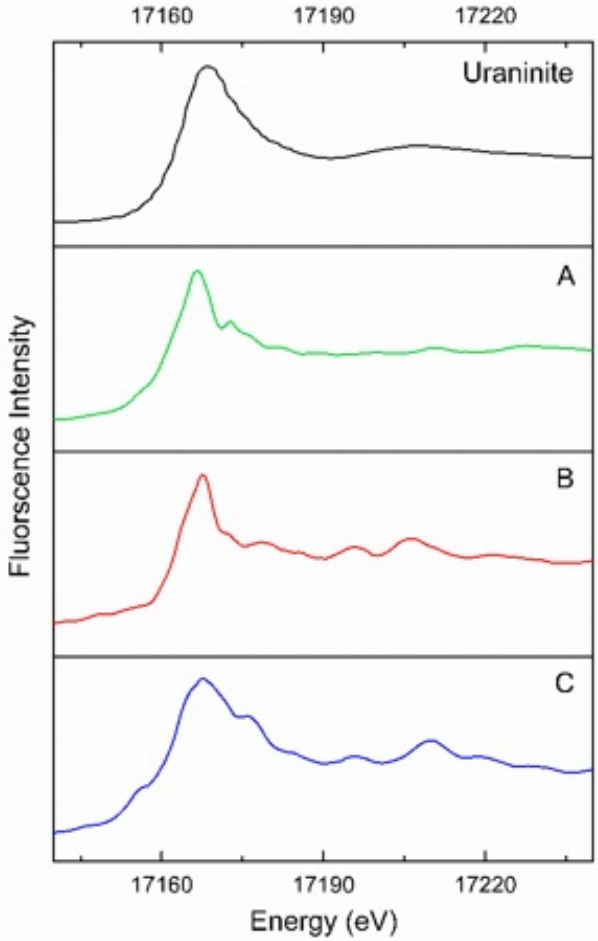
Gaining novel insight into Fukushima Daiichi Nuclear Power Plant derived fallout.

*Martin George Peter[1], T B Scott[1], D A Richards[2], Yosuke Alexandre Yamashiki[3]

1.Interface Analysis Centre, School of Physics, University of Bristol, 2.School of Geographical Sciences and Bristol Isotope Group, University of Bristol, University Road, 3.Graduate School of Advanced Integrated Studies in Human Survivability, Kyoto University

It has now been five years since the events at the Fukushima Daiichi Nuclear Power Plant (FDNPP) contaminated a large portion of eastern Japan with a range of radionuclides. Despite the wide assortment of reactor products released by the incident - the medium-lived fission product isotopes of caesium; ^{134}Cs and ^{137}Cs, with half-lives of 2.065 and 30.2 years respectively have been the subject of the most scientific study due to the significant yield produced and their contribution to the activity measured. Lesser studied have been the longer-lived fission and activation products as well as the actinide fuel material itself. With the levels of caesium in the environment declining rapidly, it is expected attention will shift to these “more exotic” or “frequently forgotten” radionuclides.
Work at the University of Bristol has focused on the analysis of surface materials contaminated with these species from the FDNPP. Sediment and other organic material (mosses and grass) from various locations around Fukushima Prefecture have first been analysed using automated high-resolution scanning electron microscopy and energy dispersive spectroscopy (EDS) to identify particles of interest. Fragments of material found within these samples were then individually removed using a nano-manipulation system onto supporting tungsten or glass capillary needles, attached via an electron-beam hardening adhesive (described fully in Martin et al, 2016 [1]). With samples removed from the bulk material it has been possible to conduct a range of analytical techniques on these particles. Of primary interest currently are those particles containing the chemically toxic and slowly decaying alpha emitter, uranium. Results from X-ray absorption spectroscopy (XAS) conducted at the Diamond Light Source Synchrotron (UK) have confirmed the nano- and micron-scale particulate to be analogous to uraninite (UO_2) with a sub-angular appearance. These fragments are comparable in size to those analysed in work by Abe et al [2] found to exist at the centres of spherical cesium-bearing particles, suggesting their common provenance from the Fukushima Daiichi plant.
Currently undertaken are a range of dissolution experiments on these extracted particles to assess their stability and behaviour within the natural environment. Through the use of high resolution-transmission electron microscopy (HR-TEM), additional current work will seek to examine the physical structure of the ejecta material and how this will impact on its environmental behaviour. Results of these current analyses will be presented. The analysis of initial particles sourced from Iitate Village (Grid Ref) via ICP-MS has highlighted a non-natural compositon of material, strongly supporting its provenance from one of the FDNPP reactors.
The small size of these uranium particles has strong implications for both humans and animals in the affected regions, with their size being conducive to inhalation and the internal exposure. Future work will seek to analyse material sourced from additional areas as well as the other radionuclides present within the samples. As for the case at Chernobyl, as the levels of radiocesium begin to subside, the focus will turn to the other, longer lasting, but still concerning isotopic species.
[1] P. G. Martin, I. Griffiths, C. P. Jones, C. A. Stitt, M. Davies-Milner, J. F. W. Mosselmans, Y. Yamashiki, D. A. Richards, and T. B. Scott, “In-situ removal and characterisation of uranium-containing particles from sediments surrounding the Fukushima Daiichi Nuclear Power Plant,”

Spectrochim. Acta Part B At. Spectrosc., vol. 117, pp. 1–7, Mar. 2016.
[2] Y. Abe, Y. Iizawa, Y. Terada, K. Adachi, Y. Igarashi, and I. Nakai, “Detection of uranium and chemical state analysis of individual radioactive microparticles emitted from the Fukushima nuclear accident using multiple synchrotron radiation X-ray analyses.,” Anal. Chem., vol. 86, no. 17, pp. 8521–5, Sep. 2014.



Results of μ-XANES analysis of three particles (A, B and C) at the uranium L_3 edge demonstrating the presence of uranium in all three particles. A reference spectra for uraninite (UO_2) is shown for comparison [1].

