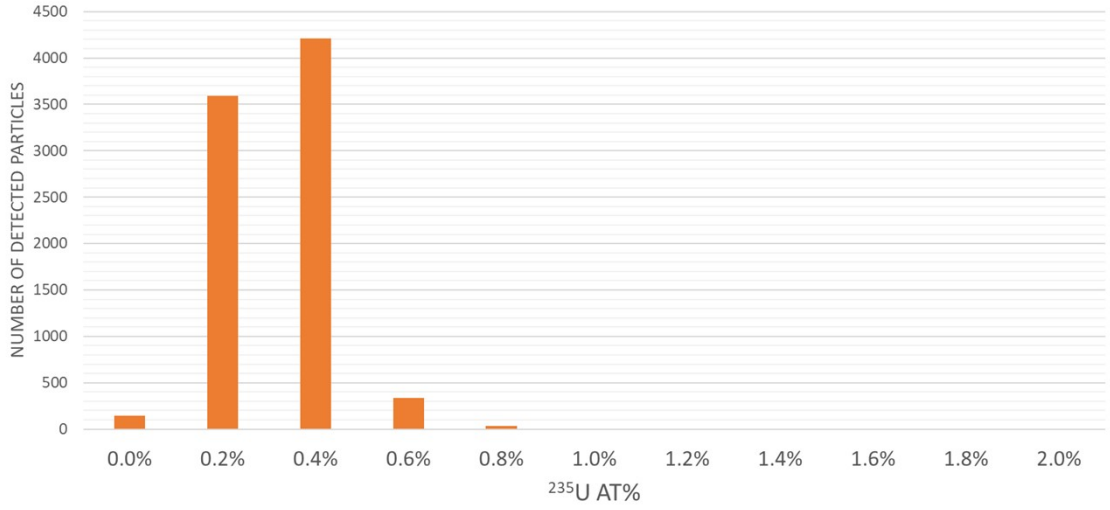
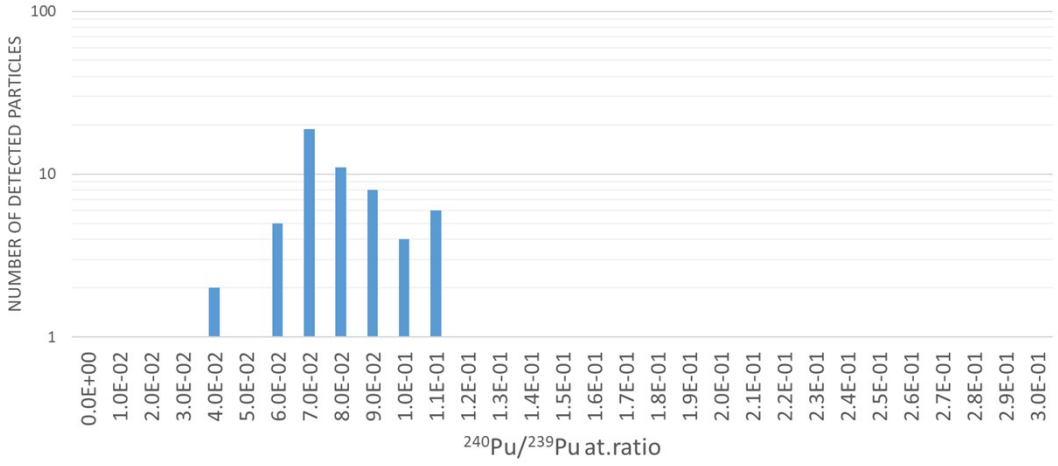
Supplementary information

