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Synergistic enhancement of urban haze by nitrate uptake into transported hygroscopic particles in the Asian continental outflow

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Text S1. Statistical reconstruction of the NH_3 data for Seoul

We reconstructed NH_3 concentrations of Seoul for the analysis period (210 days from October 2012 to June 2014) using previously reported NH_3 data measured at the Gwangjin site (37.545°N, 127.096°E; Fig. S1) over 346 days from September 2010 to August 2011 (Phan et al., 2013). The average ($\mu_{\mathrm{NH_3}}$) and standard deviation ($\sigma_{\mathrm{NH_3}}$) of the measured NH_3 concentrations was 10.9 ± 4.25 ppb, and there were significant linear relationships with three meteorological variables [T, RH, and wind speed (WS)] and three gaseous pollutants (SO_2, NO_2, and CO). To construct a multiple linear regression model of the estimated (reconstructed) NH_3 time series ($[\mathrm{NH_3}]_{est}$), we used the average (μ_x) and standard deviation (σ_x) of the six dependent variables (X) as the normalized form ($[X]_{nor} = (X - \mu_X)/\sigma_X$) for the target period (January 2012 to December 2014), as follows:

$$[\mathrm{NH_3}]_{est} = \alpha\left(c_{\mathrm{SO_2}}[\mathrm{SO_2}]_{nor} + c_{\mathrm{NO_2}}[\mathrm{NO_2}]_{nor} + c_{\mathrm{CO}}[\mathrm{CO}]_{nor} + c_{\mathrm{T}}[\mathrm{T}]_{nor} + c_{\mathrm{RH}}[\mathrm{RH}]_{nor} + c_{\mathrm{WS}}[\mathrm{WS}]_{nor}\right) + \mu_{\mathrm{NH_3}}$$

To obtain $[\mathrm{NH_3}]_{est}$, which retains some of the statistical characteristics of the measured NH_3, we assumed that (i) the average and standard deviation of $[\mathrm{NH_3}]_{est}$ should equal the measured value (10.9 ± 4.25 ppb; Phan et al., 2013) and (ii) the variance explained by each independent variable (or the relative size of R^2 between NH_3 and each independent variable) should be the same as for the measured data (Phan et al., 2013). To find a value of $[\mathrm{NH_3}]_{est}$ that satisfies the first and second assumptions, we carefully adjusted the coefficients (c_x) for each independent variable (Table S1) and obtained the coefficient α (= 3.23). The statistically reconstructed NH_3 time series is represented in Fig. S4d.

Table S1: The coefficients for the dependent variables used in the multiple regression model to predict NH_3.

Variables (X)	Correlation coefficient (r) [Phan *et al.*, 2013]	Coefficient of determination (R^2) [Phan *et al.*, 2013]	Mean (μ) and standard deviation ($\pm\sigma$) [This study]	Coefficients (c_X)
SO_2	-0.179^{**}	0.032	5.57 ± 1.87 (ppb)	−0.621
NO_2	$+0.108^{*}$	0.011	39.9 ± 12.0 (ppb)	−0.274
CO	$+0.151^{**}$	0.023	0.60 ± 0.21 (ppm)	+1.377
T	$+0.487^{**}$	0.237	12.8 ± 11.1 (°C)	+1.000
RH	$+0.505^{**}$	0.255	59.9 ± 15.2 (%)	+0.377
WS	-0.243^{**}	0.059	2.74 ± 0.86 (m s^{-1})	−0.053

* Significant correlation at the 95% or higher level ($p < 0.05$)
** Significant correlation at the 99% or higher level ($p < 0.01$)

