Electronic supplementary information

Quantification anomalies in single pulse LA-ICP-MS analysis associated with laser fluence and beam size

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Throughout the work, a LA-ICP-MS system was used with a HelEx ablation cell, either with or without Aerosol Rapid Introduction System (ARIS), to provide a fundamental basis for single pulse behavior creating a setup with two particle washout regimes and intrinsic analytical characteristics. The results of the two aerosol delivery systems cannot be compared directly but only the experiments performed for the individual systems.

Tables

Table S1 Parameters used to evaluate the signal intensity and noise contributions as a function of laser fluence (using ARIS).

	Gelatin	NIST 612
Laser fluence (J cm^{-2})	0.5-9	1-9
Repetition rate (Hz)	40	40
Scanning mode	Line scan, fixed dosage	Line scan, fixed dosage
Dosage*	1	1
Isotopes measured	^{75}As, ^{157}Gd	^{75}As, ^{157}Gd
Dwell time (ms)	25	25
Beam size (µm)	20	20
Mask shape	square	square

**Shots per position*

Table S2 Parameters to evaluate the influence of laser fluence and beam size on the signal intensity normalized to crater volume for both ARIS and HelEx.

Aerosol delivery system	ARIS	HelEx
Laser fluence (J cm^{-2})	0.5, 4 or 8	0.5, 4 or 8
Repetition rate (Hz)	20	2
Scanning mode	Line scan, fixed speed	Line scan, fixed speed
Isotopes measured	^{75}As, ^{157}Gd	^{75}As, ^{157}Gd
Dwell time (ms)	50	500
Beam size and mask shape (scanning speed)	5 µm square (300 µm s^{-1}), 10 µm square (600 µm s^{-1}), and 20 µm square (1000 µm s^{-1} [b])	35 µm square (150 µm s^{-1}), 65 µm circle[a] (260 µm s^{-1}), and 80 µm square (320 µm s^{-1})

[a]No square mask was available in the size range between 35 and 80 µm, hence a circular mask was chosen with an area (3318 µm^2), approximately half of the 80 µm square mask area (6400 µm^2).
[b]Maximum available speed of LA stage travel.

Table S3 Parameters to evaluate the effect of fluence and beam size on single shot peak profiles.

Aerosol delivery system	ARIS	HelEx
Laser fluence (J cm^{-2})	0.5 and 4	0.5 and 4
Repetition rate (Hz)	1	1
Scanning mode	Spot	Spot
Dosage (shots per position)	1	1
Isotopes measured	^{66}Zn, ^{60}Ni, ^{75}As, ^{128}Te, ^{139}La and ^{157}Gd	^{66}Zn, ^{60}Ni, ^{75}As, ^{128}Te, ^{139}La and ^{157}Gd
Dwell time (ms)	1	10
Beam size (µm)	5, 10 and 20	35, 65 and 80
Mask shape	Square	Square (35 and 80 µm), circle (65 µm)

Figures

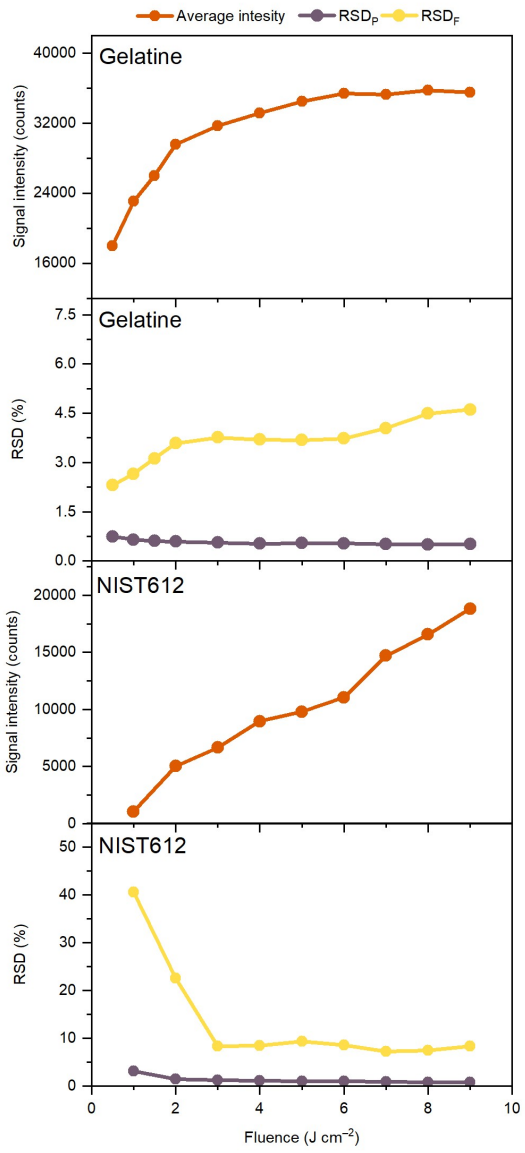

Fig. S1 Average line intensity, Poisson noise and Flicker noise as a function of fluence for single dosage experiments with ca. 50 µg g^{-1} As in gelatin and NIST SRM 612 glass standards (see Table S1 for operational conditions).

