

Supplementary Material for

Source Apportionment of PM_{2.5} using DN-PMF in Three Megacities in South Korea

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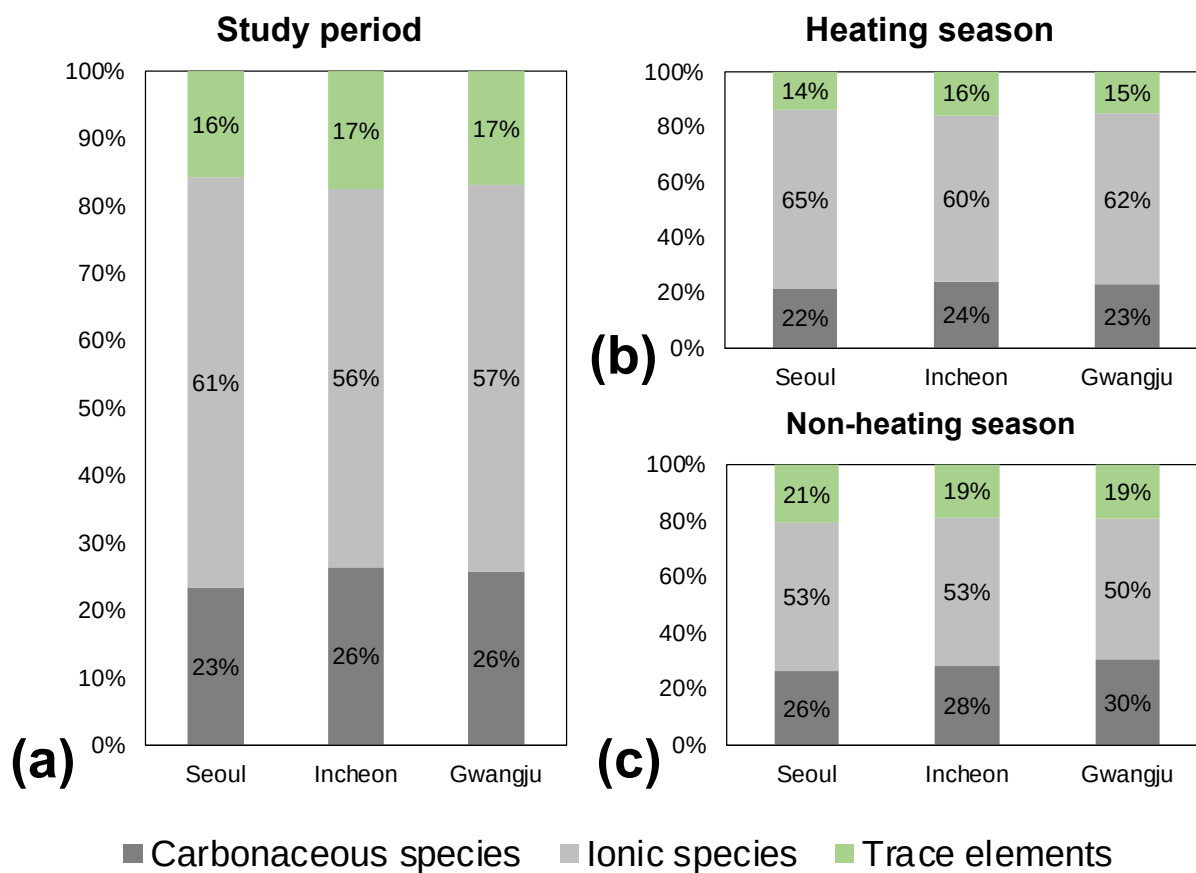


Figure S1. Chemical fractions of PM_{2.5} by (a) study period, (b) heating season, and (c) non-heating season.

