Supplement of Atmos. Chem. Phys., 15, 4117–4130, 2015
http://www.atmos-chem-phys.net/15/4117/2015/
doi:10.5194/acp-15-4117-2015-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

Characteristics of trace metals in traffic-derived particles in Hsuehshan Tunnel, Taiwan: size distribution, potential source, and fingerprinting metal ratio

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This Supporting Information material contains three tables and three figures. Table S1 reveals potential sources of airborne PM metals in different sized PM reported by previous studies. Table S2 lists the recoveries and precisions of each PM metal analyzed by ICP-MS, while Table S3 shows correlation matrix of metals in submicron PM. Three figures contained in Supporting Information include (1) average size distributions of Al, Ca, Mg and K as well as La and Ce collected at both the inlet and outlet sites of Hsuehshan Tunnel (Figure S1), (2) size distributions of traffic-related elements in all sets collected at both the two sites of Hsuehshan Tunnel (Figure S2), and (3) size distributions of some crustal and REEs in all sets collected at both the two sites of Hsuehshan Tunnel (Figure S3).

Table S1 Potential sources of airborne particulate metals in different particle sizes reported in the literatures.

Metals	Particle Size	Potential Source	Reference[a]
Si, Fe, Ca, Na, Mg, Al and K	Coarse	Road dust	1,2,3
Ca and K	Submicron	Tail pipe emission *-lubricating oil* *-volatile from K-compound*	4,5,6
Pb, Zn, Ni and V	Submicron	Pipe emissions *-gasoline and diesel engines*	3,7,8
Fe, Ba, Mn, Cu, Mo, Cd, Sb, Ti, V, Ga, Zn and Pb	Coarse	Wear dust *-brake linings* *-tire wear*	9,10,11,12
La, Ce, Pr ,Nd, Sm	Fine	Automobile catalyst	13,14

[a]1.Zhang et al. (2014); 2.Ho et al. (2003); 3.Lin et al. (2005); 4.Hee and Filip, (2005); 5.Iijima et al. (2007); 6.Kuo et al. (2009); 7.Wang et al. (2003); 8.Shafer et al. (2012); 9.Lough et al. (2005); 10.Grieshop et al. (2006); 11.Thorpe and Harrison (2008); 12. Wåhlin et al. (2006); 13.Huang et al. (1994); 14.Kulkarni et al. (2006).

Table S2

Recoveries and precisions of trace metals analyzed by ICP-MS compared to certified or reference values of SRM 1648.

Elements[a]	Certified value (μg/g)	Measured value (μg/g, N=7)	Accuracy (%, N=7)	Precision (%, N=7)	MDLs (ng/m^3, N=7)
Al	34200	35345 ± 894	103	3	2.09
Fe	39100	38603 ± 534	99	1	2.67
Na	4250	4353 ± 457	102	1	7.19
Mg*	8000	8377 ± 474	105	6	0.57
K	10500	9982 ± 119	95	1	3.36
Ba*	737	734 ± 26	100	4	0.03
Ti*	4000	3962 ± 113	99	3	0.04
Mn	786	787 ± 37	100	5	0.08
Co*	18	18± 0.4	98	2	0.003
Ni	82	82 ± 2	100	3	0.07
Cu	609	577 ± 11	95	2	0.18
Zn	4760	4411 ± 114	93	3	0.31
Cd	75	73 ± 3	98	4	0.009
Sb*	45	49 ± 0.5	109	1	0.08
Pb	6550	6621 ± 293	101	4	0.05
V	127	126 ± 2	99	2	0.03
Cr	403	389 ± 5	96	1	0.22
As	115	126 ± 2	110	1	0.07
Se	27	27 ± 1.4	100	5	0.05
Cs*	3	3 ± 0.1	105	3	0.01
Ce*	55	54 ± 2.6	98	5	0.006
U	5.5	5.5 ± 0.3	100	6	0.00004

[a.] * Reference value reported by NIST.

Table S3

Correlation matrix of selected elements in submicron particles observed in Hsuehshan Tunnel. Correlation coefficient higher than 0.8 is marked in bold.

