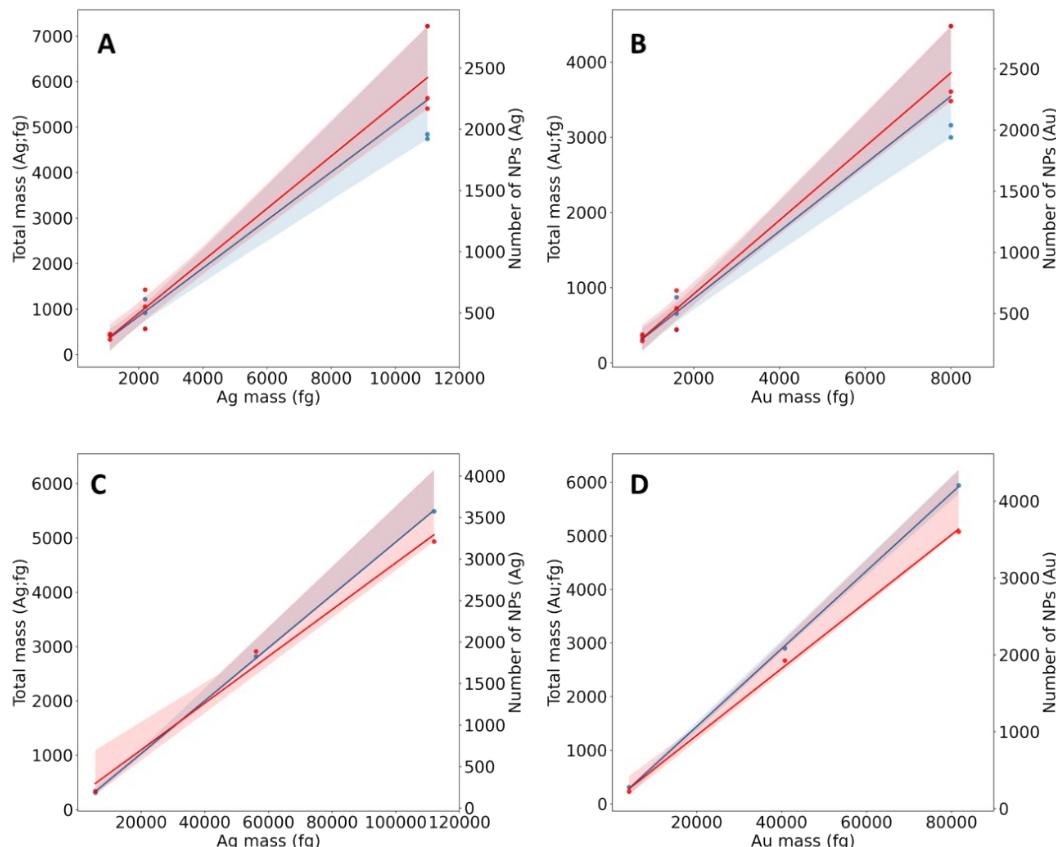


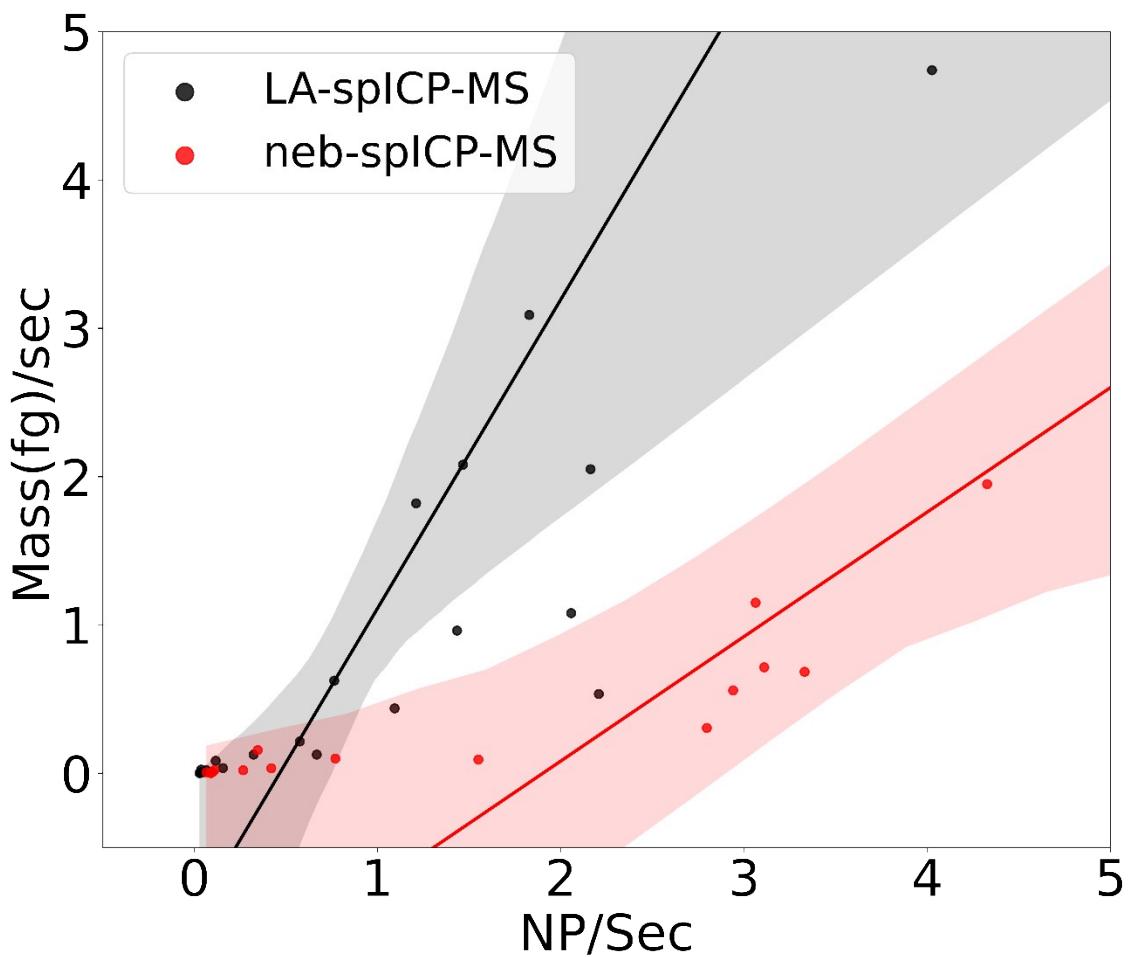
Supplementary information



SI Figure 1. Au and Ag Linearity determination of laser and nebulizer using AgAu 80nm core shell particles. Red: Number of NPs, Blue: Total mass of NPs Top laser from FFF membrane (Top A: Ag mass and number; Top B Au mass and number), Bottom Nebulizer (Bottom C: Ag mass and number; Bottom D Au mass and number. Y-axis-left: Total mass, Y-axis-right: Number of NPs,X-axis: Elemental mass measured

SI Table 1. mass limits of detection of elements chosen for analysis. LODs are reported in fg and analyzed isotopes are given in the element columns

