SUPPLEMENTARY MATERIAL

1. The National Air Pollutants Emission Service (http://airemiss.nier.go.kr/) of the National Institute of Environmental Research provides annual emission data in South Korea from 1999 to 2013. This data includes information of regions (cities, counties, and wards of some cities), type of emission sources (power plants, industrial complexes, vehicles, fire, etc.), fuels (coal, oil, natural gas, etc.), and the amount of emitted air pollutants (CO, NO_x, SO_x, TSP, PM_{10}, $PM_{2.5}$, VOC, and NH_3). Since $PM_{2.5}$ emission quantity is recorded since 2011, we used emission data for NO_x, SO_x, and NH_3, precursors of secondary $PM_{2.5}$, for the period 2005−2013. In Figure S2 and Table S2, NO_x, SO_x, and NH_3 emission quantities decreased in Seoul. However, in many other regions, NO_x and SO_x emission quantities decreased, whereas NH_3 emission quantities increased. Because NO_x and SO_x react with NH_3 to form nitrate and sulfate in the process of secondary formations, a decrease of NO_x and SO_x emission quantities in most regions implies a decrease of secondary $PM_{2.5}$. Figure S3 and Table S3 show the slopes of emission quantities in cities and counties of Gyeonggi-do province, which are located around Seoul, such that air pollutants emitted from these cities and counties could easily affect the air quality of Seoul under proper atmospheric conditions. NO_x and SO_x emission quantities decreased in most regions; NH_3 emission quantities decreased in regions closer to Seoul, whereas they increased in suburban regions. Although the seasonal trends cannot be analyzed because only annual data are available, these results may suggest that emissions did not cause the slow decrease in $PM_{2.5}$ concentration.

2. The National Institute of Environmental Research has been measuring dry and wet deposition of air pollutants in South Korea (National Institute of Environmental Research, 2016). Dry deposition is measured for one day (24 hours) per 6 days and wet deposition is measured once a day at the occurrence of precipitation. After measurements, pH, electrical conductivity, concentrations of ionic components, etc., are analyzed by using ion chromatography. Since nitrate, sulfate, and ammonium are dominant components of $PM_{2.5}$ especially in summer in Seoul (Kim *et al.*, 2007), we used NO_3^-, SO_4^{2-}, and NH_4^+ concentrations data for the period 2005−2015. Concentrations of two sites in Seoul, one in Eunpyeong-gu and the other in Gwangjin-gu (measurements began in 2008 at the site in Gwangjin-gu), were averaged. In Tables S4 and S5, concentrations of NO_3^-, SO_4^{2-}, and NH_4^+ increased significantly at the 95% confidence level in June-July-August, except for NO_3^- in non-precipitation days. Since the main sink of sulfate is dry and wet deposition and those of nitrate is wet deposition (Seinfeld and Pandis, 2016), it is inferred that secondary sulfate and nitrate increased in summer for the analysis period. There is a limit to compare these results with analysis of PMs concentrations because measurements of dry and wet deposition were not performed daily. Nevertheless, these results may imply that increased secondary pollutants are related to the slow decreasing tendency of $PM_{2.5}$ in summer.

Table S1. Annual mean concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ (μg m^{-3}).

Year	$PM_{2.5}$	$PM_{2.5-10}$
2005	28.21	27.19
2006	28.44	26.08
2007	29.48	27.99
2008	25.40	27.63
2009	25.86	25.38
2010	24.59	22.05
2011	23.90	19.93
2012	22.49	18.40
2013	24.75	19.52
2014	23.42	20.60
2015	22.39	18.36
2016	26.12	20.53
Mean	25.42	22.81
Standard deviation	2.32	3.76
Slope	−0.47	−0.90
P-value	0.00	0.00

Table S2. Longitudes and latitudes marked in Figure S2 and the slopes of emitted NO_x, SO_x, and NH_3 (ton year^{-1}) in metropolitan cities and provinces for the period 2005−2013. Blue and red represent negative and positive values, respectively. Bold denotes the slope significant at the 95% confidence level.