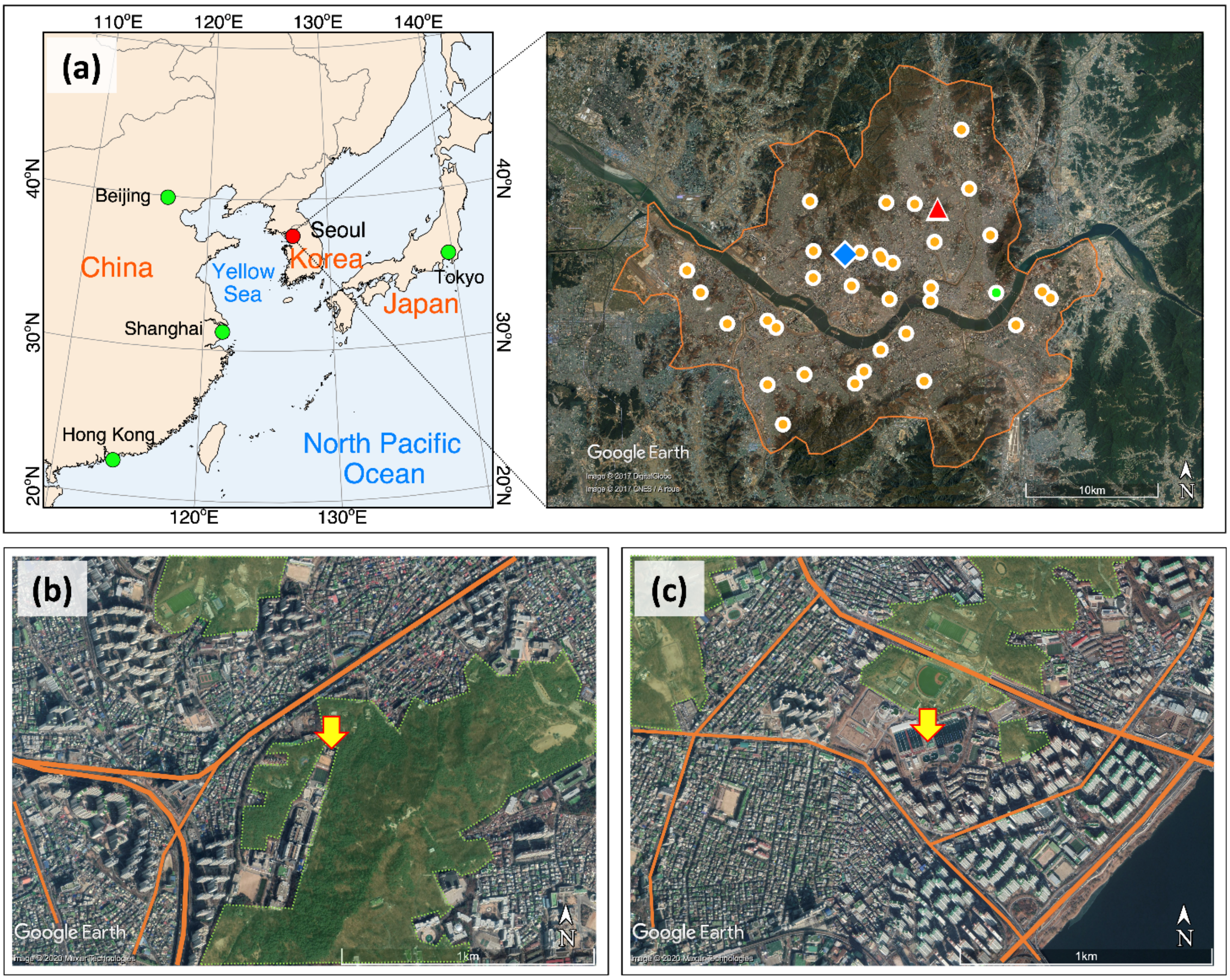

Figure S1: (a) The locations of Seoul (left panel) and the Korea Institute of Science and Technology (KIST) $PM_{2.5}$ sampling site (red triangle), the Korea Meteorological Administration (KMA) weather station (blue diamond), and Korea Ministry of Environment (KMOE) air quality monitoring sites (34 solid circles in yellow and green) in Seoul (right panel). The solid circle in green indicates the Gwangjin site at which a year-long NH_3 measurement was conducted by Phan et al. (2013). (b–c) Satellite maps of (b) the KIST $PM_{2.5}$ sampling site (37.603°N, 127.047°E) utilized in this study and (c) the Gwangjin site (37.545°N, 127.096°E) for NH_3 measurement in Phan et al. (2013). Yellow arrows are the sampling locations, areas shaded by light green are urban green areas, and lines in orange are the main roads. The background satellite images are courtesy of Google Earth.