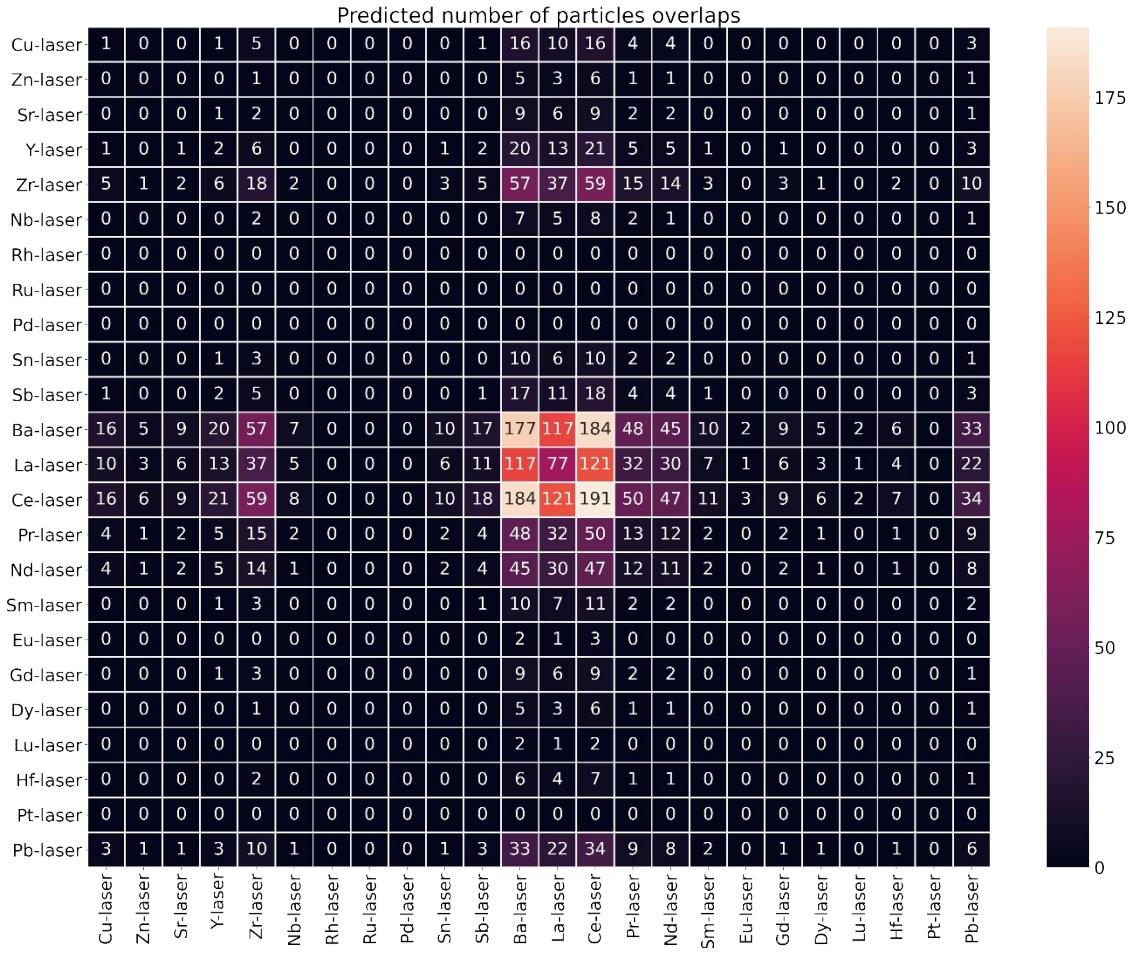
element	LOD (fg)	element	LOD (fg)
[63Cu] ⁺	0.78	[139La] ⁺	0.066
[64Zn] ⁺	0.99	[140Ce] ⁺	0.070
[88Sr] ⁺	0.17	[141Pr] ⁺	0.11
[89Y] ⁺	0.16	[144Nd] ⁺	0.056
[90Zr] ⁺	0.21	[152Sm] ⁺	0.094
[93Nb] ⁺	0.051	[153Eu] ⁺	0.091
[103Rh] ⁺	0.020	[156Gd] ⁺	0.096
[104Ru] ⁺	0.078	[163Dy] ⁺	0.088
[105Pd] ⁺	0.16	[175Lu] ⁺	0.050
[118Sn] ⁺	0.19	[178Hf] ⁺	0.036
[121Sb] ⁺	0.054	[195Pt] ⁺	0.032
[138Ba] ⁺	0.039	[208Pb] ⁺	0.0078



SI Figure 2. Comparison of the mass of particles per second vs the number of nanoparticle events per second between laser and suspension. Black: Laser ,Red: Nebulizer, X-axis: Nanoparticle per second detected, Y-axis: Sum mass of particles detected per second

SI Table 2. Statistical values for figure 3 in manuscript of the box and whisker plot. Elements are placed side-by-side for each method for comparison and describe all particle events shown in the manuscript