Region		Longitude	Latitude	Slope of NO_x	Slope of SO_x	Slope of NH_3
Metropolitan cities	Seoul	126.98	37.57	**−5896**	**−404**	−249
	Busan	129.08	35.18	**−2249**	−344	−102
	Daegu	128.60	35.87	**−2163**	**−411**	−92
	Incheon	126.71	37.46	**−4361**	345	−71
	Gwangju	126.85	35.16	−416	**−36**	−71
	Daejeon	127.38	36.35	**−1111**	−114	−60
	Ulsan	129.31	35.54	−887	−219	**−869**
Provinces	Gyeonggi-do	127.52	37.41	**−4124**	**−2351**	396
	Gangwon-do	128.16	37.82	1856	1244	142
	Chungcheongbuk-do	127.70	36.80	**1398**	**−475**	**570**
	Chungcheongnam-do	126.80	36.52	−5932	2359	**1819**
	Jeollabuk-do	127.15	35.72	**−892**	**−714**	**1193**
	Jeollanam-do	126.99	34.87	**1404**	1240	**849**
	Gyeongsangbuk-do	128.89	36.49	1514	772	**770**
	Gyeongsangnam-do	128.21	35.46	**−8200**	−1821	**925**
	Jeju-do	126.50	33.49	134	−5	**402**

Table S3. Longitudes and latitudes marked in Figure S3 and the slopes of emitted NO_x, SO_x, and NH_3 (ton $year^{-1}$) in cities and counties in Gyeonggi-do for the period 2005−2013. Blue and red represent negative and positive values, respectively. Bold denotes the slope significant at the 95% confidence level.

region	Longitude	Latitude	Slope of NO_x	Slope of SO_x	Slope of NH_3
Suwon	127.03	37.26	**−650**	**−138**	**−224**
Seongnam	127.14	37.44	**−520**	**−44**	**−125**
Uijeongbu	127.03	37.74	**−158**	**−21**	2
Anyang	126.96	37.39	−161	**−40**	−16
Bucheon	126.77	37.50	**−587**	−75	−70
Gwangmyeong	126.86	37.48	**−209**	**−23**	3
Pyeongtaek	127.11	36.99	−135	−15	−26
Dongducheon	127.06	37.90	**−65**	**−11**	28
Ansan	126.83	37.32	**−813**	**−755**	**−70**
Goyang	126.83	37.66	**−309**	**−44**	−27
Gwacheon	126.99	37.43	**−38**	**−2**	**−92**
Guri	127.13	37.59	**−49**	**−7**	**−34**
Namyangju	127.22	37.64	272	11	53
Osan	127.08	37.15	−134	−203	**−41**
Siheung	126.80	37.38	60	−139	−18
Gunpo	126.94	37.36	**−179**	**−39**	**13**
Uiwang	126.97	37.34	−129	−4	**−910**
Hanam	127.21	37.54	139	**−5**	−1
Yongin	127.18	37.24	−318	**−80**	178
Paju	126.78	37.76	29	**−27**	**114**
Icheon	127.43	37.27	**−144**	**−82**	233
Anseong	127.28	37.01	12	−58	**259**
Gimpo	126.72	37.62	136	**−13**	−29
Hwaseong	126.83	37.20	195	**−29**	283
Gwangju	127.26	37.42	−7	**−16**	−9
Yangju	127.05	37.79	−144	**−203**	172
Yeoju	127.64	37.30	−57	**−123**	**453**
Yeoncheon	127.07	38.10	−39	−88	118
Pocheon	127.20	37.89	**−66**	**−53**	**127**
Gapyeong	127.51	37.83	−31	−12	−14
Yangpyeong	127.49	37.49	−25	−11	66

Table S4. Mean, standard deviation, slope, and p-value of concentrations of ionic components (μg m^{-3}) in non-precipitation days for the period 2005−2015 in Seoul. Bold denotes a slope significant at the 95% confidence level.

		MAM	JJA	SON	DJF
NO_3^-	Mean	9.081	1.975	4.394	11.097
	Standard deviation	3.792	1.161	1.910	5.173
	Slope	0.318	−0.156	0.266	**1.412**
	P-value	0.480	0.169	0.152	0.000
SO_4^{2-}	Mean	12.281	12.055	7.385	9.093
	Standard deviation	4.283	3.241	2.273	4.889
	Slope	**0.906**	**0.682**	0.382	**1.321**
	P-value	0.046	0.017	0.075	0.000
NH_4^+	Mean	6.887	5.078	3.583	6.176
	Standard deviation	3.075	1.901	1.290	3.567
	Slope	**0.834**	**0.485**	**0.306**	**1.005**
	P-value	0.004	0.001	0.004	0.000

Table S5. Mean, standard deviation, slope, and p-value of concentrations of ionic components (μg l^{-1}) in precipitation days for the period 2005−2015 in Seoul. Bold denotes a slope significant at the 95% confidence level.