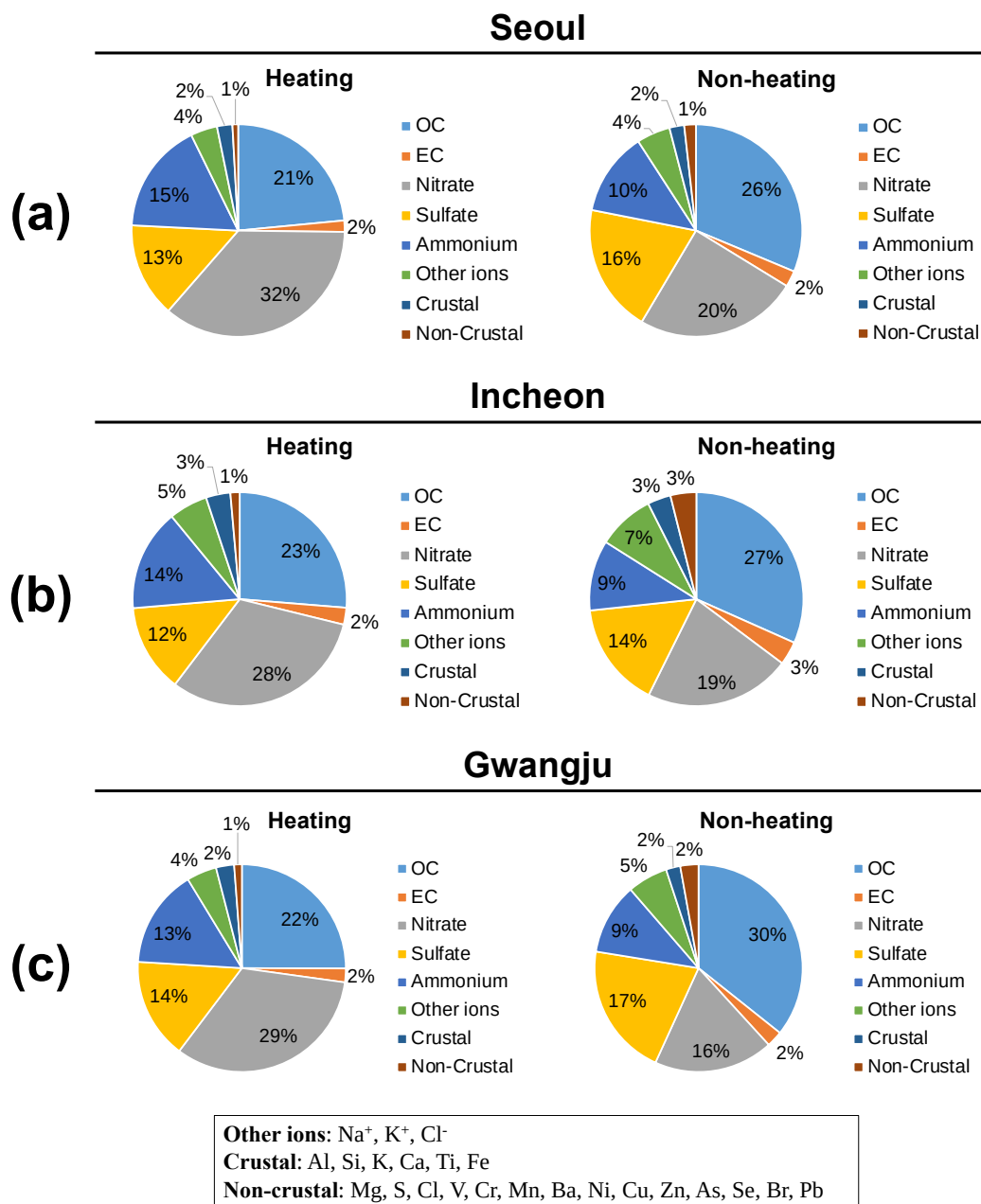


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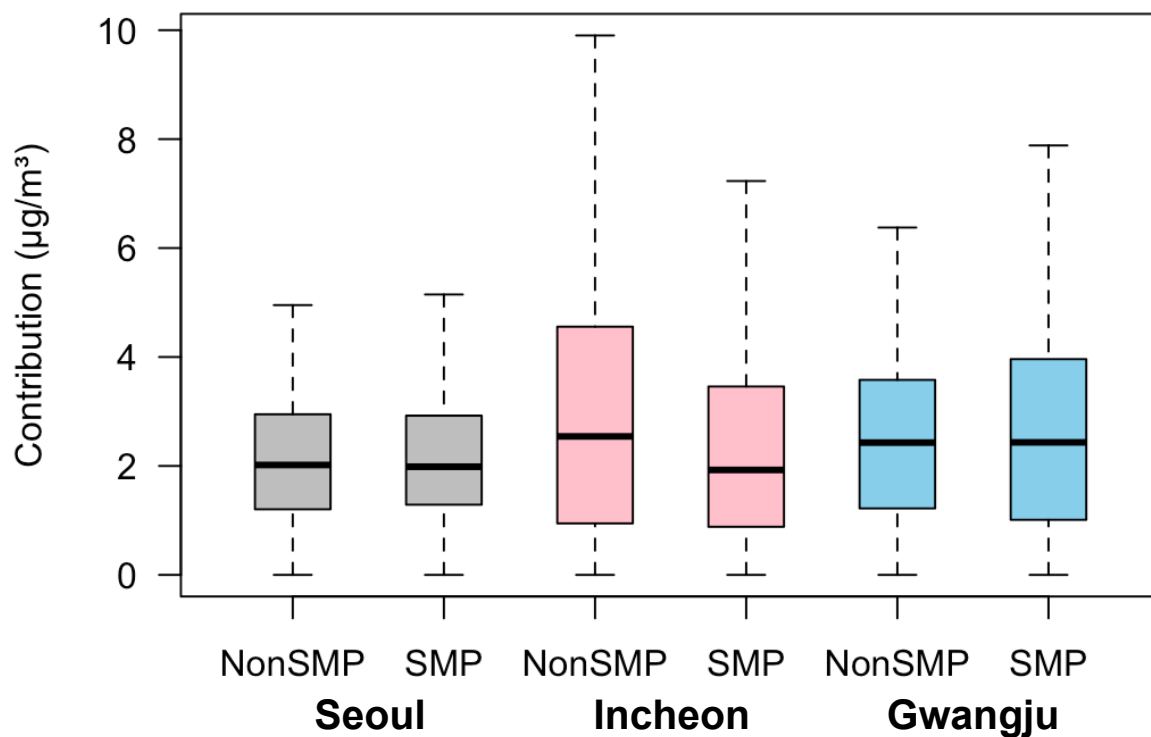


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