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ORIGINAL ARTICLE

Urinary isotopic analysis in the UK Armed Forces: no evidence of depleted uranium absorption in combat and other personnel in Iraq

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Objectives: To assess the distribution and risk factors of depleted uranium uptake in military personnel who had taken part in the invasion of Iraq in 2003.

Methods: Sector field inductively coupled plasma-mass spectrometry (SF-ICP-MS) was used to determine the uranium concentration and $^{238}\text{U}/^{235}\text{U}$ isotopic ratio in spot urine samples. The authors collected urine samples from four groups identified a priori as having different potential for exposure to depleted uranium. These groups were: combat personnel (n = 199); non-combat personnel (n = 96); medical personnel (n = 22); and "clean-up" personnel (n = 24) who had been involved in the maintenance, repair or clearance of potentially contaminated vehicles in Iraq. A short questionnaire was used to ascertain individual experience of circumstances in which depleted uranium exposure might have occurred.

Results: There was no statistically significant difference in the $^{238}\text{U}/^{235}\text{U}$ ratio between groups. Mean ratios by group varied from 138.0 (95% CI 137.3 to 138.7) for clean-up personnel to 138.2 (95% CI 138.0 to 138.5) for combat personnel, and were close to the ratio of 137.9 for natural uranium. The two highest individual ratios (146.9 and 147.7) were retested using more accurate, multiple collector inductively coupled plasma-mass spectrometry (MC-ICP-MS) and found to be within measurement of error of that for natural uranium. There were no significant differences in isotope ratio between participants according to self-reported circumstances of potential depleted uranium exposure.

Conclusions: Based on measurements using a SF-ICP-MS apparatus, this study provides reassurance following concern for potential widespread depleted uranium uptake in the UK military. The rare occurrence of elevated ratios may reflect the limits of accuracy of the SF-ICP-MS apparatus and not a real increase from the natural proportions of the isotopes. Any uptake of depleted uranium among participants in this study sample would be very unlikely to have any implications for health.

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Depleted uranium is a by-product from the manufacture of enriched uranium. Uranium as it occurs naturally in the earth's crust comprises two main isotopes, ^{238}U and ^{235}U , in a ratio of approximately 137.9:1. Enriched uranium contains a higher proportion of the more radioactive ^{235}U , and is used as a nuclear fuel. Conversely, the depleted uranium that remains after extraction of enriched uranium from natural uranium contains proportionately less ^{235}U with a $^{238}\text{U}/^{235}\text{U}$ ratio in the order of 490:1.¹ It is a weakly radioactive alpha emitter (40% less radioactive than natural uranium), but has a high density (approximately 70% greater than lead), which has been exploited in specialist engineering applications. It has also been used for military purposes, in particular in armour-piercing rounds. The first major use of depleted uranium weapons was by US and UK forces in the 1991 Gulf War, and they were subsequently used also in the Balkans campaign and in the 2003 Iraq War.

When a depleted uranium round strikes an armoured target, it undergoes spontaneous combustion, a substantial proportion being converted to a fine aerosol of largely (>65%) insoluble uranium oxides. Exposure to this material, either through inhalation or through ingestion of contaminated food or water, could pose a hazard from its radioactivity or chemical toxicity. Thus, while depleted uranium weapons are effective in battle, there is an urgent need to establish whether their use carries important long-term risks to health.

As with any toxic material, risk will depend on the route and level of exposure. Estimates can be made of the radioactive dose associated with a given uptake of uranium oxides, and also of the maximum kidney concentration and resultant risk of renal damage from chemical toxicity (the kidney is the most sensitive target organ). Thus, knowledge of the uptakes of depleted uranium as a consequence of its military use would enable assessment of any resultant long-term risks to health.

