Isotope ratio mapping of depleted uranium contamination from the NLI Colonie site

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Introduction

A depleted uranium round, discarding its sabot during flight. The kinetic energy of the DU dart penetrates armoured targets. (Federation of American Scientists 1999)

The use of depleted uranium (DU) munitions by US and British forces has been highly controversial. On impact with armoured targets, the dense pyrophoric metal combusts, shedding uranium oxide particulate that can be inhaled into the lungs. Depleted uranium is both weakly radioactive and chemically toxic, and it is frequently cited in the grey literature as a cause of Gulf War Syndrome. However, under the scrutiny of peer-review, no link has been established between low-level inhalation exposure and veterans' ill health.

This research project aims to improve understanding of the behaviour of DU particulate in the environment, using a case study of a site that is heavily contaminated by historic aerosol emissions. The research will help predict the long-term fate of pollution from uranium oxide particulate. This poster focuses on defining the present distribution of contamination.

British Challenger II battle tank. © Crown Copyright/MOD 2008

National Lead

National Lead Industries (NLI) operated a plant at Colonie, NY, USA from 1958 - 1984 [1] (see map, right). Uranium metals were fabricated into radiation shielding, counterweights and kinetic penetrators (armour-piercing munitions).

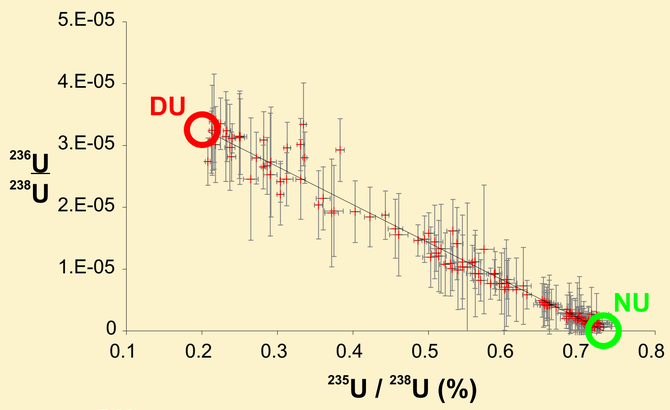
Scrap uranium metal was converted to oxide in a furnace that vented to the atmosphere (left).

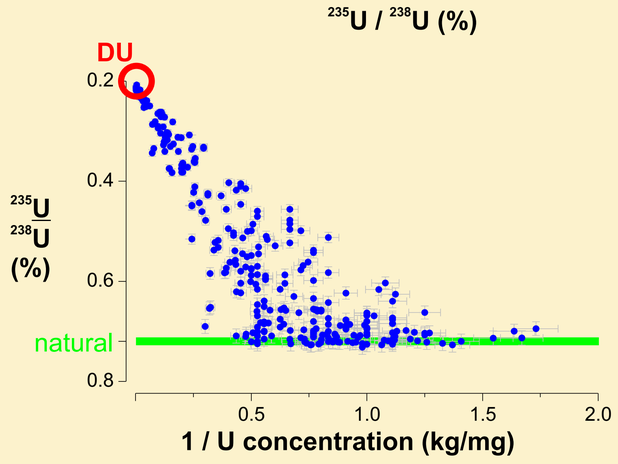
The plant (down and right) was closed by the authorities in 1984 for excessive uranium emissions to the environment. The site has now been remediated with the removal of 180,000 tonnes of contaminated soil [2] (below), reportedly costing more than $ 175,000,000 [3].

Uranium contamination is evident in a sediment core from downstream of NLI [4]. We estimate that in the order of five tonnes of uranium was deposited within 1 km^2 of NLI. This area includes residential and commercial properties. Employees and some residents excrete depleted uranium, more than 25 years after inhalation exposure [5].

For this research project, more than 300 environmental samples have been collected from 120 locations within 12 km of NLI. Soils, dusts and vegetation have been analysed by mass spectrometry.