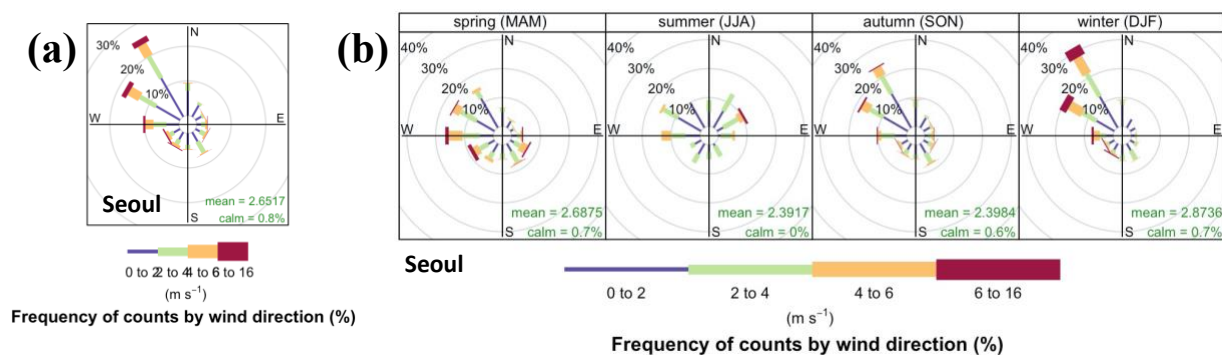


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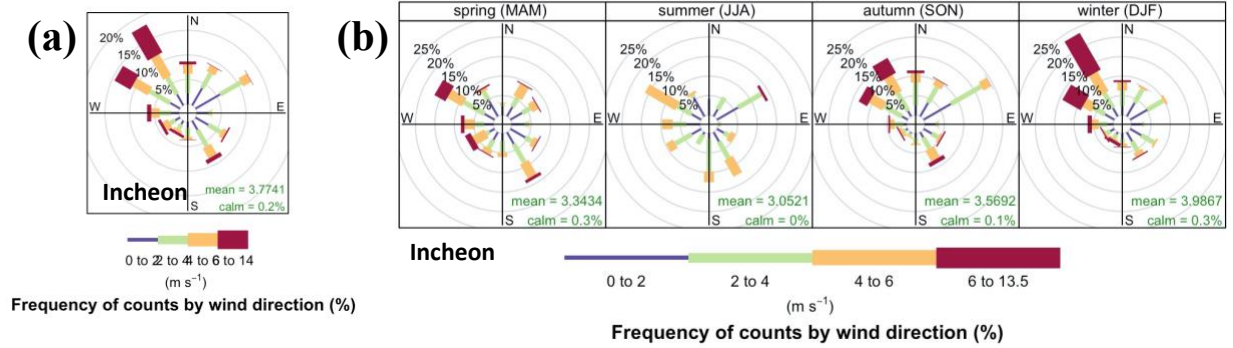


