

# Quantitative Analysis of the Concentration and Ratio of Uranium Isotopes in the US Military Personnel Deployed at Samawah, Iraq during Operation Iraqi Freedom\*

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The aim of this study was to analyze the concentration and isotopic composition of four uranium isotopes ( $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{236}\text{U}$  and  $^{238}\text{U}$ ) in the urine samples of US soldiers deployed in Samawah, Iraq during Operation Iraqi Freedom.

Nine US soldiers, the members of Military Police unit 442, deployed during the Iraq military operations in March 2003, presented with non-specific symptoms of headaches, fatigue, fever, musculoskeletal pains, respiratory impairment, neurological, and affect alterations. Twenty-four hour urine samples of each subject were analyzed together with control samples consisting of internal urine standards. The analytical methodology included preconcentration of the urine samples using either co-precipitation or evaporation, oxidation of organic matter, uranium purification by ion-exchange chromatography, and mass spectrometry analysis. For determination of total uranium concentration a 2ml aliquots, precisely weighed to 0.1%, were spiked with a certified  $^{233}\text{U}$  tracer solution. A larger aliquot of 500ml unspiked urine were used for precise determination of the  $^{238}\text{U}:$  $^{235}\text{U}$ ,  $^{234}\text{U}:$  $^{238}\text{U}$ , and  $^{236}\text{U}:$  $^{238}\text{U}$  ratios. All specimens were analyzed in duplicate, using an Aridus desolvation system and a double-focusing Thermo Finnigan Neptune multi-collector ICP-MS equipped with a retarding potential quadrupole lens and a secondary electron multiplier for ion counting. The reproducibility of the  $^{238}\text{U}:$  $^{235}\text{U}$ ,  $^{234}\text{U}:$  $^{238}\text{U}$ , and  $^{236}\text{U}:$  $^{238}\text{U}$  ( $6.8 \times 10^{-8}$ ) for an 8ppb NBS950a solution (n=14) over two days were, before applying any corrections, 0.13, 0.6 and 2.6%, respectively. Limits of detection for  $^{238}\text{U}$  are about 1 pg/L and analytical blanks were below 6 pg.

The mean concentration of total uranium was  $3.2 \pm 0.6$  ng/L. Five of the nine soldiers have a  $^{238}\text{U}:$  $^{235}\text{U}$  ratio of natural uranium. Three subjects of this group had detectable levels of  $^{236}\text{U}$ . Four soldiers were clearly identified as positive for depleted uranium excretion. The  $^{234}\text{U}:$  $^{238}\text{U}$  ratio varied from  $5.7 \times 10^{-5}$  to  $7.2 \times 10^{-5}$  and correlates negatively with the  $^{238}\text{U}:$  $^{235}\text{U}$  ratio. Urinary  $^{236}\text{U}$  concentrations of these four individuals vary from 1.4 to 12.2 fg/L and their  $^{236}\text{U}:$  $^{238}\text{U}$  ratio correlates positively with the ratio of  $^{238}\text{U}:$  $^{235}\text{U}$ .

Our findings demonstrate depleted uranium contamination of military personnel deployed in the radioactive battlefield and suggest a need of sustained follow up for potential somatic and genetic consequences. Our current studies of military and civilians contamination with isotopes of uranium and plutonium are in progress for the risk assessment of actinides in the biosphere of post-conflict Iraq.

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