	Al	Fe	Mg	K	Ca	Sr	Ba	Ti	Mn	Ni	Cu	Zn	Mo	Cd	Sn	Sb	Pb	V	Cr	Rb	Cs	Ga	La	Ce	Pr	Nd
Al	1.00																									
Fe	0.20	1.00																								
Mg	0.23	0.77	1.00																							
K	0.17	0.69	0.59	1.00																						
Ca	0.39	0.71	**0.82**	0.39	1.00																					
Sr	0.41	0.54	0.58	0.31	0.75	1.00																				
Ba	0.20	**0.91**	0.79	0.53	0.70	0.63	1.00																			
Ti	0.09	0.76	0.56	0.43	0.51	0.35	0.74	1.00																		
Mn	0.08	0.63	0.32	0.61	0.36	0.14	0.27	0.38	1.00																	
Ni	0.43	-0.06	0.09	0.06	0.15	0.34	-0.02	0.04	-0.13	1.00																
Cu	0.06	**0.90**	0.74	0.51	0.62	0.53	**0.97**	0.78	0.29	-0.07	1.00															
Zn	-0.02	0.58	0.26	0.58	0.27	0.08	0.23	0.41	**0.98**	-0.15	0.26	1.00														
Mo	0.04	**0.88**	0.73	0.51	0.66	0.63	**0.94**	0.71	0.29	-0.06	**0.96**	0.27	1.00													
Cd	0.05	0.77	0.44	0.70	0.43	0.30	0.49	0.60	**0.91**	0.00	0.52	**0.93**	0.51	1.00												
Sn	0.06	**0.87**	0.72	0.52	0.67	0.63	**0.94**	0.78	0.27	-0.05	**0.96**	0.24	**0.95**	0.52	1.00											
Sb	0.07	**0.87**	0.69	0.57	0.55	0.51	**0.92**	0.72	0.29	-0.03	**0.94**	0.25	**0.88**	0.53	**0.93**	1.00										
Pb	0.23	0.68	0.44	0.68	0.50	0.30	0.38	0.40	**0.84**	-0.02	0.35	0.77	0.35	**0.82**	0.40	0.45	1.00									
V	-0.04	-0.12	-0.16	0.01	-0.25	0.01	-0.04	-0.04	-0.24	0.50	-0.02	-0.18	-0.03	0.00	0.01	0.05	-0.18	1.00								
Cr	-0.10	0.16	-0.03	0.18	-0.02	-0.06	0.11	0.44	0.16	-0.10	0.15	0.33	0.09	0.36	0.15	0.11	0.16	0.11	1.00							
Rb	0.31	0.64	0.65	0.72	0.57	0.32	0.43	0.38	0.65	0.09	0.37	0.56	0.36	0.65	0.40	0.47	**0.89**	-0.15	0.07	1.00						
Cs	0.21	0.32	0.64	0.31	0.43	0.26	0.28	0.17	0.15	0.24	0.23	0.11	0.19	0.23	0.21	0.32	0.42	0.05	-0.02	0.73	1.00					
Ga	0.13	**0.92**	**0.82**	0.56	0.74	0.64	**0.98**	0.75	0.33	-0.01	**0.95**	0.29	**0.92**	0.54	**0.93**	**0.91**	0.43	-0.02	0.13	0.48	0.32	1.00				
La	0.39	0.78	0.68	0.43	**0.80**	0.71	**0.80**	0.56	0.30	0.06	0.74	0.22	0.79	0.41	0.76	0.67	0.45	-0.10	0.02	0.46	0.23	**0.80**	1.00			
Ce	0.11	0.73	0.65	0.39	0.71	0.72	**0.86**	0.57	0.08	-0.04	**0.84**	0.02	**0.90**	0.27	**0.89**	0.79	0.29	-0.11	0.01	0.32	0.15	**0.84**	**0.84**	1.00		
Pr	0.26	0.49	0.42	0.38	0.38	0.28	0.50	0.35	0.11	0.03	0.53	0.07	0.58	0.18	0.45	0.47	0.17	-0.16	-0.04	0.24	0.09	0.46	0.64	0.55	1.00	
Nd	0.16	0.76	0.67	0.41	0.74	0.73	**0.88**	0.59	0.10	-0.01	**0.85**	0.04	**0.90**	0.29	**0.89**	**0.80**	0.31	-0.11	0.01	0.34	0.16	**0.86**	**0.87**	**1.00**	0.57	1.00