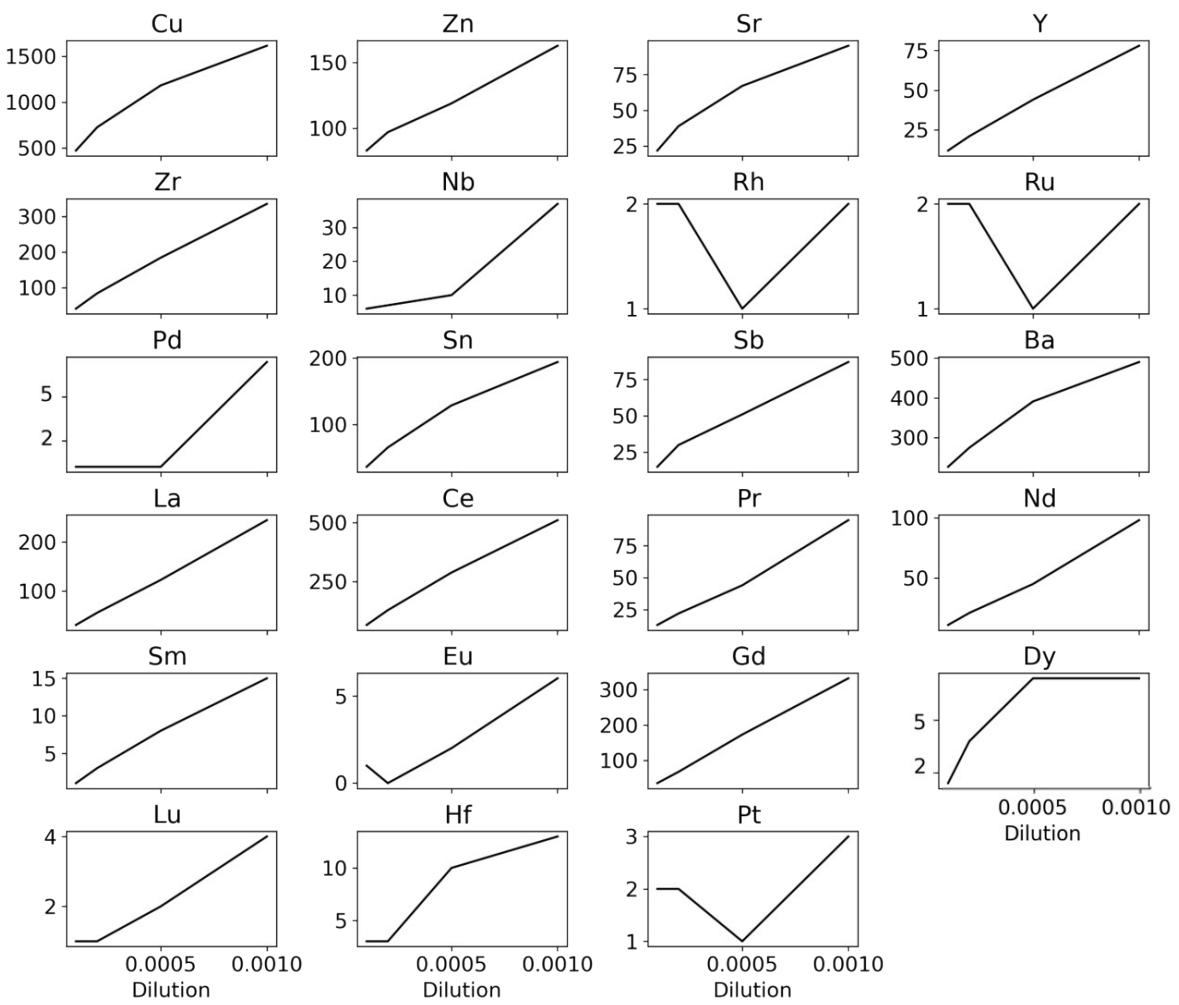
	Cu-laser	Cu-neb	Zn-laser	Zn-neb	Sr-laser	Sr-neb	Y-laser	Y-neb	Zr-laser	Zr-neb	Nb-laser	Nb-neb	Rh-laser	Rh-neb	Ru-laser	Ru-neb
count	893	1093	358	142	554	36	1250	99	3290	263	430	25	6	2	10	2
mean	1.78	1.51	1.78	1.88	0.35	0.29	0.50	0.63	0.78	0.62	0.17	0.17	0.03	0.02	0.30	0.19
std	1.71	0.87	1.11	1.12	0.47	0.10	0.68	0.85	1.06	0.63	0.30	0.25	0.01	0.00	0.32	0.07
min	0.78	0.78	0.99	0.99	0.17	0.17	0.16	0.16	0.21	0.21	0.05	0.05	0.02	0.02	0.08	0.14
25%	1.01	0.94	1.18	1.17	0.20	0.20	0.21	0.22	0.29	0.28	0.06	0.07	0.02	0.02	0.12	0.16
50%	1.31	1.21	1.43	1.44	0.25	0.26	0.30	0.36	0.45	0.39	0.09	0.08	0.02	0.02	0.19	0.19
75%	1.95	1.73	1.97	2.10	0.37	0.36	0.48	0.61	0.84	0.63	0.16	0.15	0.02	0.02	0.31	0.21
max	36.10	6.98	14.30	8.02	9.85	0.53	8.70	6.34	17.60	4.73	4.05	1.14	0.04	0.02	1.14	0.24
	Pd-laser	Pd-neb	Sn-laser	Sn-neb	Sb-laser	Sb-neb	Ba-laser	Ba-neb	La-laser	La-neb	Ce-laser	Ce-neb	Pr-laser	Pr-neb	Nd-laser	Nd-neb
count	20	3	612	160	986	349	10519	1222	6847	845	10765	1539	2879	333	2655	299
mean	0.64	0.59	0.60	0.50	0.16	0.17	0.39	0.12	0.99	0.69	1.38	0.91	0.66	0.52	0.57	0.44
std	1.02	0.32	0.77	0.48	0.19	0.27	0.97	0.12	1.79	1.23	2.90	2.11	0.88	0.62	0.75	0.52
