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## Supplementary Materials:

# Comparisons of Spatial and Temporal Variations in PM<sub>2.5</sub>-Bound Trace Elements in Urban and Rural Areas of South Korea, and Associated Potential Health Risks

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### Text S1. Temporal variation of PM<sub>2.5</sub>

The daily average concentrations of PM<sub>2.5</sub>, S, K, soil, and other elements throughout the study period are shown in Figure S2. The inverse relationship between PM<sub>2.5</sub> levels and wind speed velocity observed at both locations was attributed to increased dispersion caused by high wind velocities. Additionally, during the study period, three pollution events with PM<sub>2.5</sub> concentrations greater than 35 µg/m<sup>3</sup> were measured in Seoul and Seosan (Figure S2). The S, K, and soil increased similarly during days with high PM<sub>2.5</sub> concentrations, suggesting that these components may have impacted the increased concentrations during pollution events.

The PM<sub>2.5</sub> concentrations at both study sites were comparable to those from a previous study of South Korea, but lower than those from a previous study of China (Table S3). S was found to be a major component in both Seoul and Seosan, and this is consistent with the findings of studies of other locations in China, including Shandong, Beijing, Guzheng, and Chifeng (Table S3). For crustal elements (Si, Ca, Fe, and Ti), the concentrations in Beijing have been recorded several times higher than in Seoul and Seosan. Furthermore, the concentrations of Se and Br were also several times higher in Beijing than at the sites in this study (Table S3). It is known that these elements, along with S, are emitted during coal combustion.

### Text S2. Estimation of the origins of the elements

To estimate the origins of the elements in PM<sub>2.5</sub>, PCA, which explains statistical variance in a dataset using the fewest possible meaningful components, was applied using trace element concentrations as input data. This procedure replaces several intercorrelated variables with more minor independent variables via orthogonal transformations. The values of the factor loadings for each element assessed for each primary rotational component were maximized/minimized using a varimax-normalized rotation. Factor loadings above 1 are often regarded as excellent.

During the study period, potential trace element sources and their transport pathways to the study locations were tracked using the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) backward air-mass trajectory model. The repository of 1°-resolution meteorological data from the Global Data Assimilation System of the National Center for Environmental Prediction was used to compute the trajectories, which were calculated using the Real-Time Environmental Applications and Display System's online interface (<https://ready.arl.noaa.gov/HYSPLIT.php>) [1,2]. When computing the

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five-day backward trajectories, we used a height of 500 m AGL (above ground level) for each sampling location to avoid the impacts of topography on air-mass movement.

## Supplementary Tables

**Table S1.** Method detection limits (ng m<sup>-3</sup>) of trace elements analyzed using Xact 605i.

Element	Atomic Number	Detection Limits
Al	13	100
Si	14	17.8
P	15	5.2
S	16	3.16
Cl	17	1.73
K	19	1.17
Ca	20	0.3
Ti	22	0.16
V	23	0.12
Cr	24	0.12
Mn	25	0.14
Fe	26	0.17
Co	27	0.14
Ni	28	0.1
Cu	29	0.079
Zn	30	0.067
As	33	0.063
Se	34	0.081
Br	35	0.1
Ag	47	1.9
Cd	48	2.5
In	49	3.1
Sn	50	4.1
Sb	51	5.2
Ba	56	0.39
Hg	80	0.12
Tl	81	0.12
Pb	82	0.13

**Table S2.** Values to be used as guidelines for estimating the health risks posed by PM<sub>2.5</sub>-bound trace elements via various pathways.

