

Supplementary material

Atmospheric Signature and Potential Sources of Rare Earth Elements in Size-resolved Particulate Matter in a Megacity of China

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Table S1 Concentrations of REE in TSP, PM₁₀ and PM_{2.5} collected at urban site in Hangzhou. (Unit: ng m⁻³)

Species	ZH		
	TSP	PM ₁₀	PM _{2.5}
Y	1.27±0.59	0.94±0.41	0.32±0.13
La	2.45±0.91	2.03±0.80	0.64±0.23
Ce	4.51±1.77	3.64±1.56	1.08±0.47
Pr	0.50±0.21	0.41±0.16	0.21±0.00
Nd	1.99±0.89	1.49±0.68	0.38±0.14
Sm	0.46±0.20	0.39±0.15	0.20±0.01
Eu	0.07±0.03	0.05±0.02	0.01±0.01
Gd	0.31±0.14	0.23±0.10	0.05±0.02
Tb	0.04±0.02	0.03±0.01	0.01±0.00
Dy	0.21±0.09	0.16±0.07	0.04±0.02
Ho	0.04±0.02	0.03±0.01	0.01±0.00
Er	0.11±0.04	0.08±0.04	0.03±0.01
Tm	0.01±0.01	0.01±0.01	0.01±0.00
Yb	0.09±0.04	0.07±0.03	0.03±0.01
Lu	0.01±0.01	0.01±0.01	0.01±0.00
∑REE	11.98±4.87	9.37±4.02	2.24±1.07
LREE	9.89±3.98	7.80±3.37	2.02±0.89
HREE	0.82±0.35	0.63±0.27	0.12±0.07
LREE/HREE	12.27±1.56	12.38±1.80	20.31±13.46
LREE/∑REE	0.83±0.02	0.83±0.02	0.91±0.06
La/Ce	0.55±0.06	0.57±0.10	0.57±0.18
La/Sm	6.09±1.45	6.62±1.87	4.49±0.64

Table S2 Correlations matrix for REEs in TSP samples collected at ZH and ZWY site.

	Y	La	Ce	Pr	Nd	Sm	Eu	Ga	Tb	Dy	Ho	Er	Tm	Yb	Lu	
ZH	Y	1	.886**	.896**	.937**	.931**	.820**	.977**	.935**	.928**	.961**	.953**	.963**	.955**	.961**	.964**
	La		1	.947**	.917**	.911**	.757**	.883**	.913**	.920**	.913**	.909**	.911**	.913**	.900**	.895**
	Ce			1	.956**	.952**	.778**	.896**	.965**	.957**	.956**	.947**	.955**	.946**	.928**	.922**
	Pr				1	.996**	.849**	.933**	.987**	.953**	.977**	.981**	.973**	.955**	.949**	.947**
	Nd					1	.834**	.933**	.987**	.952**	.971**	.973**	.967**	.945**	.940**	.936**
	Sm						1	.804**	.812**	.811**	.849**	.814**	.806**	.799**	.786**	.797**
	Eu							1	.935**	.923**	.948**	.933**	.951**	.930**	.923**	.939**
	Ga								1	.969**	.981**	.976**	.977**	.958**	.951**	.945**
	Tb									1	.979**	.967**	.967**	.969**	.957**	.948**
	Dy										1	.986**	.990**	.985**	.976**	.974**
	Ho											1	.985**	.984**	.979**	.980**
	Er												1	.985**	.978**	.980**
	Tm													1	.985**	.981**
	Yb														1	.982**
	Lu															1
ZWY	Y	1	.877**	.912**	.955**	.963**	0.498	.986**	.974**	.973**	.985**	.986**	.982**	.985**	.982**	.979**
	La		1	.958**	.843**	.904**	0.356	.892**	.898**	.922**	.915**	.913**	.882**	.907**	.888**	.886**
	Ce			1	.912**	.947**	0.378	.921**	.940**	.929**	.942**	.936**	.924**	.933**	.924**	.916**
	Pr				1	.996**	.607*	.953**	.981**	.968**	.972**	.973**	.969**	.962**	.956**	.923**
	Nd					1	.617*	.968**	.992**	.978**	.983**	.982**	.971**	.964**	.967**	.952**
	Sm						1	0.501	.640*	.551*	.536*	0.512	0.485	0.474	0.505	0.461
	Eu							1	.970**	.969**	.975**	.972**	.967**	.968**	.959**	.954**
	Ga								1	.971**	.986**	.986**	.981**	.971**	.975**	.967**

Tb									1	.987**	.979**	.969**	.970**	.967**	.965**
Dy										1	.989**	.989**	.983**	.979**	.982**
Ho											1	.985**	.984**	.991**	.984**
Er												1	.986**	.976**	.982**
Tm													1	.975**	.984**
Yb														1	.986**
Lu															1

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

Table S3 Correlations matrix for REEs in PM₁₀ samples collected at ZH and ZWY site.