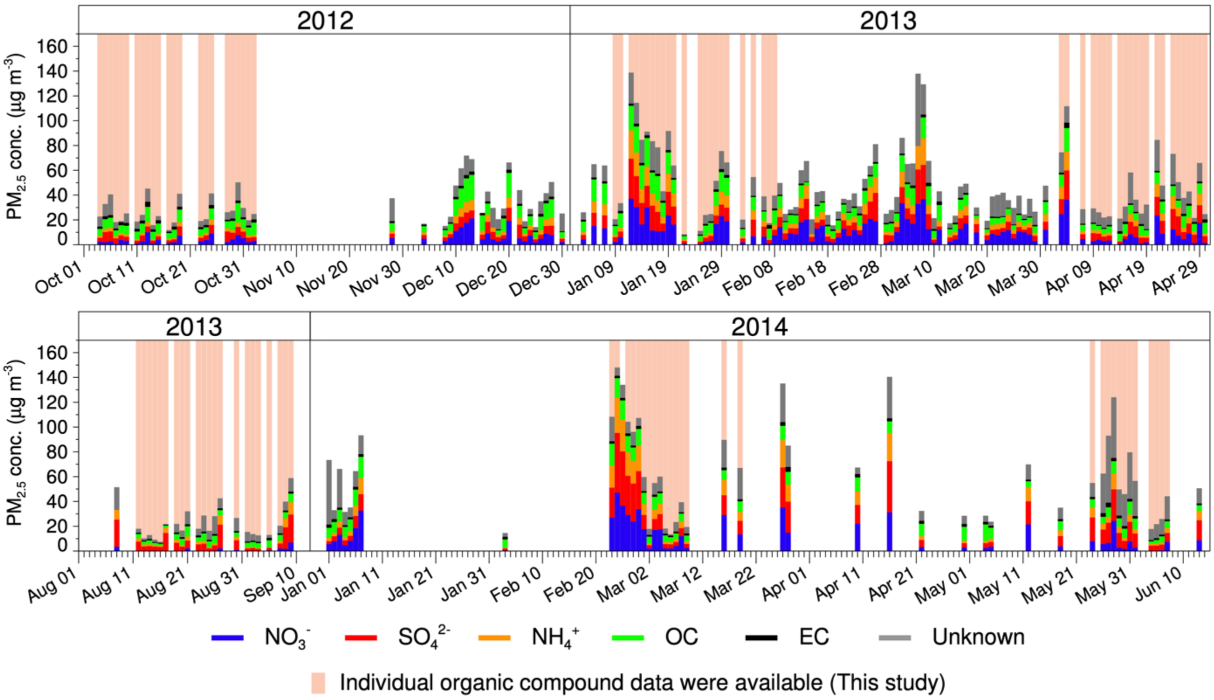

Figure S2: Time series of the chemical composition of $PM_{2.5}$ measured at the KIST site.