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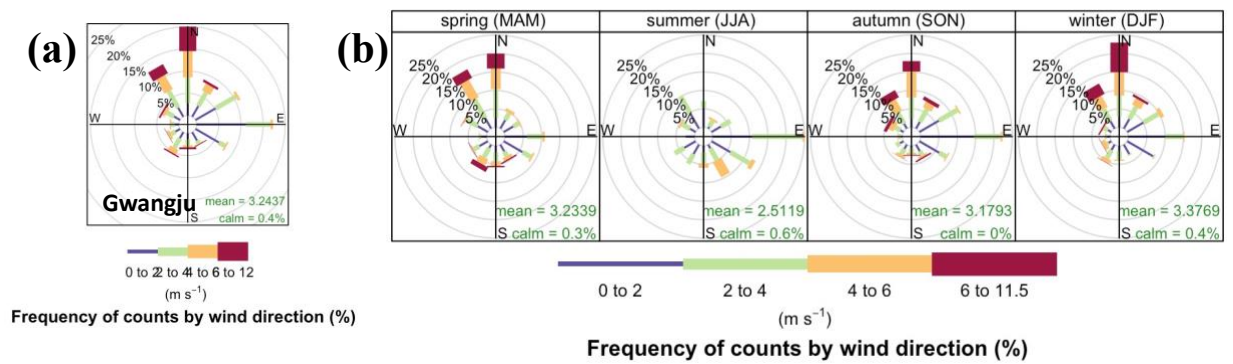


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Table S1. Uncertainty calculation of each species

Uncertainty calculation	
PM _{2.5}	$4 \times conc$
Carbonaceous species	$\sqrt{((0.05 + E) \times conc + IDLs)^2 + (S.D. of Blank)^2}$
Ionic species	$\sqrt{(global\ unc \times conc)^2 + (S.D. of Blank)^2 + (E \times conc)^2}$
Trace element species	$\sqrt{((0.1 + E) \times conc)^2 + (0.5 \times MDL)^2}$
Flow error (E)	$\frac{ (total\ flow) - 16.7 }{16.7}$

Table S2. Dates of HCEs at each site

Site		
Seoul	Incheon	Gwangju
2020/10/19	2020/10/19	2020/10/27
2020/10/26	2020/10/26	2020/12/07
2020/10/27	2020/10/27	2020/12/22
2020/11/14	2020/11/12	2021/01/14
2020/11/16	2020/11/14	2021/02/06
2020/11/26	2020/11/16	2021/02/20
2020/12/10	2020/11/26	2021/03/10
2020/12/11	2020/12/10	2021/03/29
2020/12/22	2020/12/11	2021/05/07
2020/12/26	2020/12/22	2021/11/20
2020/12/28	2021/01/11	2021/12/16
2021/01/04	2021/01/15	2022/01/09
2021/01/11	2021/02/08	2022/01/23
2021/01/13	2021/03/10	2022/02/10
2021/01/15	2021/03/11	2022/02/12
2021/01/21	2021/03/12	2022/03/04
2021/01/27	2021/03/15	2022/03/08
2021/02/01	2021/03/18	2022/03/22
2021/02/06	2021/03/20	
2021/02/19	2021/03/26	
2021/02/20	2021/03/29	
2021/03/09	2021/05/07	
2021/03/10	2021/11/04	
2021/03/11	2021/11/18	
2021/03/13	2021/11/20	
2021/03/15	2021/12/06	
2021/03/18	2021/12/08	
2021/03/20	2021/12/16	
2021/03/26	2021/12/20	

2021/03/29	2021/12/28
2021/05/07	2022/01/05
2021/11/04	2022/01/10
2021/11/14	2022/01/15
2021/11/18	2022/01/21
2021/11/20	2022/01/25
2021/12/16	2022/02/10
2021/12/20	2022/02/12
2021/12/28	2022/02/18
2022/01/09	2022/03/04
2022/01/19	2022/03/10
2022/01/23	
2022/01/25	
2022/02/10	
2022/02/12	
2022/02/14	
2022/02/26	
2022/03/08	
2022/03/16	

Table S3. PM_{2.5} mass concentration and its chemical constituents in Seoul, Incheon, and Gwangju

