Institution: University of Leicester

Unit of Assessment: 7 Earth Systems and Environmental Sciences

Title of case study: Uranium isotope ‘forensic’ testing in relation to Gulf War Illness

1. Summary of the impact

Professor Parrish at Leicester developed a unique high sensitivity urine, soil and particle isotope assay for detection of DU pollution and applied this to Gulf War veterans to quantify exposure to DU munitions. None of the Gulf War veterans tested in a UK MoD study had detectable DU; this allowed the UK government to conclude that DU exposure was limited, and that the harm to veterans was small, although residual environmental issues of chronic exposure have yet to be quantified. The test was also applied to munitions’ factory workers and nearby residents, and allowed the exposure to DU to be quantified in individuals and environmental materials. This latter study gave rise to a $0.5M exposure and health study near by the New York State Dept. of Health to better assess the health impacts of DU aerosol exposure.

2. Underpinning research

Underpinning research: Geochemical research in the Department of Geology by Parrish, Brewer and PhD student Lloyd from 2004-2009 used isotope geochronology and geochemistry methods in collaboration with the NERC Isotope Geosciences Laboratory (NIGL) that Parrish leads on secondment from Leicester, to advance uranium isotope measurement innovation (**5**, **7**). These uranium isotope method advancements, with their origins in earth science and geochronology, were adapted and applied to environmental contamination (**1**, **2**, **4**) and environmental health topics in order to address important problems. Urine and other matrices (soils, organic material, micron sized particles) are often difficult to work with, but medically and environmentally important; mass spectrometry methods were adapted in 2004-2008 for these matrices and are ongoing. For urine, the work consisted of co-precipitating, concentrating, and purifying tiny amounts (as small as 100 picograms) of uranium from the other undesired elements and compounds contained in urines, and measuring the precise isotope composition of uranium to attempt to detect and quantify anthropogenic uranium. This was demanding since the expectation of the MoD was that samples of urine from ‘DU-contaminated’ soldiers might only have 1-5% of the uranium in the samples consisting of DU, this being the expected result if a significant contamination arose up to 15 years prior to taking a urine sample. A 1-2% deviation of isotope ratio of $^{238}U/^{235}U$ was required for total quantity of uranium of as little as 1 nanogram; in addition the anthropogenic isotope ^{236}U was required to be measured at <50 femptogram level of abundance. This had never been achieved before in either medical or geochemical research. This R&D was validated by blind externally-administered proficiency tests, administered by the EU IRMM to stand up to scrutiny (**5**). In parallel a method of solid particle (<10µm in size) isotope determination using laser ablation sampling was developed (**3**), allowing forensic tracing of particles back to their source, a procedure used by the IAEA to monitor uranium enrichment worldwide using different and more costly methods. All of these method innovations have been linked by the programme of environmental, technical, and health research undertaken by the Leicester team.

The Leicester team consists of: Professor Randall Parrish (1996-present, partly seconded to lead NIGL); Dr Tim Brewer (Senior Lecturer 1994-2007; deceased 2007); Dr Nick Lloyd, PhD student 2006-2009; Prof S Hainsworth (Engineering).

3. References to the research

1. Lloyd, N. S., Chenery, S. R. & Parrish, R. R. (2009). "The distribution of depleted uranium contamination in Colonie, NY, USA." Science of the Total Environment 408(2): 397-407.

2. Lloyd, N. S., Mosselmans, J. F. W., Parrish, R. R., Chenery, S. R., Hainsworth, S. V. & Kemp, S. J. 2009a. The morphologies and compositions of depleted uranium particles from an environmental case-study. Min. Magazine, 73 (3), 493-508.

3. Lloyd NS, Parrish RR, Horstwood MSA, Chenery SRN. 2009 [Precise and accurate isotopic analysis of microscopic uranium-oxide grains using LA-MC-ICP-MS](), J. Anal. At. Spectrom., 24, 752-758 DOI**:** [10.1039/b819373h]()

4. Parrish, R. R., Horstwood, MSA, Arneson, J, Chenery, S., Brewer, T., Lloyd, N., Carpenter, D. 2008. Depleted uranium contamination by inhalation exposure and its detection after ~20 years: implications for human health assessment. Science of the Total Environment v. 390, 58-68; doi:10.1016/j.scitotenv.2007.09.044.