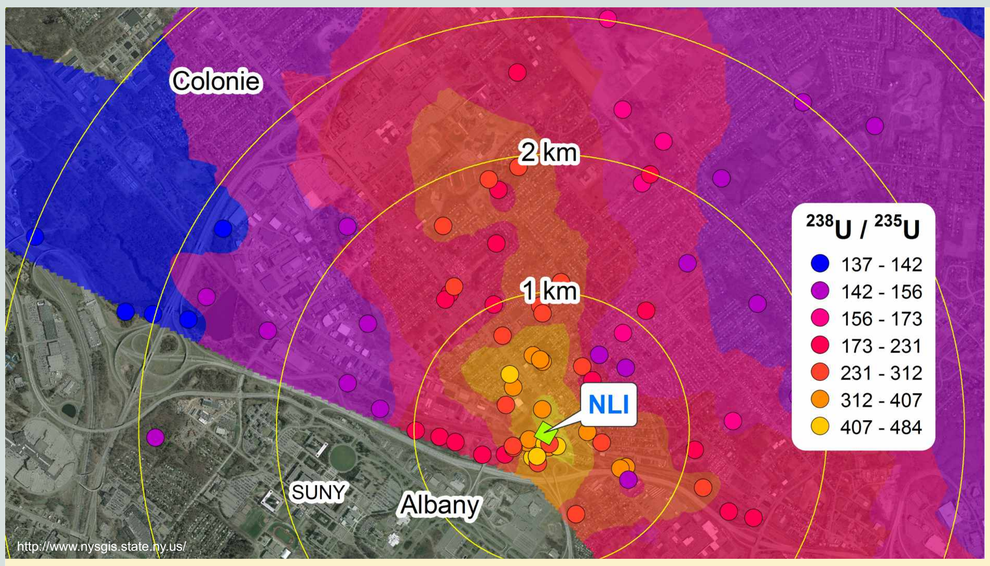
Isotope mixing lines

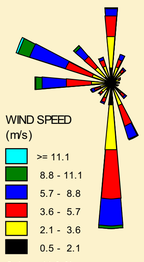




The graph on the left shows the relationship between ^{236}U:^{238}U and ^{235}U:^{238}U. Natural uranium (NU) comprises 0.72 % ^{235}U, and negligible ^{236}U. The data from the analysis of Colonie soil samples by quadrupole ICP-MS, are compatible with a two component mixing line. Background natural uranium in the soils mixes with a depleted uranium end-member, which is estimated to comprise 0.2 % ^{235}U and 3.2 x10^{-05} ^{236}U. Similar values are reported in the literature [6].

The natural background concentration of uranium in the sandy Colonie soils can be estimated using uranium isotope ratios. The graph on the left plots ^{235}U:^{238}U against one over uranium concentration. The high concentration, contaminated samples, approach the depleted uranium end-member at 0.2 % ^{235}U (top-left of graph). Approaching natural uranium isotopic composition of 0.72 % ^{235}U (green line), the data spreads between 0.7 and 2.2 mg/kg. Isotope ratios are useful for detecting low-level contamination, because with only one known exception (Oklo, Gabon), natural uranium has a fixed ^{235}U:^{238}U ratio (cf. background concentration, which is variable).





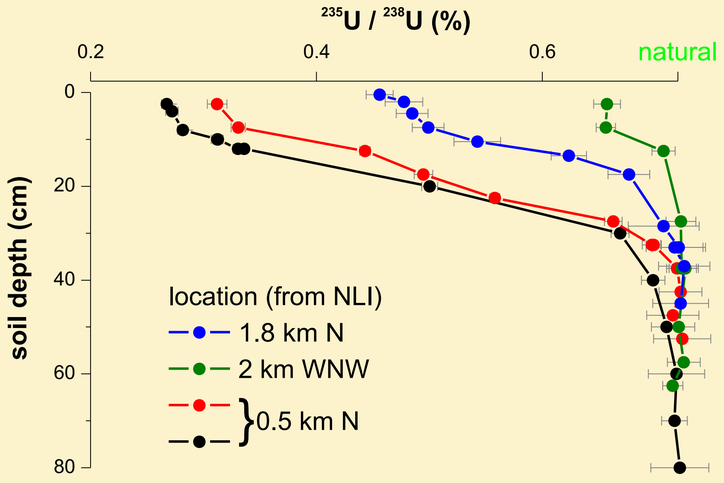
Above: wind rose (blowing from), Albany Country Airport data 1961-1984.

Left: map of surface soil uranium isotope ratios. Natural uranium has a ^{238}U:^{235}U ratio of 137.88, 142 is distinguished from natural with uncertainty. A crude interpolation of the data depicts the distribution of contamination. Sampling locations were dictated by access to undisturbed land within an urban area.

The distribution of depleted uranium contamination revealed by isotope ratios

Soil and dust samples have been analysed by quadrupole inductively-coupled-plasma mass-spectrometry (ICP-MS) at the British Geological Survey.