min	0.17	0.24	0.19	0.19	0.05	0.05	0.04	0.05	0.11	0.14	0.10	0.14	0.13	0.15	0.12	0.13
25%	0.21	0.46	0.26	0.23	0.07	0.08	0.08	0.08	0.27	0.21	0.31	0.23	0.25	0.20	0.22	0.19
50%	0.27	0.68	0.35	0.32	0.10	0.09	0.14	0.09	0.48	0.32	0.58	0.35	0.39	0.30	0.34	0.26
75%	0.63	0.76	0.63	0.53	0.17	0.15	0.31	0.12	0.96	0.60	1.29	0.72	0.69	0.55	0.58	0.47
max	4.70	0.85	8.49	3.02	2.86	2.49	39.80	1.82	32.60	18.00	75.90	35.20	11.80	5.24	9.61	5.42
	Sm-laser	Sm-neb	Eu-laser	Eu-neb	Gd-laser	Gd-neb	Dy-laser	Dy-neb	Lu-laser	Lu-neb	Hf-laser	Hf-neb	Pt-laser	Pt-neb	Pb-laser	Pb-neb
count	575	65	164	19	566	319	309	50	140	17	426	24	39	5	1980	734
mean	0.29	0.23	0.20	0.16	0.31	0.44	0.29	0.28	0.14	0.12	0.07	0.05	0.09	0.26	0.15	0.08
std	0.25	0.12	0.12	0.05	0.26	0.63	0.25	0.31	0.08	0.05	0.06	0.01	0.12	0.38	0.21	0.07
min	0.11	0.11	0.11	0.12	0.12	0.13	0.12	0.12	0.08	0.08	0.04	0.04	0.03	0.03	0.01	0.04
25%	0.15	0.14	0.13	0.13	0.17	0.17	0.16	0.15	0.09	0.08	0.04	0.04	0.04	0.05	0.05	0.05
50%	0.20	0.19	0.16	0.15	0.22	0.26	0.20	0.19	0.11	0.10	0.05	0.05	0.05	0.08	0.08	0.06
75%	0.32	0.31	0.22	0.17	0.34	0.44	0.31	0.29	0.14	0.13	0.08	0.06	0.07	0.20	0.15	0.08
max	2.20	0.71	0.91	0.33	2.02	6.76	2.03	1.99	0.63	0.24	0.59	0.07	0.71	0.93	2.59	0.67



