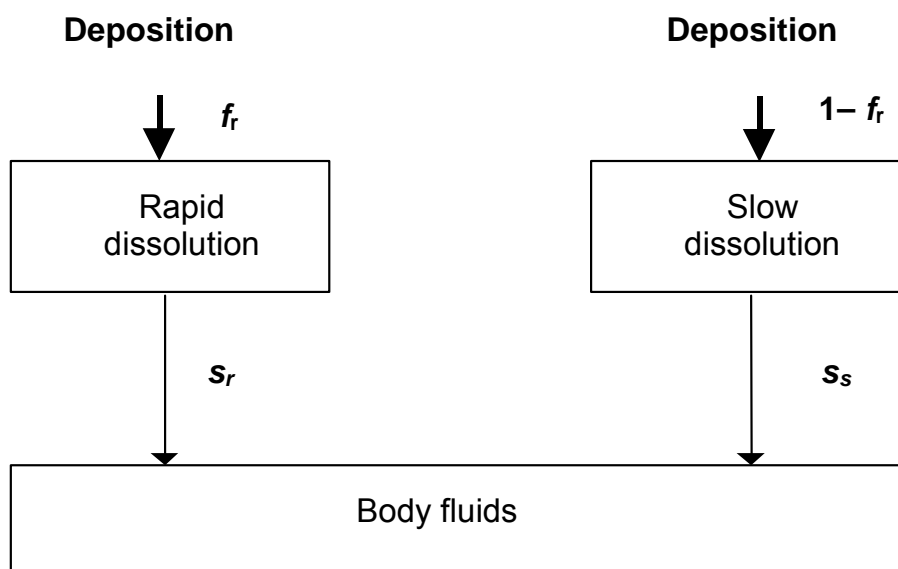
Fractional clearance rates (whether absorption or particle transport) vary with time. However, in order to simplify calculations, most models represent clearance by combinations of compartments that clear at constant rates.

### 3. Modelling absorption

Absorption to blood is a two-stage process: dissociation of the particles into material that can be absorbed into blood (dissolution); and absorption into blood of soluble material and of material dissociated from particles (uptake). Both stages can be time-dependent. In practice, it is found that dissolution of most materials can be represented by a simple two-compartment model (figure 1). A fixed fraction of deposited material,  $f_r$ , is available for rapid dissolution at a rate  $s_r$ , while the remaining fraction  $(1 - f_r)$  dissolves more slowly, at a rate  $s_s$ . Uptake to body fluids of

dissolved material can usually be treated as instantaneous. (When this is not the case, the concept of a “bound state” is employed; see Annex A, Royal Society, 2001).

**Figure 1. Compartment model describing absorption to blood.**



Thus, the absorption behaviour of most materials can be described by making a suitable choice of values for the three absorption parameters,  $f_r$ ,  $s_r$ , and  $s_s$ . Some materials are very soluble in the lungs and are absorbed almost instantaneously (eg caesium chloride), and so have an  $f_r$  value of 1. Other materials are very insoluble in the lungs (eg plutonium dioxide) and have a very low  $f_r$  value (typically 0.001). Three default absorption types have been defined for use when material-specific information is not available, known as Type F (“fast” absorption), Type M (“moderate” absorption) and Type S (“slow” absorption). Absorption parameter values for the three default Types are given in Table 1.

**Table 1. Default absorption parameter values for Type F, M, and S materials (ICRP 1994)**

Type		F(fast)	M (moderate)	S (slow)
Fraction dissolved rapidly	$f_r$	1	0.1	0.001
Dissolution rates:				
Rapid ( $d^{-1}$ )	$s_r$	100	100	100
Slow ( $d^{-1}$ )	$s_s$	-	0.005	0.0001

A rate constant of  $100 d^{-1}$  corresponds to a half time of  $\sim 10$  minutes; a rate constant of  $0.0001 d^{-1}$  corresponds to a half time of  $\sim 7000$  days.

#### 4. Absorption of depleted uranium

A number of studies have been conducted to determine absorption parameter values for uranium oxides produced during the manufacture of nuclear fuel. Results are summarised in Table 2.

**Table 2. Summary of absorption parameter values for uranium oxides**

Compound <sup>a</sup>	Absorption parameters			Reference
	$f_r$	$s_r$ (d <sup>-1</sup> )	$s_s$ (d <sup>-1</sup> )	
UO <sub>4</sub>	0.87	0.93	0.024	Ansoborlo et al 2001
UO <sub>3</sub>	0.75	14	0.02	Bailey et al 1998
UO <sub>3</sub>	0.92	1.4	0.0036	Hodgson et al 2000
UO <sub>3</sub>	0.71	0.28	0.0011	Ansoborlo et al 2001
U <sub>3</sub> O <sub>8</sub>	0.044	0.49	0.00035	Hodgson et al 2000
U <sub>3</sub> O <sub>8</sub>	0.046	2.3	0.0012	Ansoborlo et al 2001
U <sub>3</sub> O <sub>8</sub>	0.03	2.1	0.00038	Ansoborlo et al 2001
UO <sub>2</sub> – Non-ceramic	0.011	0.95	0.00061	Hodgson et al 2000
UO <sub>2</sub> – Ceramic	0.008	1.3	0.00026	Hodgson et al 2000
UO <sub>2</sub>	0.03	1.3	0.0015	Ansoborlo et al 2001
UO <sub>2</sub>	0.01	nd	0.00049	Ansoborlo et al 2001
UO <sub>2</sub>	0.01	nd	0.00058	Ansoborlo et al 2001

Note. See (Royal Society, 2001) for full references.

It can be seen that the different oxides have a very wide range of absorption characteristics. The chemical form of particulate DU produced as a result of its use in munitions depends on the conditions of formation. Particles formed by impacts are reported to be a mixture of U<sub>3</sub>O<sub>8</sub> and UO<sub>2</sub> (but predominantly U<sub>3</sub>O<sub>8</sub>), while combustion produces an oxide which is almost entirely U<sub>3</sub>O<sub>8</sub> (Royal Society, 2001). However, there remains uncertainty as to the absorption behaviour of DU formed as a result of its use in munitions, because of factors such as particle size distribution and the presence of other elements.

Human Respiratory Tract Model (HRTM) (ICRP, 1994) parameter values appropriate for a wide range of possible DU exposure scenarios are discussed in the RSWGDU report (Royal Society, 2001). Central estimates of DU parameter values appropriate for Level II or Level III inhalation of resuspension aerosols (impact or combustion) occurring within a vehicle are shown in Table 3. Also shown are parameter values describing the likely upper and lower limits of absorption. Broadly, the parameter values given in Table 3 for “Low absorption” and “High absorption” are equivalent to the values given in Table 15, Appendix 1 of the RSWGDU report for “Worst-case (radiation)” and “Worst-case (chemical toxicity)”, respectively. However, there are some small differences in the data given in the two Tables because in this note we are concerned with predicting the upper and lower limits of urinary excretion per unit intake, whereas Table 15, Appendix 1 of the RSWGDU report is concerned with predicting upper limits on dose per unit intake and chemical toxicity.

**Table 3. HRTM model parameters for Level II / III inhalation of resuspension DU aerosols (see text)**

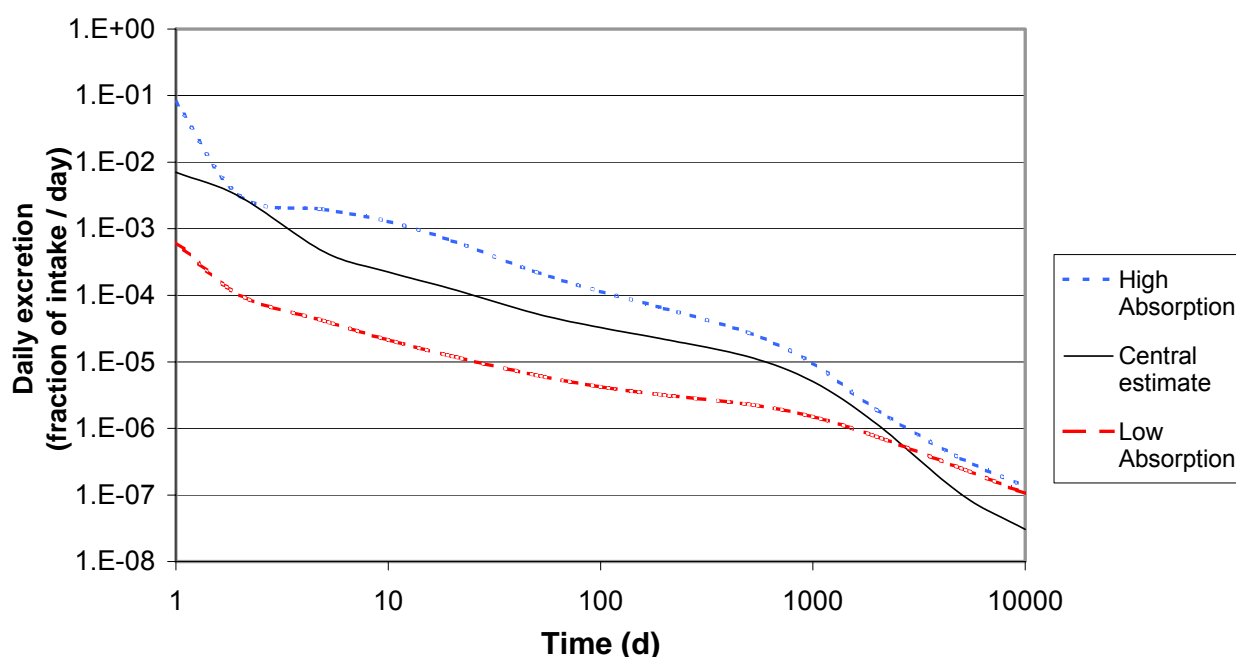
Parameter	Central estimate	“Low” absorption	“High” absorption
Rapid dissolution fraction, $f_r$	0.2	0.005	0.5
Rapid dissolution rate, $s_r$ , d <sup>-1</sup>	1	0.4	14
Slow dissolution rate, $s_s$ , d <sup>-1</sup>	0.001	0.0001	0.0015
Subject exercise level	Heavy worker	Heavy worker	Heavy worker
Aerosol activity median aerodynamic diameter, $\mu\text{m}$	5 (default workplace)	5	1
Aerosol geometric standard deviation	2.5 (default)	2.5	4
Particle density $\rho$ , g cm <sup>-3</sup>	9	9	11
Gut uptake factor, $f_1$	0.002	0.002	0.02

## 5. Urinary excretion of DU

Figure 2 shows daily urinary excretion calculated using the HRTM for the three scenarios described in Table 3. As can be seen, the range spans almost two orders of magnitude at earlier times, but the rates converge at times later than about 500 days.

This figure illustrates the effect on urinary excretion of variability or uncertainty in HRTM model parameters. An additional source of uncertainty in urinary excretion is uncertainties in systemic model parameter values. NRPB is currently planning a study to investigate uncertainties in DU urinary excretion arising from uncertainties and variability in both HRTM and systemic model parameter values.

**Figure 2. DU urinary excretion rates, calculated using the HRTM using the parameter values given in Table 3.**



## References

ICRP (1994). Human Respiratory Tract Model for Radiological Protection. ICRP Publication 66, Annals of the ICRP, **24**, No. 1-3. Oxford, Pergamon Press.

Royal Society (2001). The Health Hazards of Depleted Uranium Munitions, Part 1. Policy Document 6/01. London, Royal Society.



## APPENDIX D

### ASSESSMENT OF RADIATION DOSE AND MAXIMUM KIDNEY CONCENTRATION FROM MEASUREMENTS OF DU IN URINE

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(September 2004)

#### Summary

This note presents the relationship between urinary excretion of DU and radiation dose. It also presents the relationship between urinary excretion of DU and maximum kidney concentration ( $^{max}[U]_k$ ). These calculations were performed using current ICRP models with "best estimate" model parameter values. There is a significant level of uncertainty in these calculations, and it is judged that dose or  $^{max}[U]_k$  assessed from urine measurements could be under- or over-estimated by up to a factor of about 10. Therefore, data are also presented for a "worst case" scenario where dose per unit excretion and maximum kidney concentration per unit excretion are both a factor of 10 higher than "best estimate" values. The levels of dose and maximum kidney concentration below which exposures could be judged to be low are briefly discussed. Finally, an upper limit on daily DU urinary excretion is proposed that could be used to identify veterans for whom exposures were low.

#### 1. Urinary excretion, radiation dose and maximum kidney concentration

The relationship between urinary excretion rate and committed effective dose, and between urinary excretion rate and maximum kidney concentration, can be predicted using ICRP's Human Respiratory Tract Model for Radiological Protection (HRTM) (ICRP, 1994), together with the systemic model for uranium (ICRP, 1995). The HRTM describes the processes of inhalation, deposition, clearance and uptake to blood in the respiratory tract, while the systemic model describes uptake to organs from blood and subsequent excretion. Table 1 shows the daily urinary excretion 12.5 y after exposure that would result from an intake by inhalation giving rise to a committed effective dose ( $E_{50}$ )<sup>†</sup> of 1 milli-sievert (mSv). Table 1 also shows the maximum kidney concentration ( $^{max}[U]_k$ ) that would result from the same intake.

Values of daily urinary excretion and  $^{max}[U]_k$  corresponding to other committed effective dose values can be obtained by linear scaling of the values given in Table 1. Thus the daily DU urinary excretion corresponding to  $E_{50} = 20$  mSv would be  $20 \times 2.1 = 42$  ng/day. Similarly, if the daily DU excretion was 1 ng/day, the corresponding  $^{max}[U]_k$  would be  $0.09/2.1 = 0.043$   $\mu$ g uranium per g kidney, and the corresponding dose would be  $1/2.1 = 0.48$  mSv.

---

<sup>†</sup> The "committed" dose is the radiation dose received during the 50 year period after the intake. Committed equivalent doses are calculated to all the important organs. Each organ dose is multiplied by a "tissue weighting factor" that represents the sensitivity of the organ to radiation, compared to that of the whole body. The sum of these weighted organ doses is the committed effective dose, which is therefore a measure of the overall radiation risk from the intake.

It should be noted that a daily urinary excretion of about 0.4 ng/day DU should be detectable in people who are excreting 10 ng/day of natural uranium if mass spectrometric measurements can detect an increase in  $^{238}\text{U}$ : $^{235}\text{U}$  atomic ratio from 137.9:1 to 142:1.

**Table 1. Best estimate of DU daily urinary excretion corresponding to a committed effective dose of 1 mSv**

Daily DU urinary excretion, ng/day <sup>1,2</sup>	DU intake by inhalation, mg <sup>4</sup>	Maximum kidney concentration ( $^{\text{max}}[\text{U}]_k$ ), $\mu\text{g}$ uranium per g kidney <sup>3</sup>	Committed effective dose ( $E_{50}$ ), mSv <sup>5</sup>
2.1	16	0.09	1

Notes.

1. Exposure assumed to be 12.5 y before urine sample taken
2. 1 nanogram (ng) =  $1 \times 10^{-9}$  gram (g) (1,000,000,000 nanograms = 1 gram)
3. 1 microgram ( $\mu\text{g}$ ) =  $1 \times 10^{-6}$  gram (g) (1,000,000 micrograms = 1 gram)
4. 1 milligram (mg) =  $1 \times 10^{-3}$  gram (g) (1000 milligrams = 1 gram)
5. Using ICRP's probability coefficient, 1 mSv results in a risk factor of 1 in 20,000 for fatal cancers

The model parameter values used in this calculation were taken from the report of the Royal Society Working Group on the Health Hazards of Depleted Uranium Munitions (RSWGDU) (Royal Society, 2001), and are shown in Table 2. The values used were those presented in the RSWGDU report for Level II or Level III inhalation of resuspension aerosols (impact or combustion) occurring within a vehicle. (Further information on exposure scenarios is given in Appendix 1 of the RSWGDU report).

**Table 2. HRTM model parameters (Table 15, Appendix 1, Royal Society, 2001)**

Parameter	Central estimate
Subject exercise level, type of breathing	Heavy work (7 h light exercise + 1 h heavy exercise)
Aerosol activity median aerodynamic diameter, $\mu\text{m}$	5 (default workplace)
Aerosol geometric standard deviation	2.5 (default)
Particle density $\rho$ , $\text{g cm}^{-3}$	9
Rapid dissolution fraction, $f_r$	0.2
Rapid dissolution rate, $s_r$ , $\text{d}^{-1}$	1
Slow dissolution rate, $s_s$ , $\text{d}^{-1}$	0.001
Gut uptake factor, $f_1$	0.002

There is a significant level of uncertainty in these calculations. (For example, Figure C1 of Annex C of the RSWGDU report (Royal Society 2001) shows that at 10 years after inhalation of 1 gram of DU, the calculated daily urine excretion rate lies between about 0.1 microgram per day and 0.4 microgram per day, depending on different assumptions about the solubility of the inhaled material, and the exposure conditions.) NRPB is planning an investigation of uncertainties in doses assessed from DU-in-urine measurements, but this has not yet commenced, and it is not yet possible to make a reliable estimate of confidence intervals on assessed dose. For the purposes of the work of the DUOB, it is judged that dose or  $^{\text{max}}[\text{U}]_k$  could be under- or over-estimated by up to a factor of about 10. It will be possible to refine this estimate of uncertainty once the NRPB study is completed.

Table 3 shows the daily urinary excretion 12.5 y after exposure, and the maximum kidney concentration ( $^{max}[U]_k$ ), for a “worst case” scenario where dose per unit excretion and maximum kidney concentration per unit excretion are each a factor of 10 higher than “best estimate” values. Values for committed effective dose and maximum kidney concentration estimated from a measurement of daily urine excretion using the conversion factors of Table 3 would thus be a factor of 10 higher than those estimated using the conversion factors of Table 1.

**Table 3. “Worst case” estimate of DU daily urinary excretion corresponding to a committed effective dose of 1 mSv**

Daily urinary excretion, ng/day <sup>1</sup>	Maximum kidney concentration ( $^{max}[U]_k$ ), $\mu\text{g}$ uranium per g kidney	Committed effective dose ( $E_{50}$ ), mSv
0.21	0.09	1

Notes.