		MAM	JJA	SON	DJF
NO_3^-	Mean	11.161	5.090	8.389	15.366
	Standard deviation	5.627	2.262	4.599	7.122
	Slope	0.929	**0.588**	0.769	1.231
	P-value	0.081	0.001	0.076	0.065
SO_4^{2-}	Mean	11.818	5.454	8.149	14.524
	Standard deviation	5.468	1.406	3.409	5.159
	Slope	0.595	**0.292**	0.487	0.638
	P-value	0.275	0.019	0.141	0.210
NH_4^+	Mean	4.065	2.490	2.693	5.014
	Standard deviation	2.039	1.380	1.291	1.817
	Slope	**0.378**	**0.373**	**0.312**	**0.464**
	P-value	0.044	0.000	0.003	0.001

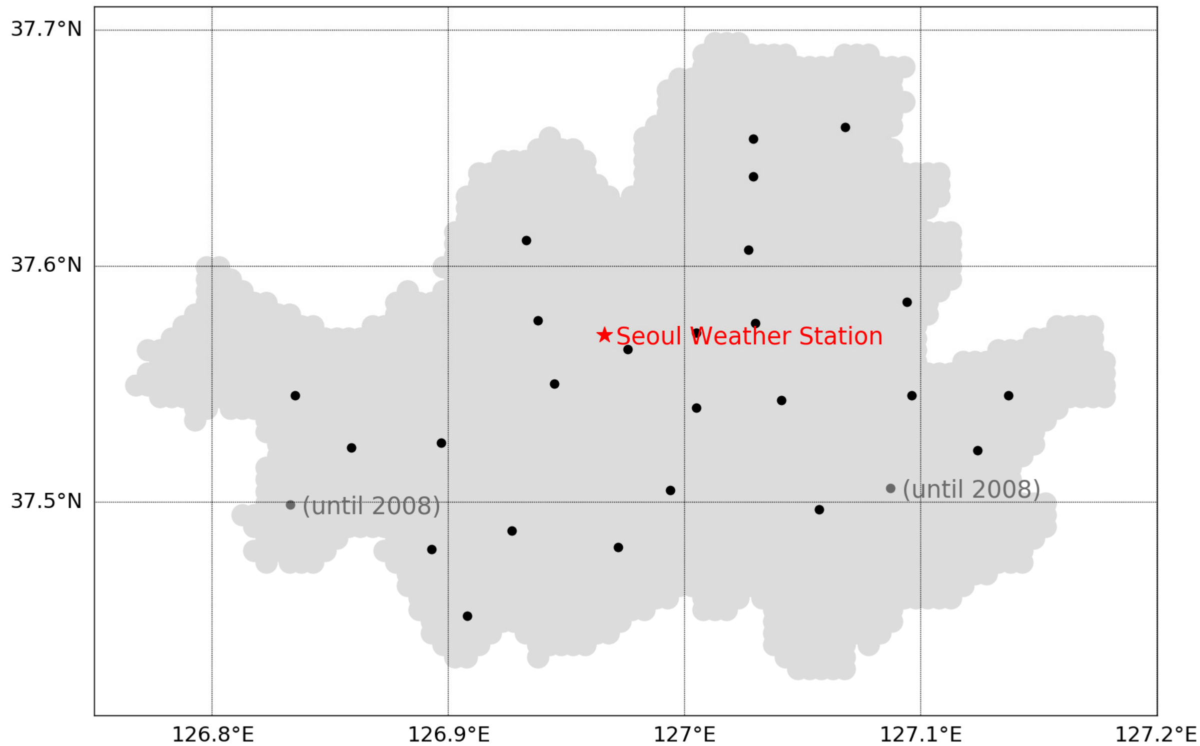

Figure S1. The ground monitoring sites in Seoul. Black dots denote the 25 sites in which $PM_{2.5}$ and PM_{10} concentrations are observed. Gray dots denote two sites which were located until 2008. The red star denotes the location of the Seoul weather station.