Parameter	Symbol	Unit	Value	Reference
Elemental concentrations	C	$\mu\text{g m}^{-3}$		This study
Ingestion rate	IngR	$\text{mg day}^{-1}$	Children (200), adults (100)	USEPA Part A Volume 1, 1989
Exposure duration	ED	Years	Children (6), adults (24)	USEPA Part E, 2004
Average body weight	BW	kg	Children (15), adults (70)	USEPA Part E, 2004
Exposure time	ET	$\text{hour day}^{-1}$	12	This study
Exposure frequency	EF	$\text{days year}^{-1}$	180	USEPA Part E, 2004
Conversion factor	CF	$\text{kg mg}^{-1}$	$10^{-6}$	USEPA Part E, 2004
Skin adherence factor	AF	$\text{mg cm}^{-2}$	Adults and children (0.2)	USEPA Part A, E, F. 2011
Averaging time	AT	days	Non-carcinogens (AT = ED × 365), Carcinogens (AT = 70 × 365)	USEPA Part E, 2004
Surface area	SA	$\text{cm}^2$	Adults (3300), children (2800)	USEPA Part A, E, F. 2011
Dermal absorption factor	ABS	$(\text{hours day}^{-1})$	0.03 for As, 0.1 for Pb and remain 0.01 for remaining elements	USEPA Part E. 2011
Average time	ATn	Non-carcinogenic (Year × hours), Carcinogenic (days × hours)	Non-carcinogenic (ED × 365 days × 24 hours/day) Carcinogens (70 year × 365 days/year × 24 h)	USEPA Part E, 2004 [3]
Inhalation unit risk	IUR	$(\mu\text{g m}^{-3})^{-1}$	Cr (0.012), Ni (0.0043), As (0.00024) Pb (0.000012),	User's Guide and Background Technical Document for US EPA Region 9's Preliminary Remediation Goals (PRG) Table. <a href="http://www.epa.gov/reg3hwm/risk/human/rb-concentrationtable/usersguide.htm">http://www.epa.gov/reg3hwm/risk/human/rb-concentrationtable/usersguide.htm</a> . [4]
Inhalation reference concentrations	RfCi	$(\text{mg m}^{-3})$	Cr (1.00E-04) Mn, Ni (5.00E-05) Ni (5.00E-05) As, (1.50E-05) Cr (5.00E-01)	USEPA Part A, E, F. 2011, [5]
Oral slope factor	SFo	$((\text{mg kg}^{-1} \text{ day}^{-1})^{-1})$	As (1.50E+00) Pb (2.80E-01)	USEPA Part A, E, F. 2011, [5]
Gastrointestinal absorption factor	GIABS		Cr (0.025) Ni (0.04)	USEPA Part A, E, F. 2011, [5]
Inhalation rate	InhR		Cu, Pb, Zn, Mn, As (1) Adults (15), children (7.5)	

Note: The USEPA citations in Table S2 were obtained from the USEPA website: <http://www.epa.gov>.

**Table S3.** Comparison of PM<sub>2.5</sub> (µg/m<sup>3</sup>) and trace element (ng m<sup>-3</sup>) concentrations at the study sites with those from other studies.

Element and PM <sub>2.5</sub>	PM <sub>2.5</sub>	Si	S	K	Ca	Ti	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Br	Ba	Pb	References
Seoul, South Kora	23	237.5	1447	251	83.3	10.7	0.6	1.2	13.1	185	0.5	4.4	49.2	4.6	1	7.3	1.7	21	This study
Seosan, South Korea	31.2	306.1	1265	295	101.1	11.5	0.5	1.2	15.8	210	0.7	4.1	50.6	3.8	1.4	8.8		21	This study
Chifeng, North-east China	36.2			1012	454.3	26.9	1.8	2.4	16.6	390	1.2	16.5	83.7	4.6			24	51	[6]
Gucheng, China	149.86			255	87.5	124.77	0.5	13.7	18.8	204	11	6.58	36.5	0	5.06		30	25.8	[7]
Shandong Province, China	133.33	2050	6270	5370	1160	80	17	20	90	1300	9.1	30	700	20	20	0	70	0	[8]
Seoul Metropolitan Area, Korea	25.2	481				1380	8.2	15.8	22.3	482	9.5	25.9	123	3	17.1		22	20.1	[9]
Shanghai (Baoshan), China	84					61	17	58	113	1187	20	54	681	32	4.5		15	149	[10]
Beijing China	122.1	2180	5100	2500	1340	90				1380		50	550	70	120	150		210	[11]