1. Exposure assumed to be 12.5 y before urine sample taken

## 2. Dose limits and chemical toxicity limits

The DUOB wishes to set an upper limit on daily DU urinary excretion that would apply for veterans with a positive DU-in-urine measurement, but for whom the amount inhaled is well below the level that would cause any important radiation risk, or any chemical poisoning. It is suggested that an appropriate dose limit or chemical toxicity limit should be chosen, and the corresponding daily DU urinary excretion then determined from Table 1 or Table 3. There is no dose limit or dose level that is clearly intended for the exposure conditions under consideration. Nevertheless, there are a number of dose levels that could be considered. However, these are annual dose levels and are usually defined on the assumption that such doses could be incurred year after year over the lifetime of an individual, whereas exposure to DU during a military operation is likely to be a once-in-a-lifetime occurrence. In order of increasing value, these dose levels are:

1. The annual dose limit for members of the public, ie 1 mSv per year
2. The average annual dose from natural background radiation in the UK, ie 2.24 mSv per year. (Note that about half of this dose results from alpha-particle irradiation of the lungs from inhaled radon and the radioactive atoms into which radon decays.)
3. The annual dose level above which workers must be Classified, and routine individual monitoring is required (Ionising Radiations Regulations, 1999), ie 6 mSv per year
4. The UK Action Level for radon in homes, (the radon concentration above which NRPB recommends that action is taken to reduce it) which corresponds to about 10 mSv per year
5. The annual dose limit for occupational exposure, ie 20 mSv per year

A number of chemical toxicity levels should also be considered:

6. The threshold level for transient biochemical indicators of renal dysfunction (RSWGDU report, Part 2, Table 1.2), ie  $\sim 1 \mu\text{g}$  uranium per gram kidney
7. The threshold level for protracted effects on the kidney, ie  $\sim 3 \mu\text{g}$  uranium per gram kidney
8. The threshold level for clinical symptoms, ie  $\sim 30 \mu\text{g}$  uranium per gram kidney

Radiation dose is the limiting factor as long as an  $E_{50}$  dose threshold less than  $\sim 10$  mSv is chosen. However, the  $1 \mu\text{g}$  uranium per gram kidney chemical toxicity level would be exceeded if a dose threshold greater than 10 mSv is chosen.

### 3. Conclusions

Using the best estimate of DU daily urinary excretion, an upper limit on DU excretion of 2 nanograms per day would correspond to a *life-time* radiation dose of  $\sim 1$  mSv. This dose is numerically equal to the *annual* radiation dose limit for members of the public. (Note that a member of the public receiving an annual dose equal to the annual dose limit of 1 mSv would receive a lifetime dose of  $\sim 70$  mSv). This level of urinary excretion corresponds to an intake that would be expected to have no adverse consequences on the kidney, and should be well within the sensitivity of the methods used by the DUOB. Even if the “worst case” estimate of DU daily urinary excretion applies, this level of urinary excretion would correspond to a lifetime radiation dose of only 10 mSv, and the threshold for transient effects on the kidney would still not be exceeded.

### References

ICRP (1994). Human Respiratory Tract Model for Radiological Protection. ICRP Publication 66, Annals of the ICRP, **24**, No. 1-3. Oxford, Pergamon Press.

ICRP (1995). Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 3 Ingestion Dose Coefficients. ICRP Publication 69, Annals of the ICRP, **25**, No. 1. Oxford, Pergamon Press.

Royal Society (2001). The Health Hazards of Depleted Uranium Munitions, Part 1. Policy Document 6/01. London, Royal Society. <http://www.royalsoc.ac.uk/policy/>

Royal Society (2002). The Health Hazards of Depleted Uranium Munitions, Part 2. Policy Document 5/02. London, Royal Society. <http://www.royalsoc.ac.uk/policy/>

## APPENDIX E

### LABORATORY METHODS

#### Harwell Scientifics

A method has been developed for the determination of uranium concentration and isotope ratios in urine by Sector Field Inductively Coupled Plasma-Mass Spectrometry (SF-ICP-MS).

From receipt until analysis is commenced, samples are controlled in accordance with the company policy as described in a separate procedure. A unique laboratory identification number is allocated to each sample. The samples are stored cold (nominally at +4° C), unless alternative storage is requested by the customer.

The first stage of the analytical procedure is the addition of a  $^{233}\text{U}$  standard of known concentration to the samples, which are then left to equilibrate. Calcium phosphate is added at this stage to ensure precipitation at the second stage. The sample is then co-precipitated with the addition of ammonium hydroxide and centrifuged for 4 minutes. The supernate is discarded and after washing the calcium phosphate precipitate, this is dissolved in nitric acid and diluted to the original sample volume with ultra-pure water. The samples are then measured using SF-ICP-MS.

The SF-ICP-MS used is a Micromass (formerly VG) PlasmaTrace2 operated with a CETAC ultrasonic nebuliser. The maximum resolution in high-resolution mode is approximately 10000. For the purpose of this work the instrument is operated in low-resolution mode, approximately 300. Typical sensitivity in low-resolution mode is approximately 100 million counts per ppm. Typical limits of detection for  $^{235}\text{U}$  and  $^{236}\text{U}$  are  $0.2 \text{ fg ml}^{-1}$ , and  $11 \text{ fg ml}^{-1}$  for  $^{238}\text{U}$ .

#### Sample preparation methodology

As soon as possible after receipt of the sample, a sub-sample is mixed well and transferred to a pre-cleaned (rinsed with 5 ml 5% nitric acid and twice with ca. 5 ml Elga water) Falcon container. The container is filled to the 50 ml mark and recapped.  $100 \mu\text{l}$   $0.5 \text{ ng ml}^{-1}$   $^{233}\text{U}$  spike solution and 1 ml  $1000 \text{ mg ml}^{-1}$  calcium phosphate are pipetted into the container and mixed by inversion<sup>1</sup>. Both the original sample and the sub-sample are stored below +4°C.

A sub-sample is prepared as follows:-

- a) After the spike has been allowed to equilibrate (at least 12 hours), 3.5 ml  $\text{NH}_4\text{OH}$  is added. The solution is mixed well; if the sample does not smell strongly of ammonia, a further 1 ml  $\text{NH}_4\text{OH}$  is added (if the sample needed to be acidified with 2.0% acid before precipitation to dissolve solids, an extra 2 ml is added).
- b) The sample is left to stand for 1 hour, and then centrifuged for 4 minutes at 3500 rpm.
- c) The supernate is carefully poured off, leaving the calcium phosphate precipitate. The drop left at the top of the tube is shaken off if necessary.

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<sup>1</sup> Before January 2005, calcium phosphate was only added to samples where this was considered to be indicated on the basis of their appearance and creatinine concentration.

d) 50 ml Elga water and 100  $\mu\text{L}$   $\text{NH}_4\text{OH}$  are added and the sample shaken to produce a fine suspension. The mixture is centrifuged for 4 minutes at 3500 rpm. The supernate is carefully poured off, leaving the calcium phosphate precipitate. The drop left at the top of the tube is shaken off if necessary.

e) 500  $\mu\text{l}$  c.  $\text{HNO}_3$  (Romil UpA or equiv.) is added. The sample is heated at ca. 60-80° C for 1 hour in a water bath or hot block with a few ml of water added to each block hole.

f) The sample is diluted back to the 50 ml mark at the top of the container with Elga water and mixed well.

### Equipment calibration

The balances and pipettes used are maintained and calibrated according to the company procedure. The volumes dispensed by the pipettes are checked when transferred to use for this work from other uses and when altered to dispense a different volume. The check is carried out by ensuring the correct weight of water is dispensed within the tolerances given in the procedure.

### Preparation of blanks

To a pre-cleaned Falcon tube is added 100  $\mu\text{l}$   $^{233}\text{U}$  spike, 500  $\mu\text{l}$   $\text{HNO}_3$  plus Elga water to 50 ml mark and the sample container labelled "BLK".

### Preparation of QC samples

The preparation procedure for the samples from the first step above is followed, replacing the in-house QC urine for the sample (two in-house QC urines are prepared - a low uranium concentration of natural isotope ratio and a spiked urine with depleted isotope ratio).

When preparation of a batch of samples is complete, the "Preparation" box on the Analytical Request form is initialled and dated by the responsible analyst. An analytical batch for measurement usually includes 10 samples, 2 blanks and 2 QC samples (1 natural and 1 spiked) run at intervals throughout the procedure. The number of duplicate samples analysed depends upon the volume of sample supplied and customer requirements.

### Reagent and standard preparation

The company policy on use of reagents and standards is set out in a separate procedure.

### Preparation of uranium standards

The concentration usually required for the uranium standard is 10  $\text{ng ml}^{-1}$ . A solution at this concentration is prepared by three serial dilutions (two 100-fold and one 10-fold) in 5% nitric acid of 1000  $\mu\text{g ml}^{-1}$  stock standard ensuring the standard used is within its marked re-test date. An aliquot of the standard is transferred to a clean screw top Falcon tube (50 ml). Details of this preparation are recorded, including which 1000  $\mu\text{g ml}^{-1}$  stock standard is used. Working standard solutions prepared as above may be stored at room temperature in ambient light.

## Preparation of instrument wash solution

An instrument wash solution containing approximately 2% nitric acid is prepared.

## Measurement procedure

The SF-ICP-MS instrument is prepared for use in accordance with the manufacturer's instructions and company procedure. Using the quantitative acquisition software, the instrument is set to scan the mass range 233-238.

## Data acquisition

The uptake tube is inserted into the sample solution. The length of time of wash between samples depends on the uranium level in the immediately preceding sample.

In theory, samples blanks and standards may be run in any sequence on the ICP-MS. In practice, imperfect wash out of one sample may affect the result for the following sample, in particular if the following sample has a uranium content several orders of magnitude lower than the first. Normal good practice is to acquire data for several blanks first, followed by samples and QCs. Extra blanks may be run at any stage at the discretion of the analyst.

## File naming convention

For customer-derived samples, spectra are given a filename corresponding to their laboratory identity number (eg MA11326 or for duplicates MA11326D). For samples generated within the laboratory (e.g. validation samples or QCs) alternative file naming conventions may be used.

When it is necessary to make a repeat measurement on a sample, filenames are assigned as follows: repeat measurement on a single prepared solution is identified by the addition of "M" onto the filename eg MA11326M. Where repeat dilution of the original sample is required, the spectrum is identified by an "R" suffix eg MA11326R. Should further repeat dilutions of the original sample be required, they are designated by "R2", "R3" etc. sequential suffices e.g. MA11326R2.

The operator notes the instrument identity and the filenames and sequence in which samples, blanks, QCs and standards are run. A proforma exists for this purpose (NB the date and time at which a spectrum is recorded is stored as part of the spectral data and may be accessed later). When measurement of a batch of samples is complete, the "Measured" box on the Analytical Request Form is initialled and dated by the responsible analyst.

## Calculations

The raw instrument signals as stored on disk are converted into peak-integrals using standard Micromass software supplied with the instrument used. Integration of the peaks may be checked before the data is exported either manually or to floppy disk.

The analytical data presented by the instrument is batched in sets of one-minute scans. It is normal to run the instrument for a total of ten scans (ten minutes) to collect sufficient data for robust interpretation. Any deviations from this normal procedure are noted by the analyst.

## Computerised spreadsheet calculation

Raw data files are integrated and the results documented or output to a text file. This text file is then loaded into Excel (documented data can be input direct to Excel) and the uranium

concentrations and  $^{236/238}\text{U}$  and  $^{235/238}\text{U}$  isotope ratios in the measured solutions are calculated. Uranium concentrations are calculated from the actual concentration of the  $^{233}\text{U}$  spike used. The isotope ratio is corrected if necessary for mass bias, measured using a standard solution of an appropriate standard with a certified value (this is carried out only if mass bias >1%). Ratios are reported in mass% and atom%, or as required by the customer.

A full printout of each Excel workbook is produced in order that all operator input may be checked. This printout is defined as the raw data and signed and dated by the analyst generating it. When calculation is complete the "Calculation" box on the Analytical Request form is initialled and dated by the responsible analyst.

### Deviations from this method

If variations from this procedure are necessary, they are approved in writing by the laboratory manager or other responsible person. All cases of variation from this procedure would be fully recorded.

### Reporting

Results may be reported to the client after the calculation step. When the final report has been prepared it is independently checked by a second analyst along with the raw data and the study records, and any discrepancies are resolved.

### Records

All study records, raw data etc. as defined in the company procedure are stored in the trace metals record store or other premises as directed by the client.

### Safety

All staff handling human body fluids wear lab coats, eye protectors and gloves at all times. The dispensing and dilution of such samples is ideally carried out in a fume-cupboard. The same personal protection precautions apply when handling concentrated acids. All dispensing of concentrated hydrochloric acid is carried out in a fume cupboard. Further details are in the relevant company procedure.

All solid waste (disposable test-tubes, pipette tips etc.) associated with preparation and measurement of biological fluids are disposed of in yellow "Biohazard" bags, which are eventually consigned for incineration. Full details of the handling of biological tissues and fluids are given in a separate procedure, with which the analyst is familiar before commencing work. Risk and hazards have been assessed for all relevant processes and are documented.

### Environmental considerations, including disposal

All reagents used are kept to a minimum. Samples are diluted further with water and treated with disinfectant granules before disposal down the waste drain. When not in use, all electrical equipment is switched off.



## Data calculation

Data are exported from the instrument as a CSV file and saved in the relevant file on the company computer network including only integrated data from the procedure. The data are copied to a new work sheet and called "Calculations". For a ten minute analytical run the data consist of ten sets of instrumental data for the uranium isotopes. It is possible from this to calculate manually or via a macro the mean, standard deviation (95% confidence), total of the run and standard error of the mean and percent error relating to the mean of the counts. An uncorrected  $^{235}/^{238}$  ratio and concentration may be calculated at this stage for reference.

In order to improve the errors of measurement it is normal practice to reject 2 of the individual readings per isotope. These are the instrumental data that deviate furthest from the mean. This accommodates sudden drops in sensitivity or spikes that occur in the normal operation of the instrument.

Totals for the run are translated into a worksheet called "Results". This is used to calculate final ratios and concentrations including blank subtraction and any mass bias correction if needed. The total error for the sample is calculated as the square root of the sum of the squares of all the data that are used in the error calculations of the final ratios.

## Interpretation

Interpreted data are presented as atomic ratios.

Levels of natural uranium present in the body are assumed to be of a concentration between 0-30 ng l<sup>-1</sup> (ppt) in urine and have a  $^{238}/^{235}$ U isotope ratio of 137.9. Isotope ratios with values calculated between 133.8 and 142 are reported as "No DU present".

Total errors calculated should not exceed 5% for the analysis of a single sample to be accepted. Samples giving errors between 5-8% are re-analysed to give greater certainty. Samples giving errors >8% are repeated in order to increase recovery and reduce matrix effects.

Samples giving low isotope ratios (<133.8) are due to possible interferences on  $^{235}$ U measurement, and they are diluted before re-analysis.

Isotope ratios with values calculated between 142 and 145 are treated as possibly having DU present and reported as such. The raw analytical data are scrutinised with regard to error of measurement. The  $^{236}/^{238}$ U ratio is checked and reported if found to be significant.

Isotope ratios with values exceeding 145 with good precision are regarded as DU present and reported as such, with an estimation of  $^{236}/^{238}$ U ratio.

Results are rejected if concentration of uranium is  $\leq$  1ppt unless standard error of measurement for the sample is within 5%. The sample is reported as "containing insufficient uranium for accurate analysis".

## NIGL

The isotopic analysis of uranium for the main DUOB retrospective testing contract consists of three principal aspects: namely, chemical extraction and purification of uranium from the sample with previously added uranium tracer, the isotopic analysis of the sample by multi-

collector ICP-MS mass spectrometry using both faraday cups and an ion counting detector, and the data reduction and error propagation of the data to achieve the final results.

Unless the sample quantity is very small (<200 ml), the laboratory uses a co-precipitation procedure to concentrate U from urine. For very small samples, evaporation may instead be used.

### Co-precipitation procedure

Concentrated nitric acid is added to samples as the first step in the procedure, to achieve ~2% HNO<sub>3</sub> concentration, with concentrations checked to ensure a pH<1.0. Following this a <sup>233</sup>U tracer is added to the acidified sample and mixed to homogenise the uranium isotopes, and the uranium is separated and purified by being co-precipitated after addition of ammonia from the larger volume of liquid as CaPO<sub>4</sub> at a pH of ~9. With urine, it is inevitable that a considerable quantity of organic material also forms a precipitate at this stage, and all solids are separated and washed by centrifuging in water. The precipitate (CaPO<sub>4</sub> and organic material) is then wet-ashed in an open quartz beaker on a hot plate by repeated exposure to concentrated HNO<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> until all of the organic material is destroyed and the sample becomes white in colour. This white solid is then re-dissolved in weaker nitric acid and loaded onto an Eichrom UTEVA® resin column, washed with nitric acid to remove all elements aside from actinides, and then removed from the column using weak hydrochloric acid. Further evaporations with concentrated nitric acid and hydrogen peroxide are followed by re-dissolving the sample in 2% HNO<sub>3</sub>, which is the preferred medium for introduction into the mass spectrometer for uranium isotope measurements.

### Evaporation procedure (if used)

A portion of the sample is evaporated to dryness on a hot plate with a large quantity of coloured deposit resulting. The warm sample is then wet-ashed in an open beaker on a hot plate by repeated exposure to concentrated HNO<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> until all of the organic material is destroyed and the sample becomes white in colour. This white solid is then re-dissolved in weaker nitric acid and loaded onto an Eichrom UTEVA® resin column, washed with nitric acid to remove all elements aside from actinides, and then removed from the column using weak hydrochloric acid. Further evaporations with concentrated nitric acid and hydrogen peroxide are followed by re-dissolving the sample in 2% HNO<sub>3</sub>, the preferred medium for introduction into the mass spectrometer for uranium isotope measurements.

### Description of the mass spectrometer

The instrument used is the ThermoElemental Axiom double focusing ICP mass spectrometer equipped with an autosampler (Cetac ASX-100), a desolvating nebuliser (Cetac Aridus or Cetac MCN6000) and a multiple array of faraday cups (one axial, 4 low mass, 4 high mass) and one ion counting detector. The overall capabilities and performance of this instrument have been well described in the geochemical literature. It is capable of multiple ion detection and high precision isotope ratio measurement of a wide variety of elements of the periodic table, and has been used widely for uranium isotope measurements in the nuclear industry and in the geochemical-geological research community. During the period of the contract, the laboratory also has available the use of a ThermoFinnigan Neptune MC-ICP-MS.

