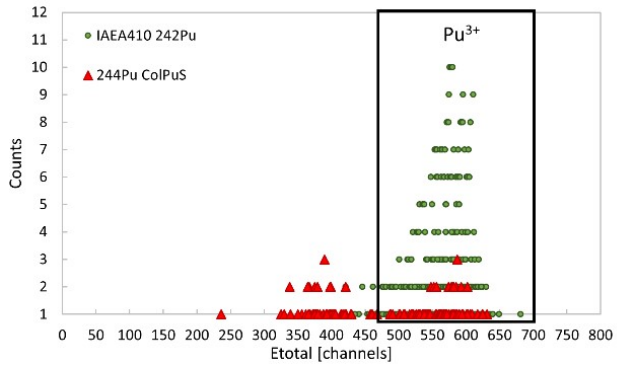
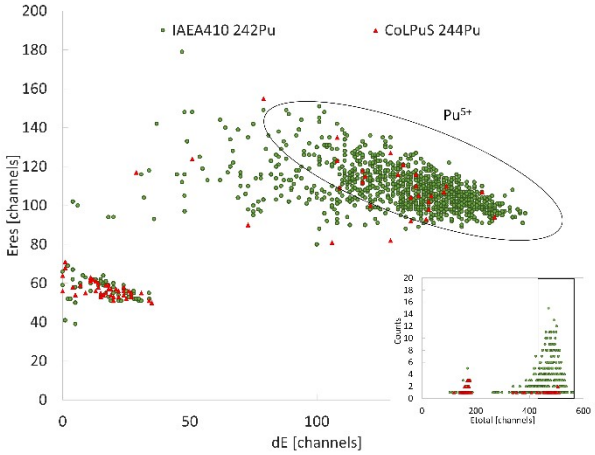
Supplementary Fig. 1 Total energy loss spectrum, obtained from GIC of the 1MV AMS system, for 2.53 MeV $^{242}Pu^{5+}$ (for IAEA 410 sediment sample, green circles) and $^{244}Pu^{5+}$ ions (for ColPuS Pu standard, red triangles). The isotopes are used to set the region of interest



Supplementary Fig. 2 Two dimensional Eres – dE and Total energy (minimized) spectrums, obtained from GIC of the 3MV AMS system, for 14.37 MeV $^{242}Pu^{5+}$ (for IAEA 410 sediment sample) and $^{244}Pu^{5+}$ ions (for ColPuS Pu standard)



Supplementary Table 1 Radiochemical recoveries of Pu (ED – Electrodeposition, MP – Microprecipitation)

Sample	Measurement time	Measured Activity [Bq]	Chemical yield [%]
AS2 ED, only ^{242}Pu	6.9h	0.016 ± 0.003	76.6
AS7 MP, only ^{242}Pu	6.9h	0.021 ± 0.004	94.9
AS10 MP, IAEA 410 spiked with ^{242}Pu and TEVA separation	30.3h	0.018 ± 0.001	85.7
AS16 MP, only ^{242}Pu and TEVA separation	6.9h	0.054 ± 0.006	94.3

Supplementary Table 2 The isotopic ratios for $^{244}Pu/^{239}Pu$ with 2σ uncertainties (using the confidence level given in [1]) and $^{240}Pu/^{239}Pu$ with 1σ uncertainties

Sample/ Code	$^{244}Pu/^{239}Pu\ (\cdot 10^{-4})$	$^{240}Pu/^{239}Pu$
IAEA 410.1 **IFIN-HH, this work**	3.38 ± 0.65	0.250 ± 0.024
IAEA 410.2 **IFIN-HH, this work**	$3.42\ ^{+1.87}_{-1.36}$	0.265 ± 0.026
IAEA 410.3 **IFIN-HH, this work**	4.98 ± 0.98	0.259 ± 0.024
IAEA 410.4 **IFIN-HH, this work**	$4.50\ ^{+2.79}_{-2.02}$	0.218 ± 0.023
IAEA 410.5 **IFIN-HH, this work**	$2.50\ ^{+1.89}_{-1.33}$	0.226 ± 0.022
IAEA 410.6 **IFIN-HH, this work**	$3.38\ ^{+2.29}_{-1.63}$	0.290 ± 0.069
IAEA 410.7 **IFIN-HH, this work**	$1.97\ ^{+1.91}_{-1.24}$	0.265 ± 0.064
Bikini Atoll, Island, BL5 [2]	$3.1\ ^{+5.4}_{-2.0}$	0.319 ± 0.026
Bikini Atoll, Island, BL6 [2]	5.4 ± 1.6	0.323 ± 0.011
IAEA 410, CNA Seville, Spain [3]	/	$0.257 \pm 0.0_{23}$
IAEA 412.1 **IFIN-HH, this work**	$1.22\ ^{+1.11}_{-0.77}$	$0.194 \pm 0.02_{1}$
IAEA 412.2 **IFIN-HH, this work**	$1.34\ ^{+1.22}_{-0.85}$	0.191 ± 0.018
IAEA 412.3 **IFIN-HH, this work**	$2.65\ ^{+1.69}_{-1.17}$	0.156 ± 0.016
IAEA 412.4 **IFIN-HH, this work**	$1.23\ ^{+1.20}_{-0.78}$	0.168 ± 0.017
IAEA 412, CNA Seville, Spain [3]	/	$0.182 \pm 0.0_{15}$
Pacific Ocean, deep-sea manganese crust [4]	1.0 ± 0.3	/

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