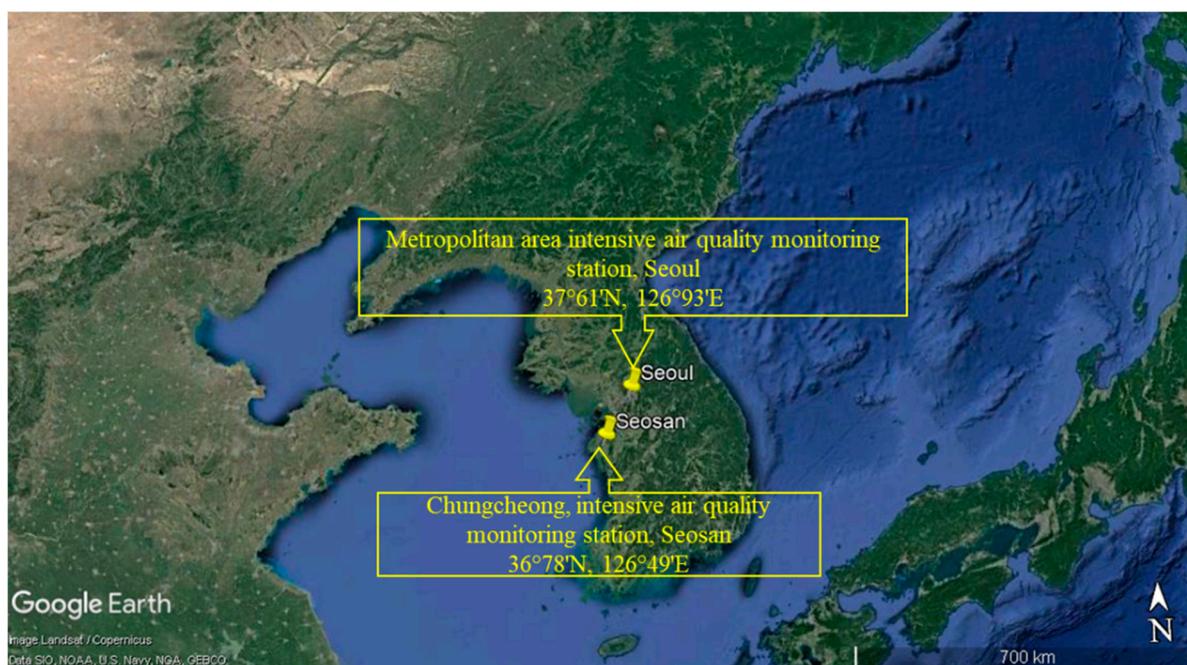
**Table S4.** One-way ANOVA with Tukey's test and paired *t*-tests of daily mean PM<sub>2.5</sub> and chemical species concentrations in Seoul and Seosan.

	One-way ANOVA with Tukey's test	Paired <i>t</i> -test
PM <sub>2.5</sub>	0.01	0.00
Si	0.53	0.09
S	0.27	0.01
K	0.10	0.00
Ca	0.53	0.01
Ti	0.76	0.37
V	0.31	0.06
Cr	0.88	0.79
Mn	0.26	0.21
Fe	0.45	0.05
Ni	0.09	0.00
Cu	0.60	0.52
Zn	0.83	0.83
As	0.08	0.05
Se	0.11	0.00
Br	0.12	0.01
Ba	NA	NA
Pb	0.99	0.99

**Table S5.** One-way ANOVA with Tukey's test and paired t-tests of health and exposure associated with PM<sub>2.5</sub>-bound trace elements in Seoul and Seosan.