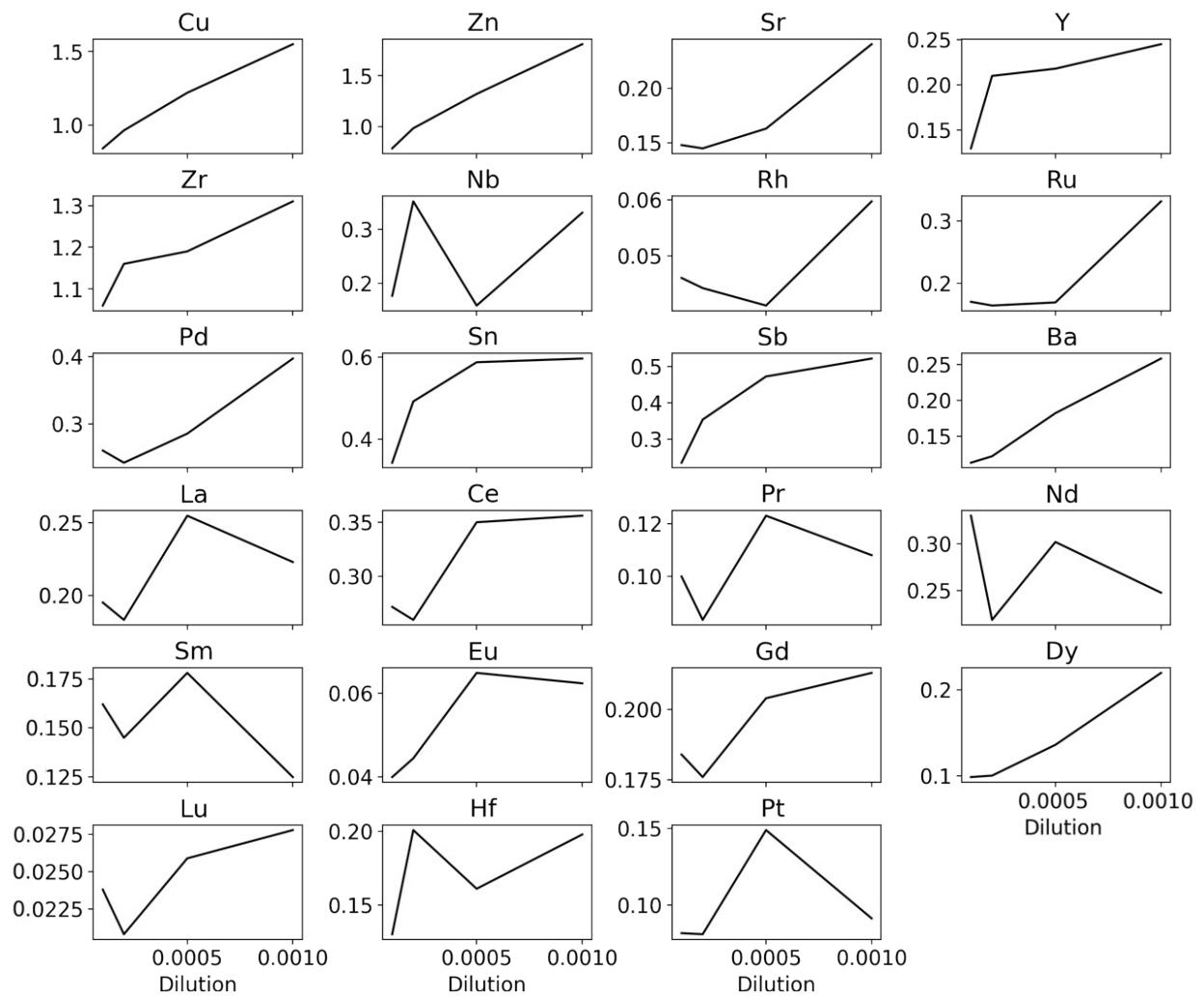
SI Figure 3. Concurrency analysis of all measured elements using random Poisson distributions. The figure depicts a square matrix with the diagonal showing the calculated number of double events for the laser undiluted extracted sediment. The intersection cells describe the likelihood of two random elemental events overlapping.