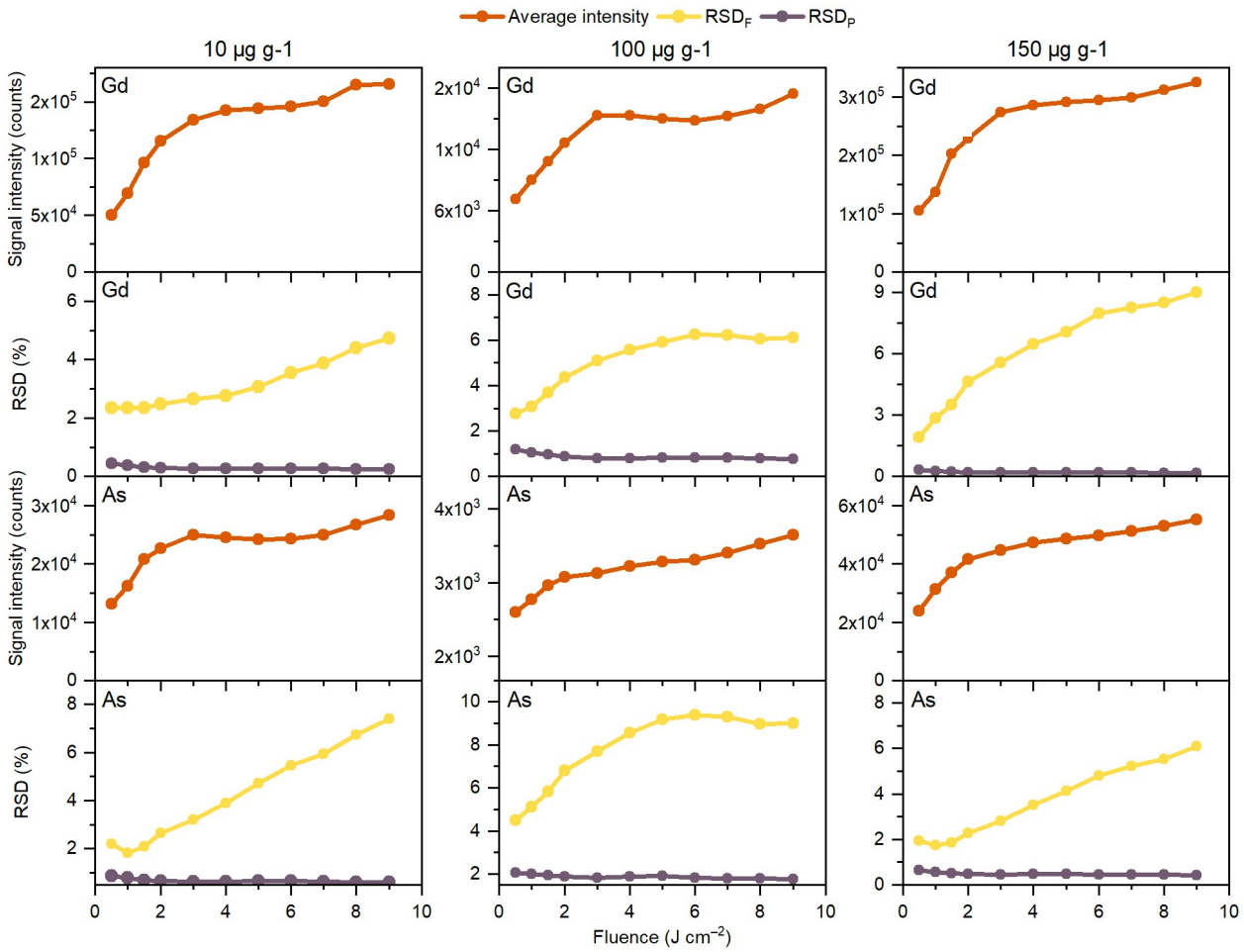

Fig. S2 Average line intensity, Poisson noise and Flicker noise as a function of the fluence for single dosage experiments for 10, 100 and 150 µg g^{-1} Gd and As in gelatin standards (see Table S1 for operational conditions).