	One-way ANOVA with Tukey's test	Paired t-test
Exposure through ingestion in adults	0.00	0.12
Exposure through ingestion in children	0.16	0.12
Exposure through dermal in adults	0.92	0.86
Exposure through dermal in children	0.92	0.86
Exposure through inhalation in adults	0.28	0.22
Exposure through inhalation in children	0.28	0.22
Adult hazard index	0.78	0.63
Child hazard index	0.62	0.44

## Supplementary Figures



**Figure S1.** Study sites in South Korea.

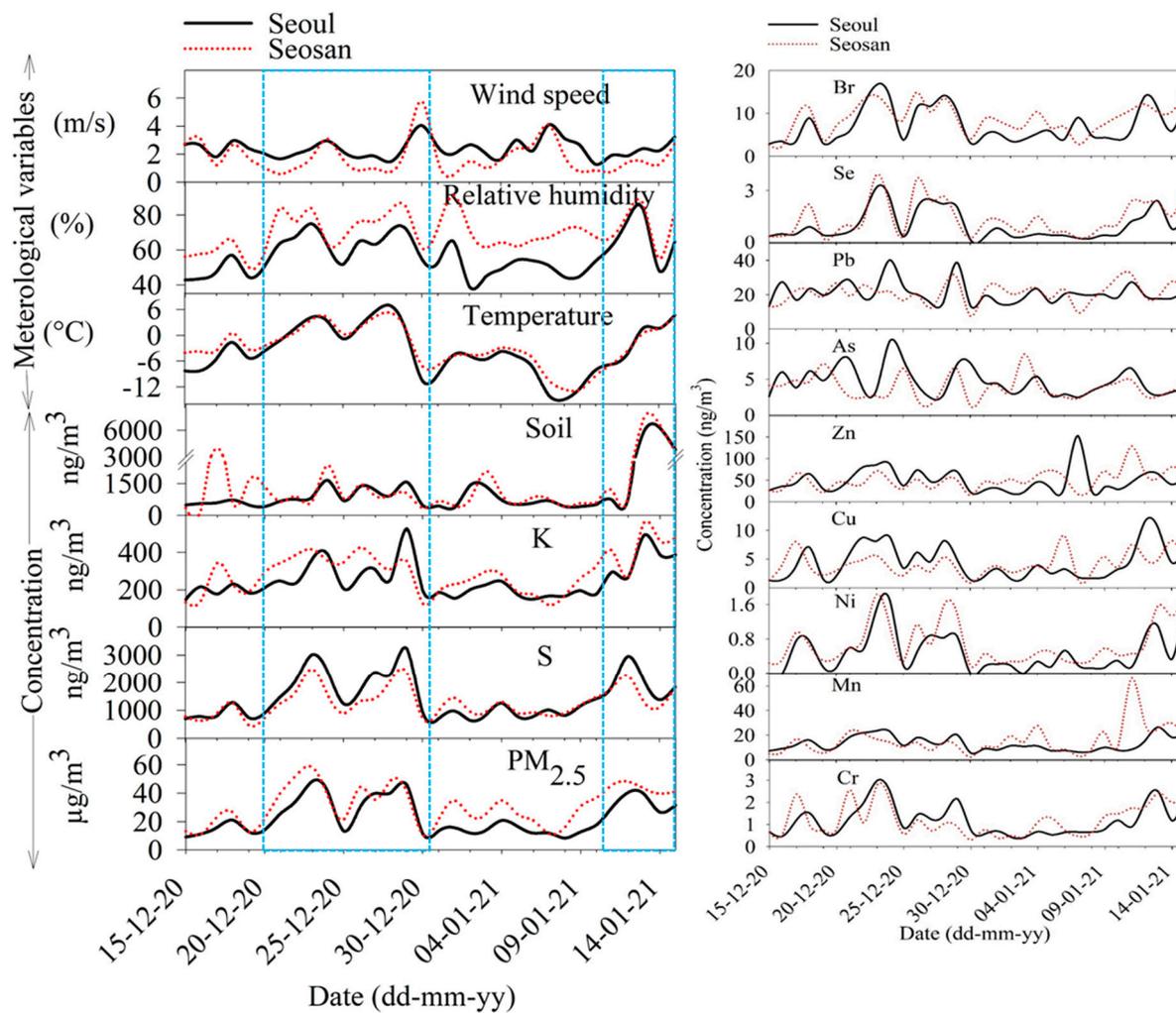