Like quadrupole ICPMS instruments, the instrument includes a liquid sample introduction system with argon carrier gas, a quartz glass torch, an RF coil to produce a plasma of the argon gas, a series of sample introduction cones to facilitate ion acceleration, and a series of lenses to focus the ion beam. Where it differs from quadrupole ICP-MS instruments is in the

electrostatic and magnetic double focusing analysers. Focused and accelerated ions are guided first through an electrostatic analyzer (ESA) followed by a magnetic sector analyzer, in turn followed by impact against an array of multiple detectors. The instrument used contains multiple faraday cup detectors lying in the focal plane whose positions can be changed, with 4 on the low mass side, 4 on the high mass side, and one in the axial position, the latter of which can be retracted to allow the axial ion beam to progress further down the flight tube, into an ion counting detector, the latter being used in this work.

Ions produced from the sample, its carrier acid components, and the argon gas all enter the high vacuum region of the mass spectrometer, kept at c.  $6 \times 10^{-9}$  mbar pressure. The mass resolution of the instrument is c. 400, with flat top peaks (i.e. 100% of the ions travelling in the high vacuum region entering the cup) and wide baselines separating peaks of unit mass throughout the mass range of all elements of the periodic table. The magnetic field can be varied to steer different combinations of ions into the cup array. By a combination of multiple detection and peak switching, it is possible to measure all of the masses of U by faraday or ion counting detectors, in order to formulate isotopic ratios over a very large dynamic range of signal intensities in a single c. 20 minute measurement. The nature of the plasma is that there are temporal fluctuations in the ion beam intensity that cannot be avoided, and therefore multiple collection of ion beams confers a major precision advantage in the analysis over single collector instruments that entirely rely on peak switching. The sensitivity of the instrument is in the neighbourhood of  $3 \times 10^9$  counts/second/ppm for U using a  $10^{-11}$  Ohm resistor. By a combination of faraday and ion counting detectors, this instrument can measure signals as small as a few counts/s simultaneously with signals of more than  $10^8$  counts/s.

The instrument, the auto-sampler, and the data acquisition are all under computer control, with resultant data written to Excel-readable files and in-house data reduction programmes.

### Set-up of cups, detectors, and measurement cycle

For all measurements of small peaks on the ion counting detector (i.e. 234, 235, 236 measured using ion counting) the signal is ratioed to either 233 or 238 measured in the faraday cup, in order to correct for signal instability. The 233/238 is measured using faraday detectors. Finally, the gain of the ion counting is calibrated for non-linearity by comparison of the 238F/233F (sequence 1) with the 238IC/233F or by external measurement of known standards of variable intensities.

### U standards measured in the analysis for QA of instrument performance

Repeated measurement of the CRM112 natural uranium and CRM U010 doped U solution are made to calibrate the hydride contribution, abundance sensitivity effect, mass bias, and multiplier gain and non-linearity and to ensure that the machine is properly functioning.

### Limits of detection for each uranium isotope

If 3 standard deviations are accepted as the indicator of ability to measure signals and equate this to the limit of detection, then the limits of detection are approximately:

$^{238}\text{U}$  ~600 counts/second, equating to 0.2 ppt in the measured solution

$^{235}\text{U}$  ~20 counts/second, equating to 0.01 ppt in the measured solution

$^{236}\text{U}$  ~3 counts/second, equating to 0.002 ppt in the measured solution

## Uncertainty in the uranium concentration of urine

Using isotope dilution in aqueous liquids (acid, water) the laboratory is able to measure uranium concentration to a precision of better than 0.2%. The failure to achieve this level of precise agreement for urine (actual variation amongst duplicates of 0.1 – 5%) clearly indicates anomalous behaviour for urine. It is most likely that this relates to lack of homogenization of uranium between tracer and urine (dissolved and particulates) and is due to particulates. In all likelihood this could be avoided only by total evaporation of the sample in the presence of the tracer. Once the sample is aliquoted by pouring there may be a fractionation of uranium introduced between aliquots. Experience indicates that the conservative limit of confidence for asserting a single measurement accuracy on concentration measured at this laboratory is  $\pm 3\%$ .

## Propagation of uncertainties in $^{238}\text{U}/^{235}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$

At this laboratory the finalisation of U isotope results consists of *measuring isotope ratios and their uncertainties* using multicollector mass spectrometry on the chemically-separated uranium, *making corrections* to these measured ratios, and *propagating uncertainties* (usually weighted quadratic addition) to the corrections to arrive at final answers, the latter of which contains an element of external reproducibility.

Measurement of ratios consists of two principal aspects – the measurement of standards and unknowns that are interspersed.

## Mass spectrometry measurements of unknowns

Samples are measured in the multicollector mass spectrometer by integrating the signals (using the above peak hopping procedure) of the U isotopes over say ~40-50 separate fixed time intervals, providing ~40-50 separate measurements; >2 sigma outliers (10%) are rejected and the average ratio and its standard deviation re-calculated. From this the standard error of the mean is calculated by dividing the standard deviation by the root of n (n being the number of integrations used). Thus standard error is obtained for the initial measurement. The value in percent of the standard error can vary from ~0.01% to over 1%, depending usually on the signal to noise ratio, in turn a function of the amount of uranium analysed and the sensitivity of the instrument(s). Once primary measurements are made on samples a variety of corrections are then made as described below.

## Corrections

*Measurement of standards (CRM112)*. It is essential to measure standards to monitor instrument performance and for use in normalisation. This is done with a relatively concentrated U signal in order to monitor the instrument's stability, precision, and performance. An important measurement here is the **mass bias**, a precise measure of the extent to which the instrument discriminates against lighter isotopes. For example, a ratio of  $^{238}\text{U}/^{235}\text{U}$  of one will be measured >1 due to the tendency in plasma instruments to favour the heavier isotope. A measure of this through a series of standards interspersed throughout the unknowns is made.

For natural U, the  $^{235}\text{U}/^{238}\text{U}$  is  $1/137.88$  or 0.007253, but in a typical set of standard measurements it will be for example

0.0070839 +/- 0.0000013 (std deviation) or +/- 0.019% (std deviation)

It is appropriate to use the std deviation here rather than standard error since the deviation reflects a time-dependent instrument stability variation. A correction is applied to normalise the data (on standards and unknowns) in line with the natural uranium ratio of the standard, and an uncertainty of +/-0.019% (for the above example) is incurred in so doing. This uncertainty applies equally to all measurements of unknowns during an analytical session and it is quadratically added to the measured ratios of unknowns with a weighting of 1. Thus the propagated uncertainty of this example after this is

$$\text{Uncertainty} = \sqrt{(\% \text{ measured uncertainty})^2 + (0.019\%)^2}$$

The above gives an example of the general procedure for error propagation – the weighted quadratic additions of sources of uncertainty deriving from corrections to measurements. Further sources of uncertainty are added quadratically to the above example equation. These are listed below.

*Abundance sensitivity:* This is the tendency of ions to scatter by collisions in the high vacuum region due to an imperfect vacuum and by imperfect ion optics; when a large peak (say mass  $^{238}\text{U}$ ) is measured there will be a ‘tailing’ of this signal both up and down mass, with some counts registering on adjacent masses. Measurements are made on mass 237 in the knowledge that there is a complete absence of actual peak there, and the count rate gives the tailing. This is relevant particularly for the contributions on 236 arising from tailing from both 235 and 238. These corrections are small and are applied, but an error propagation could be made nevertheless. In practise, the additional uncertainties to the  $^{236}\text{U}/^{238}\text{U}$  error are very small and are usually neglected unless the abundance sensitivity is particularly poor.

*Hydride formation:* There is a small tendency for the ion  $\text{UH}^+$  to form such that the 236 mass would consist of both  $^{235}\text{UH}^+$  and  $^{236}\text{U}^+$ . This is monitored by measuring the 239 mass, and the value of the hydride formation is also less than 1 part in 1,000,000 generally. Again this is so small that the additional uncertainty can be effectively neglected.

*On-peak zero counts correction:* When measuring very small signals on  $^{236}\text{U}$ , it can be important to ascertain whether the instrument is recording counts on that mass without any sample U being analysed. This can be done by aspirating 2% nitric acid (the same as samples are dissolved in) and counting on each mass. Normally this is not done prior to each individual unknown (although this has been done from time to time) but instead the background counts are made periodically for 235, 236 and 238, and in between unknowns the count rate from 238 is required to decline below a nominally low value, usually a few hundred counts/second maximum. This allows the on-peak zero counts to remain below a certain threshold in order to limit its contribution to the next sample to a level that is small in comparison with other sources of uncertainty. Where it is felt that such a correction can confidently be made (for instance when on peak zeros are measured frequently and quantitatively), a weighted uncertainty is quadratically added, the weighting being proportional to the ratio of the correction to the signal intensity of the same mass on unknowns.

*Multiplier gain and non-linearity correction.* Any electron multiplier behaves to a certain extent in a non-linear fashion, and each must be calibrated, usually with the corrected counts being a polynomial function of signal intensity. Measurements of U standards over a wide range of count rates allow this correction equation to be defined, and the scatter about the curve to be estimated. The relative uncertainty about the best fit curve (0.1-0.4% depending on the degree of non-linearity) is then added quadratically to the propagated error for all ratios that include ion counting. For some analytical runs, more frequent ion counting

measurements of uranium standards are made with a level of intensity similar to unknowns, to assist in normalisation.

*Background counts on 236.* When measuring the 236/238 ratio, it is necessary to take account of the noise on the 236 mass arising from instrumental factors. This is done usually by aspirating a blank solution and recording the net count rate on 236 above that produced by abundance sensitivity and hydride corrections. This amount varies according to analytical session but could range from 0.1 up to 5 counts/second. This value is then subtracted from the count rate on 236 on samples, corrected for abundance sensitivity, spike, non-linearity, and hydride, and the net count rate for 236 is then ratioed to the 238 count rate to arrive at a final measured 236/238. The uncertainty of this correction for background counts on 236 is estimated by taking  $\pm\sqrt{\text{background counts}}$  and propagating this into the correction. Quite often the net 236/238 and its uncertainty will overlap with zero and include negative values of 236/238, which is normal and would indicate that 236/238 cannot be distinguished from zero.

*Spike and blank subtraction.* Once the final isotopic ratios are arrived at, there is a correction for the contribution to the final ratio arising from the contamination in the procedure due to blank (i.e. reagents) and from the addition of the  $^{233}\text{U}$  spike (which contains very small amounts of other U isotopes). By far the most important in practice is the blank, since uncertainty arising from the spike is  $\ll 0.1\%$  (because a very high purity spike is used). For the blank, one or two blanks per batch of samples are measured, and the quantity of the blank, its variability amongst the replicate blanks, and its isotopic composition. Usually a realistic uncertainty to the quantity of the blank would be assigned (for example  $\pm 30\%$  of the total blank) and the mean measured isotopic composition of the blank used (usually near the natural value or slightly enriched due to probable small interferences on the 235 isotope). The mean and upper and lower limits to the blank are taken and the upper and lower amounts of the blank subtracted from the measured total quantity of U isotopes in the sample, in order to arrive at the % deviation from the mean arising from the uncertainty in the quantity of blank. This additional deviation is then propagated into the final uncertainty for the isotopic ratio of the sample. The magnitude of the blank correction is generally small, but can rise to significant levels if the amount of U in a sample is very small. Following this step, the final isotopic composition of the sample is derived, along with its uncertainty.

The above corrections comprise the steps to arrive at the final, reportable isotopic ratios.

### Identification of a non-natural isotopic composition of U in urine

The laboratory's procedures of uncertainty propagation appear to be consistent with its measured reproducibility of duplicates and the vast majority of samples that are likely to contain only natural uranium. Because of the precision of measurement and the small magnitude of corrections, it is possible to distinguish non-natural U in a urine sample when the range of 95% confidence quoted on the measured ratio falls outside of the range 137.0-139.0. For example, if the measurement is  $140.2 \pm 0.4$  (95% confidence: range 139.8-140.6) then this would mean that this sample contained DU (albeit small amounts). On the other hand if the measurement returned  $140.2 \pm 1.4$  (95% confidence: range 138.8-141.6) then this would mean that this sample overlaps the range 137.0-139.0 and therefore the conclusion would be that this sample cannot be distinguished from the natural value, though it also cannot be proven NOT to contain DU.

## Fate of the samples and laboratory recording

Once samples are processed, there is residual material left. This is retained until such time as the measurements are deemed satisfactory, after which the samples are discarded, unless otherwise requested by the DUOB.

## Quality assurance procedures

Quality assurances and reliability rest on the assessment of procedures and records, the robustness of the procedures, the reliability of the data and its reproducibility, and the evaluation of repeated analyses of standards and samples.

The laboratory's measurements are traceable to uranium standards, specifically CRM112 natural uranium metal. This metal was purchased for the prior pilot studies, and prepared into a gravimetric solution of known uranium concentration with certified isotopic value.

The principal evidence of reliability of the data is the degree to which analyses on internal urine standards and certified isotopic standards can be reproduced on a repeated basis, the quantity of such repetitions in relation to the unknowns, and the agreement of duplicates of unknowns. All of this applies not only to isotopic ratio, but also measured/calculated uranium concentration.

In the current programme instrument performance is monitored by the analysis of certified U standards 950A (CRM112, natural U). The analysis of these standards is the basis of mass bias corrections and precision of that correction, and corrections on small signals (i.e.  $^{236}\text{U}$ ) for ion counting non-linearity.

The magnitude of blank and spike U contributions to the samples is assessed by isotope dilution analysis of several total procedural blanks, but also by isotope dilution analysis of all of the individual components to the procedure; knowledge of the quantities of all reagents used in each procedure allows for the calculation of a reasonably accurate 'synthetic' blank which can be compared to the total procedural blanks that are measured. In general there is good agreement, and the uncertainties on the blank corrections are propagated through the overall error assessment of sample U isotopic ratios. Each variation in the chemical procedure has its own assigned blank. These are listed in worksheets of respective spreadsheets.

The composition of the  $^{233}\text{U}$  tracer was verified during the course of the pilot study and is precisely known. In practise, the tracer is sufficiently isotopically pure >99.7% that corrections to other isotopic ratios are nearly zero.

The overall quality assurance confidence relies on the internal consistency of replicates, on the general consistency of most analyses with the natural U isotopic value, the resulting agreement of the laboratory's natural uranium urine standards with the natural uranium value, and in part with the observed correlations between 236/238 and 235/238 that are observed with DU-positive samples.

Records and notes are kept of all procedures on the samples, and are contained in a laboratory notebook dedicated to this work.

The laboratory's mass spectrometers receive regular maintenance by its own trained staff supplemented by visits from the manufacturer.

## APPENDIX F

### INFORMATION FOR THOSE SEEKING A TEST FOR DEPLETED URANIUM (DU)

#### What is the test?

- The test measures the levels and types of uranium in a person's urine. This can tell us about a person's past exposure to depleted uranium (DU).

#### How much can the test tell me about my exposure to DU?

The urine test can:

- Detect past exposure to DU in the Persian Gulf or the Balkans at levels well below those which most medical experts believe would carry any significant risk to health.

The urine test cannot:

- Tell you for certain when or where a person's exposure to DU happened.
- Detect very low exposures to DU especially if they happened many years ago.

#### Will my urine be tested for anything else?

- A chemical called creatinine will also be measured. This is to help take account of the fact that some people drink more fluids than others, and therefore have more dilute urine.

#### Can the test tell me anything about my health?

- No, the urine test only gives information about exposure to DU. The test cannot show up the presence or absence of disease.

#### Who is running the test?

- The Depleted Uranium Oversight Board (DUOB) has developed the testing programme. The Board is an independent committee that includes representatives of the Royal British Legion, Gulf veterans, health professionals from within and outside MOD, independent scientists, and representatives from non-governmental organisations (NGOs).
- The day-to-day administration of the testing programme and analysis of urine samples are carried out by independent organisations which have been appointed on the recommendation of the DUOB.



## **Who is eligible for testing?**

Individuals who were:

- members of the UK armed forces on deployment; or
  - civilian employees of the UK Ministry of Defence; or
  - civilians working under contract to the UK Ministry of Defence; or
  - civilians employed by other UK Government departments or non-governmental organisations in support of UK military operations;
- a) in the Persian Gulf area between August 1<sup>st</sup> 1990 and July 31<sup>st</sup> 1991; or
- b) in the former Republic of Yugoslavia on or after August 5<sup>th</sup> 1994.

## **How can I apply for a test?**

- You should have received an application form with this factsheet. If not, please write to the following address or email: [info@duob.org.uk](mailto:info@duob.org.uk).

DUOB Secretariat  
Veterans Policy Unit  
Floor 7 Zone H  
MoD Main Building  
Whitehall  
London SW1A 2HB

## **Is there a time limit for applying for the test?**

Yes: the closing date for receipt of applications is January 31<sup>st</sup> 2006.

## **Will I have to pay for the test?**

- The MOD will pay for the testing of veterans, contractors and non-governmental (NGO) personnel who were deployed with military units in the Persian Gulf or Balkans, although it is in no way involved in the testing itself or in the reporting and interpretation of individual test results.
- If MOD is paying for your test, it will also reimburse all reasonable travel expenses that are incurred. You will be given a claim form once the Veterans Policy Unit has confirmed your eligibility for testing.
- NGO personnel who were not attached to military units will also have access to testing, but they or their organisations may be asked to pay.

### **What does the test involve?**

- You will receive an appointment to attend a clinic at an NHS hospital.
- Before you attend the clinic, you will have been sent a container to collect your urine over a 24-hour period.
- You will be given guidance on how and when to collect the urine.
- Before your test you will be asked to complete a questionnaire about yourself and your service or work in the Persian Gulf and/or Balkans.
- This information will be used to help a doctor interpret the results of your test. With your agreement, it will also be used (in an anonymous way) to give statistical information about the relationship of depleted uranium exposure to different aspects of service in the Persian Gulf and/or Balkans.
- At the clinic, you will hand in your urine sample and completed questionnaire. The clinic staff will discuss any difficulties you may have had collecting the sample or completing the questionnaire.
- If you wish, you will be given a copy of the completed questionnaire for your own use.
- Your sample will then be sent to an independent laboratory for analysis.
- You will receive a letter telling you the results of the analysis (usually within 6 weeks) and what the results mean. In case you have any questions about the test or your results, a telephone helpline will be available.
- The test results can be sent to your GP if you wish.

