Chemical Forensic Detective Work: The search for depleted uranium in biological and environmental samples*

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In nature, uranium has three naturally occurring isotopes, ^{238}U, ^{235}U, and ^{234}U, with abundances 99.2745%, 0.7200%, and 0.0055% respectively. The key ratio of interest is the ^{238}U:^{235}U ratio, measured to be 137.88 for naturally occurring uranium. The nuclear industry refines mined U_3O_8 and removes the majority of ^{235}U and ^{234}U for use in nuclear fuel rods. This enrichment process creates a "left-over" form of uranium with substantially less ^{235}U at 0.20% abundance and increased ^{238}U abundance of approximately 99.79%. This "left-over" uranium is referred to as depleted uranium and one of its uses is the manufacture of armaments. The ^{238}U:^{235}U ratio measured from one particular piece of shrapnel is 491.87 ± 0.16 (2 σ). Every person will have trace amounts of naturally occurring uranium in their system from eating vegetables, drinking ground water, and inhaling or ingesting small amounts of dust and dirt. What isn't expected is to find a ^{238}U:^{235}U ratio higher than 137.88 unless there has been exposure to depleted uranium.

The Atlantic Universities Radiogenic Isotope Facility (AURIF) analyzes geological samples, i.e. minerals and whole rock powders. To separate uranium and lead, AURIF uses a standard hydrobromic acid / anion exchange resin technique developed by Manhes et al. (1978). Dealing with biological and environmental samples meant modification of this existing technique to accommodate the different types of samples being analyzed such as urine and human tissue. The pre-concentrated and purified uranium fractions were analyzed by isotopic dilution thermal ionization mass spectrometry using a Finnigan MAT 262V mass spectrometer. Uranium isotopic ratios were obtained using peak jumping routines utilizing a secondary electron multiplier - ion counting system with 32 second counting times on each isotope. Data from the preliminary group of British gulf war veterans showed ^{238}U:^{235}U ratios ranging from natural for at least half of this group, 137.80 ± 0.13 (2 σ), up to a ratio of 231.67 ± 0.014 (2 σ). This indicates the presence of up to 30 to 50% of depleted uranium mixed with natural uranium.

As the study progressed, faster new analytical techniques were required to process these samples. Urine samples were difficult and time-consuming to pre-concentrate and process due to the presence of large amounts of solid organic materials. A new technique for precipitation of uranium from urine has been developed that reduces the 10 day evaporation procedure to 2 days. Ion exchange chemistry is still required once the uranium has been precipitated, but the development of a new element-specific resin from EICHROM Industries has further sped up the process while maintaining low uranium blanks (0.15 picograms).

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