Site		Seoul		Incheon		Gwangju	
Species	Unit	Mean	SD	Mean	SD	Mean	SD
PM _{2.5}	µg/m ³	24.2	21.4	24.6	20.0	18.8	16.5
OC	µg/m ³	4.79	2.22	5.82	2.71	4.45	2.14
EC	µg/m ³	0.36	0.17	0.59	0.30	0.36	0.22
NO ₃ ⁻	µg/m ³	6.54	7.03	6.27	6.83	4.82	4.84
SO ₄ ²⁻	µg/m ³	2.97	1.97	2.94	1.97	2.73	1.58
NH ₄ ⁺	µg/m ³	3.11	2.94	3.06	2.81	2.33	1.98
Cl ⁻	µg/m ³	0.45	0.38	0.77	0.57	0.43	0.31
Na ⁺	µg/m ³	0.20	0.28	0.41	0.63	0.25	0.35
K ⁺	µg/m ³	0.17	0.11	0.18	0.10	0.13	0.11
ΣTrace elements	µg/m ³	3.48	5.50	4.23	5.56	3.13	5.22
Crustal	ng/m ³	1448.9	4252.4	1763.2	4160.9	1277.3	4240.8
Non-crustal	ng/m ³	2029.9	1246.9	2465.2	1397.9	1856.0	982.4
Mg	ng/m ³	72.8	133.0	74.5	138.0	68.9	149.6
Al	ng/m ³	178.4	665.4	208.1	676.7	167.4	705.8
Si	ng/m ³	513.8	1996.2	617.6	1996.1	490.2	2067.6
S	ng/m ³	1410.6	939.5	1395.8	842.2	1265.2	690.6
Cl	ng/m ³	453.4	414.6	864.1	653.9	453.4	401.6
K	ng/m ³	302.4	476.1	326.3	456.2	246.1	452.2
Ca	ng/m ³	156.8	348.4	205.4	323.8	127.5	319.9
Ti	ng/m ³	18.4	61.4	21.1	60.5	16.1	58.4
V	ng/m ³	0.4	1.6	0.4	1.6	0.3	1.3
Cr	ng/m ³	1.6	1.0	5.2	6.3	1.3	2.8
Mn	ng/m ³	15.0	20.8	27.5	26.8	12.0	19.3
Ba	ng/m ³	5.0	4.7	5.6	5.8	4.7	4.9
Fe	ng/m ³	278.9	727.8	392.6	700.5	230.0	663.6
Ni	ng/m ³	0.9	0.8	3.1	3.9	0.7	0.9
Cu	ng/m ³	3.2	3.5	6.7	11.8	3.3	4.0
Zn	ng/m ³	40.0	35.4	58.4	36.2	28.5	28.7
As	ng/m ³	4.6	6.4	5.4	8.4	1.9	2.4
Se	ng/m ³	0.9	0.9	0.8	0.8	0.9	0.8
Br	ng/m ³	6.1	4.1	8.9	7.4	5.5	6.9
Pb	ng/m ³	15.5	10.6	20.2	15.2	9.4	7.3

Table S4. Explained values of chemical species for each source in Seoul

Species	Sources									
	SN	SS	BB	Mobile	Soil	Waste	CC	Ind/Oil	AGS	Ind
PM _{2.5}	33.592	15.013	13.622	10.051	7.817	6.825	3.981	2.275	2.275	4.550
OC	13.319	11.096	21.560	34.052	3.807	4.578	6.615	0.819	1.649	2.506
EC	4.559	0.000	17.716	38.019	14.365	12.477	10.567	0.000	0.000	2.297
NO ₃ ⁻	67.861	6.341	15.732	0.000	0.828	4.126	2.176	0.000	0.000	2.936
SO ₄ ²⁻	18.929	49.909	7.106	3.747	0.000	0.000	4.158	4.378	3.256	8.517
NH ₄ ⁺	55.254	25.181	9.591	3.738	1.115	1.414	2.475	0.000	1.035	0.197
Cl ⁻	8.251	0.501	0.000	0.000	0.000	80.388	4.440	6.421	0.000	0.000
Na ⁺	6.258	0.000	0.877	9.479	0.000	3.394	0.000	0.207	79.786	0.000
K ⁺	0.000	0.000	73.424	0.000	0.000	5.315	3.245	18.016	0.000	0.000
Mg	0.000	11.297	4.677	0.000	61.498	0.000	6.464	0.000	8.722	7.341
Al	1.197	6.788	0.000	0.000	77.576	0.234	8.759	3.978	1.468	0.000
Si	0.000	3.540	4.482	0.133	75.091	1.611	9.519	5.624	0.000	0.000
Ca	1.356	0.000	10.120	12.209	59.002	6.160	7.146	2.146	1.595	0.266
Ti	8.219	3.365	6.145	15.187	51.332	0.347	6.464	4.315	3.038	1.587
V	0.000	11.707	0.000	0.000	9.329	6.745	9.494	60.086	2.639	0.000
Cr	17.597	0.751	4.977	12.922	14.746	0.000	0.000	11.072	0.000	37.935
Mn	20.579	4.046	0.332	13.314	28.532	3.029	1.122	5.810	1.326	21.910
Ba	0.000	0.000	0.000	50.425	0.000	0.000	22.488	18.081	0.000	9.006
Fe	12.263	2.375	1.275	17.545	36.985	3.239	6.853	8.873	0.055	10.537
Ni	24.394	0.000	17.508	0.386	14.533	12.984	0.000	27.224	2.970	0.000
Cu	13.421	0.000	0.000	23.294	0.000	0.000	9.016	54.269	0.000	0.000
Zn	23.665	6.135	0.000	10.959	6.211	0.220	1.113	8.036	0.495	43.165
As	0.000	0.000	10.229	8.466	0.000	0.000	74.317	4.858	2.131	0.000
Pb	5.914	0.000	22.698	1.022	3.297	1.279	17.273	0.000	0.285	48.231