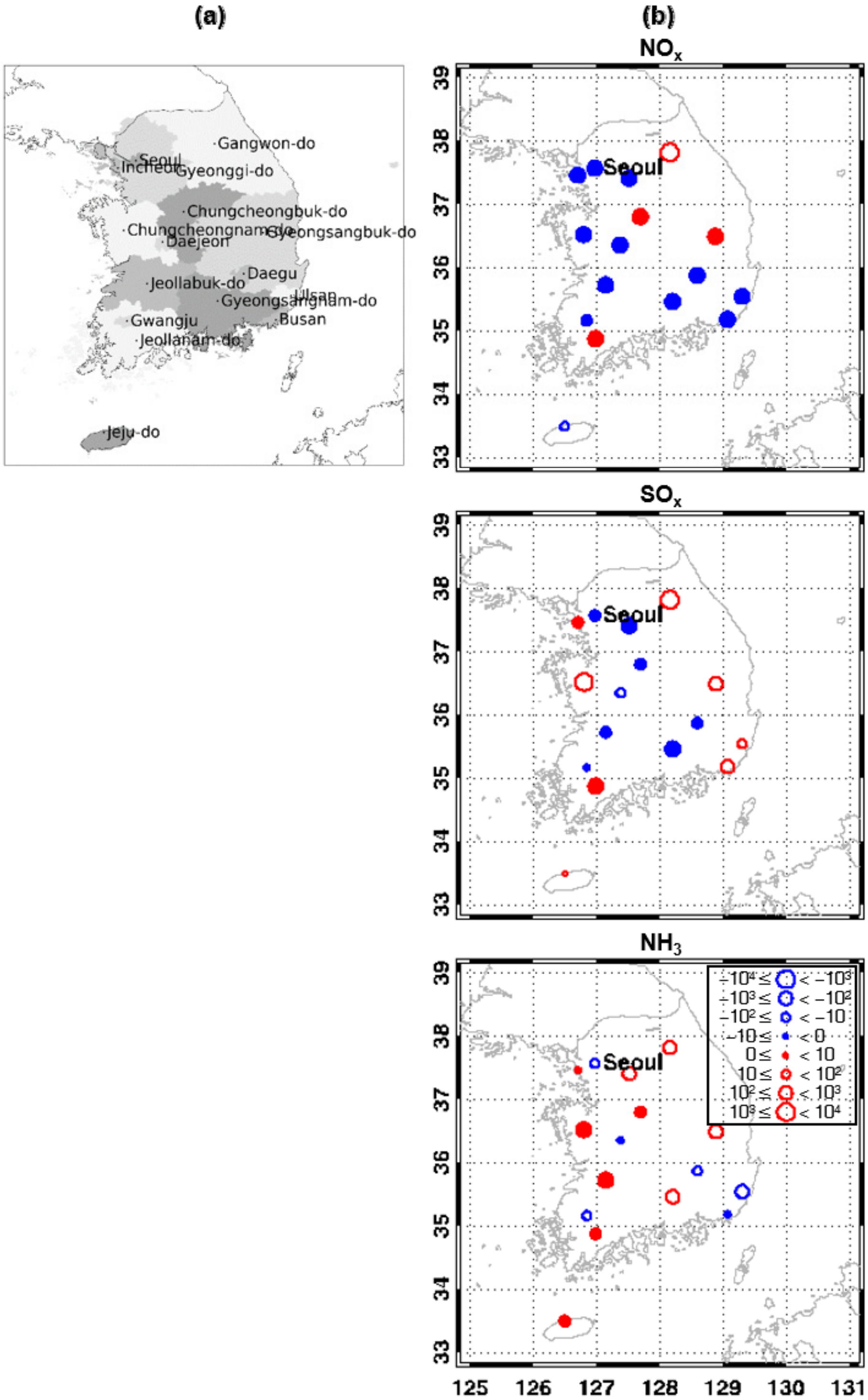

Figure S2. (a) Locations of metropolitan cities and provinces and (b) the slopes of emitted NO_x, SO_x, and NH_3 (ton year^{-1}) in metropolitan cities and provinces for the period 2005−2013. Closed circles represent the slopes significant at the 95% confidence level.