5. Parrish, RR, Thirlwall, M, Pickford, C, Horstwood, MSA, Gerdes, A., Anderson, J., and Coggan, D., 2006, Determination of 238U/235U, 236U/238U and uranium concentration in urine using SF-ICP-MS and MC-ICP-MS: An inter-laboratory comparison. Health Physics v.90 (2), p. 127-138.

6. Parrish, RR, Impacts of Depleted Uranium to the natural environment: A report commissioned by the Natural Environment Research Council for the UK Ministry of Defence, submitted to NERC and MoD 2010.

7. DUOB (Depleted Uranium Oversight Board). DUOB Testing Programme: final report; 2007. http://www.duob.org.uk/final_report_feb2007.pdf or www.bandepleteduranium.org/en/docs/27.pdf

4. Details of the impact

Background to Impact. Two major wars in the Persian Gulf area in 1991 and 2003-2008 involved more than 1M allied military personnel and the use of DU munitions; subsequent to the 1991 conflict, ~250,000 UK, US, and Canadian veterans developed Gulf War Illness, the cause of which remains elusive. Possible culprits are the nerve gas Sarin, DU, excessive pesticide use during the conflict, and post-traumatic stress. During the period 1997-2008 there was considerable concern that exposure of military personnel (and collateral exposure of civilians) to DU munitions might contribute to illness of veterans (Gulf War Illness, cancer, birth defects, etc) and have other deleterious environmental effects. These concerns were documented by the Royal Society and the United Nations in 2001-2002. In 2005 the UK Government appointed the Depleted Uranium Oversight Board (DUOB) to commission a study to both develop a high sensitivity assay and to apply this to veterans to find out if DU exposure was widespread (**7**); the MoD also commissioned a joint NERC research programme on the environmental impacts (**6**). No such test was available worldwide at that time. At the same time, sites of DU pollution from manufacturing munitions were poorly studied and understood in terms of their environmental footprint. A common thread in all of this science is that a sensitive, reliable test to quantify DU exposure that took place many years earlier had been lacking, unavailable to the environmental health community. No DU-exposed human cohort had ever been studied in terms of aerosol DU particle exposure, and this question was also addressed by the Leicester group.

Beneficiaries: Beneficiaries of this work are primarily the UK, Canadian and US governments who have learned that the DU exposure sustained by Gulf War veterans is more limited than thought. Other benefits are directly to those ~250,000 US, UK, and Canadian veterans with Gulf War Illness who can achieve more clarity about whether DU played a role in their illness. Other beneficiaries are New York Department of Health who have adopted the Leicester test and are conducting a more comprehensive exposure and health study on the DU munitions factory site studied by Leicester personnel. As a follow up to this work, the Atomic Weapons Establishment (AWE Aldermaston) sought collaboration with the team and NIGL colleagues to develop a laser ablation ICP-MS method to analyse uranium isotopes in tiny (~1 μm) particles to reduce the cost of nuclear forensic testing in uranium enrichment establishments for the IAEA and the UK government.

Direct Impacts. The urine-testing research showed that no tested veteran had detectable DU (none out of 466 tested, documented in final report of the DUOB (**7** section 4), allowing the UK government to conclude the study without costly subsequent follow up and be satisfied that significant exposure to veterans by DU could not be substantiated in terms of a large group of veterans. This allowed the UK MoD to largely draw a line under this particular issue, and reduce future expenditure related to DU-related issues. This is substantiated by a letter from the lead author on the 2001-2002 Royal Society Report on DU, stating "*The Depleted Uranium Oversight Board were particularly impressed by the high performance of NIGL in testing for uranium isotopes ....which has resulted in a much clearer view of the levels of exposure to DU on the battlefield (which subsequently have been supported by uranium isotope measurements by Harwell on urine from veterans of the 2001 Gulf War) and have very largely eliminated further consideration of DU exposure as a contributor to Gulf War diseases*" (**C**).