Uptake of depleted uranium can be estimated retrospectively by measurement of uranium isotopes in urine. At a rate depending on its chemical speciation, the depleted uranium that is taken up by the body will be dissolved in tissue fluids, redistributed, and excreted via the kidney. Within the urine, it will be mixed with natural uranium arising from normal dietary exposures, and this will manifest as an elevation of the $^{238}\text{U}/^{235}\text{U}$ ratio above the value of 137.9 for natural uranium. Based on empirical data, a biokinetic model has been developed which predicts the level of depleted uranium excretion in the urine at a given interval after exposure to a specified inhaled dose of uranium oxides similar in chemical composition to that produced when a depleted uranium round impacts on an armoured target.²

Abbreviations: MC-ICP-MS, multiple collector inductively coupled plasma mass spectrometry; SF-ICP-MS, sector field inductively coupled plasma mass spectrometry

Such exposure may occur in military units that use depleted uranium munitions, such as armoured brigades, or those tasked with "cleaning up tanks" that have been attacked with depleted uranium munitions. The Royal Society (2001) report on depleted uranium munitions distinguished three levels of exposure that might occur in battlefield scenarios.³ The highest exposures (through inhalation of depleted uranium aerosols and injury by shrapnel fragments) would be expected in combat personnel who were in a vehicle when it was struck by a depleted uranium round or entered it immediately afterwards (level 1). Level 2 exposures (by inhalation or ingestion following hand-to-mouth transfer) would occur in personnel working in or on contaminated vehicles after combat, and the lowest exposures (level 3) would occur in other circumstances (for example, through dispersal of depleted uranium oxides downwind of fires). In theory, medical personnel might sustain cross-contamination from injured personnel, vehicles or ground when providing aid to casualties of depleted uranium munitions.

In this paper we report the urinary uranium excretion of members of the UK Armed Forces who took part in the 2003 Iraq War, in which depleted uranium munitions were used. Our study focussed on specific groups identified as being at particular risk of exposure, including a group with potential for level 2 exposures, together with a low-risk control group. All participants were selected from a large epidemiological study of the war in Iraq on the health of the UK Armed Forces.⁴

The main aim of the present study was to assess the distribution and risk factors of any uranium exposures resulting from the use of depleted uranium weapons, using isotopic analysis of urine as the main index of exposure.

METHODS

Study design and participants

The sample for the present study was obtained from a cohort study that monitored the physical and psychological outcomes for UK service personnel who served in the recent Iraq War.⁴ The cohort study⁴ investigated approximately 17 500 Royal Navy, British Army and Royal Air Force personnel, approximately 40% of whom deployed in the Iraq War of 2003. The UK Ministry of Defence's Defence Analytical Services Agency (DASA) identified participants and generated a list of personnel excluding Special Forces. We selected subjects from those that deployed during the major combat phase between 18 January 2003 and 28 June 2003, also known as Operation TELIC 1. From this initial sampling frame, we selected three groups for the

present study, with differing potential for depleted uranium exposure in battlefield scenarios as delineated in the Royal Society report.³ These groups were: combat personnel; non-combat personnel; and a group referred to as "clean-up" which included personnel identified by DASA as having taken part in the maintenance, repair or clearance of potentially contaminated vehicles in theatre. In addition, we also studied medical personnel in view of their potential for exposure to depleted uranium when treating casualties whose clothes may have been contaminated by uranium oxides. The non-combat group served as the control group.

The study received ethical approval from the Ministry of Defence (Navy) personnel research ethics committee and the King's College Hospital local research ethics committee.

Data collection

Urine sample collection was conducted during visits to military establishments within the UK and Germany as part of the data collection for the main study.⁴ Visits were assigned on the basis of a high concentration of TELIC-deployed personnel, within the main sample, by location. We sought volunteers in each of the four pre-identified groups to take part in the depleted uranium study until further visits to increase group sizes were deemed impractical. We provided a short verbal brief about the depleted uranium study, and obtained written consent (separate from that for the main study) from those who responded. Spot urine samples were collected in 100 ml square-form "Kartell" containers, and were stored at 4°C before transport to the analysing laboratory. Recruitment and collection of urine samples was completed between August 2004 and July 2005.

A supplementary self-administered questionnaire was used to record circumstances of possible exposure to depleted uranium in four battlefield scenarios covering all recent deployments (in addition to Iraq in 2003) by UK Armed Forces where depleted uranium weapons were employed (1991 Gulf War, Bosnia 1994–5, Kosovo 1999 and Afghanistan 2001).