ZH	Y	La	Ce	Pr	Nd	Sm	Eu	Ga	Tb	Dy	Ho	Er	Tm	Yb	Lu
Y	1	.868**	.902**	.943**	.952**	.792**	.969**	.960**	.950**	.968**	.966**	.978**	.970**	.972**	.958**
La		1	.905**	.838**	.868**	.635**	.860**	.857**	.850**	.873**	.889**	.892**	.868**	.885**	.901**
Ce			1	.912**	.940**	.711**	.909**	.933**	.925**	.930**	.946**	.940**	.910**	.921**	.915**
Pr				1	.995**	.853**	.923**	.992**	.961**	.980**	.981**	.980**	.957**	.970**	.964**
Nd					1	.841**	.943**	.993**	.967**	.980**	.984**	.980**	.957**	.970**	.955**
Sm						1	.777**	.823**	.802**	.826**	.833**	.827**	.767**	.782**	.780**
Eu							1	.952**	.954**	.961**	.960**	.963**	.946**	.952**	.940**
Ga								1	.971**	.981**	.984**	.981**	.962**	.973**	.958**
Tb									1	.984**	.976**	.976**	.956**	.959**	.948**
Dy										1	.992**	.993**	.983**	.981**	.972**
Ho											1	.989**	.973**	.977**	.971**
Er												1	.982**	.985**	.974**
Tm													1	.975**	.973**

																		1	.981**
																			1
ZWY	Y	1	.834**	.919**	.924**	.975**	.792*	.992**	.987**	.964**	.976**	.987**	.982**	.954**	.983**	.973**			
	La		1	.940**	0.592	.871**	0.313	.855**	.853**	.815**	.867**	.877**	.807**	.878**	.857**	.877**			
	Ce			1	.829**	.948**	0.581	.939**	.939**	.914**	.955**	.948**	.911**	.934**	.941**	.940**			
	Pr				1	.991**	.891**	.939**	.982**	.987**	.951**	.945**	.913**	.821**	.916**	.881**			
	Nd					1	.895**	.979**	.992**	.982**	.989**	.991**	.977**	.956**	.974**	.969**			
	Sm						1	.756*	.866*	.828*	.791*	.793*	0.723	0.489	.759*	0.694			
	Eu							1	.988**	.975**	.984**	.985**	.983**	.955**	.975**	.961**			
	Ga								1	.982**	.988**	.987**	.974**	.946**	.973**	.960**			
	Tb									1	.979**	.970**	.955**	.947**	.949**	.950**			
	Dy										1	.990**	.983**	.965**	.980**	.970**			
	Ho											1	.994**	.964**	.988**	.984**			
	Er												1	.952**	.982**	.971**			
	Tm													1	.953**	.967**			
	Yb														1	.984**			
	Lu															1			

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

Table S4 Correlations matrix for REEs in PM_{2.5} samples collected at ZH and ZWY site.

ZH	Y	La	Ce	Pr	Nd	Sm	Eu	Ga	Tb	Dy	Ho	Er	Tm	Yb	Lu	
Y	1	0.403	0.512	.a	.763**	1.000**	.706**	.803**	.898**	.828**	.758**	.954**	.866**	.966**	.662**	
La		1	.737**	.a	.495**	1.000**	.582**	.553**	.599**	.654**	.636**	.763**	.566**	0.441	.661**	
Ce			1	.a	.682**	1.000**	.724**	.717**	.684**	.791**	.770**	.624*	.684**	.747**	.568**	
Pr				.a	.a	.a	.a	.a	.a	.a	.a	.a	.a	.a	.a	
Nd					1	1.000**	.917**	.959**	.818**	.942**	.914**	.948**	.905**	.945**	.827**	
Sm						1	1.000**	1.000*	1.000*	1.000*	1.000*	1.000*	1.000*	1.000*	1.000*	
								*	*	*	*	*	*	*	*	
Eu							1	.949**	.781**	.920**	.916**	.936**	.889**	.906**	.828**	
Ga								1	.848**	.953**	.932**	.928**	.891**	.902**	.857**	
Tb									1	.904**	.833**	.799**	.816**	.886**	.699**	
Dy										1	.940**	.944**	.914**	.973**	.851**	
Ho											1	.917**	.891**	.963**	.833**	
Er												1	.950**	.949**	.855**	
Tm													1	.967**	.817**	
Yb														1	.881**	
Lu															1	
ZWY	Y	1	-0.069	0.097	.a	.872**	.a	.913**	.896**	.979**	.922**	.953**	.910**	.940**	.980**	.730*
	La		1	.686**	.a	.591*	.a	.674**	.600**	.611**	.599**	.607**	-0.2	.584**	-0.286	0.445
	Ce			1	.a	.639**	.a	.749**	.718**	.696**	.640**	.742**	0.255	.518*	-0.021	0.149
	Pr				.a	.a	.a	.a	.a	.a	.a	.a	.a	.a	.a	.a
	Nd					1	.a	.950**	.950**	.883**	.951**	.934**	.872*	.911**	0.791	.758**
	Sm						.a	.a	.a	.a	.a	.a	.a	.a	.a	.a
	Eu							1	.876**	.819**	.861**	.904**	.923**	.896**	.911*	.697**