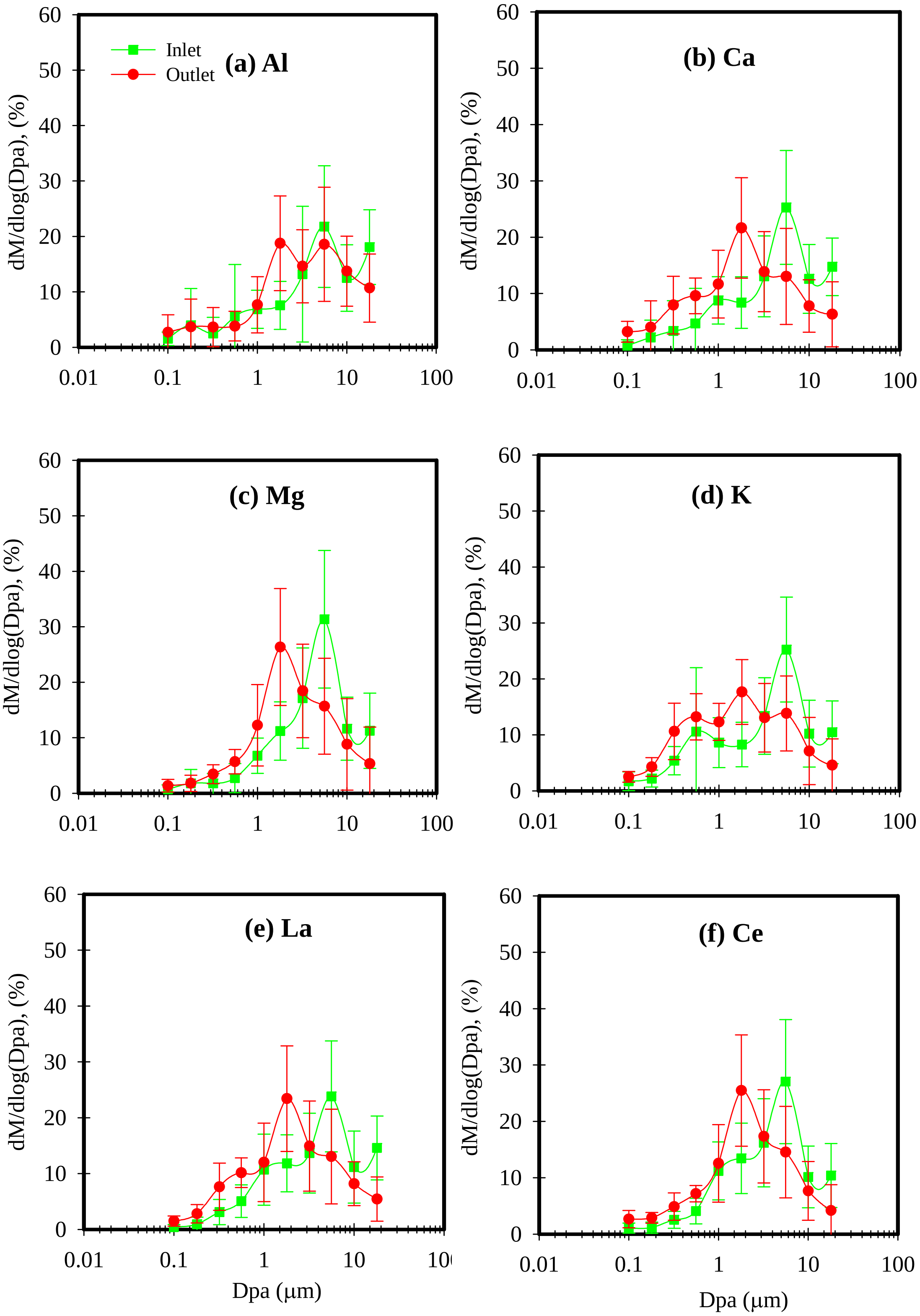

Figure S1. Average size distributions of Al, Ca, Mg, K as well as La and Ce observed at the inlet and outlet sites inside Hsuehshan Tunnel.