### **What happens if I am too ill to travel to a clinic?**

- If someone is too ill to travel to a clinic, an appointment can be made at their home. If you think this might apply to you, you should let us know when you apply for a test.

### **How will my privacy and the security of my sample be ensured?**

- All staff involved in the testing programme are subject to a strict code of patient confidentiality.
- An individual code number (which you will be told) will identify your sample and questionnaire.
- Before being sent to the laboratory, your urine sample will be closed by the clinic staff in your presence with a "tamper-evident" seal. The laboratory will alert the DUOB if it receives any samples that may have been tampered with. In the unlikely event that this happens, the sample will be discarded, the individual concerned informed, and a repeat test arranged.
- No identifiable information will be passed to the MOD or anyone outside the testing programme without prior consent from the individual concerned.
- Non-identifiable information may be used for accounting purposes.
- If you agree, a copy of your questionnaire and of your test results will be lodged with an independent solicitor. It will then be available for future reference by you or your nominated representative if necessary.

**Why is a 24-hour specimen necessary?**

- Currently a 24-hour specimen is thought necessary to ensure that the result of the test is sufficiently accurate. Smaller samples of urine have not been shown to give the required accuracy for exposures that occurred many years before testing.

**Can I continue to take my prescribed medication during the urine collection?**

- Yes.

**Can I eat and drink normally during the urine collection?**

- Yes.

**Some countries use hair samples for testing for DU; why are we using only urine samples?**

- Unlike the urine test, tests based on hair samples have not yet been shown to be sufficiently accurate or reliable.

## APPENDIX G

### DEPLETED URANIUM FACTSHEET FOR VETERANS WHO REQUIRE BASIC LEVEL INFORMATION ABOUT DU

#### What is uranium?

**Uranium** is a natural element found in soil, water, and mineral deposits. It is a heavy metal, nearly twice as dense as lead, is radioactive and chemically toxic. **Everyone** has low levels of uranium in their body, from their food, drink and the environment, and passes small amounts in their urine.

#### What is depleted uranium (also called DU)?

**Depleted uranium** or **DU** is a waste product. It is what is left after the removal of some of the more radioactive parts of **natural uranium** for use in the nuclear industry.

#### Is it radioactive?

Yes, but its radiation risk when outside the body is low. DU is slightly less radioactive than natural uranium.

#### Is it poisonous?

DU is a heavy metal and, in common with other heavy metals such as lead and mercury, it is chemically poisonous in high doses.

#### What are the military uses of DU?

Being a very dense and hard metal makes **DU** an ideal core for tank shells designed to pierce armoured vehicles.

**DU** is used in:

- Armour piercing anti-tank weapons
- Protective plating in some tanks (US "M1 Abrams")
- Some naval armaments (Phalanx)
- Ammunition of US A-10 anti-tank aircraft

#### How does it work?

DU rounds behave a bit like large self-sharpening darts. On hitting an armoured target, they go through it, break up, and partially burn with the formation of a cloud of fine dust.

#### Sources of exposure to DU in the Persian Gulf and Balkans

A small number of US soldiers in the Gulf War suffered shrapnel wounds from the use of **DU munitions** in "friendly fire" incidents. Exposure would more commonly occur by breathing in the fine dust produced when armoured targets are hit. This might happen soon after the target was struck or later, since wind and motion can disturb the settled dust, sending it up into the air again.

## Who may have been exposed to DU?

Personnel who may have been exposed to DU include:

- personnel who were inside or near vehicles struck by DU weapons
- personnel involved in removing casualties
- clean-up teams
- vehicle and aircraft recovery teams
- explosive ordnance disposal technicians
- electrical and mechanical engineers
- medical services staff

## How could DU get inside my body?

DU can enter the body by:

- Ingestion, by eating food and drink contaminated with DU, or from hand to mouth contact (e.g. when smoking)
- Breathing in the fine dust
- Contamination of cuts and wounds
- Embedded shrapnel

## Health hazards of DU inside the body

- DU dust is chemically poisonous and weakly radioactive. If the dust is breathed in, some radioactive dust particles may stay in the body for many years and increase the risk of cancer.
- Like other heavy metals such as lead, **depleted uranium** is chemically poisonous if it gets inside the body. The kidney is the organ most sensitive to uranium poisoning.

## Sources of further information:

Use search engine, type depleted uranium for links to further information from web sites:

Depleted Uranium Oversight Board: [www.duob.org.uk](http://www.duob.org.uk)

Ministry of Defence:

<http://www.mod.uk/DefenceInternet/AboutDefence/WhatWeDo/HealthandSafety/Depleted+Uranium/DepletedUraniumdu.htm>

World Health Organisation: [www.who.int](http://www.who.int)

Royal Society: [www.royalsoc.ac.uk](http://www.royalsoc.ac.uk)

Office of the Special Assistant for Gulf War Illnesses:

[http://www.gulflink.osd.mil/faq/faq\\_du.jsp](http://www.gulflink.osd.mil/faq/faq_du.jsp)

BBC News online: <http://news.bbc.co.uk/>

Health Protection Agency: [www.hpa.org.uk/radiation/faq/default.htm](http://www.hpa.org.uk/radiation/faq/default.htm)

An alternative to the mainstream scientific view can be found at:

- [www.euradcom.org](http://www.euradcom.org)
- [www.llrc.org](http://www.llrc.org)

## **INFORMATION FOR VETERANS AND THEIR MEDICAL ADVISORS: THE ASSESSMENT OF RADIATION DOSE FROM MEASUREMENTS OF DEPLETED URANIUM IN URINE SAMPLES**

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### **Summary**

This note describes in simple terms how measurements on urine samples can be used to determine the radiation dose for a person who may have breathed in depleted uranium (DU) dust a number of years earlier. It also explains how to judge the significance of radiation doses assessed in this way. It is written for a non-scientific audience, including veterans of the 1990/91 Gulf War and those involved in peace-keeping operations in the former Republic of Yugoslavia during and after 1994. It should also be of use to General Practitioners and other medically-qualified professionals who may be consulted by veterans.

### **Why are measurements being made on urine samples?**

If a person breathes in a dust containing DU, some of the inhaled particles remain in the lungs. These particles are then removed by natural processes. Particles in the upper airways may be trapped in mucus that moves to the throat and is then swallowed within a few hours. These particles are then excreted within a day or so, mainly in faeces. Particles in the deep lung are captured by cells similar to white blood cells, and are moved either towards the throat, or to lymph nodes. Removal of these particles is slow and some can remain in the lungs for years.

While the particles are being moved out of the lungs by natural processes, they also dissolve, releasing DU into the bloodstream. This is the main source of the DU that can be found in urine. The rate at which particles dissolve in the lungs depends on what they are made of<sup>2</sup>, but even insoluble materials dissolve to some extent. Most of the DU released into the bloodstream is excreted in urine within a day or so. The DU that is not rapidly excreted in this way goes into other organs of the body, mainly the kidneys and the skeleton. About 15% of the DU released into the bloodstream goes into the skeleton, from which it is removed slowly; about 1% is still present after 25 years. The slow dissolution of particles in the lungs and the slow release of material from the organs back into the bloodstream means that small concentrations of DU are present in urine many years after the DU was inhaled.

A great deal of scientific work has been carried out over the last 50 years to understand the behaviour of inhaled radioactive materials in the body. As a result, mathematical models have been developed that can predict the daily excretion of a radioactive material at any time provided the amount originally inhaled<sup>3</sup> is known. The models can also be used "in reverse", to calculate the amount originally inhaled from the results of a measurement of

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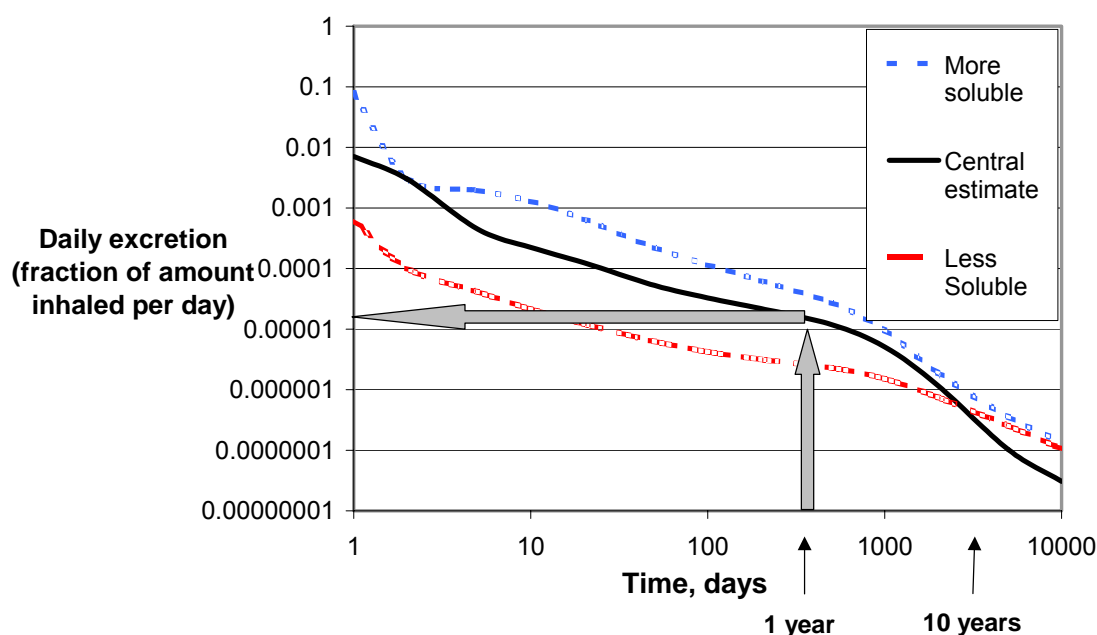
<sup>2</sup> For example, salt particles are very soluble, while particles of fine sand are very insoluble.

<sup>3</sup> The amount inhaled is sometimes called the "intake".

daily excretion. Figure 1 shows how this is done. If a measurement of daily DU excretion in urine is made 1 year after inhalation of DU took place, the graph shows that the amount excreted is expected to be about 1/100,000 of the amount inhaled. The amount inhaled is therefore approximately equal to the measured daily excretion multiplied by 100,000.

We do not have complete information on the physical and chemical properties of the DU dusts that could be inhaled, and so there is some uncertainty in the model predictions of urine excretion, as illustrated in the figure. More information on the topic of “What happens to depleted uranium inside the body” can be found on the website of the Health Protection Agency, <http://www.hpa.org.uk/radiation/fag/du/du7.htm>.

**Figure 1. DU urinary excretion predicted using ICRP<sup>4</sup> models with parameter values appropriate for DU (Royal Society, 2001)**



### How are measurements of DU-in-urine made ?

It is difficult to measure DU in urine, for two main reasons. Firstly, the amounts of DU excreted in urine several years after intake are likely to be very low, requiring a measurement technique that can measure very small amounts. Secondly, everyone excretes uranium in their urine every day, because of the presence of naturally-occurring uranium in drinking water and food<sup>5</sup>. Thus, the measurement technique must be able to distinguish between DU and natural uranium. This can be done by measuring separately the amounts of the different uranium *isotopes* using high sensitivity *mass spectrometry* techniques.

<sup>4</sup> The International Commission on Radiological Protection, a group of scientists who are experts in radiation protection.

<sup>5</sup> Uranium is found everywhere in the environment. For instance, the top metre of soil in a typical garden contains about 2 kilograms of uranium.

Natural uranium contains three isotopes: uranium-234, uranium-235 and uranium-238 <sup>6</sup>. Almost all of the weight of a natural uranium sample (more than 99%) comes from the uranium-238 isotope, while uranium-235 makes up 0.71% and uranium-234 makes up 0.005% <sup>7</sup>. In DU, most but not all of the uranium-234 and uranium-235 isotopes have been removed. Typically, DU used in munitions contains 0.2% of uranium-235 by weight. If the uranium-235 content in a sample of uranium is measured as 0.71%, then the uranium is natural and no DU is present; if the uranium-235 content is 0.2%, then the uranium is depleted and no natural uranium is present; and if the uranium-235 content is between 0.2% and 0.71%, then the uranium is a mixture of DU and natural uranium. The amounts of DU and natural uranium can be determined using simple maths.

The daily DU urine excretion can be determined directly by collecting and measuring a 24-hour urine sample, or by measuring a sample collected over a shorter time and then multiplying up to get the daily amount. This is best done by measuring the amount of a chemical called creatinine that is produced by the body and excreted in the urine. The amount of creatinine excreted each day is known and fairly constant, and so the amount measured in a urine sample can be used to determine the fraction of the daily excretion of urine that the sample represents.

Once the DU daily excretion has been determined, mathematical models can be used to calculate the amount originally inhaled, as shown in figure 1. The same models can also be used to determine how the amount of DU in different organs of the body varies over time following that intake.

### What is a radiation dose ?

All the isotopes of uranium are radioactive. This means that the nucleus <sup>8</sup> of a uranium atom can undergo a change called radioactive decay, emitting *ionising radiation*. It is this radiation that can cause the damage inside the cells of the body that might result in a cancer. The radioactive decay of uranium results in three main types of ionising radiation: *alpha*, *beta* and *gamma* radiation. Each radioactive decay of a nucleus results in the emission of an alpha particle or a beta particle and/or one or more gamma rays or X-rays, which each carry a certain amount of energy. The number of decays of a radioactive isotope in each second is a measure of the amount present, and is called the *activity* of the isotope. It is measured in becquerels, abbreviated to Bq. A becquerel is a very small amount: a thousand becquerels is a kilobecquerel, abbreviated to kBq.

Alpha and beta particles do not travel very far before being absorbed and giving up their energy. After a radioactive decay inside an organ of the body (eg the lungs), alpha and beta particles are therefore likely to deposit their energy in the same organ. Gamma rays travel further, and may deposit their energy in the same organ or in a neighbouring one. The *radiation dose* received by each organ during a particular period of time is the amount of energy deposited in that organ as a result of radioactive decay, divided by the weight of the organ. This is called the *absorbed dose*, and is measured in grays, abbreviated to Gy. The risk of harm from ionising radiation is generally thought to be related to the absorbed dose.

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<sup>6</sup> The isotopes are all forms of uranium, the difference being that the atoms of different isotopes have slightly different weights (or masses). *Mass spectrometry* actually measures the masses of single atoms in a sample of material.

<sup>7</sup> Even though almost all of the weight of uranium is associated with the uranium-238 isotope, the other isotopes still need to be considered because, weight for weight, the other two isotopes are more radioactive than uranium-238.

<sup>8</sup> The nucleus of an atom is much smaller than the atom itself, but contains almost all of the mass.



Scientific papers and reports often refer to a number of other radiation dose quantities, described briefly below.

### Equivalent dose

Some types of ionising radiation, notably alpha radiation, are more harmful than others. To take account of this, the absorbed dose is “weighted”, which means that it is multiplied by a radiation weighting factor to give the equivalent dose. For gamma-rays, X-rays and beta radiation the factor is set to 1, while for alpha radiation it is 20. Equivalent dose is expressed in a unit called the sievert, abbreviated to Sv. This is a rather large radiation dose, so the millisievert (mSv), which is one thousandth of a sievert, is more commonly used.

### Committed equivalent dose

When a person has a medical X-ray, the radiation dose is received immediately. No further dose is received after the X-ray has been taken. However, when a radioactive material is inhaled, the radiation dose continues to be received for as long as the material remains in the body. As already mentioned, a small fraction of inhaled uranium can remain in the body for many years. Thus, even though the uranium may have been inhaled over a short time, we say that the person is *committed* to receiving a dose for a long time to come. The committed equivalent dose is the dose expected to be received by a particular organ in a stated period after the intake, usually 50 years for exposures at work or up to age 70 for members of the public. The radiation dose received from a medical X-ray can be measured easily using a dose meter, because the source of the radiation is outside the body. However, when the radiation dose comes from radioactive materials that are inside the body, it cannot be measured directly. This is the reason for using mathematical models to calculate *internal* doses, as described earlier.

### Committed effective dose

It has already been mentioned that some types of radiation are more harmful than others. Scientists have also found that radiation is more harmful to some organs rather than others. For example, 1 Sv received by the lungs is about twice as likely to result in a cancer as 1 Sv received by the liver. To obtain an estimate of overall risk of harm to a person, the committed equivalent doses to each important organ are multiplied by a tissue weighting factor, and are then added together to give the committed effective dose. This is also expressed in units of Sv or mSv. The tissue weighting factor represents the risk of harm per Sv to the organ, compared to the risk of harm per Sv to the whole body. Effective dose is a useful standardised measure of the risks from exposures to radiation, especially those resulting from intakes of radioactive materials, which often result in very different doses to different organs.

### What level of radiation dose is safe?

For radiation protection purposes, we make an assumption (based on scientific evidence) that the risk of cancer in a particular organ increases as the radiation dose to that organ increases<sup>9</sup>. According to this description, no radiation dose (whether natural or artificial) is

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<sup>9</sup> This assumption is the subject of considerable debate. Most radiation protection scientists think that the assumption is either realistic or cautious, and so is adequate for the purposes of radiation protection. Some think that low radiation doses carry no risk at all, or can even have beneficial effects (reference 1), while a small number think that these risks are seriously underestimated (reference 2).

*absolutely* safe. Rather, large radiation doses result in high risks, while low doses result in small risks. One way of judging the significance of a particular radiation dose is to compare it with the average annual dose from natural background radiation in the UK, ie 2.3 mSv per year. Another way is to compare it to one of the *dose limits*. Dose limits are set so that continued exposure just above these levels would result in additional risks that in normal circumstances are judged to be unacceptable. The annual dose limit for members of the public is 1 mSv per year, while the annual dose limit for exposure at work is 20 mSv per year. A common misunderstanding is that radiation doses above the dose limit are bound to cause health problems. This is not the case. The radiation doses that would cause “radiation sickness” are more than 1000 times greater than the annual dose limit for members of the public.