SI Table 3. Calculated particle concentrations for each element for both methods for the CPE sediment sample (NPs/ml)

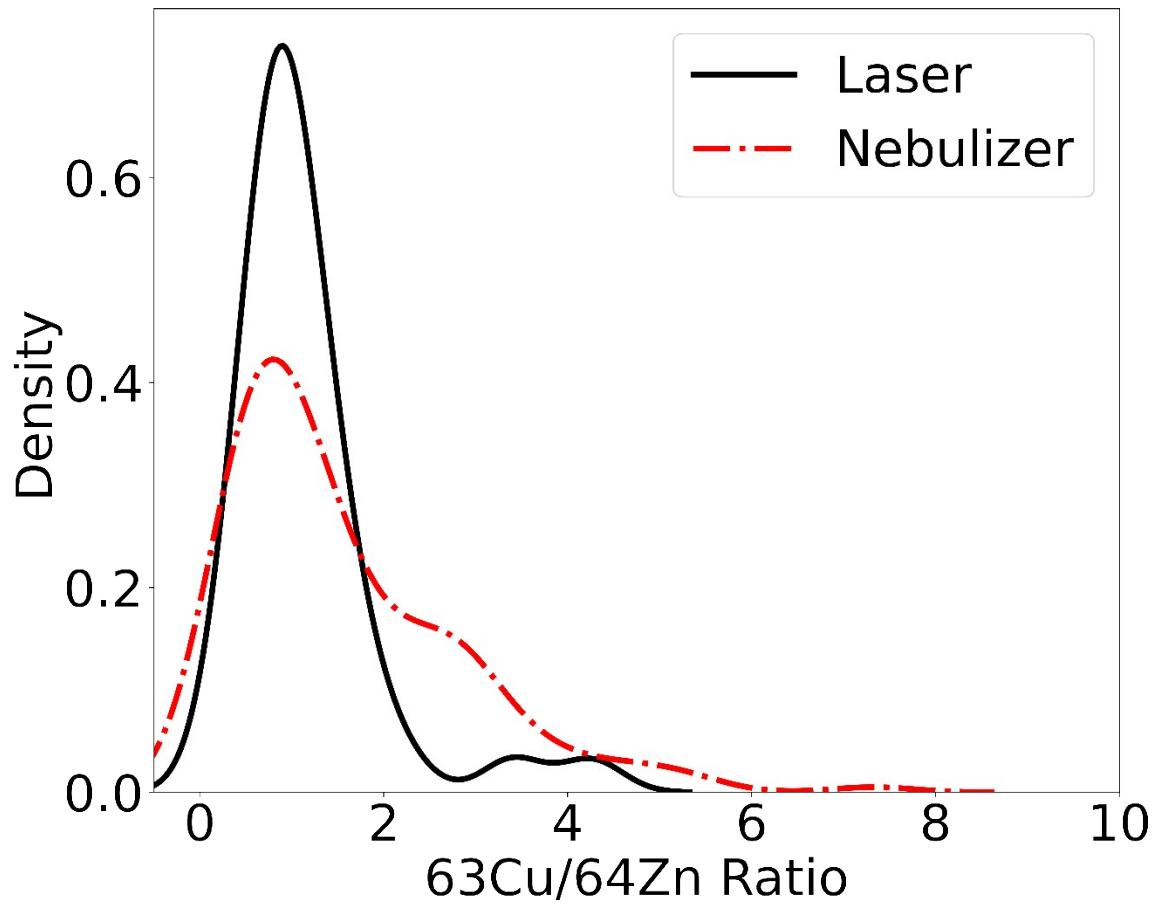
	Cu	Zn	Sr	Y	Zr	Nb	Rh	Ru
CPE-Laser	4.47E+06	1.79E+06	2.77E+06	6.25E+06	1.65E+07	2.15E+06	3.00E+04	5.00E+04
CPE-Nebulizer	4.30E+07	5.59E+06	1.42E+06	3.90E+06	1.04E+07	9.84E+05	7.87E+04	7.87E+04
	Pd	Sn	Sb	Ba	La	Ce	Pr	Nd
CPE-Laser	1.00E+05	3.06E+06	4.93E+06	5.26E+07	3.42E+07	5.38E+07	1.44E+07	1.33E+07
CPE-Nebulizer	1.18E+05	6.30E+06	1.37E+07	4.81E+07	3.33E+07	6.06E+07	1.31E+07	1.18E+07
	Sm	Eu	Gd	Dy	Lu	Hf	Pt	Pb
CPE-Laser	2.88E+06	8.20E+05	2.83E+06	1.55E+06	7.00E+05	2.13E+06	1.95E+05	9.90E+06
CPE-Nebulizer	2.56E+06	7.48E+05	1.26E+07	1.97E+06	6.69E+05	9.45E+05	1.97E+05	2.89E+07