Inlet site

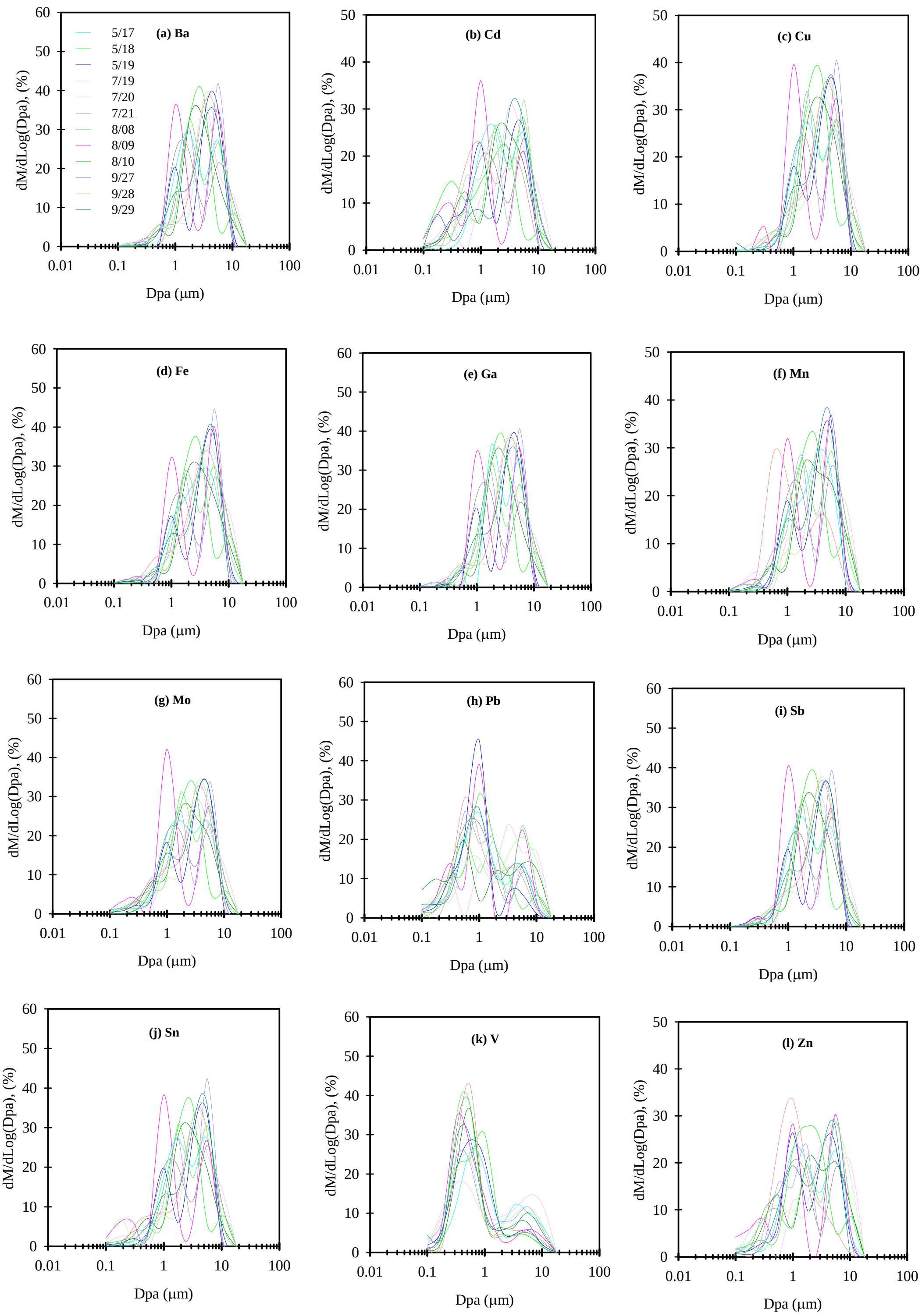

Figure S2. Size distributions of traffic-derived elements in all sets of size-resolved samples collected at both the inlet and outlet sites in Hsuehshan Tunnel.

