Determining source terms for FDNPP fallout using $^{135}Cs/^{137}Cs$ and Pu isotopes

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We aim to better establish the physico-chemical nature of the land-based fallout from the Fukushima reactor explosions and the changing distribution of both Cs-based fallout material and denser fallout particles containing U and, by implicit association, Pu. We focus on the $^{135}Cs/^{137}Cs$ atom ratio, which is indicative of the conditions that relate to the nuclear fission reaction responsible for producing the respective radiocaesium isotopes.

We present a method that can be applied to quantify $^{135}Cs/^{137}Cs$ atom ratios by using multi-collector thermal ionization mass spectrometry (ThermoTRITON) in challenging environmental samples: estuarine sediments, where $^{135,137}Cs/^{133}Cs \approx 1 \times 10^{-9}$, to soil, lichen and moss samples from FDNPP catchment. Even though Cs is effectively separated from matrix using double AMP-PAN cation exchange column followed by Sr-spec resin, it has a very low ionization potential and care has to be taken to monitor and eliminate isobaric interferences. Also, with such a high dynamic range, reflections and ion scattering are potential problems and we present strategies to consider their effect. We have successfully eliminated isobaric interferences using a glucose activator. High precision data from Fukushima are presented (e.g decay corrected $^{135}Cs/^{137}Cs$ atom ratio 0.384 ± 0.001 (n = 5) for roadside dust from Iitate region), and these are in agreement with prelimary estimates by others. These data are supplemented by additional analyses from the FDNPP fallout zone to discriminate between different source terms (see also [1]). In addition to Cs isotopes, we have adopted Pu isotope analysis by MC-ICPMS (ThermoNEPTUNE) using ion counting to add a further dimension to the forensic analysis.

We are also refining methods for particle isolation and characterisation using electron microscopy, focused ion beam (Dualbeam), nano/micro manipulators and nano-scale imaging x-ray photoelectron spectroscopy (NanoESCA).

[1] Snow et al. (2016) *Rapid Comm. Mass Spec.* **30**, 523-32