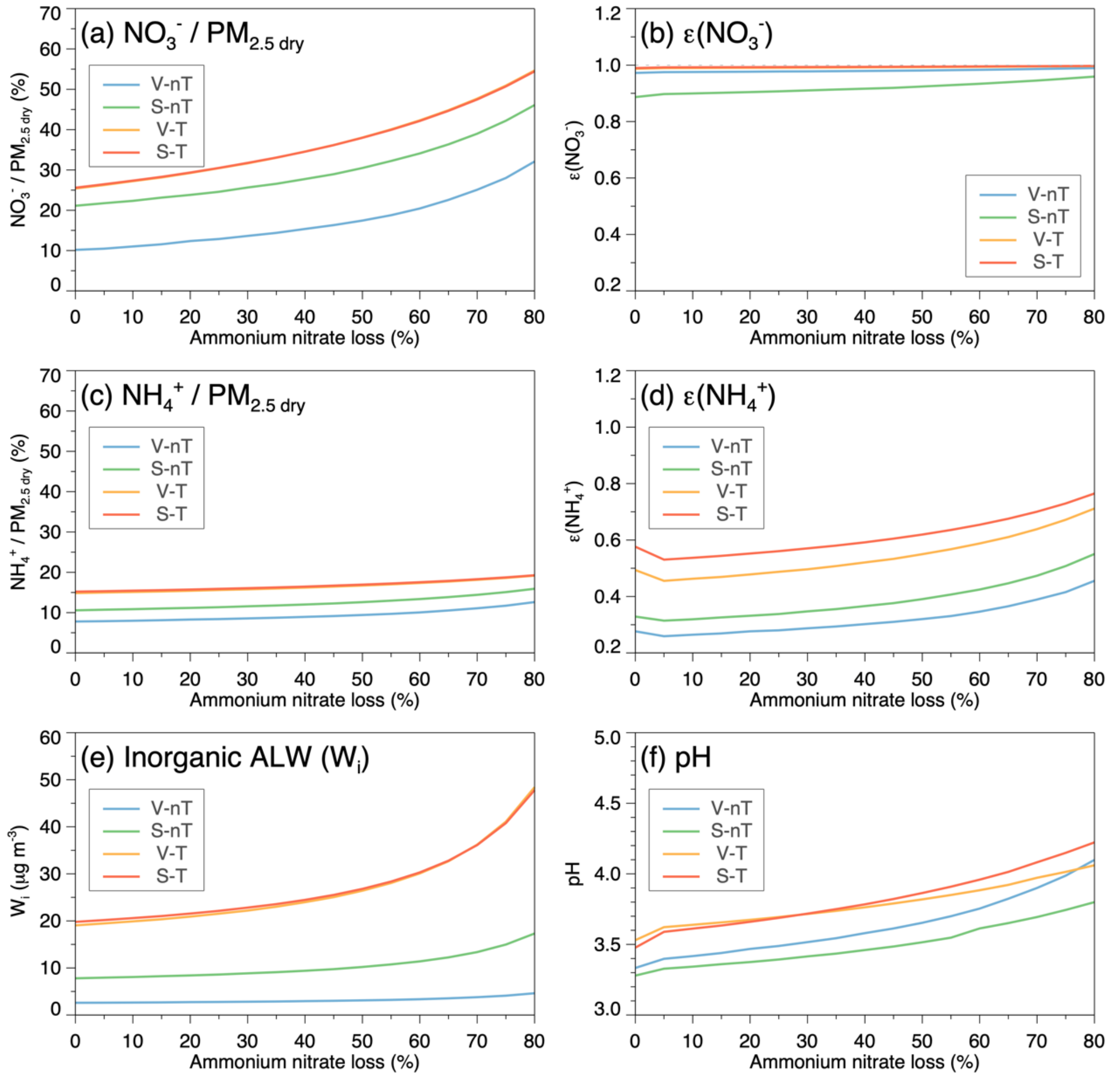

Figure S3: Sensitivity of (a) nitrate fraction in dry $PM_{2.5}$ (NO_3^- / $PM_{2.5\ dry}$), (b) nitrate partitioning ratio [$\varepsilon(NO_3^-)$], (c) ammonium fraction in dry $PM_{2.5}$ (NH_4^+ / $PM_{2.5}$), (d) ammonium partitioning ratio [$\varepsilon(NH_4^+)$], (e) inorganic ALW (W_i) content, and (f) pH to the hypothetical ammonium nitrate loss during the sampling on Teflon filters. Average $PM_{2.5}$ components (Table 2) with extrapolated concentrations of NO_3^- and excess NH_4^+ [$NH_4^+{}_{excess}$ = (NH_4^+ / SO_4^{2-} − 1.5) × SO_4^{2-}; Pathak et al., 2004], considering the hypothetical ammonium nitrate loss from 0% to 80%, were employed in the ISORROPIA simulations. Sensitivity curves in blue, green, yellow, and red colors represent the *local ventilation with no regional transport* (V-nT) group, *local stagnation with no regional transport* (S-nT) group, *local ventilation with regional transport* (V-T) group, and *local stagnation with regional transport* (S-T) group, respectively.