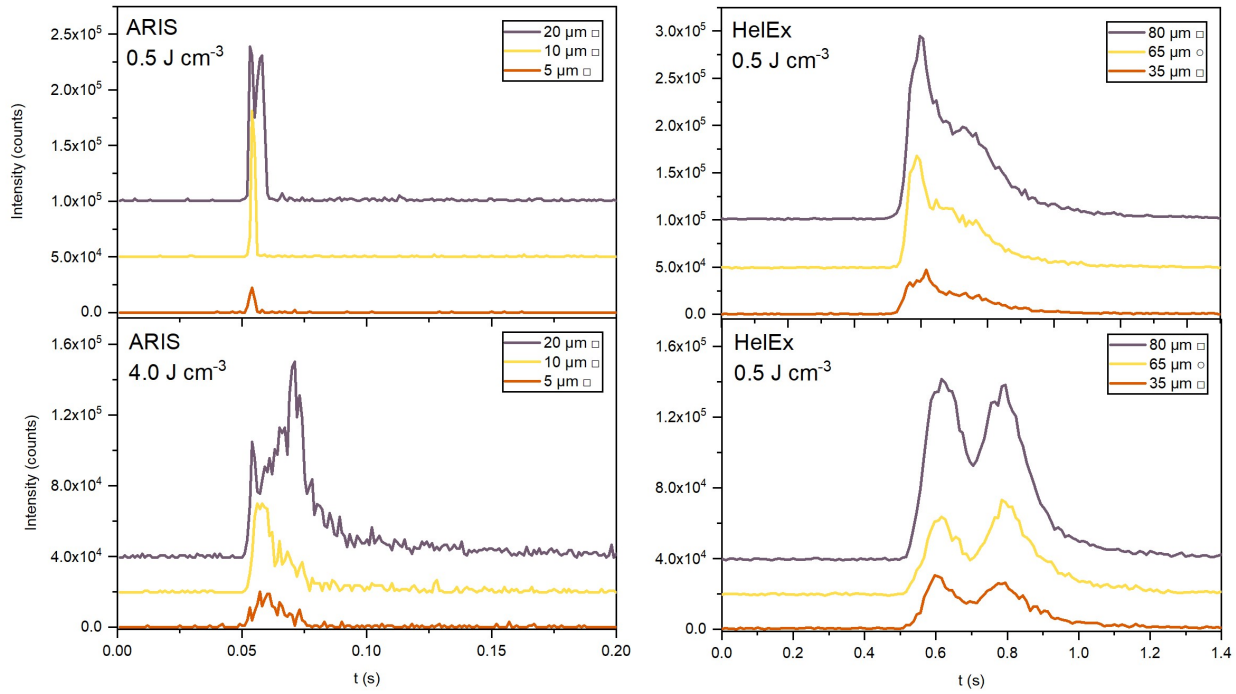

Fig. S3 Single pulse peak profiles for As in gelatin obtained using different aerosol delivery systems (ARIS of HelEx), beam sizes (5, 10 or 20 µm for ARIS and 35, 65 or 80 µm for HelEx), and laser fluences (0.5 or 4 J cm^{-2}) (see Table S3 for operational conditions).

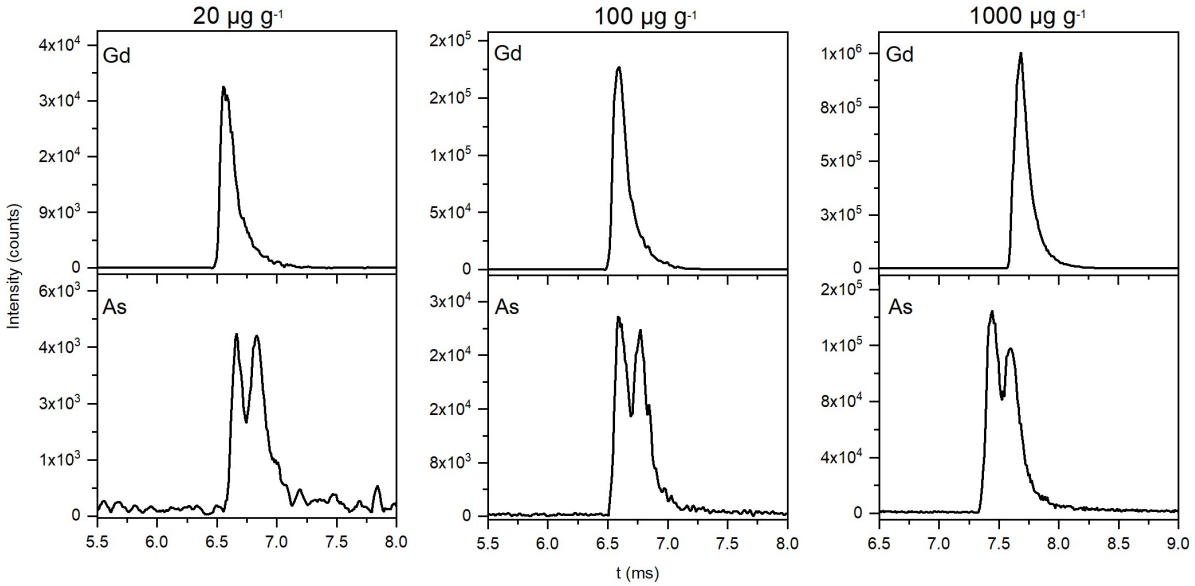

Fig. S4 Single pulse profiles for 20, 100 and 1000 µg g^{-1} Gd (single peaks) and As (double peaks) in gelatin using the HelEx setup at a fluence of 4 J cm^{-2} and a beam size of 80 µm (square mask) (see Table S3 for operational conditions).