Abbreviations: SN (Secondary nitrate), SS (Secondary sulfate), BB (Biomass burning), CC (Coal combustion), Ind/Oil (Industry/Oil combustion), AGS (Aged sea salt), Ind (Industry).

Table S5. Explained values of chemical species for each source in Incheon

Species	Sources									
	SN	SS	BB	Mobile	Soil	Waste	CC	Ind/Oil	AGS	MP
PM _{2.5}	25.694	18.373	12.805	11.136	8.898	12.361	3.897	1.670	3.341	1.825
OC	9.130	13.676	21.414	21.594	4.512	20.485	5.374	1.376	0.283	2.156
EC	0.000	0.000	22.719	34.788	11.339	21.755	5.992	0.000	0.000	3.406
NO ₃ ⁻	71.437	0.000	8.118	0.000	3.389	10.641	2.987	0.000	2.707	0.721
SO ₄ ²⁻	16.110	64.642	0.000	10.240	0.000	0.000	4.914	4.094	0.000	0.000
NH ₄ ⁺	49.651	25.624	1.178	4.186	0.000	14.044	2.558	1.683	1.076	0.000
Cl ⁻	5.209	9.626	0.000	15.371	0.477	59.572	0.255	0.634	7.414	1.442
Na ⁺	4.551	0.000	3.939	0.000	0.230	2.043	1.727	1.834	85.677	0.000
K ⁺	0.000	20.160	52.641	0.000	2.464	18.383	0.000	0.000	0.382	5.969
Mg	0.000	12.748	8.470	8.564	48.086	9.267	11.257	0.000	1.608	0.000
Al	5.187	8.019	4.894	14.730	52.081	2.683	8.198	1.985	1.526	0.696
Si	5.176	4.733	7.319	17.317	50.446	0.000	7.113	1.622	3.015	3.259
Ca	2.104	1.845	15.931	21.493	39.433	8.412	3.525	0.191	2.761	4.306
Ti	8.351	3.209	8.652	23.229	37.178	7.158	1.703	2.772	3.624	4.124
V	3.122	15.937	26.307	0.000	24.634	5.691	0.403	23.732	0.174	0.000
Cr	0.000	0.000	5.477	25.605	1.161	0.000	3.137	1.305	1.965	61.351
Mn	8.514	3.528	0.000	47.073	11.999	11.031	0.000	2.628	2.504	12.722
Ba	0.000	0.000	32.758	56.083	0.000	0.000	8.631	2.528	0.000	0.000
Fe	9.347	0.000	6.123	37.376	21.106	5.654	2.528	3.599	3.055	11.211
Ni	1.652	0.725	7.385	0.000	0.000	9.089	0.000	0.000	2.879	78.271
Cu	8.100	0.000	0.000	19.775	0.000	0.000	0.036	68.682	3.408	0.000
Zn	11.406	2.983	5.200	43.485	4.483	13.743	4.947	7.224	4.296	2.232
As	0.000	0.000	0.000	0.000	0.000	0.000	89.827	0.098	6.713	3.362
Pb	4.043	1.602	10.052	10.029	7.262	15.974	41.600	3.287	0.000	6.152

Abbreviations: SN (Secondary nitrate), SS (Secondary sulfate), BB (Biomass burning), CC (Coal combustion), Ind/Oil (Industry/Oil combustion), AGS (Aged sea salt), MP (Metal plating).