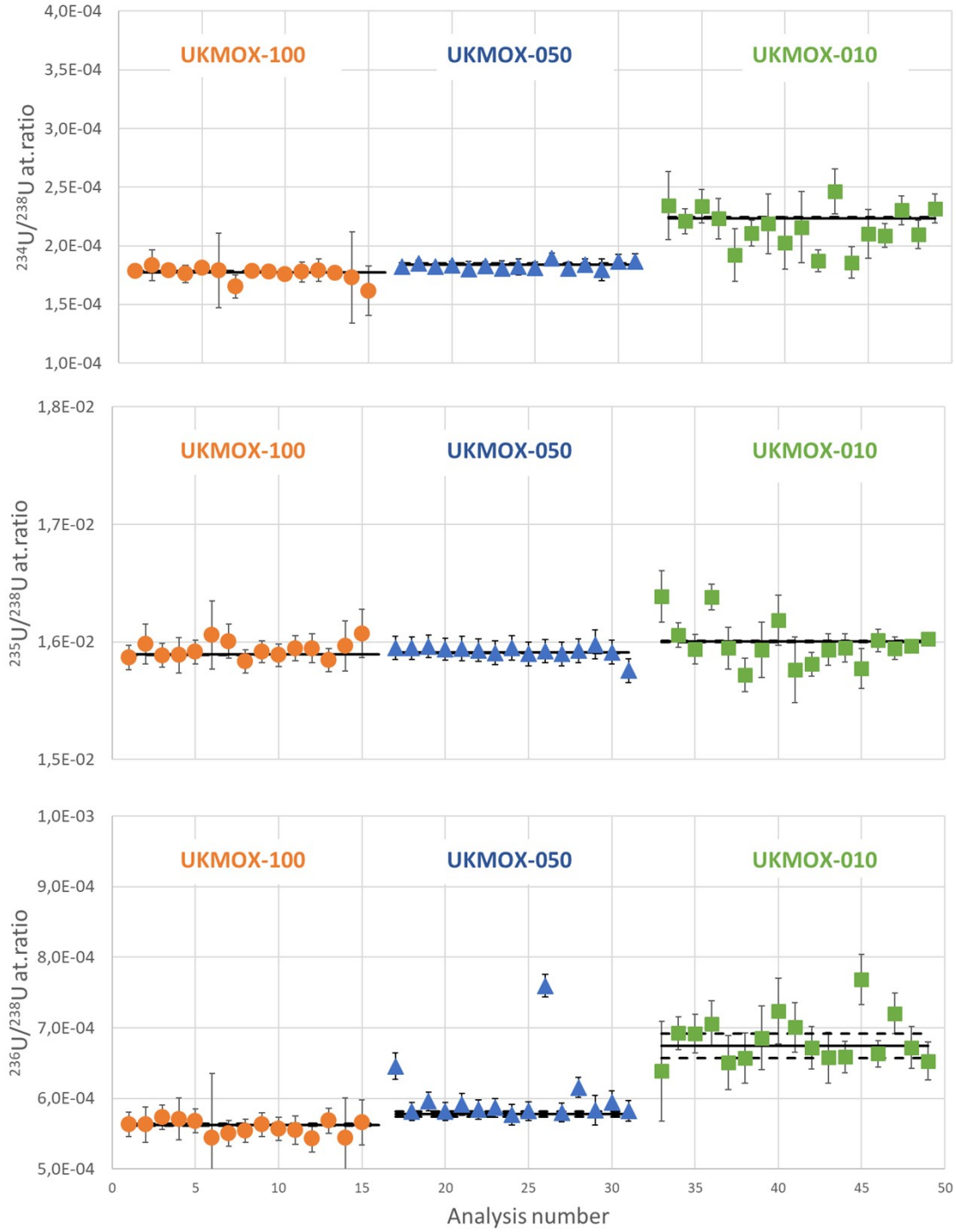
Isotopic ratio	Expanded uncertainty
$\frac{240Pu}{239Pu} = \sum_{i=3}^{n} \frac{240Pu\,i,\ t}{239Pui,\ t} \times \frac{1}{n}$ - $240Pui,t$ is the corrected signal measured in Pu or MOX particle during cycle i corresponding to the analysis time t (see equation 1) - $239Pui,t$ is the raw or corrected signal measured in Pu or MOX particle during cycle i corresponding to the analysis time t (see equation 2) - *N.B.: the time-dependent hydride rate, that is calculated using the power law, may be highly over-estimated during cycles 1 and 2. The isotopic ratio calculations did not take into account these first two cycles.*	$i_{\frac{240Pu}{239Pu}} = \frac{240Pu}{239Pu} \times \sqrt{RSE_{RI}^2 + RSD_{detector\ yield}^2 + RSD_{IMFamu}^2 + \left(i\%_{\frac{240Pu}{239Pu}}\right)_H}$ - RSE_{RI}^2 is the relative standard error of $^{240}Pu/^{239}Pu$ measurements - $RSD_{detector\ yield}^2$ is the relative standard deviation of the detector yield measurements - RSD_{IMFamu}^2 is the relative standard deviation of the IMF measurements - $\left(i\%_{\frac{240Pu}{239Pu}}\right)_H$ is the relative uncertainty due to the hydride correction.
$\frac{236U}{238U} = \sum_{i=3}^{n} \frac{236Ui,t}{238Ui,\ t} \times \frac{1}{n}$ - $236U\,i,\ t$ is the corrected signal measured in MOX particle during cycle i corresponding to the analysis time t (see equation 3)	$i_{\frac{236U}{238U}} = \frac{236U}{238U} \times \sqrt{RSE_{RI}^2 + RSD_{detector\ yield}^2 + RSD_{IMFamu}^2 + \left(i\%_{\frac{236U}{238U}}\right)_H^2}$
$\frac{239Pu}{238U} = \sum_{i=3}^{n} \frac{239Pui,\ t}{238Ui,\ t} \times \frac{1}{n}$	$i_{\frac{239Pu}{238U}} = \frac{239Pu}{238U} \times \sqrt{RSE_{RI}^2 + RSD_{detector\ yield}^2 + RSD_{IMFamu}^2 + \left(i\%_{\frac{239Pu}{238U}}\right)_H^2}$

Supplementary data S1: calculation of the $^{240}Pu/^{239}Pu$, $^{239}Pu/^{238}U$, $^{236}U/^{238}U$ isotopic ratios and corresponding uncertainties.





Supplementary data S2: histograms of the isotopic distributions of the U (top) and Pu (bottom) particles detected in CMX6 using APM methodology.



Supplementary data S3: U isotopic ratios of MOX particles from UKMOX-100 (orange circle), UKMOX-050 (blue triangle) and UKMOX-010 (green square). Each point corresponds to the analysis of one particle. Expanded uncertainties are given with a 95% confidence level. Black line corresponds to the reference value and dotted black lines correspond to the expanded uncertainties of the reference value. Reference values were calculated at the time of analysis taking into account Pu decay.