Ga	1	.902**	.956**	.964**	.949**	.896**	.836*	.675**
Tb		1	.946**	.922**	.893**	.831**	.970**	.660**
Dy			1	.968**	.992**	.919**	.925**	.698**
Ho				1	.989**	.927**	.936**	.668**
Er					1	.974**	.923**	0.387
Tm						1	.978**	.779**
Yb							1	0.58
Lu								1

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

a Cannot be computed because at least one of the variables is constant.

Table S5 The total concentrations of REEs in atmospheric particles around the world from literatures data. (Unit: ng m⁻³).

	Cities	Periods	Size	Total REE	LREE	HREE	Reference
China	Hangzhou	2011-2012	TSP	11.98	9.89	0.82	This study
	Beijing	February to June,1998	APM	53.02	49.18	3.84	(Wang, 2001)
	Wuhan		TSP	125.51	119.65	5.86	(Shuai Qin et al., 2005)
	Bayan Obo mine region	March 2013	TSP	239.64	228.96	10.68	(Wang and Liang, 2014)
Netherlands	Delft	July to September, 1997	PM ₁₀	4.35	4.04	0.31	(Wang, 2001)
Canada	Burnaby	2006–2008	PM _{2.5}	0.2434	0.2323	0.0111	(Celo et al., 2012)
	Abbotsford	2005–2008	PM _{2.5}	0.0986	0.0875	0.0111	
	Windsor	2006–2008	PM _{2.5}	0.1548	0.1412	0.0136	
	Toronto	2005–2008	PM _{2.5}	0.1514	0.1384	0.013	
	Wallaceburg	2005–2008	PM _{2.5}	0.1152	0.1036	0.0116	
	Montreal	2005–2007	PM _{2.5}	0.1431	0.131	0.0121	
	Canterbury	2005–2007	PM _{2.5}	0.059	0.0479	0.0111	
	Halifax	2006–2008	PM _{2.5}	0.3003	0.2879	0.0124	
America	E.Houston	08/30/2006-09/20/2006	PM _{2.5}	1.668	1.574	0.094	(Kulkarni et al., 2007)
Spain	Puertollano (central Spain)	22/1/2004-1/3/2005	PM ₁₀	2.11	1.95	0.16	(Querol X et al., 2004)
	Sagrera(Barcelona)	January to December 2001	PM ₁₀	3.03	2.67	0.36	
	Llodio		PM ₁₀	1.13	0.91	0.22	
	Bemantes		PM ₁₀	0.86	0.66	0.2	
	Alcobendas		PM ₁₀	1.63	1.38	0.25	
	Huelva		PM ₁₀	2.52	2.04	0.48	
	Canaries		PM ₁₀	2.52	2.24	0.28	
	Tarragona		PM ₁₀	1.6	1.28	0.32	

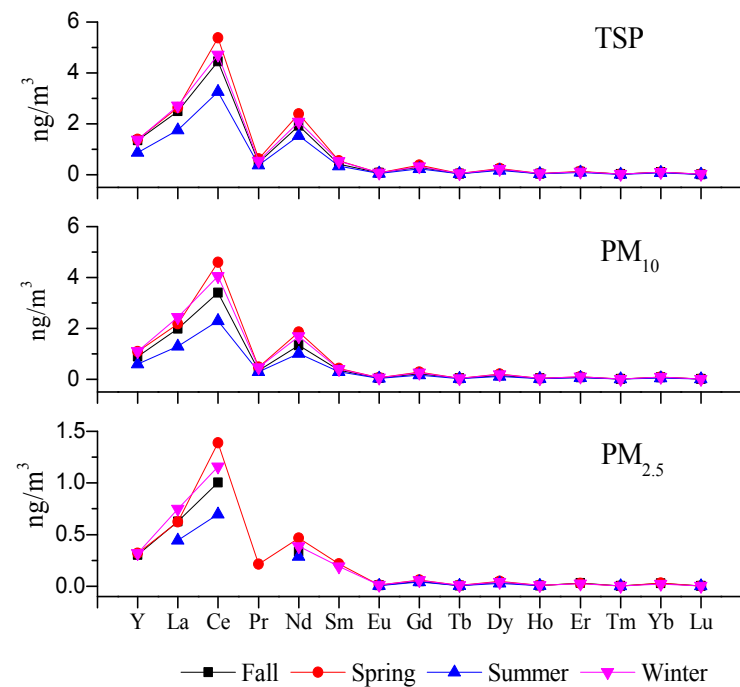


Figure S1 Seasonal variation of REE concentrations in size-resolved particle samples collected at ZH site.

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