Urinalysis

Analysis was by sector field inductively coupled plasma mass spectrometry (SF-ICP-MS), and results were presented as total uranium concentrations and values for the ²³⁸U/²³⁵U isotope ratio. Details of the analytical method and its sensitivity and validity have been published elsewhere.⁵ Pilot work had indicated that the assay was capable of detecting an elevation of the ²³⁸U/²³⁵U ratio above 144 in urine samples containing less than 5 ng/l.⁵

Table 1 The frequencies (percentages) of participants by sociodemographic background

	Combat (n = 199)	Clean-up (n = 24)	Medical (n = 22)	Non-combat (n = 96)
Service				
Army	183 (92%)	24 (100%)	21 (95%)	77 (80%)
Royal Marines	16 (8%)	0	0	19 (20%)
Royal Navy	0	0	1 (5%)	0
Rank				
Commissioned officers	8 (4%)	0	3 (14%)	6 (6%)
Ranks	191 (96%)	24 (100%)	19 (86%)	90 (94%)
Sex				
Male	196 (98%)	22 (92%)	15 (68%)	91 (95%)
Female	3† (2%)	2 (8%)	7 (32%)	5 (5%)
Age years, median (range)	28 (20–51)	31 (24–40)	28 (21–44)	30 (20–44)
Education*				
1	18 (10%)	0	0	13 (15%)
2	100 (54%)	8 (36%)	12 (55%)	47 (54%)
3	68 (36%)	14 (64%)	10 (45%)	27 (31%)

Denominators for education and age vary because some participants did not complete relevant questionnaires.

*Highest level of education achieved: 1, left school with no qualifications; 2, GCSE or equivalent; 3, A level or equivalent, degree or above.

†Defined as combat personnel by unit at the time of deployment in TELIC-1.

Table 2 Uranium concentrations and isotope ratios by category of personnel

Group	Combat (n=199)	Clean-up (n=24)	Medical (n=22)	Non-combat (n=96)
Mean $^{238}\text{U}/^{235}\text{U}$ ratio (95% CI)	138.2 (138.0 to 138.5)	138.0 (137.3 to 138.7)	138.1 (137.6 to 138.6)	138.2 (137.9 to 138.5)
Mean uranium concentration, ng/l (95% CI)	3.9 (3.4 to 4.6)	2.7 (2.0 to 3.6)	4.2 (2.9 to 5.9)	3.9 (3.4 to 4.6)

*The $^{238}\text{U}/^{235}\text{U}$ ratio for natural uranium is 137.9.

Measurements

From the main survey and supplementary questionnaire we used information on age, sex, rank, education level, service, and deployment in previous operations involving depleted uranium.⁴

The main outcome measures from urinalysis were the total uranium concentration and the $^{238}\text{U}/^{235}\text{U}$ isotope ratio as an indicator of exposure to depleted uranium. The analysing laboratory (Scientifics Ltd, Harwell, Oxon, UK) advised that ratios above 142 generally indicated possible exposure and above 145, probable exposure to depleted uranium. Isotope ratios below 142 were reported as "no depleted uranium present". However, the precision of each individual measurement was reported, as this could vary (for example, according to the total uranium content of the sample).

Statistical analyses

Analyses were done in STATA 9 and SPSS for Windows (version 13.0). As there were too few participants with a $^{238}\text{U}/^{235}\text{U}$ ratio above 142, we carried out a multiple linear regression analysis using the ratio as the dependent variable. The distribution of the ratio was checked for normality and heterogeneity of the residuals. Regression coefficients with 95% confidence intervals were calculated to express associations between groups, after adjustment for age and sex. Geometric means were calculated for the $^{238}\text{U}/^{235}\text{U}$ ratio and uranium concentration.