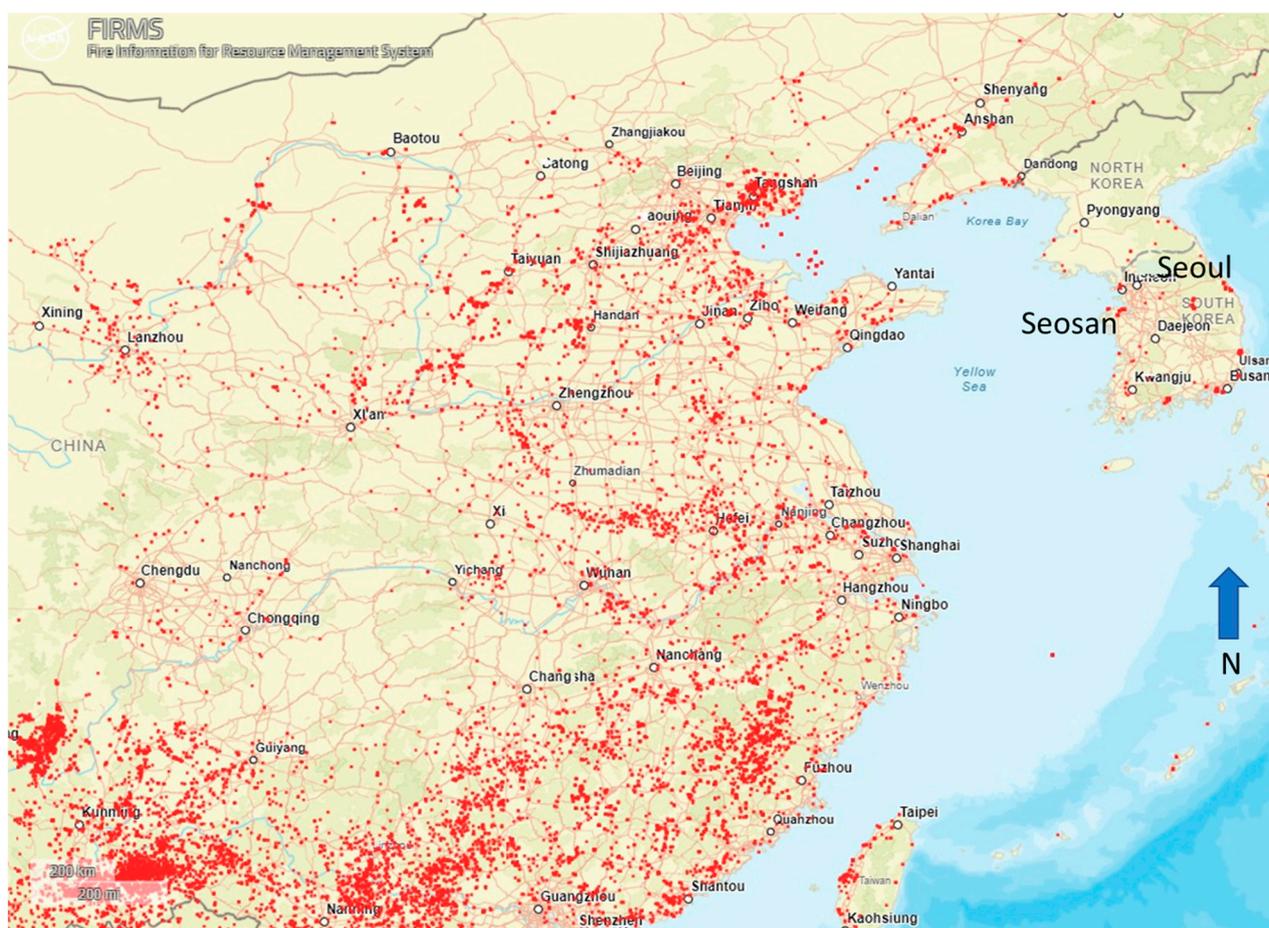
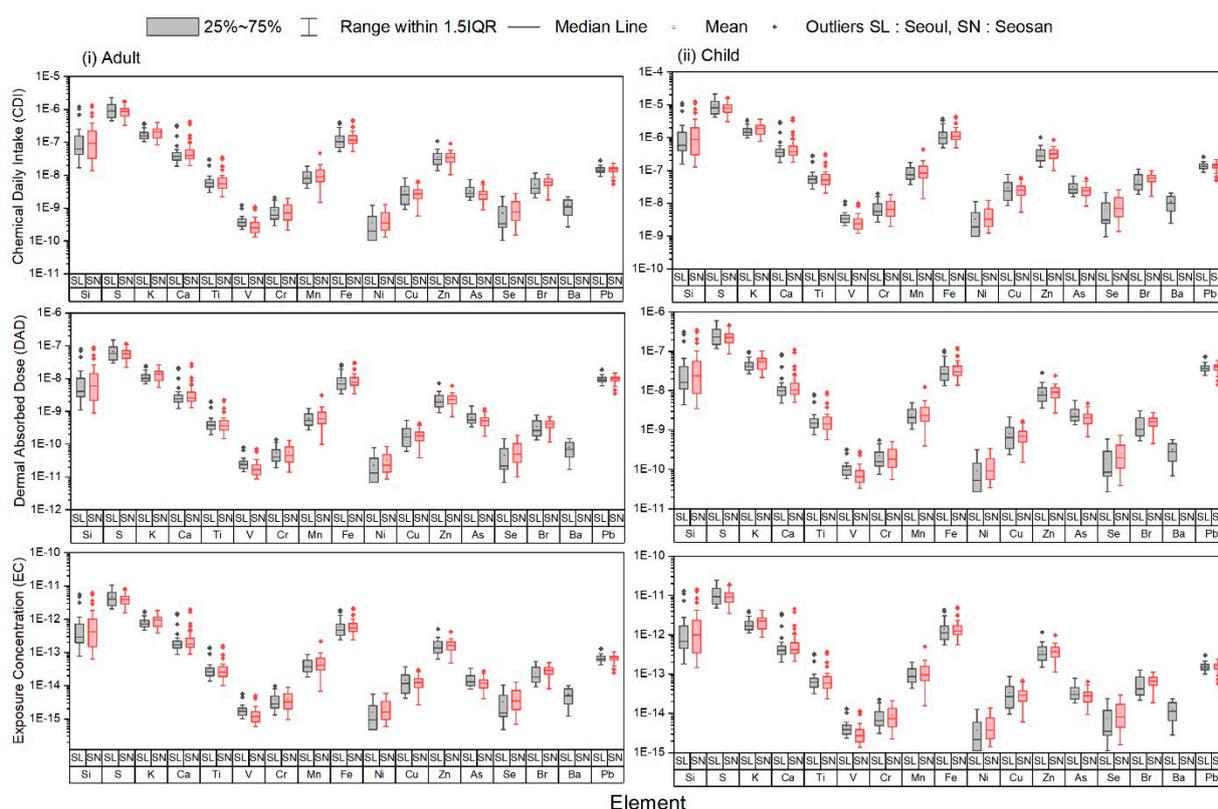


Figure S2. Daily variation of minor element in Seoul and Seosan during the winter period.



**Figure S3.** Fire spots in South Korea and neighboring countries during the sampling period.

Image retrieved from NASA's FIRMS VIIRS satellite (<https://firms.modaps.eosdis.nasa.gov/>) (accessed on 02 April 2023).



**Figure S4.** PM<sub>2.5</sub>-bound element exposure assessment over the Seoul (grey box) and Seosan (red box) locations in South Korea during the winter of 2020–2021. The values in this figure show TE exposure assessment based on daily average concentrations. Ingestion: chemical daily intake (CDI), dermal contact: dermal absorbed dose (DAD), and inhalation: exposure concentration (EC) were used to estimate exposure to elements in PM<sub>2.5</sub>.

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