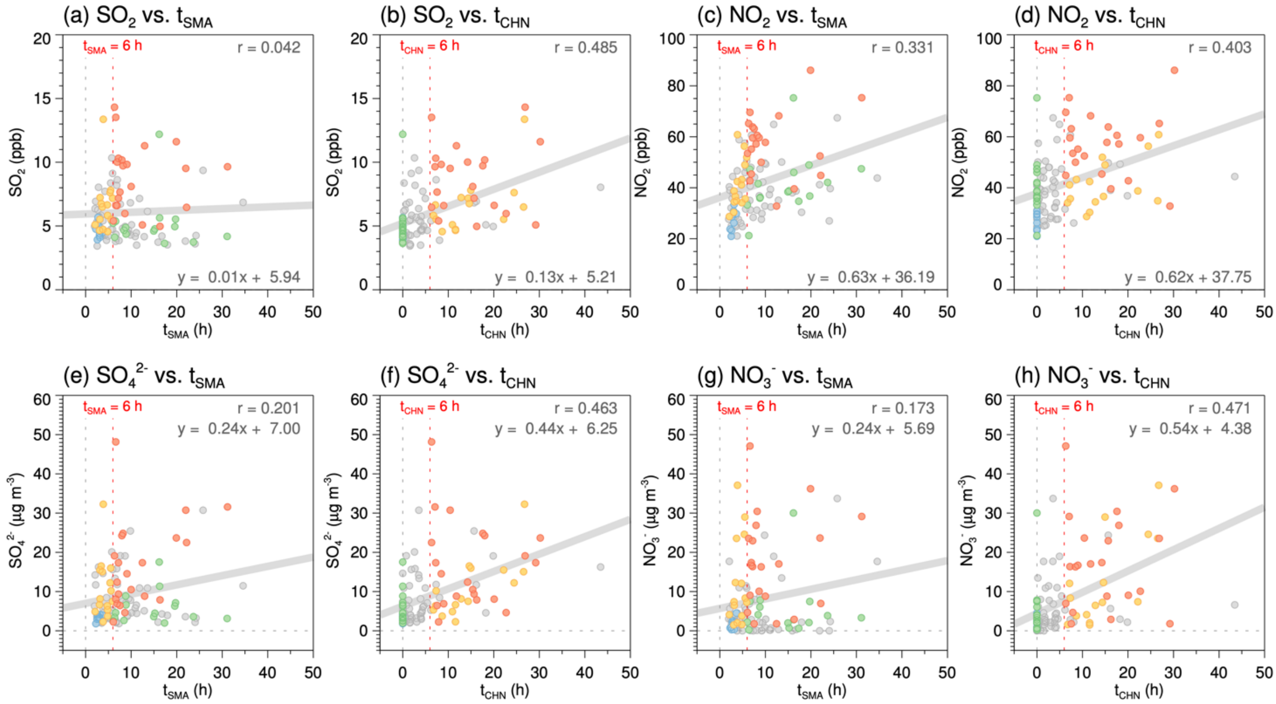

Figure S4: Scatterplots of gaseous species (SO_2 and NO_2) and $PM_{2.5}$ inorganic species (SO_4^{2-} and NO_3^-) concentrations versus the average daily residence time of backward trajectories in the SMA (t_{SMA}) and the NCP and YRD (t_{CHN}).