The environmental study in New York (**1**-**3**) documented the long term environmental legacy of DU pollution, uptake by plants and animals, slow migration in soil, and very slow corrosion of highly oxidized DU aerosol particles. None of these conclusions were known until this work was done, yet policy decisions need to rely on such evidence. The research proved as *entirely incorrect* the conclusions of the US Agency of Toxic Substances and Disease Registry (ATSDR)which stated that the DU contamination legacy surrounding the New York manufacturing plant could not be quantified and that detection in humans after 20 years would be impossible. The method was applied to exposed individuals to show that DU particulate exposure is still detectable after 20 years (**4**). Arising from this work, the US Congress Committee on Science and Technology sought testimony from Parrish on 12 March 2009 (**A,B**) concerning the research in order to learn lessons about how to quantify exposure and improve the performance of its own agency responsible for health concerns in such sites (i.e. ATSDR).

The long term health and environment effects of chronic DU-inhalation or ingestion exposure are as yet un-quantified, because no health study has been conducted on putatively-exposed humans, even though numerous animal studies show mutagenic and teratogenic effects. The Leicester study directly prompted the New York State Department of Health to conduct in 2011-13 a larger (~$0.5M; cohort of 300) targeted DU exposure and health study to address the continuing health concerns of that locality, and Parrish is advising this group with parallel analyses to assess Quality Control, advice on how best to conduct chemical separations, and in critical evaluation of method publication. He will likely be involved in interpretation of exposure once data is acquired by that laboratory on a cohort in New York.

Although the whole of the 1991 Gulf War veteran cohort is unlikely to have had significant DU-exposure, it can only be eliminated as a cause of Gulf War Illness in a proper study of veterans suffering from the illness. A direct impact of the research is that testing for DU is taking place during 2011-13 by Parrish on 160 intensively studied Gulf War Illness sufferers whose medical conditions were the subject of a 10-year, $20M study at the Southwestern Medical Centre, University of Texas. This DU exposure study will define the role, if any, of DU in this illness and conclude in 2013, of considerable medical significance.

Finally, the AWE (Atomic Weapons Establishment Aldermaston) has consulted Leicester and NIGL personnel in order to conduct collaborative development and knowledge exchange to develop a more rapid and cost-effective LA-ICP-MS method to analyse ~1µm uranium particles in nuclear forensic safeguard testing at AWE, who have a strategic regulatory national role in this area. This capability of using LA-ICP-MS for U particle isotope composition is a priority topic of the IAEA (Vienna) and the nuclear forensic safeguards community because of the very high cost and time-consuming nature of doing this work by the alternative FT-TIMS or SIMS methods, which are the only methods currently accredited by the IAEA; LA-ICP-MS offers similar sensitivity, faster turnaround and more rapid analysis including the minor uranium isotopes.

A further specific impact is that the Leicester PhD student who participated in this work was then quickly employed by a major mass spectrometry manufacturer as a key applications scientist. He is now involved in developing and marketing the type of instruments used in the study to other scientific establishments worldwide including IAEA and AWE; this is a tangible impact of the high-level training delivered as part of the on-going research.

5. Sources to corroborate the impact

Media, and letters

(A) Testimony to US Congress Subcommittee on Investigations and Oversight, Science and Technology, 12 March 2009 by R Parrish, transcript available from US Congress Archives http://www.gpo.gov/fdsys/pkg/CHRG-111hhrg47718/html/CHRG-111hhrg47718.htm

(B) Chair of US Congress Subcommittee on Investigations and Oversight.

(C) Chair of Royal Society Working Group on Depleted Uranium, and member of the Depleted Uranium Oversight Board.

Press Release

March 12, 2009 Investigations and Oversight Subcommittee Examines the Failures of the Agency for Toxic Substances and Disease Registry, US Congress Committee on Science and Technology Hearings; http://archives.democrats.science.house.gov/press/PRArticle.aspx?NewsID=2390 [there have been many other related press releases related to the DU work but are not itemised].