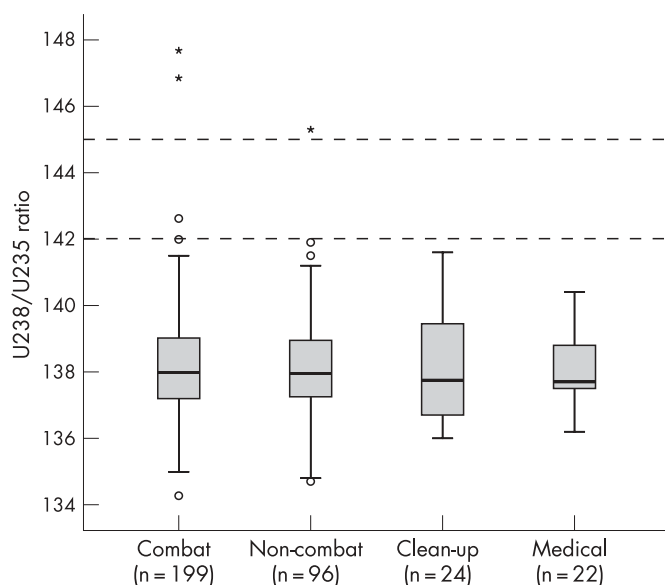


Figure 1 Boxplots summarising the spread and shape of the distribution of the $^{238}\text{U}/^{235}\text{U}$ ratio for each group. Horizontal bands indicate medians. The shaded boxes represent the 25th to 75th centiles. Circles represent outliers between 1.5 to 3 box lengths from the 25th or 75th centiles. Cases marked * indicate outliers greater than 3 box lengths from the 25th or 75th centiles. Whisker lines show the largest values that are not outliers. Horizontal dashed lines correspond to Scientifics Ltd's guidelines for reporting samples as indicating possible exposure to depleted uranium (142) and probable exposure to depleted uranium (145).

RESULTS

Baseline information

In total, 369 individuals agreed to take part in the study of depleted uranium exposure. However, the $^{238}\text{U}/^{235}\text{U}$ ratio could not be determined for 28 urine samples because the uranium content was below the limit of detection (12 samples), or although measurable, the total uranium concentration was low (generally <1 ng/l).

The majority of the participants were males below the rank of officer in the Army (table 1). The four groups sampled differed in education level, but the great majority of participants had finished compulsory education. The maximum number of personnel that could have taken part in the study was 776. The denominator was derived from a maximum 40% attendance by TELIC personnel, per location. The response rate was 48%.

In comparison to the main cohort study, participants were of slightly lower rank, similar age and qualifications reflected the trades related to the selection strategy of the sample.

$^{238}\text{U}/^{235}\text{U}$ ratio

There were no significant differences in isotope ratio between the four groups. With the exception of combat personnel (138.2, 95% CI 138.0 to 138.5), the 95% confidence intervals for the mean ratio in the other three groups included the value of 137.9 for natural uranium (table 2). Figure 1 shows the distribution of the ratio in the four groups. Three samples in the combat group and one sample in the non-combat group showed ratios above 142. Uranium concentrations for the four samples with ratios above 142 ranged from 2.1 ng/l to 4.4 ng/l. The two samples with the highest ratios (146.9, 147.7) were retested at the UK Natural Environmental Research Council Isotope Geosciences Laboratory, Nottinghamshire, using a more accurate analytical technique (multiple collector inductively coupled plasma mass spectrometry (MC-ICP-MS)).⁵ Both were found to have ratios within measurement error of that for natural uranium (136.0, 136.1).

In the multiple regression analysis there was no association of the $^{238}\text{U}/^{235}\text{U}$ ratio with any of the four groups (table 3).

Table 3 The relation between groups and the $^{238}\text{U}/^{235}\text{U}$ ratio by number of depleted uranium deployments and self-reporting of possible depleted uranium exposure

n=339	Coefficients*(95% CI)	p Value
Group		0.8
Combat	0.10 (-0.30 to 0.51)	
Clean-up	-0.21 (-0.95 to 0.53)	
Medical	0.01 (-0.79 to 0.81)	
Two or more DU deployments during service	0.25 (-0.13 to 0.63)	0.2
One or more self-reported circumstance of possible DU exposure	-0.11 (-0.49 to 0.27)	0.6

"DU" deployments refer to participants deployed in operations where depleted uranium munitions were used.

Reference groups: non-combat, 1 "DU" deployment, 0 self-reported exposure.

*Adjusted for age and sex in a single, multiple linear regression analysis.

Table 4 Self-reported frequencies of circumstances of potential depleted uranium exposure

Circumstance of potential exposure	Group			
	Combat (n = 199)	Non-combat (n = 96)	Clean-up (n = 24)	Medical (n = 22)
In a vehicle struck by a depleted uranium round	14 (7.0%)	1 (1.0%)	4 (16.7%)	0 (0%)
Entered a vehicle immediately after it had been hit by a depleted uranium round	8 (4.0%)	0 (0%)	1 (4.2%)	1 (4.5%)
Worked in or on contaminated vehicles to carry out repairs or other tasks	24 (12.1%)	5 (5.2%)	11 (45.8%)	3 (13.6%)
Treated or helped casualties that may have had depleted uranium-related injuries	5 (2.5%)	1 (1.0%)	1 (1.0%)	6 (27.3%)
Other*	42 (21.1%)	19 (19.8%)	1 (1.0%)	3 (13.6%)
Total number of possible depleted uranium exposures	93	26	18	13
Total number of potentially exposed subjects	68 (34%)	20 (21%)	11 (46%)	11 (50%)

Types of recorded exposure may be more than one per participant.