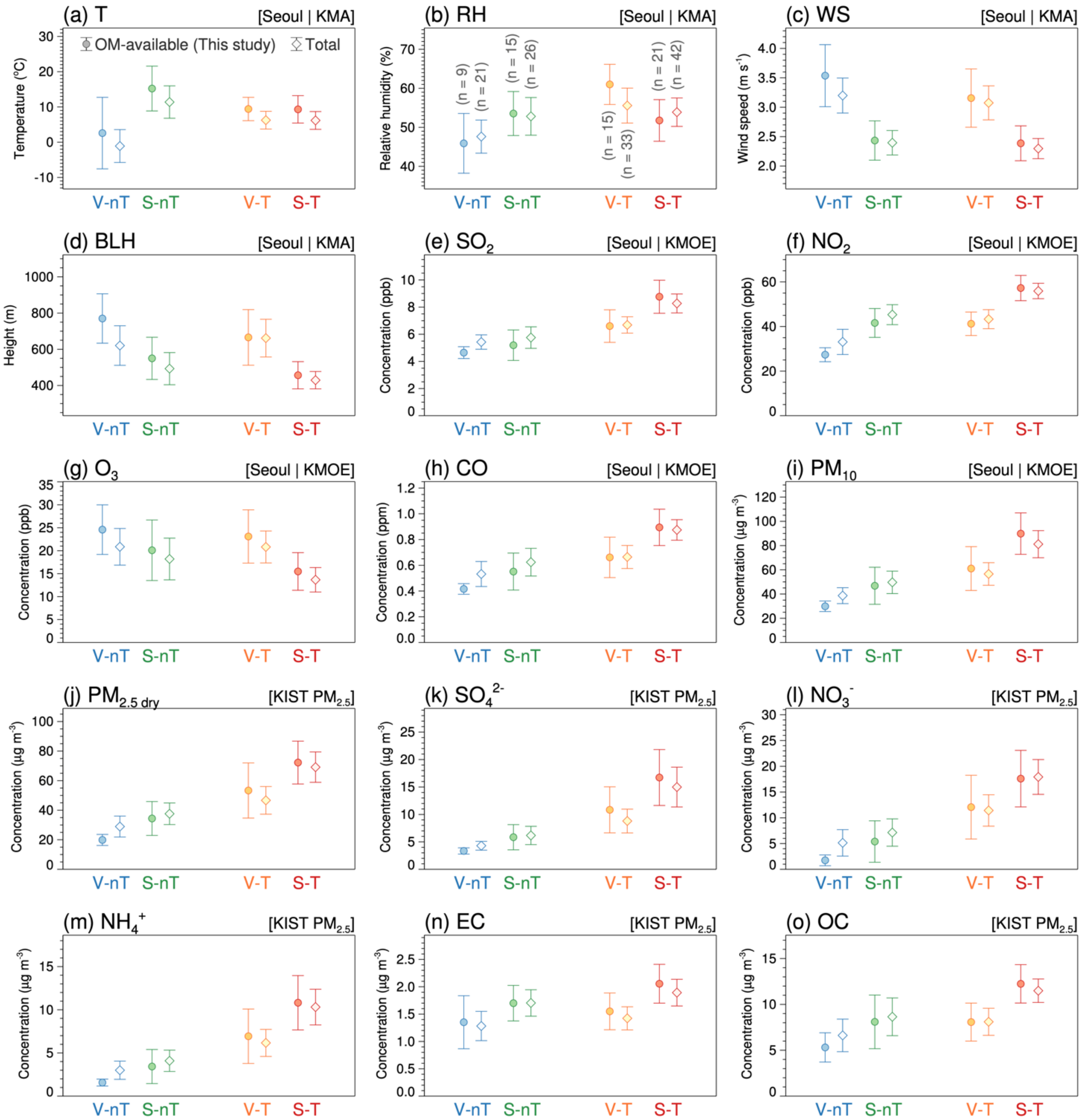

Figure S5: Comparison of average meteorological factors from the Seoul weather station ((a) temperature (T), (b) relative humidity (RH), (c) wind speed (WS), and (d) boundary layer height (BLH)), concentrations of the five representative pollutants from the KMOE air quality monitoring network ((e) SO_2, (f) NO_2, (g) O_3, (h) CO, and (i) PM_{10}), and $PM_{2.5}$ compositions from the KIST site ((j) dry mass concentration ($PM_{2.5\,dry}$), (k) sulfate (SO_4^{2-}), (l) nitrate (NO_3^-), (m) ammonium (NH_4^+), (n) elemental carbon (EC), and (o) organic carbon (OC)) categorized based on the OM-available data utilized in this study (filled circles) and based on the total measurement data (open diamonds). Error bars represent confidence interval at 95%. The numbers of data for each category group are represented in (b).

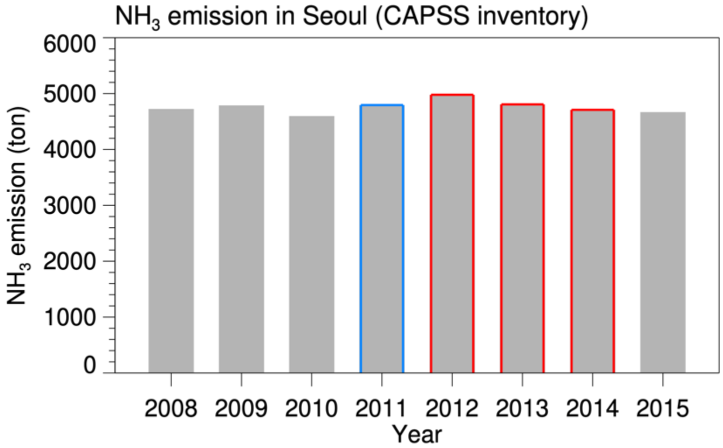

Figure S6: Estimated annual NH_3 emissions in Seoul for 2008–2015 from the Clean Air Policy Support System (CAPSS) inventory (NIER, 2018). Blue and red frames represent the years of measurement in Phan et al. (2013) and this study, respectively.