Outlet site

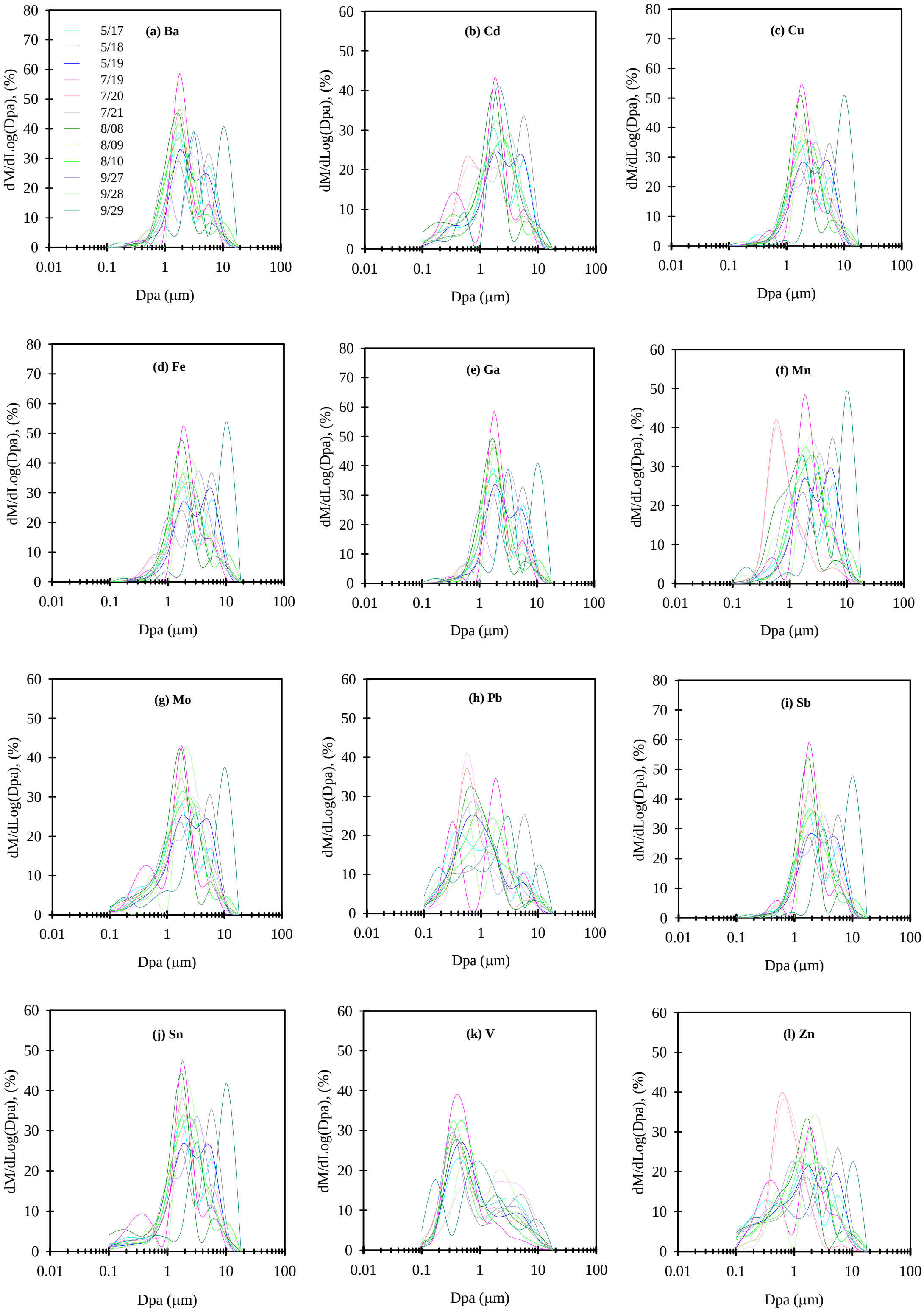

Figure S2 *(continued)*

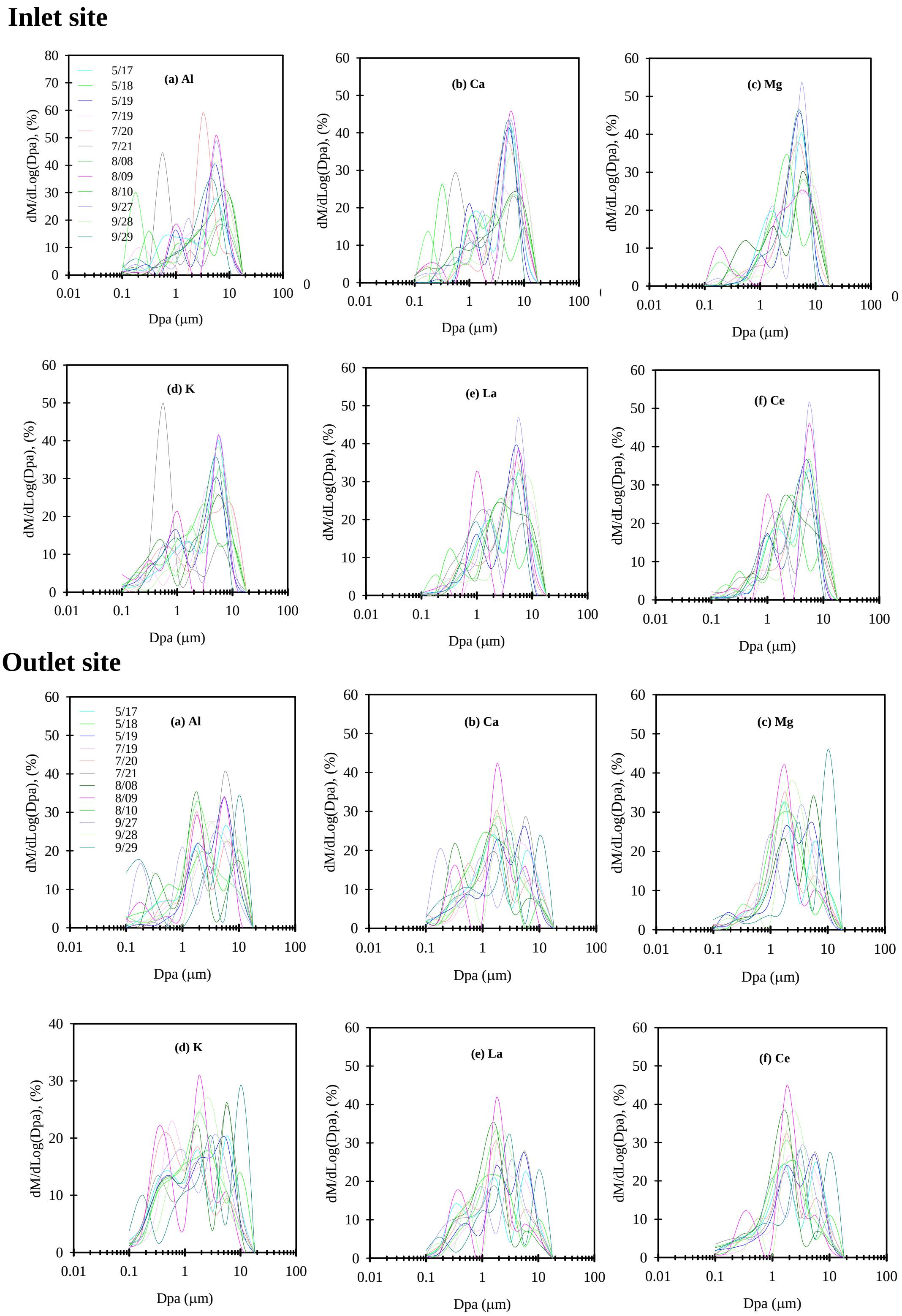

Figure S3 Size distributions of major crustal elements (Al, Ca, Mg and K) and rare earth elements (La and Ce) of all size-resolved samples collected in Hsuehshan Tunnel.

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