Table S6. Explained values of chemical species for each source in Gwangju

Species	Sources								
	SN	SS	BB	Mobile	Soil	Waste	CC	Ind/Oil	AGS
PM _{2.5}	29.152	16.353	14.931	12.430	4.209	10.666	5.932	2.844	3.484
OC	8.142	11.771	19.759	37.820	0.000	8.726	7.103	4.754	1.926
EC	0.000	0.000	20.872	51.263	1.852	16.680	9.334	0.000	0.000
NO ₃ ⁻	73.054	0.000	12.747	0.000	2.037	6.521	5.641	0.000	0.000
SO ₄ ²⁻	5.883	51.435	19.697	0.000	0.000	14.767	2.573	0.244	5.402
NH ₄ ⁺	51.612	23.471	13.491	0.251	0.000	6.360	3.640	0.053	1.122
Cl ⁻	8.973	0.000	1.983	6.074	0.000	71.600	0.915	10.454	0.000
Na ⁺	0.000	0.000	8.161	1.730	0.000	9.178	0.000	0.856	80.075
K ⁺	6.097	0.000	50.914	0.000	0.319	7.005	0.000	35.664	0.000
Mg	0.000	8.505	17.201	0.000	41.768	22.055	9.236	0.000	1.236
Al	7.677	8.532	11.740	7.754	55.907	1.729	5.735	0.000	0.927
Si	10.617	1.821	9.830	7.082	61.235	1.304	4.606	3.505	0.000
Ca	13.032	0.000	7.683	7.264	50.972	11.705	1.689	7.548	0.107
Ti	12.095	5.391	8.476	18.351	44.627	3.647	0.000	6.149	1.264
V	0.000	20.122	0.000	0.000	14.000	0.000	8.336	51.876	5.665
Cr	23.787	1.413	5.098	23.624	10.224	1.479	10.370	24.006	0.000
Mn	21.228	10.258	0.442	24.613	21.598	3.090	8.254	8.996	1.522
Fe	17.594	3.331	5.231	23.551	34.004	0.000	5.496	9.793	1.000
Ni	19.619	12.160	4.768	3.868	12.571	19.762	0.000	27.253	0.000
Cu	7.146	0.000	0.912	33.717	0.000	0.000	4.181	52.353	1.690
Zn	21.468	6.356	0.000	23.648	5.512	5.902	18.973	18.142	0.000
As	0.000	0.000	23.471	0.000	0.000	0.000	74.926	1.603	0.000
Pb	8.531	9.278	16.785	14.155	3.024	7.743	36.938	3.028	0.517

Abbreviations: SN (Secondary nitrate), SS (Secondary sulfate), BB (Biomass burning), CC (Coal combustion), Ind/Oil (Industry/Oil combustion), AGS (Aged sea salt), MP (Metal plating).

Statistical analysis

Table S7. Comparison of source contributions between the heating season and non-heating season in each site (Mann-Whitney U test at $\alpha = 0.05$)

Source	<i>p</i> -Value		
	Seoul	Incheon	Gwangju
Secondary nitrate	≤ 0.001	≤ 0.001	≤ 0.001
Secondary sulfate	0.049	0.008	0.467
Biomass burning	≤ 0.001	≤ 0.001	0.103
Mobile	0.927	0.153	0.12
Soil	0.005	0.062	0.073
Waste incinerator	≤ 0.001	≤ 0.001	≤ 0.001
Coal combustion	0.007	0.923	≤ 0.001
Industry/Oil combustion	≤ 0.001	≤ 0.001	≤ 0.001
Aged sea salt	0.257	0.223	0.011
Industry	0.023	-	-
Metal plating	-	0.021	-

Inter-site comparison of common source contributions by Kruskal-Wallis One Way Analysis of Variance on Ranks at $\alpha = 0.05$.

Table S8. Statistics of PM_{2.5} mass concentration

Group	N	Missing	Median	25%	75%
Seoul	222	0	18.300	11.500	31.850
Incheon	217	0	20.080	12.625	31.660
Gwangju	219	0	15.051	9.900	23.50

H = 18.271 with 2 degrees of freedom. ($p \leq 0.001$)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference ($p \leq 0.001$).