### What do radiation doses mean in terms of risk to health ?

ICRP has estimated the probability of contracting a fatal lung cancer as a result of radiation exposures at work as 0.68% per Sv <sup>10</sup>. This means that a committed equivalent dose of 1 Sv to the lungs would result in an additional lifetime risk of about 1 in 150 of a person contracting lung cancer. An intake of DU by inhalation giving rise to a **committed effective dose of 1 mSv** would also result in a committed equivalent dose to the lungs of about 7 mSv, and an additional lifetime risk of developing fatal lung cancer of about **1 in 20,000**. The risk of developing other types of fatal cancer would be less than 1% of the fatal lung cancer risk. These risks should be compared with the average lifetime risk of contracting a fatal cancer **from all causes** of about **1 in 4**.

### What level of DU in urine corresponds to a dose of 1 mSv?

The daily DU urine excretion that would, if measured, lead to an estimate for committed effective dose of 1 mSv can be calculated using the mathematical models discussed earlier. If a urine sample is taken 12½ years after the DU was inhaled, our best estimate for the amount of DU excreted is about 2 nanograms <sup>11</sup> per day <sup>12</sup>. Values of daily urine excretion corresponding to other committed effective dose values can be obtained by simple arithmetic. Thus the daily DU urine excretion corresponding to a committed effective dose of 20 mSv would be about  $20 \times 2 = 40$  ng per day. There is some uncertainty in the model predictions of urine excretion, and it is possible we could underestimate or overestimate the committed effective dose by a factor of ten. Thus, pessimistically, a measured DU daily excretion of 2 ng/day could possibly correspond to a committed effective dose of 10 mSv. If DU is detected in a urine sample, this type of calculation will be performed on a case-by-case basis to obtain an estimate of the radiation dose.

It should be noted that doses to veterans assessed from DU in urine measurements are committed doses resulting from a single intake that will in all likelihood not be repeated during the veteran's lifetime. On the other hand, the annual dose limits are set making the underlying assumption that intakes could be received in each year of the lifetime of an

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<sup>10</sup> ICRP's probability coefficients for workers have been determined for a population comprising equal numbers of both sexes with a wide range of ages, and are not appropriate for evaluating cancer risks for individuals. Rather, assessments of risk must be carried out on a case-by-case basis, taking into account the age and sex of the subject, as well as the available information on the nature of the exposure to DU.

<sup>11</sup> There are 1,000,000,000 nanograms (abbreviated to ng) in 1 gram. One grain of fine sand weighs about 10,000 nanograms.

<sup>12</sup> If the sample was taken later, the predicted excretion would be somewhat lower; if earlier, the predicted excretion would be somewhat higher.

individual and that (in the absence of doses from external exposure) each annual intake might result in a committed dose at the annual dose limit. Thus, by comparing an assessed dose from a single exposure to DU with the annual dose limit, and by using pessimistic model predictions, a large safety margin is built in.

### Can we measure radiation doses from inhalation of DU with enough sensitivity?

Recent work carried out by the DUOB in collaboration with the laboratories who are making the urine sample measurements shows that daily DU urine excretion of about 0.4 ng/day in people who are also excreting 10 ng/day of natural uranium should usually be detectable by mass spectrometry. Our mathematical models show that lifetime doses below both the annual public dose limit and the average annual dose from natural background radiation in the UK should be quite easily detectable.

### Further reading

National Radiological Protection Board (2004). Frequently Asked Questions; Depleted Uranium. <http://www.hpa.org.uk/radiation/faq/du/>.

Royal Society (2001). The Health Hazards of Depleted Uranium Munitions, Part 1. Policy Document 6/01. London, Royal Society. <http://www.royalsoc.ac.uk/> (Search for “depleted uranium”).

### References

1. E J Calabrese and L A Baldwin. Toxicology rethinks its central belief. *Nature*, volume 421, pages 691-2 (2003).
2. 2003 Recommendations of the European Committee on Radiation Risk. Green Audit Press, Aberystwyth (2003).

Minority review of the discussions, investigations  
and results of the  
UK Ministry of Defence  
Depleted Uranium Oversight Board  
2001-2006

Dr Chris Busby  
DUOB  
Prof. Malcolm Hooper  
DUOB

(Dr Derek Hall, DUOB  
Mr Ivor Connolly, DUOB)

## **Foreword**

The DUOB completed its work and obtained results from two laboratories which conducted urine tests for uranium isotopes on those veterans who had engaged in the process. However, there were differences in the committee over the interpretation of the results and discussions. The committee agreed that these differences were best expressed by including a dissenting review of the evidence and its interpretation from those members of the committee who disagreed with the thrust of the main DUOB report. This review follows.

## **Summary**

We believe that the evidence of enriched uranium in veterans' samples and in the environment, which emerged through the period of the DUOB measurements and discussions, makes the method unsafe. The results cannot be used to draw conclusions about veterans' contamination. In addition to this, we believe that application of the ICRP biokinetic model as basis of deciding whether historic contamination has occurred is also unsafe. This is because the model uses parameters for rats exposed to a different type of uranium and data collected over a 6 month period. The extrapolation for rats and large enriched uranium particles over 6 months to humans and sub micron ceramic uranium oxide particles over 13 years is unscientific.

There is now a great deal of evidence that the health consequences of uranium exposure are real and serious and are not predicted or explained by the radiological risk model presently employed. We have concluded that the DUOB measurement process did not obtain the information it set out to obtain and that other approaches are needed to make decisions about the veterans' contaminations and illnesses. For these reasons, we feel that it is important that no assumptions about veterans' health and exposure to depleted uranium should be made on the basis of the DUOB results. We also believe that the situation in Iraqis now addressed as fully as possible.

At the end of this process, we feel strongly that oppositional committees, like the DUOB, represent the best way society has of obtaining as close to the truth as it is possible to get in complex scientific areas. We see the DUOB as a model of excellence in this regard and thank both the members of the board, and the Chair and also the MoD administrative staff for their courtesy, forbearance and hard work.

## **1. Background**

The first study of depleted uranium, DU, in Gulf War 1 veterans arose following the concerns of Dr Asaf Durakovic, a specialist in nuclear medicine, during his time as a physician at the Veterans Administration in the USA. He has published several papers and reviews of the adverse health effects of DU (Durakovic, 1999, 2001) including a study in which UK veterans were found to have DU in their urine, (Durakovic 2002). He also carried out analyses of autopsy samples from a Canadian veteran, Terry Riordan, and detected high DU levels of DU in bone (Canadian Parliament, 2000).

It has been clearly established that the participants in the first Gulf War were not told of any particular hazards that might be associated with the use of DU munitions and were exposed without any protective measures taken or advised to these hazards, despite warnings and advice given in military manuals since 1974 in the USA, (Fahey 1996). Furthermore, the MOD admitted repeatedly failing to inform troops of these hazards, (MOD, 1991, 1993 and Watts and Norton-Taylor, 2001). Many veterans regard this failure as a breach of the duty of care owed to them by the MOD. In the States the situation led to government action and publications of data from studies on "friendly fire" events, (McDiarmid, 1999, 2000).

In the UK, a report from Atomic Energy Technology, the commercial arm of the Atomic Energy Authority, prepared in 1991 became public in 1999 (AET 1991) following parliamentary questions. It estimated 500,000 extra cancers would occur in S. Iraq over the next 10 years.

The Royal Society, (2001, 2002) was drawn into issues about DU alongside emerging, wider questions about the impact of low level radiation and the internal exposure to radioactive materials that were under discussion by CERRIE, the Committee Examining Radiation Risks of Internal Emitters, that reported to COMARE, the Committee on Medical Aspects of Radiation in the Environment, another government committee looking at the same kind of questions. In Europe and internationally similar questions were also being raised by concerned scientists and the public (ECRR2003).

The DUOB was finally established in 2001 – 10 years after the end of the First Gulf War, 1990-1991, and before the beginning of the second Gulf War, 2003, and war in Afghanistan.

Following both pressure from veterans and suggestions made by the Royal Society Committee process which examined the issue of the health effects following battlefield use of Depleted Uranium weapons, the Depleted Uranium Oversight Board (DUOB) of the Ministry of Defence was set up. The Royal Society suggested in 2001 that the Gulf War 1 veterans should be tested for contamination by Depleted Uranium and so the DUOB's main remit was to determine the best method for such examinations and to oversee the process. In addition it was to look at other ways of examining the issue, to advise on epidemiological investigations of the reports of association between uranium exposure and ill health and generally review the latter area.

The DUOB was one of the first of a type of committee that has recently been called 'oppositional' (Busby et al 2000, van den Hazel et al 2006). This is a committee which has to examine and advise on an issue which is contentious and to which there are different attitudes or interpretations of the scientific evidence. To do this it starts out by purposely including members who have diametrically opposed views on the issue so as to get to the best answer by a dialectical process. In this case, probably because there was considerable suspicion of the Ministry of Defence, the DUOB was set up to include contributions and the involvement of actors from all sides of the debate into the health effects of DU weapons. Thus members who were critical of the position taken by the military and the conventional (Royal Society) view of the radiological consequences of exposure to battlefield uranium oxide particles were included. We would like to put on record that we have nothing but praise for the committee process itself and both for the administrative support and the behaviour of the MoD officials who organised the process, prepared and circulated documents and helped with all the various complications relating to the measurements. The DUOB process could be seen as a model of excellence for the idea of the oppositional committee and support a belief that, properly and agnostically managed this is a good model for advisory committees on contentious issues. Members of the committee and the Chair were courteous, helpful and accommodating even when there were strong disagreements. The main purpose of the DUOB was to develop a test which would show past exposure to DU and oversee its application to as many Gulf War 1 veterans who wanted to be tested. As we saw our role, it was to ensure, as far as possible, that the test results would be accurate and believable. We were particularly concerned, as were many veterans, that the MoD would find some way of making the measurements but not reporting the true values. The process, in other words, had to be believable. To ensure that this happened, we argued for the introduction of many safety procedures involving blinding (coding) of samples and duplication of measurements. We also argued for the random introduction into the measurement routines of dummy samples that had been spiked with a known quantity of depleted uranium by a different laboratory and for these dummies to be tested by both of the two laboratories that had been chosen after the initial pilot studies were conducted. These safety procedures were mostly accepted and employed. Because of this we find it hard to see how there could be any suspicion that the results we have obtained are not the true results of the measurements. We do not rule it out as impossible, but there would have to be so much collusion between senior staff in so many different laboratories and institutions that the risk of this coming to light would be too dangerous a possibility. So for what it is worth, we give our seal of approval to the results that are reported. But what do these results mean, and how are they to be interpreted? And as far as the other work of the DUOB was

concerned, the process brought to light and examined various issues relating to the health effects of DU contamination, and we will review this area separately.

## **2 The urine test results for DU in the Gulf War 1 veterans and their interpretation.**

The urine test was employed in order to see which veterans had been exposed to Depleted Uranium in the first Gulf War in 1991. Epidemiological studies could then make comparisons between exposed and unexposed. But a number of questions needed to be asked before it might be assumed that the concentration and isotopic ratio of uranium in urine could be safely used to characterise exposed individuals given that some 12 years had elapsed between the testing and the exposures. In particular, the question arises as to whether the absence of evidence in the urine test means both absence of exposure to depleted uranium and absence of health effects from such exposure. We believe it is not safe to assume either of these for reasons which we shall now outline.

Natural uranium, which exists in ores in rocks or which existed on earth before the nuclear age has an atom ratio for the isotopes U238/U235 of 137.88. It is this fact that is the basis of the urine test. In this test, significant deviations in the uranium found in an individual's urine upwards from a natural isotopic ratio are used to conclude that the individual had been exposed to depleted uranium in the Gulf War or Balkans. The isotopic ratio of pure depleted is about 450, and so it is assumed, for the purposes of the conclusions, that if the individual had been exposed in the distant past to depleted uranium, some of this, from the depot stored in the body, would still be excreted daily some 12 years after the initial exposure. But there are two assumptions implicit in this reasoning:

- The isotopic ratio of all uranium in the environment of the individual since the war period of exposure is indeed the natural one of 137.88.
- The depleted uranium contamination the individual received in the war period has made available a depot of depleted uranium in the body which is being constantly excreted in the urine some 12 years later at a rate given by an accurate biokinetic model whose parameters are known for the type of uranium produced by depleted uranium weapons.

With regard to the health effects of depleted uranium exposure there are further major uncertainties which are relevant to the work of the DUOB. They are associated with the radiological risk model employed to link the uranium exposures with possible health effects and include within this the assumption of a monotonic relationship between exposure and ill health, and little or no variation in the sensitivity of individuals to such exposures. We shall return to the radiological health models, but will first consider the assumptions relating to the measurements and their interpretation. These concerns about the interpretation of the results are condensed in Table 2.1 below.

**Table 2.1.** Problems with assumption on which the interpretations of the DUOB conclusions are based.

Group/ Decision	Assumption	Problem	Cause
Exposed Veterans	Excrete DU	No longer excrete DU	Too long ago, biokinetic model false, based on rats and different uranium type.
UK population	Excrete natural U	Don't excrete natural U	Environment is contaminated with depleted and enriched uranium from various sources, weapons fallout, nuclear sites, ???
Decision base 1	Exposed if U238/U235 >142	Not true	U238/U235 is affected by presence of U235 in environment and individual (see text).
Decision base 2	U-236 presence = exposed	Various	1. Not known if all DU has U-236 2. Too long a period has elapsed 3. Sensitivity problems. 4. Only half the vets measured.

*The biokinetic modelling assumptions are invalid*

In order to interpret the results of the veterans we must know what to expect in a population living in the UK. We must also have an idea of what level of difference from an ideal control population result we might expect. The DUOB interpretation framework was entirely based on the biokinetic mathematical models for uranium excretion used by the Royal Society. These were the biokinetic organ partition models of the International Commission for Radiological Protection (ICRP). This employed factors which had been culled from animal experiments with a different type of Uranium from the type produced on the battlefield (Ansoborlo et al 1998). These authors obtained concentrations of uranium in urine from studies of 35 rats exposed to aerosols produced from a laser enrichment facility. The particles were much larger than the 'ceramic' DU oxide particles produced by battlefield weapons, more than 80% of the particles being larger than 5 µ mean diameter. DU particles from battlefield origins are much smaller, being mostly well below 1 µ diameter (Glissmeyer et al 1979,1981) and therefore likely to reach parts of the body that the larger particles would not. Second, it must be emphasized that the end point of the calculation is the predicted urinary excretion of some of the depleted uranium in a human being 12 years after contamination. This is based on a mathematical model which employs measurements made on the urine from a rat, contaminated with enriched uranium, over a period of 6 months coupled with the uranium concentration in the rat's body when it was killed at this point. We do not believe that this model can safely be used as a predictor of uranium concentration in the DUOB process. The scientific (philosophical) distance between the model and its parameters and assumptions and what it is being employed for is too great. In particular, it seems to us that it is highly likely that there is a depot of uranium oxide particles in the body, probably in the lymphatic system draining the lung, and in bone, (Canadian Parliament, UMRC, download 2007), that may be considered to be entirely insoluble.

For these reasons we do not believe the arguments of the paper on uranium biokinetics produced for the DUOB by the National Radiological Protection Board and attached as an annexe to the final report.

*There are errors due to environmental variation of the uranium isotope ratio.*

There were two parameters available from the tests. These were Total Uranium in urine and Uranium isotope ratios. Total Uranium excretion is quite variable since Uranium occurs



naturally in different amounts and is present in different foods to different extents, so the total uranium levels are not in themselves alone very useful. Depleted Uranium is natural Uranium from which much of the fissile isotope U-235 has been extracted. Natural Uranium in the environment (prior to any man made activity) has a ratio of U-238 to U-235 of 137.88. Depleted Uranium as used in weapons is believed to have an isotope ratio of U238/U235 of about 450. There is also, of course, enriched Uranium which should be confined to nuclear reactors and nuclear bombs, but as we shall see, is also present in the environment. This may be considered to have a ratio of 60 or below. However, the DUOB did not initially imagine that there could be any enriched Uranium in the environment and so the basis for the test was that past exposures could be signalled by the presence of an isotope ratio in the urine of a value significantly greater than 137.88. After some discussion about errors in the instrumental procedures it was agreed by the committee that DU presence would be indicated by a cut off level of 142. Thus any ratio of greater than 142 would be reported as showing DU presence. Following the results of a pilot study of non-veterans conducted to see what the background levels and ratios were in the UK one of us (CB) suggested a cut off point which was lower than this, but this was not adopted by the committee.

The results of the pilot study are helpful in interpreting the veteran's results. However, the measurements that were made of both veterans and the pilot study gave results that showed the existence of enriched uranium in the environment. This was confirmed at one meeting where Prof. Parrish told the committee that he had made measurements of drinking water samples which showed the presence of enriched uranium in part of the water supply of the UK. At this meeting, where a draft of this present paper was discussed, it was suggested that the statistical evidence for the presence of enriched uranium in the overall data was an artefact and due to errors in the measurements at low total uranium concentration. It was suggested that if the isotope ratios results were examined by their standard error (discriminating between low overall and higher overall uranium concentration) the evidence for enriched uranium would be seen to be artifactual. For this reason, this idea has been examined this by a further statistical analysis of the data split into groups of low and high total uranium. The result shows that there is no difference in the evidence for enriched uranium contamination by levels of tabulated error and so this argument falls. This is an important point since, as we shall show below, if there has been contamination by enriched uranium at any point in the period following the original putative depleted uranium exposure, the whole test is invalidated and the results become meaningless.