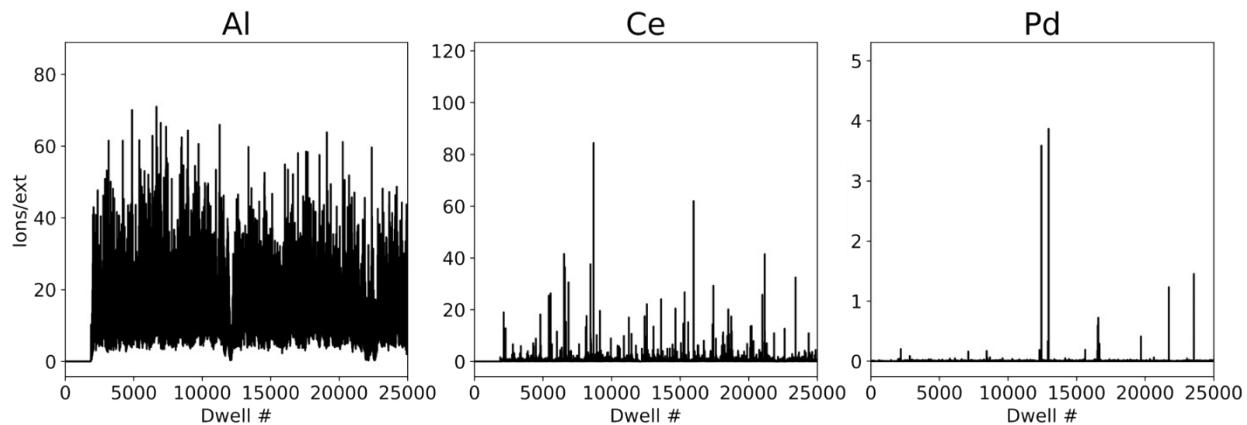
SI Figure 4 . Particle counts of sedimentation basin for dilution series of extracted nebulizer dispersion, X-axis: Dilution value, Y-axis: Nanoparticle counts



SI Figure 5. Mean particle mass (fg) of sedimentation basin for dilution series of extracted nebulizer dispersion, X-axis: Dilution value, Y-axis: Nanoparticle mean mass.