Percentages shown are to one decimal place.

*Other self-reported circumstances of potential depleted uranium exposure not matching above descriptions.

None of the other variables in the model was associated with the $^{238}\text{U}/^{235}\text{U}$ ratio.

Self-reporting of potential depleted uranium exposure

Table 4 shows the frequencies of self-reported circumstances of potential depleted uranium exposure in each group. The majority of participants, 231 (68%) out of 341, reported no likely depleted uranium exposure. Combat personnel reported the highest total number of potential exposures, including a high number of potential exposures to depleted uranium from other sources (21.1%). Working with contaminated vehicles (45.8%) and treating possible depleted uranium casualties (27.3%) were the most frequently reported circumstances of potential exposure among clean-up and medical personnel, respectively.

DISCUSSION

We did not find evidence of substantial depleted uranium exposure or statistically significant differences in $^{238}\text{U}/^{235}\text{U}$ ratio between the four groups studied. For the great majority of the participants any possible uptake of depleted uranium was below the limit of detection of the assay. Participants in groups identified a priori as having a higher potential for exposure to depleted uranium were more likely to report circumstances of possible exposure.

In our study four samples had a $^{238}\text{U}/^{235}\text{U}$ ratio above 142. However, when the two samples with ratios above 145 were reanalysed by the UK Natural Environmental Research Council Isotope Geosciences Laboratory, the ratios were consistent with natural uranium.

Where exposure to depleted uranium occurs, the risks to health will depend upon the route and extent of exposure. It has been estimated that inhalation of 16 mg depleted uranium as an aerosol of the type produced when a depleted uranium round strikes an armoured target would produce a committed effective dose of ionising radiation in the order of 1 mSv (equivalent to the current annual dose limit for members of the public in the UK), and a maximum renal concentration of 0.09 µg uranium per g kidney (which is well below the threshold for chemical toxicity).² Biokinetic calculations indicate that an inhaled dose of this magnitude could be expected to result in a daily urinary excretion of approximately 100 ng of depleted uranium after an interval of 2–3 years.⁶ Even if the daily excretion of depleted uranium were only 10 ng (that is, allowing for a tenfold error in the model), and the background excretion of natural uranium was relatively high at 30 ng per day, the $^{238}\text{U}/^{235}\text{U}$ ratio in urine would still exceed 200. This is well above any of the measured values in our study. In the few

participants whose $^{238}\text{U}/^{235}\text{U}$ ratio could not be measured, the total uranium content of the urine sample was low, and below the level that would have been expected if daily excretion of depleted uranium were as high as 10 ng.

Our findings are similar to those reported by Jones *et al* from a pilot study of normative uranium levels in the UK civilian population,⁷ and a comparative sample of serving UK personnel that did not deploy to Iraq in TELIC 1.⁸ Further, our failure to detect clinically important exposure to depleted uranium accords with the results from a voluntary testing programme for veterans of the 1991 Gulf War and the Balkans campaign.⁹

The overall distribution of uranium concentrations compare to reference values for a US sample, median 6.3 ng/l,¹⁰ and a sample of UK service personnel not deployed in the Iraq War, median, 2.3 ng/l.⁸ For military personnel retaining DU fragments, concentrations have been reported to average 80 ng/l, seven years after exposure.¹¹ The lower concentrations in the present study provide further indication there was no significant depleted uranium exposure among the participants.

A limitation of our study was that the $^{238}\text{U}/^{235}\text{U}$ ratio using the SF-ICP-MS apparatus has been reported to have a lack of precision of approximately 4%.⁵ Inter-laboratory comparison demonstrated that the accuracy using SF-ICP-MS in samples of natural isotopic composition and samples containing known quantities of depleted uranium was inferior to MC-ICP-MS (a lack of precision of 1%).⁵ In addition to the issue of precision, the MC-ICP-MS, but not the SF-ICP-MS, is able to determine ^{236}U , a contaminant generally found in depleted uranium. The reanalysis of the two outlier samples showed absence of ^{236}U , thus giving further reassurance that these participants had not been importantly exposed to depleted uranium.