### **3 The normative study results.**

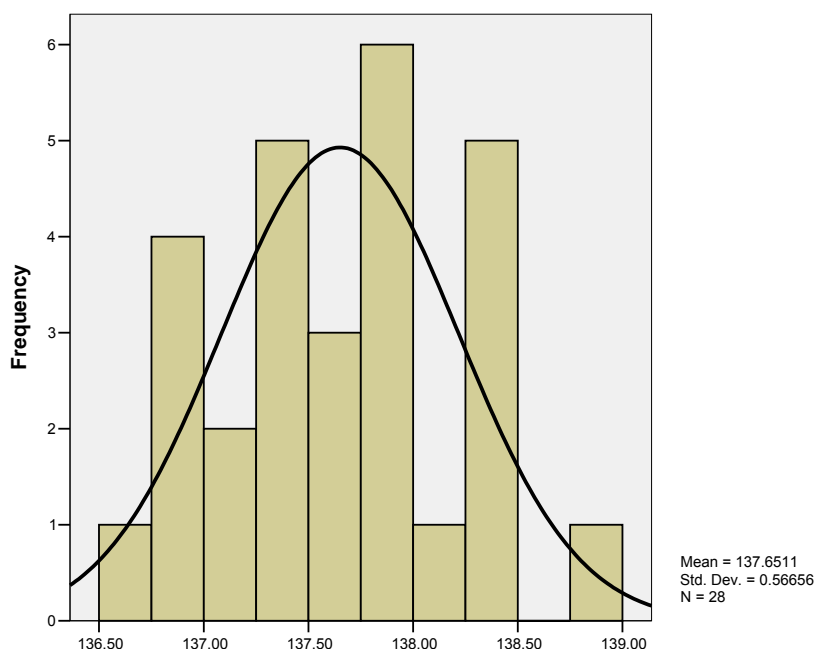
There were two laboratories which analysed the veteran's samples, Harwell and NIGL. However the instrument and procedure used by NIGL was more accurate than that used by Harwell as will be seen by my analysis of the results. A pilot normative study using volunteers from the local area was conducted by the Institute of Occupational Health in Edinburgh (an area where there might be quite high environmental Uranium due to the granite and the granite in the building materials). 28 individuals gave urine and we will now look at the results as obtained by NIGL. Table 3.1 shows the main summary statistics and Fig 3.1 shows the distribution of the isotope ratios. U-236 was also measured as this is an indicator for the presence of material that has been in a nuclear fission process. However the limits of detection of U-236 are very low and so there is a limit to how useful this measure is. In addition, we do not know that all weapons DU has U-236 in it, nor were all the vets analysed for U-236. This analysis was only available at the NIGL laboratory.

**Table 3.1** The normative study results; uran is the total concentration in urine ng/l; sigrat is the standard error of the ratio U238/U235

**Descriptive Statistics**

	N	Minimum	Maximum	Mean	Std. Deviation
Uran	28	.48	11.28	4.2804	2.99867
Ratio	28	136.72	138.96	137.6511	.56656
Sigrat	28	.42	.78	.5057	.08909
Valid N (listwise)	28				

**Fig 3. 1** The distribution of the normative study uranium ratios



In addition, we examined the shape of the normal distribution to see if it was skewed in the direction of high ratio or low ratio. The coefficient of skewness was +0.2 (SE = 0.44). That is to say, it was slightly skewed to the DU end but not significantly. It is clear that in the normative study, the mean was slightly below 137.88 and the standard deviation of the measurements made by NIGL was 0.57. Thus we can argue that a cut off of 142 for deciding that there was DU exposure was too high. However, the board also defined that in addition, the laboratory had to have no suspicion (e.g. from U-236 measurements) that there was DU present. Scientists would normally use two standard deviations from a mean to signal a statistically significant effect. For the NIGL measurements, the cut off should therefore be 139.02 for DU or 136.74 for enriched uranium (EU). There is a probability of 0.05 or 1 in 20 that a result outside these ranges could have occurred by chance. Three standard deviations from 137.88 is 139.59. There is a probability of 1 in 100 that a result above this value signalling DU exposure could have occurred by chance.

### 3.2 The NIGL and Harwell results on the veterans

Since the two laboratories had different instruments and techniques we will first deal with the final results for each laboratory. Samples were measured by the laboratories and about half the samples were measured by both laboratories as a check. In our opinion and in general there were no very large unexplained or suspicious differences between the two laboratory measurements of the same samples that could not be explained by the different instruments used and their intrinsic accuracy.

#### 3.2.1 Harwell

Table 3.2 and Table 3.3 shows statistics for the Harwell measurements whilst the distribution of the ratios is given in Fig 2.

**Table 3.2** The Harwell vets study results; uran is the total concentration in urine ng/l

#### Descriptive Statistics

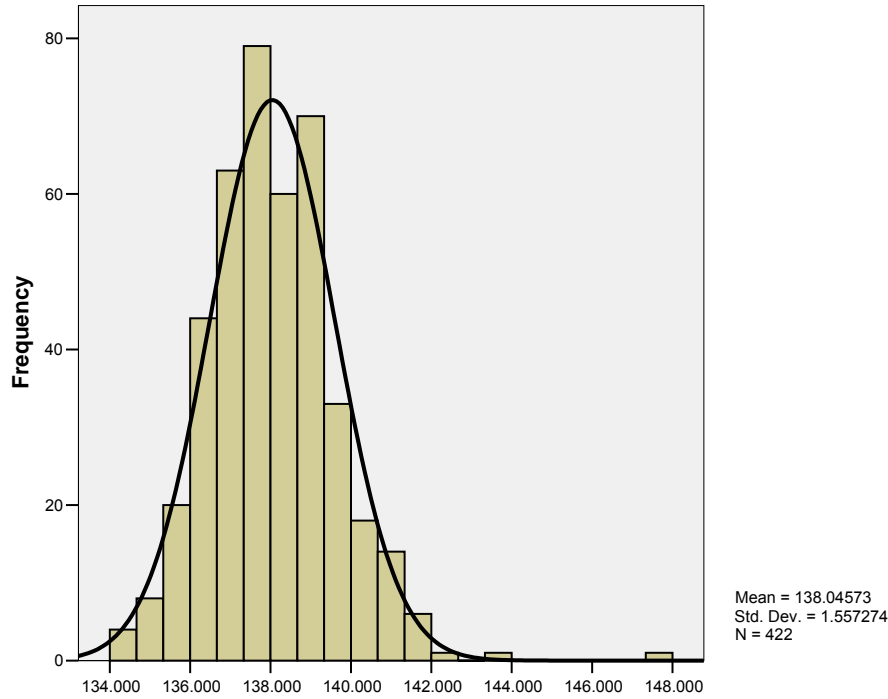
	N	Minimum	Maximum	Mean	Std. Deviation
Uran	466	.40	231.58	4.1505	12.50955
Ratio	422	134.220	147.580	138.04573	1.557274
Valid N (listwise)	422				

**Table 3.3** Descriptive Statistics for the ratio U238/U235 from Harwell results

#### Descriptives

Ratio	Mean	138.04573	.075807
	95% Confidence Interval for Mean	Lower Bound 137.89673	Upper Bound 138.19474
	5% Trimmed Mean	138.00601	
	Median	137.90500	
	Variance	2.425	
	Std. Deviation	1.557274	
	Minimum	134.220	
	Maximum	147.580	
	Range	13.360	
	Interquartile Range	2.003	
	Skewness	.730	.119
	Kurtosis	2.943	.237

**Fig 3.2** Distribution of Harwell ratio results



### 3.2.2 NIGL

Table 3.3 and Table 3.4 shows statistics for the NIGL measurements whilst the distribution of the ratios is given in Fig 3.

**Table 3.3** The NIGL vets study results; uran is the total concentration in urine ng/l

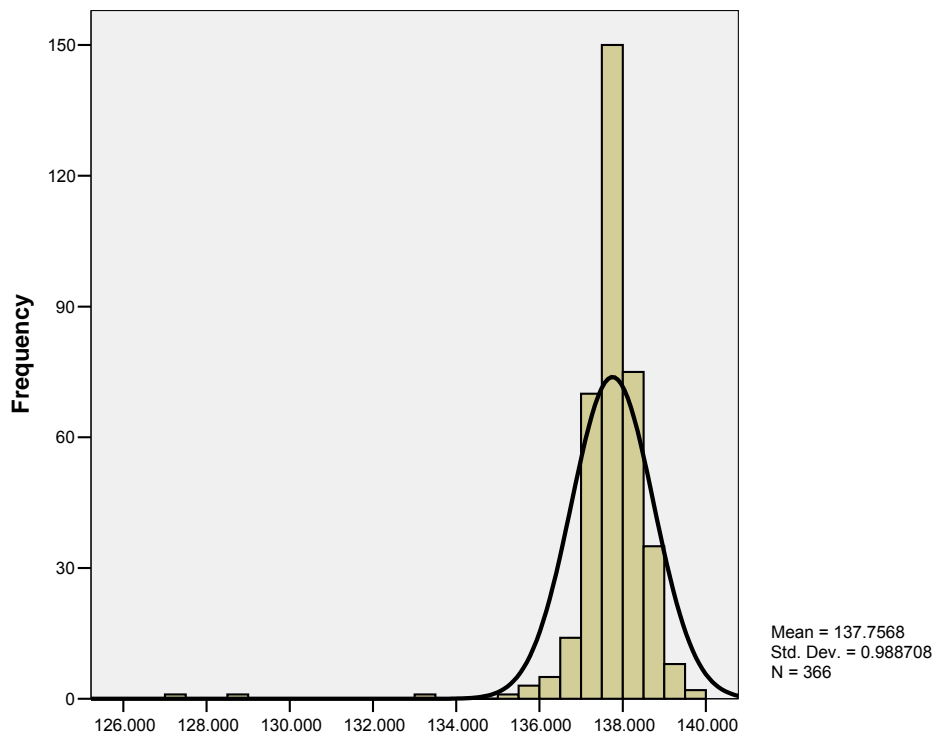
#### Descriptive Statistics

	N	Minimum	Maximum	Mean	Std. Deviation
Uran	366	.23	229.40	4.3136	13.61671
Ratio	366	127.100	139.730	137.75680	.988708
Valid N (listwise)	366				

**Table 3. 4** Descriptive Statistics for the ratio U238/U235 from NIGL results

Descriptives					
Ratio	Mean			137.75680	.051681
	95% Confidence Interval for Mean	Lower Bound		137.65517	
		Upper Bound		137.85843	
	5% Trimmed Mean			137.82342	
	Median			137.82000	
	Variance			.978	
	Std. Deviation			.988708	
	Minimum			127.100	
	Maximum			139.730	
	Range			12.630	
	Interquartile Range			.690	
	Skewness			-5.922	.128
	Kurtosis			57.524	.254

**Fig 3.3** Distribution of NIGL ratio results



### 3.3 Combining all the measurements

The combined results are given in Table 3.5 below. We have not examined them further as they are derived from two separate measurement systems. If we are to make sense of the

DUOB results we have to examine these separately. Since NIGL results were available from the background population we can compare these, and we do so in the next section.

**Table 3.5** Combined results; all measurements

Descriptive Statistics					
	N	Minimum	Maximum	Mean	Std. Deviation
Uran	834	.23	231.58	4.2140	12.98596
Ratio	790	127.100	147.580	137.91282	1.329634
Valid N (listwise)	790				

### 3.4 What do the NIGL ratio results tell us, if anything?

The normative study showed that the SD of the measurements made by NIGL in the background population in Edinburgh was 0.57 and therefore the 95% confidence range for the ratio was **136.74 to 139.02**. The 99% confidence range was 136.17 to 139.6.

Let us see how many individuals were outside the 95% uranium isotope ratio confidence interval as defined by the normative study? The result is given in Table 3.6.

**Table 3.6** Number of Veterans' results outside the 95% and 99% confidence intervals defined by two and three standard deviations as measured by NIGL in the normative study population.

Results	95% CI (136.74- 139.02)	99%CI (136.17-139.6)
Above (Depleted U)	9	2
Below (Enriched U)	18	9

These results confirm what the skew analysis of the distribution of the results from NIGL show, that there is a significant amount of enriched uranium in the veterans population. The standard deviation of the results in the vets population as measured by NIGL is much higher than the SD in the normative population. This could be because of variation in enriched uranium (EU) levels in the UK. We can look at this, since we have the data. But since the DUOB method assumes that deviation from the standard background ratio of 137.88 can be used to signal the presence of historic DU, it is clear that the presence of EU makes it impossible to use the test if the levels of DU excretion are low. We can examine the extent to which the presence of EU affects a result. We do this for some reasonable low levels of DU, EU and NU in Table 3.7. In addition one of us (CB) asked the DUOB if he could have results of the locations of the individuals to see if where they lived affected their uranium levels of isotope ratios. He has made an initial study of this which we briefly refer to here but will be published separately.

If N is concentration of normal uranium (137.88:1), E is concentration of Enriched Uranium (60:1) and D is concentration of DU (450:1), the extent to which the presence of EU as well as DU affects the isotope ratio F can be written down as follows:

$$F = A/B \text{ where}$$

$$A = N*(137.88/138.88) + E*(60/61) + D* (450/451)$$

$$B = N/(138.88) + E/ (61) + D/ (451)$$

There will generally therefore be a range of values for both DU and EU for each value of N which will cancel each other out to give a result that is in the range of 95% confidence interval uncertainty such that  $136.74 < F < 139.02$ . We have made this calculation for some

scenarios and give the results below in Table 3.7. We also calculate the initial contamination based upon the relationship given by Etherington to the DUOB which in turn relied upon the biokinetic equations and parameters employed by the Royal Society. On the basis of the fractional excretion of DU after 13 years of  $5 \times 10^{-8}$  we can say roughly that the initial contamination in milligrams is 20 times the final DU excretion in nanograms. It is clear that the decision about DU presence is dependent on total uranium and on enriched uranium.

**Table 3.7** Final measured isotope ratios for various amounts of normal, depleted and enriched Uranium excretion. Column DUOB gives the conclusion that the DUOB has made on the result based on the cut-off of 142. It is clear that we can have significant DU contamination but with EU contamination the result is considered negative. Calculation based on fractional excretion relationship given by Etherington and 13 year lag gives the initial contamination in mg.

ng Natural	ng Enr	ngDU	Allu238	Allu235	Fratio	DUOB	initial contamination mg
2	0	0.1	2.085377	0.014728	141.5967	OK	2
2	0	0.2	2.185156	0.01495	146.1661	DU	4
2	0	0.3	2.284934	0.015172	150.6017	DU	6
2	0	0.4	2.384712	0.015394	154.9092	DU	8
2	0	0.5	2.48449	0.015616	159.0942	DU	10
2	0.1	0.1	2.183738	0.016394	133.2014	OK	2
2	0.1	0.2	2.283516	0.016616	137.4248	OK	4
2	0.1	0.3	2.383295	0.016839	141.5367	OK	6
2	0.1	0.4	2.483073	0.017061	145.5415	DU	8
2	0.1	0.5	2.582851	0.017283	149.4434	DU	10
2	0.2	0.1	2.282099	0.018061	126.3556	EU	2
2	0.2	0.2	2.381877	0.018283	130.2772	EU	4
2	0.2	0.3	2.481655	0.018505	134.1046	OK	6
2	0.2	0.4	2.581433	0.018728	137.8412	OK	8
2	0.2	0.5	2.681212	0.01895	141.4902	OK	10
5	0	0.1	5.063776	0.036486	138.7882	OK	2
5	0	0.2	5.163554	0.036708	140.6662	OK	4
5	0	0.3	5.263333	0.03693	142.5215	DU	6
5	0	0.4	5.363111	0.037152	144.3547	DU	8
5	0	0.5	5.462889	0.037375	146.1661	DU	10
5	0.1	0.1	5.162137	0.038152	135.3034	OK	2
5	0.1	0.2	5.261915	0.038375	137.12	OK	4
5	0.1	0.3	5.361693	0.038597	138.9157	OK	6
5	0.1	0.4	5.461471	0.038819	140.6908	OK	8
5	0.1	0.5	5.56125	0.039041	142.4457	DU	10
5	0.2	0.1	5.260497	0.039819	132.1103	EU	2
5	0.2	0.2	5.360276	0.040041	133.869	EU	4
5	0.2	0.3	5.460054	0.040263	135.6083	OK	6
5	0.2	0.4	5.559832	0.040486	137.3285	OK	8
5	0.2	0.5	5.65961	0.040708	139.0299	OK	10
10	0	0.1	10.02777	0.072749	137.8406	OK	2
10	0	0.2	10.12755	0.072971	138.7882	OK	4
10	0	0.3	10.22733	0.073194	139.73	OK	6
10	0	0.4	10.32711	0.073416	140.6662	OK	8
10	0	0.5	10.42689	0.073638	141.5967	OK	10
10	0.1	0.1	10.12613	0.074416	136.0752	OK	2
10	0.1	0.2	10.22591	0.074638	137.0069	OK	4
10	0.1	0.3	10.32569	0.07486	137.933	OK	6

10	0.1	0.4	10.42547	0.075082	138.8537	OK	8
10	0.1	0.5	10.52525	0.075305	139.769	OK	10
10	0.2	0.1	10.22449	0.076082	134.3871	OK	2
10	0.2	0.2	10.32427	0.076305	135.3034	OK	4
10	0.2	0.3	10.42405	0.076527	136.2143	OK	6
10	0.2	0.4	10.52383	0.076749	137.12	OK	8
10	0.2	0.5	10.62361	0.076971	138.0204	OK	10
13	0	0.1	13.00617	0.094507	137.6211	OK	-2
13	0	0.2	13.10595	0.094729	138.3516	OK	4
13	0	0.3	13.20573	0.094952	139.0786	OK	6
13	0	0.4	13.30551	0.095174	139.8022	OK	8
13	0	0.5	13.40529	0.095396	140.5225	OK	10
13	0.1	0.1	13.10453	0.096174	136.2589	OK	2
13	0.1	0.2	13.20431	0.096396	136.9799	OK	4
13	0.1	0.3	13.30409	0.096618	137.6975	OK	6
13	0.1	0.4	13.40387	0.09684	138.4119	OK	8
13	0.1	0.5	13.50365	0.097063	139.123	OK	10
13	0.2	0.1	13.20289	0.09784	134.9431	OK	2
13	0.2	0.2	13.30267	0.098063	135.6548	OK	4
13	0.2	0.3	13.40245	0.098285	136.3633	OK	6
13	0.2	0.4	13.50223	0.098507	137.0686	OK	8
13	0.2	0.5	13.60201	0.098729	137.7707	OK	10

### 3.5 Conclusions on the meaning of the ratio results

Thus there are two major discoveries that tell us that it is unsafe to use the urine results to make firm statements about historic exposures. The main one is that there is enriched uranium in the population. The distribution of the results from the more accurate NIGL analyses are skewed toward enriched uranium. From Table 3.6 we see that 18 of the 366 results (5%) show statistically significant enrichment at the  $p = 0.05$  level. Table 4 shows a skewness of -5.9 i.e.towards EU which is significantly greater than the SD for the sample population. This level of EU contamination can be masked by DU contamination but the presence of EU signals fit well with the kind of scenario adopted for the calculation in table 7. We cannot know that the true level of **enrichment** (skewness) is not greater since we do not know the level of **depletion**.

The suggestion that these results were due to errors in the method at low total uranium concentration can be laid to rest. Table 3.5.1 shows the mean isotopic ratios for the samples measured by NIGL split between two groups on the basis of the mean errors ( $2\sigma$ ) tabulated by NIGL for the individual measurements.