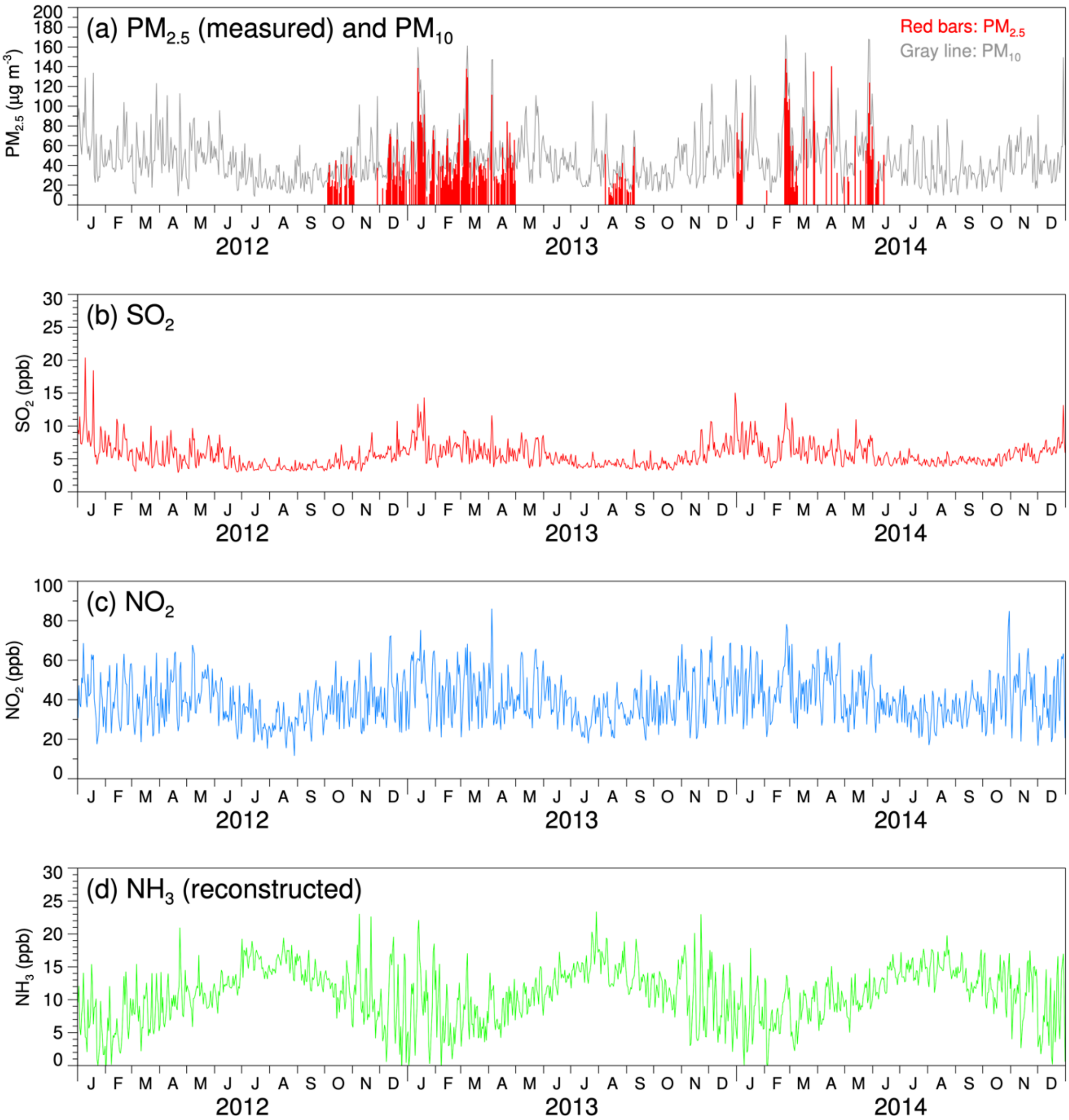

Figure S7: Daily time series of (a) PM_{10} and measured $PM_{2.5}$ (b) SO_2, (c) NO_2, and (d) statistically reconstructed NH_3 concentrations. The $PM_{2.5}$ concentrations are measured for samples collected at the KIST site (37.603°N, 127.047°E). The PM_{10}, SO_2, and NO_2 concentrations are the average daily concentrations for 34 air quality monitoring sites in Seoul. The NH_3 concentrations are reconstructed using the measured data for Seoul (Gwangjin site; 37.545°N, 127.096°E) from September 2010 to August 2011 (Phan et al., 2013).

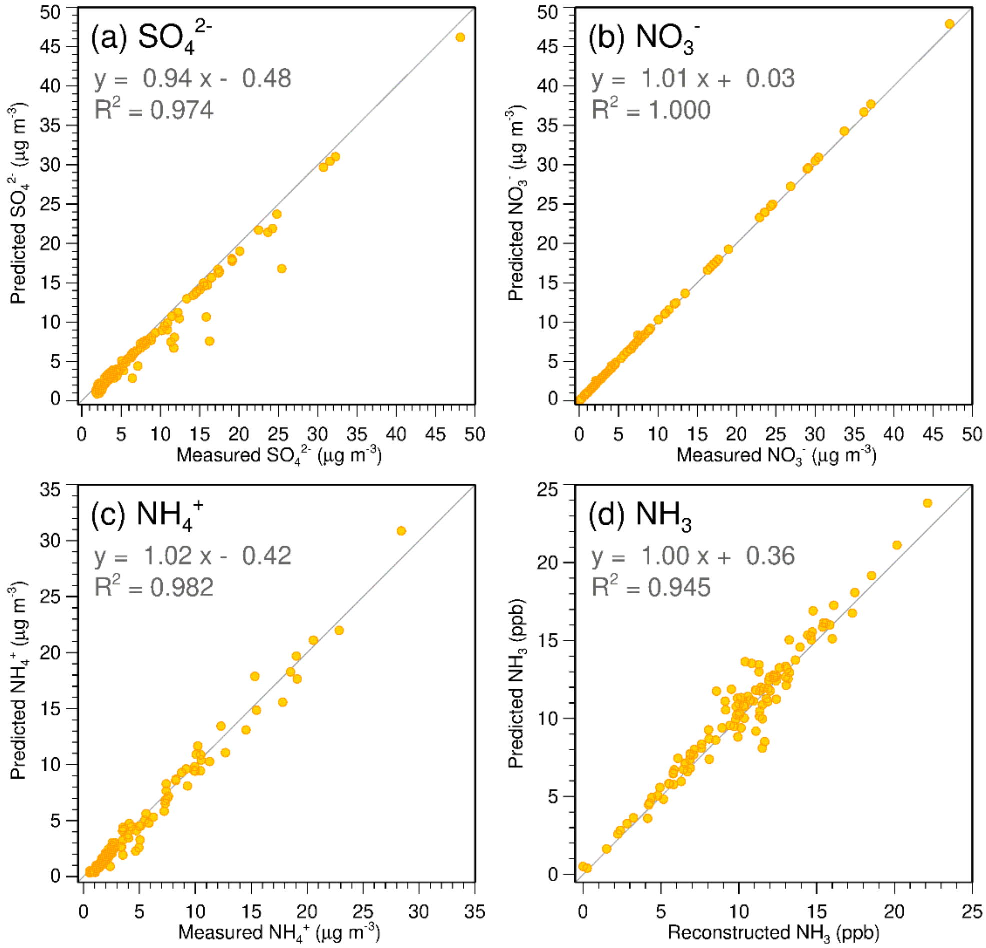

Figure S8: Evaluation of the thermodynamic model. Comparisons of the predicted and measured (a) SO_4^{2-}, (b) NO_3^-, (c) NH_4^+, and (d) NH_3 concentrations. Gray solid lines indicate a 1-to-1 relationship. Note that reconstructed NH_3 concentrations are used in this study due to the absence of NH_3 measurements for Seoul during the analysis period.