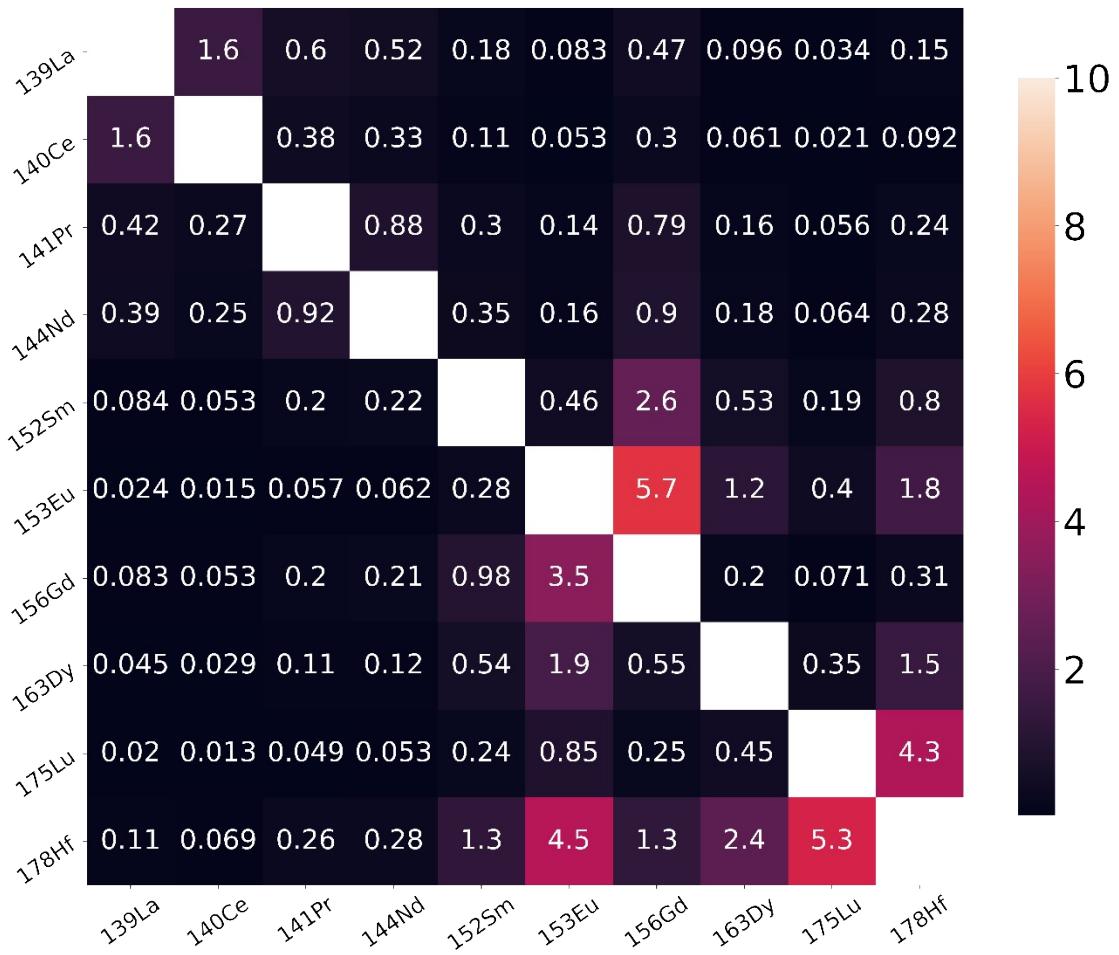
SI Figure 6. Kernel density distribution of PMD for dual elemental particles consisting of Cu and Zn. X-axis: Mass ratio of Cu to Zn in each particle, Y-axis: Calculated kernel density values.



SI Figure 7. Elemental trace of elements from sediment laser ablation without prior cloud point extraction for Al, Ce, and Pd. X-axis: dwell number, Y-axis: ions per extraction.

SI Table 4. Bulk concentrations measured by ICP-MS of digested particle extracted sediment. ug/g of soil

Al	Ti	V	Cr	Mn	Co	Cu	Zn	Sr
164.10 ± 4.1	8.60 ± 0.27	0.50 ± 0.17	3.33 ± 0.16	2.24 ± 0.051	0.12 ± 0.0034	6.67 ± 0.17	6.99 ± 0.23	0.81 ± 0.0075
Y	Zr	Mo	Nb	Rh	Ru	Pd	Ag	Cd
0.57 ± 0.0024	1.04 ± 0.029	0.75 ± 0.017	0.17 ± 0.0017	0.04 ± 0.00066	0.43 ± 0.026	0.90 ± 0.076	2.57 ± 0.15	0.20 ± 0.00003
Sn	Sb	Ba	La	Ce	Pr	Nd	Sm	Eu
1.14 ± 0.0075	0.60 ± 0.019	3.19 ± 0.017	0.63 ± 0.0072	0.93 ± 0.015	0.49 ± 0.050	0.63 ± 0.14	0.50 ± 0.0025	0.45 ± 0.040
Gd	Dy	Lu	Hf	W	Pt	Pb	U	
0.61 ± 0.0030	0.37 ± 0.0056	0.26 ± 0.0061	0.24 ± 0.0018	0.39 ± 0.021	0.01 ± 0.0023	2.22 ± 0.0049	0.34 ± 0.010	



SI Figure 8. Dual half square matrix of elemental ratios for LA-splCP-ToF-MS for extracted sediment proxy sample (Bottom left half of the matrix), and unextracted sediment sample (Top right half of the matrix). Color indicates the ratio of particle counts to its intersection point