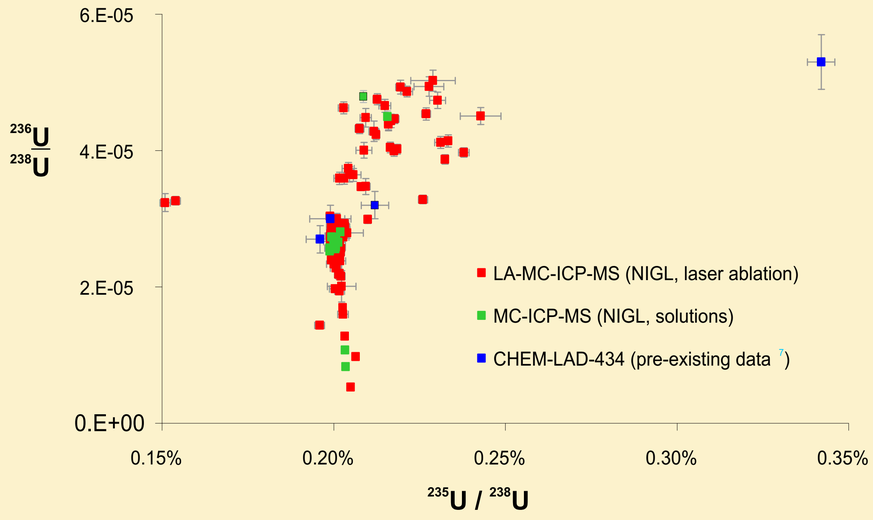
The map above shows the spatial distribution of depleted uranium contamination in surface soils surrounding the former NLI plant. It is apparent that prevailing winds (see wind rose, above right) controlled this distribution. Contamination has been traced as far as 5.8 km from NLI (limit of detection ^{238}U:^{235}U ~ 142 or c. 4% DU component).

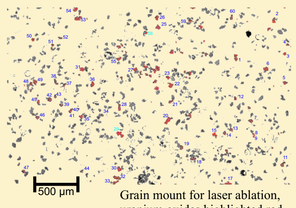


The graph on the left shows depth profiles from four sampling pits. The profiles show an exponential decrease in contamination with depth, reaching natural composition around 40 cm below surface. There is increased homogenisation in the surface samples, but there is no strong association to the organic rich surface layer (0 - 20 cm). Primary particles survive as UO_2 in surface soil samples (unpublished EXAFS data). Particulates may have been physically transported by wash through and bioturbation, with only limited dissolution and reprecipitation.

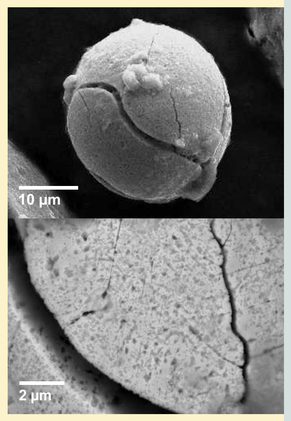
Uranium isotope ratios from individual particles

We analysed the uranium isotope ratios of individual uranium oxide particles (in the 30 to 60 µm diameter range). The particles were concentrated from contaminated soil and dust samples using heavy liquids. Grain mounts were sampled using laser ablation (New Wave Research LUV266 with 10 µm spot), and solutions using a desolvating nebuliser (Cetac Aridus). High precision isotope ratios were measured using multi-collector ICP-MS (VG Elemental AXIOM) at the NERC Isotope Geochemistry Laboratory (NIGL).





Grain mount for laser ablation, uranium oxides highlighted red.



High resolution images of a uranium oxide sphere from a dust sample. These particles survive as UO_2 in soil and dust samples for more than 25 years.

The graph shows that individual particles are isotopically distinct, reflecting variations in the UF_4 feedstocks used at NLI. There are variations in both the ^{236}U and ^{235}U abundance, with a cluster at 0.2 % ^{235}U and 2.7 x10^{-05} ^{236}U This could have implications for the interpretation of contamination in bulk samples. However, the bulk sample data, shown in the frame above, are compatible with a 2 component mixing line; demonstrating that these samples have effectively aggregated and averaged the signatures. Enriched uranium is not seen in these samples.

Summary

- Depleted uranium contamination is evident in soils and dusts surrounding the former NLI site.
- The spatial distribution of contamination was controlled by prevailing winds.
- Contamination can be traced to at least 5.8 km from NLI, and to 40 cm depth in soils closer to NLI.
- The background concentration of natural uranium in the sandy Colonie soils ranges 0.7 to 2.2 mg/kg.
- The isotope ratios from bulk samples fit a mixing line between natural and depleted uranium.
- On average, the uranium emitted from NLI comprised 0.2 % ^{235}U and 3.2 x10^{-05} ^{236}U.
- However, individual UO_2 grains have distinct isotopic signatures, reflecting variations in DU feedstock.
- Uranium oxide particulate survives in soils and dusts as UO_2 for more than 25 years.

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