**Table 3.5.1** Mean isotope ratios and standard deviations tabulated by NIGL for two groups of veterans based upon the mean error in the individual measurements. The cut point is  $2\sigma = 1.0$

Group	Number of samples	Mean isotope ratio (SD)
All NIGL results	365	137.7562 (.99000)
Low concentration $2\sigma > 1.0$	169	137.7697 (1.13683)
High concentration $2\sigma < 1.0$	200	137.7445 (.83624)



The second piece of evidence is that the distribution of ratios and uranium levels across the country does not appear to be a simple function of the area of residence of the individual. Thus the historic exposure in 1991 may now be affected by the background uranium intake of the individual and the absence of DU cannot be automatically inferred from the isotope ratios at the levels of DU likely to be in the veterans after 12 years have passed.

### 3.6 Background levels in the sample population

The excretion of uranium in the total veteran population is given in Table 5 as 4.2ng/l with a SD of 12.9ng/l. The laboratories agree well on these overall excretion levels. The normative results analysed by NIGL gave (Table 1) 4.28 SD = 2.99. Thus we can see that the veterans were excreting roughly the same amount of uranium as the background population but that the standard deviation in the veterans was far greater owing to a few outliers. This will become of interest when I look at the Gulf War 2 (OpTelic) veterans results.

### 4. The OpTelic results

Measurements were made by Harwell on the concentrations of uranium, and the isotope ratios in (spot samples, note much greater SD, Tab 4.1A from) the Gulf War 2 veterans. Since Uranium weapons were used in GW2 these results would be useful to see what the concentrations were in troops shortly after the conflict. The process was not overseen by DUOB although the board asked for this to be the case. One of us (CB) asked for these results, and after some discussion the committee agreed to ask the MoD for them. He obtained the result which had been filtered by the MoD for 'troops who were injured in friendly fire incidents involving DU'. The military operation was named *OpTelic*. There were measurements from Harwell for 337 individuals. The statistics of these results are given in Table 4.1a and Table 4.1b and Fig 4.1.

**Table 4.1A** OpTelic data results

#### Descriptive Statistics

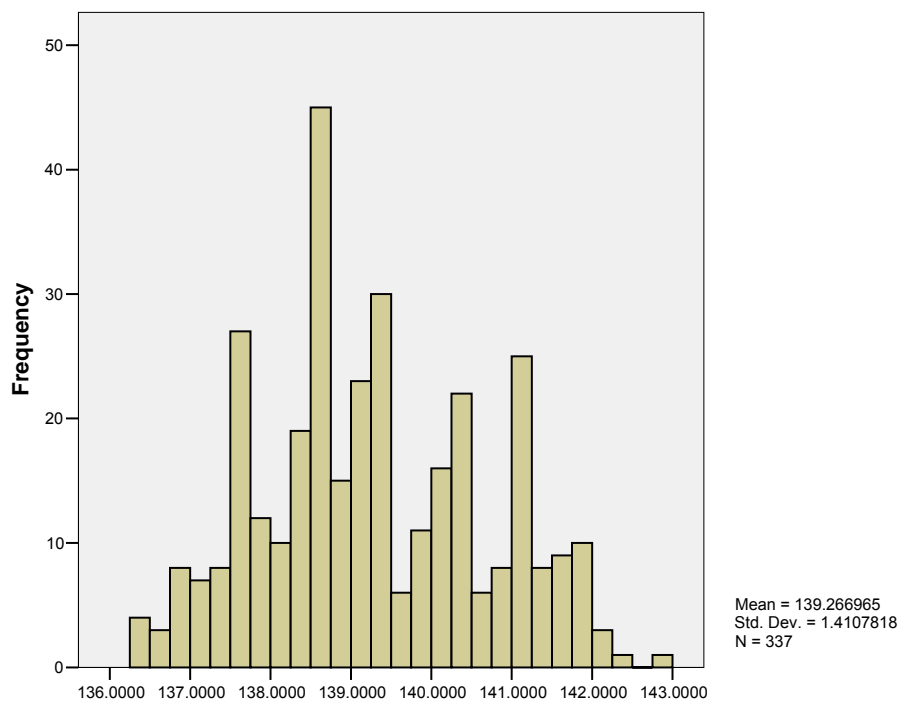
	N	Minimum	Maximum	Mean	Std. Deviation
Measratio	337	136.4256	142.8571	139.26696	1.4107818
Nglitre	339	.100000	450.00000	12.082064	41.086441290
Valid N (listwise)	337				

**Table 4.1B** OpTelic isotope ratio statistics

**Descriptives**

measrat	Mean		.007181	.0000040
	95% Confidence Interval for Mean	Lower Bound	.007173	
		Upper Bound	.007189	
	5% Trimmed Mean		.007182	
	Median		.007190	
	Variance		.000	
	Std. Deviation		.0000726	
	Minimum		.0070	
	Maximum		.0073	
	Range		.0003	
	Interquartile Range		.0001	
	Skewness		-.183	.133
	Kurtosis		-.768	.265

**Fig 4.1** OpTelic data isotope ratio distribution



It is clear from these data that the levels of Uranium in the OpTelic veterans is much higher than that in the background population. Table 4.1a gives a mean of about 12ng/l with a large Standard Deviation of 41. This can be compared with the normative background rate in the UK of around 4.2 SD 2.99. Thus the OpTelic troops are significantly contaminated with Uranium compared with the normative study individuals. They are also contaminated compared with the DUOB veterans. Using the Harwell measurements the mean levels in the GW1 veterans were much the same as the normative study levels i.e. around 4.2ng/l. And what about the isotope ratios in the OpTelic sample? It is clear from the distribution that there are two sub distributions, one of which peaks at about 141.5. We can use the Harwell measurements to look at this. If we examine the Harwell DUOB vets, the SD in the DUOB isotope ratio test is 1.55. So assuming this is a background (as we did for the total uranium) then 2 SDs is about 3. How many of the OpTelic troops are more than 3 units from 137.88, i.e. 140.88? The answer is that 57 individuals had ratios above 140.88 and therefore were 95% likely to have been contaminated with DU assuming that no enriched Uranium was present. This is about 17% of the sample, or about the same proportion that suffered Gulf War syndrome in GW1. This also assumes that only DU was used in the 2<sup>nd</sup> Gulf War which is far from certain for reasons that one of us (CB) has given elsewhere and which is covered using new data below. If we take out the Harwell outlier at 148, the number increases to about a third of the group (this equates to the proportion of USA veterans who are judged to have developed Gulf war illness, RACGWI, 2004). If we use the normative study as a background (i.e.  $F > 139.02$ ) then the number of OpTelic individuals who were exposed rises to 179. This is the right-hand sub distribution and is described in Table 4.2

**Table 4.2** Descriptive statistics of OpTelic troops with isotope ratios greater than 139.02 which is the upper 95%CI for the normative study.

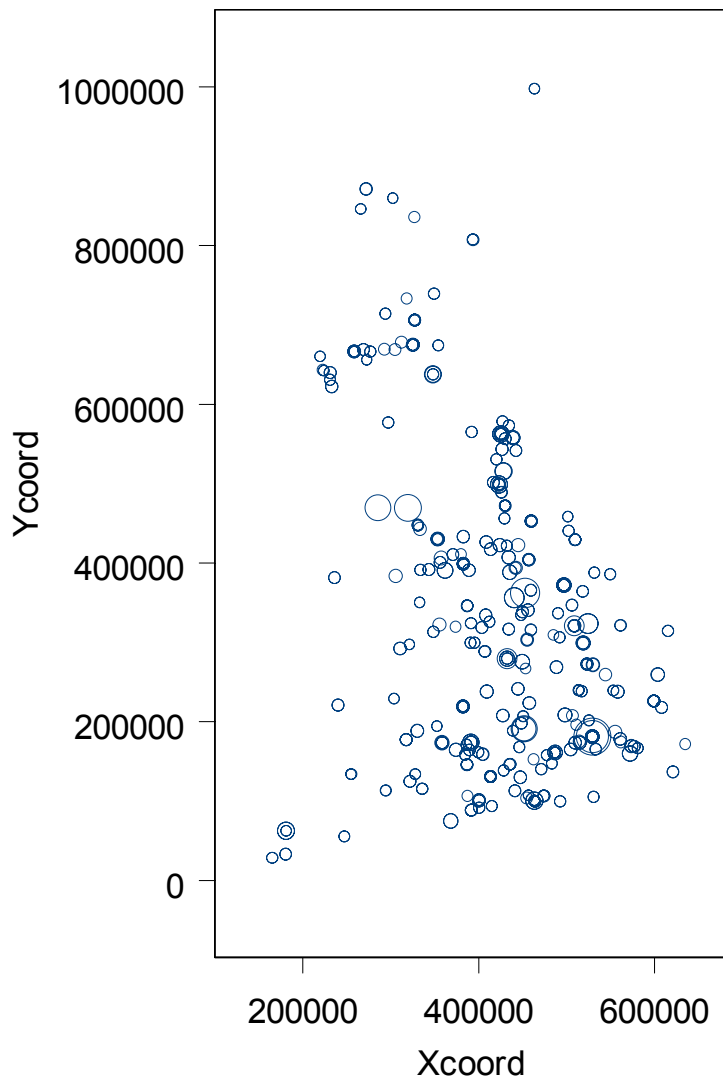
**Descriptive Statistics**

	N	Minimum	Maximum	Mean	Std. Deviation
Measratio	179	139.0821	142.8571	140.34553	.9370751
Valid N (listwise)	179				

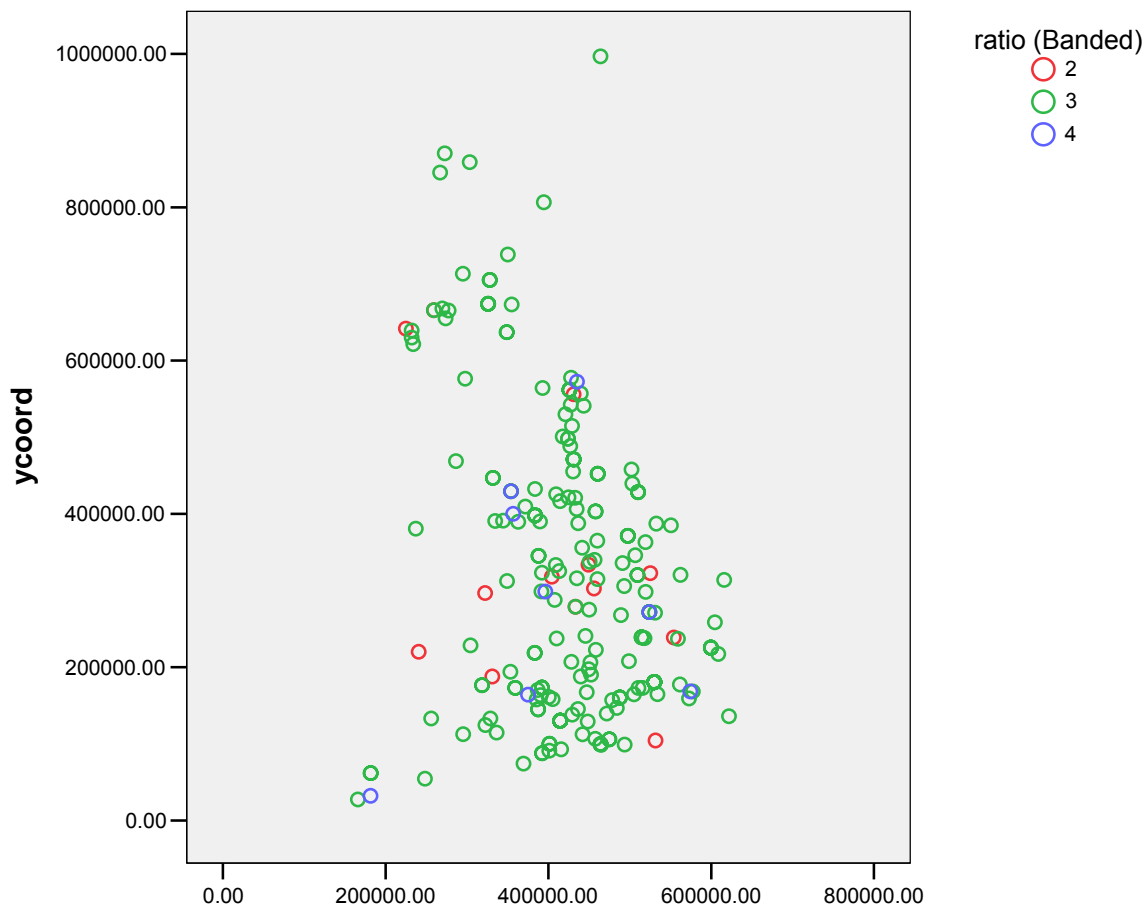
**5. The study of parameters by area of residence.**

One of us (CB) was given the towns of residence of a sample of about 400 individuals for whom the tests had been carried out. He converted the towns into coordinates using the Ordnance Survey XY metric system and examined the distribution of total uranium excretion and also the isotope ratios by area (Busby and de Messieres 2006). There was a significant variation of both total uranium and isotope ratios by area, and the statistical analyses of these data is not yet quite complete. Fig 5.1A shows the variation in concentration plotted as a bubble plot on an XY coordinate system in which X represents metres East and Y represents metres north of Ordnance Survey Origin (about 50km West of Penzance in Cornwall). Work on the area distribution of the isotope ratio results is under way and will be reported elsewhere (Busby, de Messieres and Thomas 2006). However the variation in concentration appears to show no unusual area pattern (see Fig 5.1A) with slightly high levels where they might be expected i.e. near nuclear sites e.g. Sellafield. Variation in ratio seems to be randomly distributed around the country on the basis of the NIGL results. Fig 5.1B shows locations of samples coloured according to whether they are within, (green) above (blue, DU) or below (red, EU) the 95% CI for the NIGL results for the normative population.

**Fig 5.1A** Bubble plot showing towns where samples were sent from and concentrations measured for total Uranium. Size of bubble indicates concentration. Sellafield is at P(3000,5000). (Busby, de Messieres and Thomas 2006).



**Fig 5.1B** Uranium isotope ratios in the NIGL results for individuals by town of residence in the UK plotted as Ordnance Survey metric positions. Cut points for scatter define above (blue) or below (red) the green points which represent normal uranium i.e  $136.74 < R < 139.02$  (Busby, de Messieres and Thomas 2006).



## 6. The ICRP biokinetic model and its data as applied to the DUOB results.

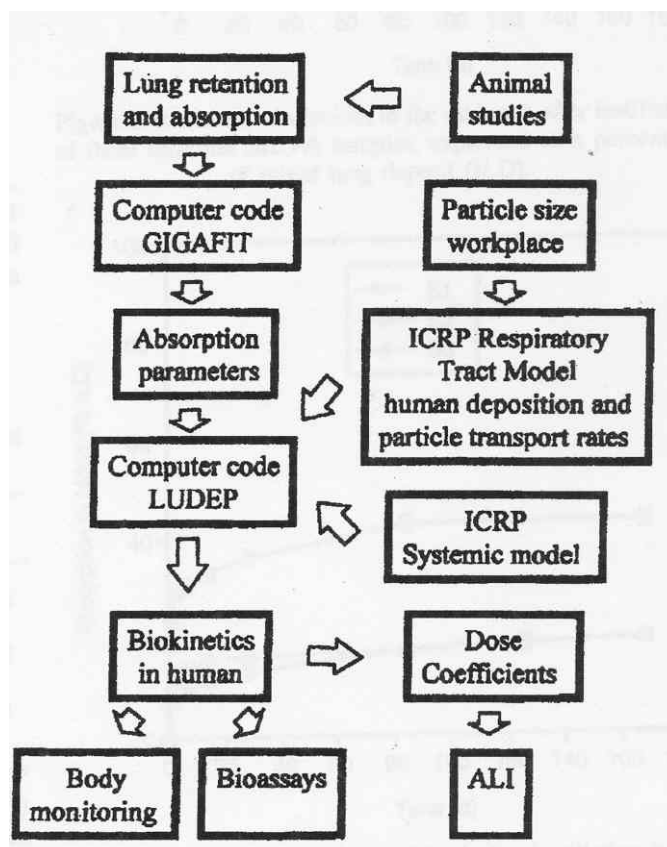
### 6.1 Limits to detection of DU imposed by the model employed.

It has already been argued elsewhere that the ICRP biokinetic model is unsafe (Busby 2001, IRSN 2006) However, it seems that even if we employ it, the question if whether we would be likely to find anything some 12 years after an exposure remains open. The model was applied by G Etherington and BG Spratt to calculating the extent to which exposures to DU in GWI might still produce urinary levels of DU which might be picked up by the testing some 14 years after the event. These calculations were based on parameters presented from research data employed by the Royal Society committee and presented in their reports (RS 2001, 2002). Calculations were also made on the basis that there might be uncertainties as high as a factor of 10 in the model. Under those circumstances Etherington and Spratt calculate that the ingestion or inhalation associated with a calculated mean committed effective dose of 1mSv was 0.2ng/day if the interval was 12.5 years between exposure and measurement. 0.2ng of DU represents an ability to accurately measure  $4.5 \times 10^{-4}$  ng of U-235.

## 7.2 Limits to detection imposed by errors in the parameters used in the model

A representation of the ICRP model is shown in Fig 7.2.1 below, taken from Ansoborlo et al, 1998 the study on which the DUOB uranium biokinetic considerations were based.

**Fig 7.2.1** Human respiratory tract model (from Ansoborlo et al, 1998)



Each stage of this complex partition model requires data and assumptions about the rate of movement of uranium between the various components. We argue that to expect a useful answer to the question of urine excretion after 12 years following a small contamination whilst employing rat data obtained over 6 months from exposures involving a different type and particle size of uranium is bordering on religious faith and has little to do with science.

The ICRP model employed clearance from the body on the basis of three types of solubility for the Uranium ingested and inhaled. The parameters for solubility and transport for the slowest dissolving of these were based on measurements on animals of the excretion of a type of Uranium oxide and chemical identity ( $U_3O_8$ ) which is not the same as the oxide particles produced in battlefield conditions when ceramic  $UO_2+U_3O_8$  is formed owing to the oxygen limitations imposed by the explosion. The model is therefore unsafe. DU ceramic particles may in fact be totally insoluble *in vivo* since they may become encysted in a layer of cells which they continuously kill by alpha decay or photoelectron emission. Other substances in the body are totally insoluble (e.g. asbestos fibres) and there is yet no evidence that all Uranium from DU use has some finite level of solubility as is assumed by the HRPT model and its parameters. We know of no mass balance studies that indicate all the absorbed uranium is finally excreted.