A study of civilians by the Institute of Occupational Medicine, Edinburgh, UK used the same laboratory to analyse ^{238}U and ^{235}U in spot and 24-h sample collections, and observed a similar proportion (2 out of 139) of elevated ratios in low concentration spot samples.⁷ These elevated ratios were unconfirmed when reassessed. These data suggest that occasionally elevated ratios can occur in spot samples where concentrations are relatively low (<1 ng/l).⁷ However, we believe that for surveillance purposes SF-ICP-MS can be sufficiently accurate for initial assessment. Moreover, although the precision of individual measurements may be limited when uranium concentrations are low, the reliability of the group means, as reflected in their 95% confidence intervals, is such that average isotope ratios can be meaningfully compared between groups.

Another weakness of our study was that we were unable to collect urine samples over a 24-h period, and had to rely on spot samples. We wanted to maximise the level of collaboration

Main messages

- No evidence for significant depleted uranium contamination in UK forces exposed to depleted uranium munitions.

Policy implications

- The estimates reported here would be an appropriate comparison for future studies of Service personnel returning from operational deployments in which depleted uranium munitions may be used.

from participants and 24-h collection was unfeasible within brief visits to military units and the nature of military duties. However, there is evidence that uranium concentrations in spot and 24-h collections are correlated.^{11 12} Furthermore, any distortion of total uranium concentration from unusually low or high concomitant excretion of water would not extend to the ²³⁸U/²³⁵U ratio.

The use of isotope ratios was a particular strength of our study design. Uranium concentration alone may be an ambiguous indicator of urinary depleted uranium. Gwiazda *et al* showed an overlap in the spread of uranium concentrations for personnel exposed to depleted uranium and a non-exposed reference group.¹³ Uranium concentrations for the two highest samples measured in this study were >200 ng/l, and compare to the 95th to 99th percentile in a sample of non-deployed personnel.⁸ Isotopic analysis can diminish the potential confounding effects of dietary uranium, of which the primary source is drinking water.¹⁴ In our study, the ²³⁸U/²³⁵U ratios for samples with the highest concentrations of uranium were all within measurement error of that for natural uranium, and unusually high ingestion of natural uranium is the most likely explanation for these higher concentrations.

Another strength of our study was that participants were recruited as part of a larger epidemiological survey.⁴ It is possible that subjects were more inclined to volunteer if they thought that they had been exposed to depleted uranium, but the effect of any resultant bias would have been to exaggerate rather than diminish the prevalence of measured exposures. Previous studies have carried out urinary isotopic analysis in self-selected participants concerned about their exposure,¹⁵ or among personnel with verifiable exposure to depleted uranium.^{1 13}

In our study most participants did not recall circumstances of potential exposure to depleted uranium in Iraq or during previous operations. However, report of possible exposures was more common in the groups that a priori were expected to be at greater risk. Our study showed that perceived depleted uranium exposure at level 2 in the Royal Society³ classification was unrelated to depleted uranium uptake at detectable levels.

In conclusion, although our findings cannot be extrapolated to civilians, they should allay concerns about possible widespread exposure to depleted uranium among UK Armed Forces personnel in Iraq. We cannot exclude the possibility that a small number of service personnel absorbed larger quantities of depleted uranium. However, any uptake of depleted uranium in

our study sample would be very unlikely to have any implications for health.

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Competing interests: D Coggon is chair of the independent Depleted Uranium Oversight Board (DUOB), established by the UK Ministry of Defence (MoD). The DUOB consists of technical experts in the fields of epidemiology, toxicology, screening, mass spectrometry and representatives of UK Armed Forces veterans. S Wessely is Honorary Civilian Consultant Advisor to the British Army. N Greenberg is a full-time active service medical officer who has been seconded to King's College Centre for Military Health Research as a liaison officer; although paid from Ministry of Defence funds he was not directed in any way by the Ministry in relation to this publication. All other authors declare that they have no conflict of interest.

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