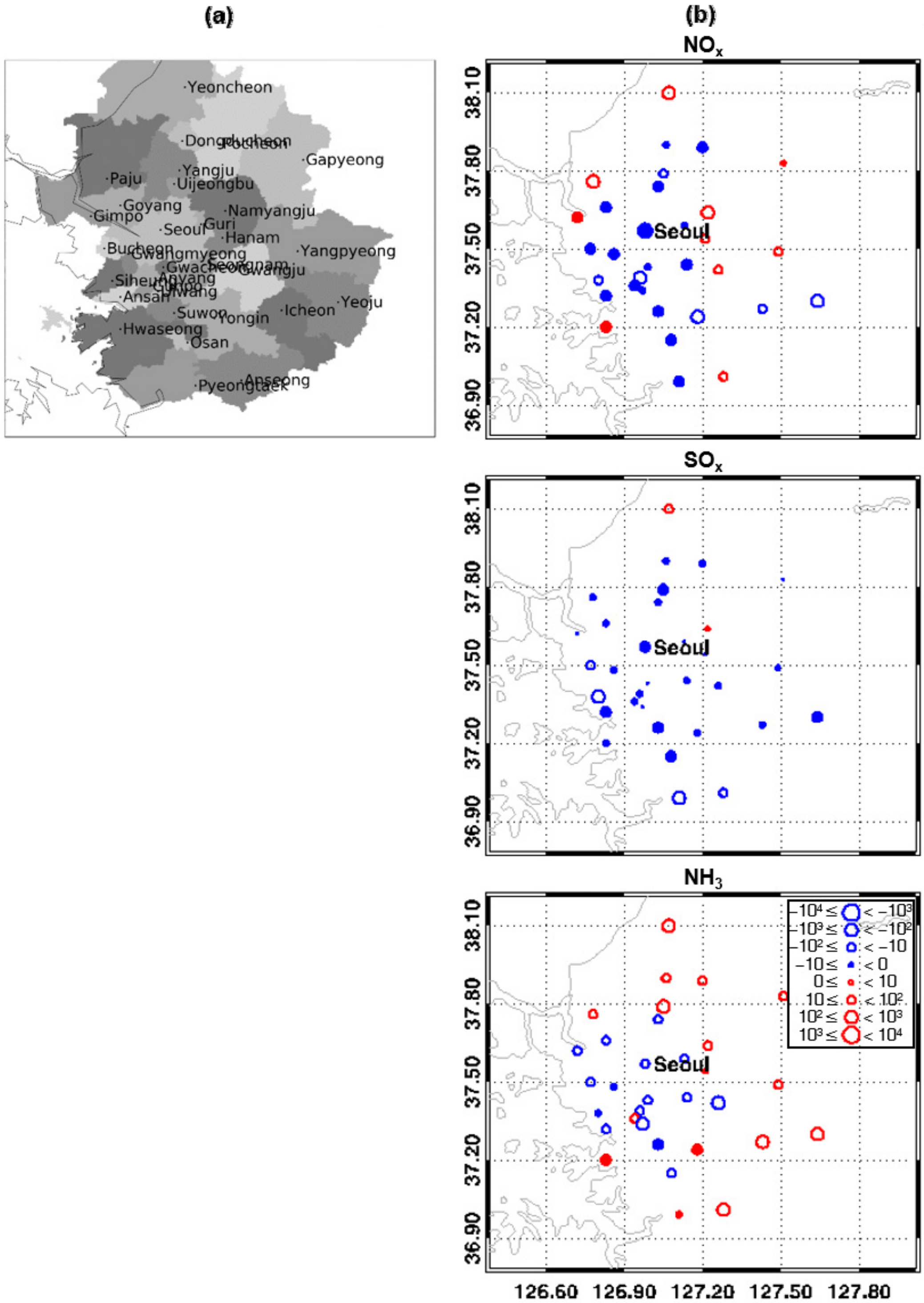

Figure S3. (a) Locations of cities and counties in Gyeonggi-do and (b) the slopes of emitted NO_x, SO_x, and NH_3 (ton year^{-1}) in cities and counties in Gyeonggi-do for the period 2005−2013. Closed circles represent the slopes significant at the 95% confidence level.