## 8. Other considerations

There is no doubt that the GW1 and GW2 (OpTelic) veterans inhaled DU and or Uranium particles. This is because we know from other studies that large amounts of Uranium were used in both conflicts. There were about 370 tons used in GW1 and perhaps as much as 1800 tons in GW2, if we assume the use of uranium in bunker busting bombs and guided bombs and missiles (see below). DU was also employed in the Balkans.

We know also from many studies that the smaller particles are extremely mobile and may even travel over global distances (e.g. Busby and Morgan 2005). Therefore to argue from the measurements that have been made some 14 years after the exposure that there was no exposure seems to me to be absurd. One of us (CB) has personally found DU in both Iraq and Kosovo and has seen measurements of urinary Uranium made by Japanese scientists in Iraq. The OpTelic data show that those troops were contaminated. If we employ the equations of the HRTM for the period of 100 days (which we assume was the approximate lag in their cases) then the difference between their mean excretion and the mean excretion in the UK was about 9ng/l or 18ng/day assuming that the contamination was from natural Uranium. Application of the HRTM relationship gives a central estimate for contamination in Iraq at  $10^{-5}$  of their initial absorption. This would put their initial contamination at a mean value of  $18\text{ng} * 10^5 = 0.9 \text{ mg}$ . There will be many who have received significantly greater contamination.

There have been arguments that the US may have begun using natural Uranium in their large weapons. This followed from the finding of high levels of normal Uranium in Afghan individuals by Durakovic's team. Whilst this may have seemed highly speculative at the time, one of us (CB) has recently reported elsewhere measurements of Uranium isotopes in soil from a crater caused by a US supplied guided bomb used by the Israeli army in Khiam Southern Lebanon. These measurements were made by Harwell and showed that Enriched Uranium with an isotope ratio of 108 was used in this weapon (Busby and Williams 2006a, 2006b, 2007).

## 9. Health effects of exposure to Uranium from weapons.

At the beginning of the 5 year period of the DUOB and over the period of the Royal Society committee investigations we were almost alone in arguing that DU particles could not be safely modelled using the IRCP radiological system. However, the Royal Society and the DUOB have persisted in employing this model even though the evidence that it is wildly incorrect has continuously accrued since 2001. The model, with its dependence on the average quantity 'absorbed dose' is patently incorrect when applied to anisotropic exposures from hot particles or uranium oxide particles. It has not been difficult to demonstrate this. Despite this, all the official agencies of the UN and the national risk agencies continue to blindly rely on a model that is bankrupt. The evidence comes from many sources, from *in vitro* cell biology, from theoretical analyses and from epidemiology, from sick Gulf veterans and recent troops who look to the DUOB and the Royal Society and official organisations like WHO to protect them and not least from the dead and dying in the countries which have been contaminated. Our position and that of Baverstock and others has been well discussed in the course of the meetings of the DUOB. We will not review this area here as it would take up too much space. We have placed some articles in the reference section.

## 10. Other analytical methods

Given that the uranium measurements were to form the basis of an epidemiological investigation clearly it is impossible to use them in this way, for all the reasons given above. Under these circumstances we suggest that a way forward would be to develop a chromosome aberration analysis, (Schroder et al, 2003, Rossner et al, 2005), and use it to examine veterans who believe they have been contaminated. In addition, we would propose that bone biopsy analyses and autopsy sample analyses be organised to see if the biokinetic parameters employed are indeed correct. In particular we should examine uranium particles in the tracheobronchial lymph nodes and other tissues of autopsy specimens.

## 11. Conclusions and recommendations

1. The measurement methods and techniques were accurate
2. The measurement system was unbiased and the results represented the true measurements of uranium in urine
3. The biokinetic model employed to interpret the results was unsafe owing to uncertainties in both the input parameters which were obtained from a different type of exposure, animal and period and also the assumptions underlying the model itself.
4. The interpretation algorithm (decision process about whether a veteran had been exposed based upon the urine isotope ratio result) of the DUOB was flawed first because it used too wide a confidence interval and second because it also assumed that there was no Enriched Uranium in the UK population. For this latter reason the true levels of DU contamination could not be determined from an isotope ratio.
5. For these reasons, we feel that it is important that no assumptions about veterans' health and exposure to depleted uranium should be made on the basis of the DUOB results. Epidemiological studies should be carried out on the basis of other appropriate covariates e.g. biophysical markers like chromosome tests.
6. There are variations in uranium isotope ratios across the UK for reasons which are not clear at present
7. The OpTelic Gulf War 2 veterans have been contaminated with Uranium and Depleted Uranium, contrary to the statements of the MoD. They should be followed up with regard to health effects and exposure, and this must be done independently through some committee like the DUOB with independent members and over many years at least 20 to include slow growing cancers and other chronic conditions.
8. The health effects of battlefield Uranium exposure are increasingly seen to be serious and are not predicted or explained by the present ICRP model used by the Royal Society and other risk analyses employed by the MoD.
9. The dispersion of battlefield uranium particles is global and uranium weapons therefore have indiscriminate effects on non combatants.
10. Since it is now seen that non-depleted uranium is employed in weapons, isotope ratios can no longer be seen as safe and other methods will have to be developed to determine historic exposure. The use of enriched uranium in US supplied weapons in Lebanon calls for an investigation into the use of such weapons in Afghanistan and Iraq and begs for an inquiry into the types of bomb and guided missile that are involved and also a re-examination of the OpTelic 2<sup>nd</sup> Gulf war conclusions.
11. The only way we can now use all the mass of data which has been obtained at great cost is to examine the health effects of the GW1 veterans reported in the questionnaires and carry out a statistical analysis of these veterans by uranium isotope ratio and ill health category. Despite the argument that they may be a biased sample we can see if, within that biased sample, there is a relationship between uranium data results and ill health. This might not show anything, but it would be wrong and wasteful not to look.
12. We believe that there is a moral, medical and scientific imperative to monitor the health of people in Iraq, especially children.
13. Oppositional committees are the best way we have of obtaining as close to the truth as it is possible to get in complex scientific areas.



## References

- Ansoborlo E, Hodgson A, Stradling GN, Hodgson S, Metiviert H, Henge-Napoli MH, Jarvis NS, Birchall A (1998) Exposure implications for uranium aerosols formed at a new laser enrichment facility: application of the ICRP respiratory tract and systemic model. *Radiation Protection Dosimetry* Vol 79 Nos 1-4 p 23-27
- Atomic Energy Authority Technology. Kuwait-Depleted Uranium Contamination, April 1991. This report was only uncovered through a Parliamentary answer to a question from the Countess of Mar on 2<sup>nd</sup> March 1998.
- Baverstock K., Mothersill C and Thorne M. Radiological Toxicity of DU. WHO Document 5 Nov 2001.
- Baverstock K. Presentation to European Parliament 23<sup>rd</sup> June 2005.
- Baverstock, K., Science, politics and ethics in the low dose debate. *Medicine, Conflict and Survival*, 2005. 21: p. 88 - 100.
- Bertell R (1999) in Metal of Dishonor, International Action Center, ISBN: 0-9656916-0-8.
- Busby (1995) *Wings of Death. Nuclear Pollution and Human Health* Aberystwyth: Green Audit
- Busby C (2001RP) *Health risks following exposure to aerosols produced by the use of Depleted Uranium weapons. Presentation to Res publica International Conference, Prague Nov 2001.* Occasional Paper 2001/12 (Aberystwyth: Green Audit)
- Busby C (2001RS) *Science on Trial: On the biological effects and health risks following exposure to Depleted Uranium weapons. Invited presentation to the Royal Society London July 19th 2000 and also given at the International Conference on DU in Manchester Nov.4th 2000.* Occasional Paper 2000/11 (Aberystwyth: Green Audit)
- Busby C (2001UN) *Depleted Uranium in Kosovo: Review of the UNEP Report of 13 Mar 2001* Occasional Paper 2001/3 (Aberystwyth: Green Audit).
- Busby C (2002) *Lymphoma incidence in Italian Military Personnel involved in operations in Bosnia and Kosovo* Occasional Paper 2002/2 (Aberystwyth: Green Audit)
- Busby C (2002BN) *High risks at low doses.* Proceedings of the British Nuclear Energy Society International Conference on risks from low doses of ionising radiation, Oxford September 2002 (London :BNES)
- Busby C (2002CC) *The Health effects of depleted Uranium weapons. Written Evidence to the US Congressional Subcommittee on National Security, Veterans Affairs and International Relations Hearing, London 18 June 2002.* Occasional Paper 2002/3 (Aberystwyth: Green Audit)
- Busby C (2002HO) Review of the Home Office statement on the health consequences of DU in Kosovo. Occasional paper 2002/2 (Aberystwyth: Green Audit).
- Busby C (2005) Depleted Uranium Weapons and Radiation Dose. *European Journal of Biology and Bioelectromagnetics* Vol 1 No 1 p82-93
- Busby C (2005) Does Uranium contamination amplify natural background radiation dose to DNA? *European Journal of Biology and Bioelectromagnetics* Vol 1 No2 p120-131
- Busby C, Scott Cato M, (2000) 'Increases in leukaemia in infants in Wales and Scotland following Chernobyl: evidence for errors in risk estimates' *Energy and Environment* 11(2) 127-139
- Busby C and Fucic A (2006) Ionizing Radiation and Children's Health: Conclusions of the PINCHE Project *Acta Paediatrica* 95 S453 81
- Busby C and Morgan S (2006) Did the use of Uranium weapons in Gulf War 2 result in contamination of Europe? Evidence from the measurements of the Atomic Weapons Establishment, Aldermaston, Berkshire, UK. *European Journal of Biology and Bioelectromagnetics* Vol 1 No 5 p650-668
- Busby C and Williams D (2006) Evidence of Enriched Uranium in guided weapons employed by the Israeli Military in Lebanon in July 2006 Preliminary Note. Occasional paper 2006/6 Aberystwyth: Green Audit.
- Busby C, 2003 *Depleted Science: Health consequences and mechanisms of exposure to fallout from Depleted Uranium Weapons. Contribution to International Conference Hamburg Oct*

16th-19th 2003 p28 in *The Trojan Horses of Nuclear War* ed. Kuepker Marion and Kraft Dave (Hamburg: GAAA)

- Coryell V and Stearns D (2006) Molecular analysis of hprt mutations generated in chinese hamster ovary EM9 cells by uranyl acetate, by hydrogen peroxide and spontaneously. *Molecular Carcinogenesis* 45: 60-72
- Craft E.S, Abu Quare A, Flaherty MM, Garofolo MC, Rincavage HL and Abouu Donia MB (2004) Depleted and Natural Uranium: Chemistry and Toxicological Effects *J Toxicol and Env. Health Part B* 7: 297-317
- De Sutter E. Too many babies without eyes. *Dutch J Med Sci* 2001, **145**, 1024.
- Duncan K. Pensions Appeal Tribunals (Scotland). 2<sup>nd</sup> Feb.2004.
- Durakovic A, Horan P, Dietz L. Quantitative Analysis of Depleted Uranium Isotopes in British, Canadian, and U.S. Gulf War Veterans. *Military Medicine* 2002;167:620-7.
- Durakovic A. Medical effects of internal contamination with uranium. *Croatian Med J* 1999, **40**, 49-66.
- Durakovic A. On Depleted Uranium: Gulf War and Balkan Syndrome. *Croatian Medical Journal* 2001, **42**, 130-4.
- ECRR2003 (2003) 2003 recommendations of the European Committee on Radiation Risk. The health effects of ionising radiation exposure at low doses for radiation protection purposes. Regulators Edition ed-Chris Busby, Rosalie Bertell, Inge Schmitz-Feuerhake, Alexey Yablokov (Brussels: ECRR)
- Fahey D. Case Narrative: Depleted Uranium (DU) Exposures, 1998. Available at National Gulf War Resource Center, Inc. 1224 M St, NW Washington, DC 20005, USA. <http://www.gulfweb.org/ngwrc> This is a comprehensive documentation of the sources of the information on DU. It is particularly useful for its references to Military studies which have not been reported in the published literature.
- Fahey D. SCIENCE OR SCIENCE FICTION? *Facts, Myths and Propaganda In the Debate Over Depleted Uranium Weapons* March 12, 2003 download [www.antenna.nl/wise/uranium/pdf/dumyths](http://www.antenna.nl/wise/uranium/pdf/dumyths)
- Glissmeyer JA and Mishima J (1979) Characterisation of airborne uranium from test firing of XM774 ammunition. Pacific Northwest Laboratory, Richland, Washington 99352; US Army Document PNL-2944
- Glissmeyer JA, Mishima J, Bamberger JA (1985) Prototype Firing Range Air Cleaning System *Proceedings of the 18<sup>th</sup> DOE Nuclear Airborne Waste Management and Air Cleaning Conference, Baltimore Maryland 12-16 Aug 1984*. Ed- First, Melvin CONF 840806
- Hei T K, Wu L-J, Liu S-X, Vannais D, Waldren C A and Randers-Pehrson G. Mutagenic effects of a single and an exact number of alpha particles in mammalian cells. *Proc. Natl. Acad. Sci. USA*.1997;94:3765 -70.
- Hoffmann W and Schmitz-Feuerhake I (1999) 'How radiation specific is the discentric assay?' *Journal of exposure analysis and Environmental Epidemiology* 2, 113-133
- Italian Report, (2001) *Seconda Relazione Della Commissione Istituita Dal Ministro Della Difesa Sull' Incidenza di Neoplasie Maligne tra I Militari impiegati in Bosnia 28 Maggio 2001* Rome: Ministry of Defence
- Iyer R and Lehnert BE. Radiation – Induced Effects in Unirradiated Cells. *Science and Medicine*. 2000; Jan/Feb 54 – 63.
- Lorimore S A, Coates P J and Wright E G. Radiation–induced genomic instability and bystander effects: inter – related nontargeted effects of exposure to ionizing radiation. *Oncogene*.2003;22: 7058–69.
- McDiarmid MA, Hooper FJ, Squibb K, McPhaul K. The utility of spot collection for urinary uranium determination in depleted uranium exposed Gulf War veterans. *Health Phys* 1999, **77**, 261-264.
- McDiarmid MA, Keogh JP, Hooper FJ, McPhaul K, Squibb K, Kane R, DiPino R, Kabat M, Kaup B, Anderson L, Hoover D, Brown L, Hamilton M, Jacobson-Kram D, Burrows B, Walsh M. Health effects of depleted uranium on exposed Gulf War veterans. *Environmental Research* 2000, **82**, 168-180.

- Miller A C, Stewart M, Brooks K, Shi L and Page N. Depleted uranium–catalyzed DNA damage: absence of significant alpha particle decay. *J. Inorg. Biochem* . 2002;91:246–252.
- Miller AC, Brooks K, Stewart M, Anderson B, Shi Lin, McLain D and Page N (2003) Genomic Instability in human osteoblast cells after exposure to depleted uranium: delayed lethality and micronuclei formation. *J. Env. Radioact.* 64 247-259
- Miller, A.C., et al., Effect of the militarily-relevant heavy metals, depleted uranium and heavy metal tungsten-alloy on gene expression in human liver carcinoma cells (HepG2). *Mol Cell Biochem*, 2004. 255(1-2): p. 247-56.
- Miller, A.C., et al., Genomic instability in human osteoblast cells after exposure to depleted uranium: delayed lethality and micronuclei formation. *J Environ Radioact*, 2003. 64(2-3): p. 247-59.
- Miller, A.C., et al., Potential late health effects of depleted uranium and tungsten used in armor-piercing munitions: comparison of neoplastic transformation and genotoxicity with the known carcinogen nickel. *Mil Med*, 2002. 167(2 Suppl): p. 120-2.
- MOD 1991. Declassified Signal R 252240Z Feb 91 Precautionary measures[with regard to DU.
- MOD 1993. Letter to Mr McGinley, 2nd Aug 1993. "...we are aware of the hazards of depleted uranium."
- Monleau M, de Meo M, Paquet F, Chazel V, Dumenil G and Donnadiou-Claraz M (2006) Genotoxic and inflammatory effects of Depleted Uranium particles inhaled by rats. *Toxicological Sciences* 89 (1) 287-95
- Nagasawa H, Little JB. Induction of sister chromatid exchanges by extremely low doses of alpha-particles. *Cancer Research* 1992;52:6394-6396.
- political action, but such a resolution appears distant by years if not decades."
- Risk of Cancer. *Environ Health Perspect* 2005;113:517–520.
- Schroder H, Heimers A, Frenzel Beyme R, Schott A and Hoffmann W (2003) 'Chromosome aberration analysis in peripheral lymphocytes of Gulf War and Balkan War veterans.' *Rad. Prot.Dosim.* 103(3) 211-219
- UMRC Uranium Medical Research Centre download 28-1-2007, <http://www.umrc.net/riordon.aspx>
- UNEP (2001) 'Depleted Uranium in Kosovo. Post Conflict Environmental Assessment. Geneva: UNEP (These UNEP environmental reports are available in a modified form on the internet. The Annex tables which are no longer available, are available from Green Audit.)
- Van der Hazel P, Zuurbier M, Bistrup ML, Busby C, Fucic A (2006) Policy and Science in children's health and environment: recommendations from the PINCHE project. *Acta Paediatrica* 95 S453 114
- Watt RS and Norton-Taylor, 2001 R. Troops not told of shells' toxic risk. *Guardian* Feb 8th 2001. Includes admission that in Iraq, 1991, Kosovo, 1999, troops were not told of hazards of DU.
- Wu L-J, Randers-Pehrson G, Xu A, Waldren C A et al. Targeted cytoplasmic irradiation with alpha particles induces mutation in mammalian cells. *Proc. Natl. Acad Sci USA.* 1999;96:4959–4964.
- Yacoub A, Al-Sadoom I, Hasan J. The evidence for causal association between exposure to depleted uranium and malignancies among children in Basrah by applying epidemiological criteria of causality, in Conference Proceedings, The Effects of the Use of Depleted Uranium, Baghdad, 2002, p. 87-97.