## Article "Capabilities of laser ablation – ICP-TOF-MS coupling for isotopic analysis of individual uranium micrometric particles", A.L. Ronzani, F. Pointurier, M. Rittner, O. Borovinskaya, M.Tanner, A. Hubert, A.C. Humbert, J. Aupiais, N. Dacheux

## **Electronic Supplementary Information**

Table S1. Synthesis of performance reported in literature for isotopic analysis by LA-ICPMS of µm-sized U particles for safeguards.

Ref.	Instruments, methods	Samples, particles	Performance for <sup>235</sup> U/ <sup>238</sup> U IR		Performance for <sup>234</sup> U/ <sup>238</sup> U IR	
			Best relative IP	Best relative EP	Best relative IP	Best relative EP
11	ns-UV (213 nm) laser, quadrupole-based ICPMS, particle localization by SEM or FT	U particles $\Phi$ <1 $\mu$ m for most of them, particulate CRM IRMM "Nusimep-6" (NU) and an IAEA sample (HEU)	~10%	~20%	n.m.	n.m.
13	ns-UV (213 nm) laser, quadrupole-based ICPMS, particle localization by SEM or FT	U particles Φ ~0.6 μm, particulate CRM IRMM "Nusimep-7" (LEU) and an IAEA sample (HEU)	~7%	~10%	n.m.	n.m.
15	fs-IR (1030 nm) laser, quadrupole-based ICPMS, particle localization by SEM or FT	U particles $\Phi$ ~1-2 $\mu m,$ not certified (LEU)	~6%	n.r.	n.m.	n.m.
17	fs-UV (213 nm) laser, sector- field SC ICPMS, fast rinsing ablation cell	U particles Φ ~0.3-1.5 μm, isotopic CRM <u>NBS U010</u> (LEU), NBS U950A (NU)	~12% ~1%*	~26% ~5%*	~8% ~2%*	~48% ~10%*
12	ns-UV (193 nm) and fs-IR (795 nm) lasers, ICPMS/MC, no particle localization.	Glass particles doped with U, $\Phi$ ~10-20 $\mu m$	0.6%	-	0.7%	-
14	ns-UV (193 nm) and fs-IR (795 nm) lasers, ICPMS/MC, no particle localization (raster)	U particles Φ ~0.3-5 μm, isotopic CRM IRMM "Nusimep-7" (LEU), NBS U010 (LEU), NBS U500 (HEU), etc.	1-3%	n.m.	n.m.	n.m.
16	fs-IR (1030 nm) laser, ICPMS/MC, particle localization with optical microscope	NU particles Φ ~1-3.5 μm	0.9%	2.1%	1.2%	2.5%

Notes:

\*: by means of a correction method for sharp variations of intensity (SRF – Sharp Rejection Filter), which "smooths" the signals. Outlying count rates (the
ones that are significantly lower or higher than the previous and next ones) are replaced by average values of preceding and following count rates.
Therefore, measurement cycles which are supposedly responsible for biases on IR and poor precisions are not taken into account and replaced by
average values of the previous and next cycles. However, this smoothing of the signals also artificially improves internal precisions by adding measured
ratios in good agreement with the other ones.

n.m.: not measured; n.r.: not reported.

Table S2. Certified IRs for isotopic CRMs NBS U005, U010 and U500 and measured IRs for U-ore concentrate "Bolet" material. Ratios are atomic ratios. Uncertainties are expanded-uncertainties (95%-confidence level).

Materials	<sup>235</sup> U/ <sup>238</sup> U	<sup>234</sup> U/ <sup>238</sup> U	<sup>236</sup> U/ <sup>238</sup> U
NBS U005	(4.919±0.003)×10 <sup>-3</sup>	(2.19±0.02)×10 <sup>-5</sup>	(4.68±0.03)×10 <sup>-5</sup>
CETAMA Bolet*	(7.257±0.008)×10 <sup>-3</sup>	(5.933±0.050)×10 <sup>-5</sup>	<10 <sup>-10</sup>
NBS U010	(1.1014±0.0010)×10 <sup>-2</sup>	(5.466±0.050)×10 <sup>-5</sup>	(6.88±0.07)×10 <sup>-5</sup>
NBS U500	0.99970±0.00071	(1.042±0.001)×10 <sup>-2</sup>	(1.519±0.005)×10 <sup>-3</sup>

\* Measurements carried out by TIMS Thermo 'Triton' using µg-amounts of material.

Table S3. Average, maximal and minimal integrated numbers of counts recorded for particles of isotopic CRMs NBS U005, U010, U500 and of CETAMA "Bolet" RM.

Reference Materials	U005	Bolet	U010	U500
Number of analyzed particles	40	30	20	16
Average numbers of counts	3.8×10 <sup>5</sup>	3.1×10 <sup>5</sup>	8.1×10 <sup>5</sup>	5.6×10⁵
Maximal numbers of counts	2.1×10 <sup>6</sup>	5.5×10⁵	5.0×10 <sup>6</sup>	9.4×10 <sup>5</sup>
Minimal numbers of counts	1.9×10 <sup>4</sup>	1.3×10 <sup>5</sup>	4.2×10 <sup>4</sup>	1.8×10 <sup>5</sup>
Ratio maximal/minimal numbers of counts	108	4	118	5



Fig. S1. Graph representing the <sup>235</sup>U/<sup>238</sup>U isotopic ratios for all measurement cycles for a U particle (from NBS U500 CRM) versus the numbers of <sup>235</sup>U counts for the corresponding measurement cycle.