Table S9. All Pairwise Multiple Comparison Procedures (Dunn's method) of PM_{2.5}

Comparison	Diff of Ranks	Q	P	P<0.050
Incheon vs Gwangju	72.985	4.008	<0.001	Yes
Incheon vs Seoul	13.397	0.738	1	No
Seoul vs Gwangju	59.588	3.291	0.003	Yes

Table S10. Statistics of secondary nitrate source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	5.449	1.449	17.041
Incheon	183	0	4.418	1.477	14.390
Gwangju	186	0	4.388	0.933	9.662

H = 5.204 with 2 degrees of freedom. ($p = 0.074$)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference ($p = 0.074$).

Table S11. Statistics of secondary sulfate source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	1.941	0.548	3.725
Incheon	183	0	2.502	1.061	4.474
Gwangju	186	0	2.163	1.071	3.853

$H = 4.765$ with 2 degrees of freedom. ($p = 0.092$)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference ($p = 0.092$).

Table S12. Statistics of biomass burning source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	2.115	1.138	3.569
Incheon	183	0	2.659	1.623	4.077
Gwangju	186	0	1.841	0.856	3.739

$H = 10.405$ with 2 degrees of freedom. ($p = 0.006$)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference ($p = 0.006$).

Table S13. All Pairwise Multiple Comparison Procedures (Dunn's method) of biomass burning source

Comparison	Diff of Ranks	Q	P	P<0.050
Incheon vs Gwangju	51.136	3.074	0.006	Yes
Incheon vs Seoul	39.826	2.387	0.051	No
Seoul vs Gwangju	11.31	0.681	1	No

Table S14. Statistics of mobile source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	2.002	1.220	2.925
Incheon	183	0	1.977	0.906	4.048
Gwangju	186	0	2.429	1.137	3.820

H = 3.751 with 2 degrees of freedom. ($p = 0.153$)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference ($p = 0.153$).

Table S15. Statistics of soil source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	1.234	0.657	2.019
Incheon	183	0	1.108	0.410	2.285
Gwangju	186	0	0.254	0.0418	0.598

H = 95.674 with 2 degrees of freedom. ($p \leq 0.001$)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference ($p \leq 0.001$).

Table S16. All Pairwise Multiple Comparison Procedures (Dunn's method) of soil source

Comparison	Diff of Ranks	Q	P	P<0.050
Seoul vs Gwangju	150.875	9.081	<0.001	Yes
Seoul vs Incheon	23.734	1.423	0.464	No
Incheon vs Gwangju	127.14	7.642	<0.001	Yes

Table S17. Statistics of coal combustion source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	0.375	0.0870	1.152
Incheon	183	0	0.475	0.206	1.225
Gwangju	186	0	0.572	0.155	1.265

$H = 2.614$ with 2 degrees of freedom. ($p = 0.271$)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference ($p = 0.271$).

Table S18. Statistics of waste incinerator source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	0.953	0.232	1.629
Incheon	183	0	2.053	0.735	3.515
Gwangju	186	0	0.957	0.215	2.107

$H = 44.673$ with 2 degrees of freedom. ($p \leq 0.001$)

The differences in the median values among the treatment groups are greater than would be expected by chance; there is a statistically significant difference ($p \leq 0.001$).

Table S19. All Pairwise Multiple Comparison Procedures (Dunn's method) of waste incinerator source

Comparison	Diff of Ranks	Q	P	$P < 0.050$
Incheon vs Seoul	102.109	6.121	< 0.001	Yes
Incheon vs Gwangju	89.504	5.38	< 0.001	Yes
Gwangju vs Seoul	12.605	0.759	1	No

Table S20. Statistics of industry/oil combustion source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	0.286	0.100	0.736
Incheon	183	0	0.242	0.0727	0.659
Gwangju	186	0	0.351	0.102	0.726

H = 0.481 with 2 degrees of freedom. ($p = 0.786$)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference ($p = 0.786$).

Table S21. Statistics of aged sea salt source contribution

Group	N	Missing	Median	25%	75%
Seoul	184	0	0.137	0.000	0.394
Incheon	183	0	0.163	0.000	0.635
Gwangju	186	0	0.249	0.000	0.609

H = 2.726 with 2 degrees of freedom. ($p = 0.256$)

The differences in the median values among the treatment groups are not great enough to exclude the possibility that the difference is due to random sampling variability; there is not a statistically significant difference ($p = 0.256$).