Supplement of

Physical and chemical constraints on transformation and mass-increase of fine aerosols in northeast Asia

Saehee Lim et al.

Correspondence to: Meehye Lee (meehye@korea.ac.kr)

Supplementary Text

Text S1. Calculations of Particle Growth Rate (GR) and Condensation Sink (CS)

Particle Growth Rate

The particle growth rate (GR) describes the change in the particle diameter within a time period when a new particle formation occurs. The GR during the period of the top 10% of EOF1 was calculated via the maximum-concentration method (Kulmala et al., 2012), using the following equation:

$$GR_{Dp_1 \sim Dp_2} = \frac{Dp_2 - Dp_1}{t_{Dp_{2,max}} - t_{Dp_{1,max}}}$$

where Dp is the particle diameter (nm), and t_{Dp} is the time when the maximum number concentration of Dp occurs.

Here, GR was defined by the linear least-squares fit of points ($t_{Dp,max}$, Dp) in the size range of 10–25 nm. Consequently, the calculated mean ± SD of $GR_{10–25}$ was 2.13 ± 1.90 nm h^{-1}.

Condensation Sink

The condensation sink (CS) describes the rate at which condensable gaseous molecules condense on pre-existing particles. The loss rate of molecules can be obtained by integrating over the particle size spectrum (Kulmala et al., 2001; Pirjola et al., 1999):

$$CS = 2\pi D \sum_{Dp} \beta_{m,Dp} Dp N_{Dp}$$

where N_{Dp} is the number concentration of Dp, D is the diffusion coefficient, and $\beta_{m,Dp}$ denotes the transitional regime correction factor. The factor $\beta_{m,Dp}$ can be calculated by applying *Fuchs theory* and the approach given below (Fuchs, 1965; Seinfeld and Pandis, 2006).

$$\beta_F = \frac{\overline{C_A} R_p}{\overline{C_A} R_p + 4D}$$

As $R_p = \frac{D_p}{2}$, the equation be rewritten as

$$\beta_F = \frac{\overline{C_A} D_p}{\overline{C_A} D_p + 8D}$$

$\overline{C_A}$ indicates the mean speed of the molecules, which can be expressed as $C_A = \sqrt{\frac{8kT}{\pi m_A}}$. We consider D as the diffusion coefficient of H_2SO_4 in air and set D_{H2SO4} = 0.104 (0.091–0.113) $cm^2\ s^{-1}$, which was calculated via the FSG/LaBas method (https://www3.epa.gov/ceampubl/learn2model/part-two/onsite/estdiffusion-ext.html). The CS values in the sizes of 10–25 nm and 100–470 nm were calculated for the top 10% of both EOF1 and EOF2. Consequently, $CS_{10–25nm}$ and $CS_{100–470nm}$ were 0.011 ± 0.020 (× $10^{-2}\ s^{-1}$) and 1.26 ± 0.86 (× $10^{-2}\ s^{-1}$), respectively, for EOF; the corresponding values for EOF2 were 0.002 ± 0.003 (× $10^{-2}\ s^{-1}$) and 1.76 ± 1.04, respectively.

Supplementary Figures

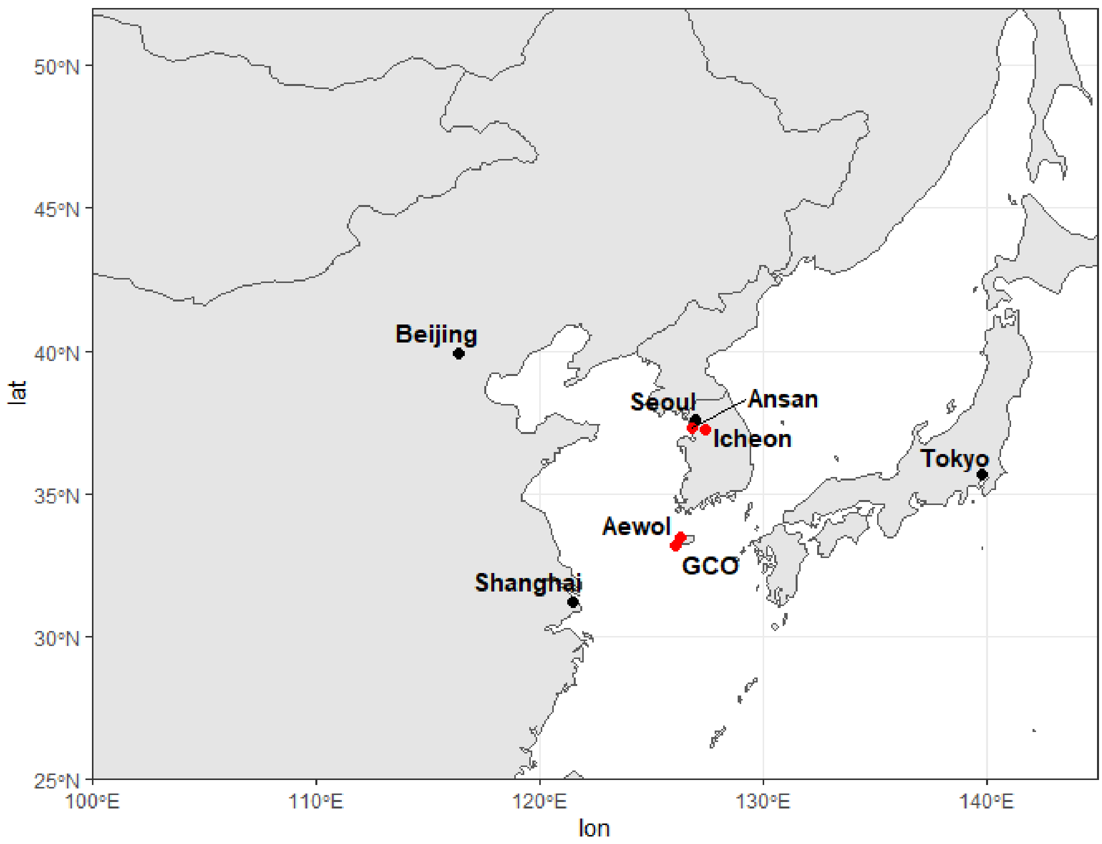
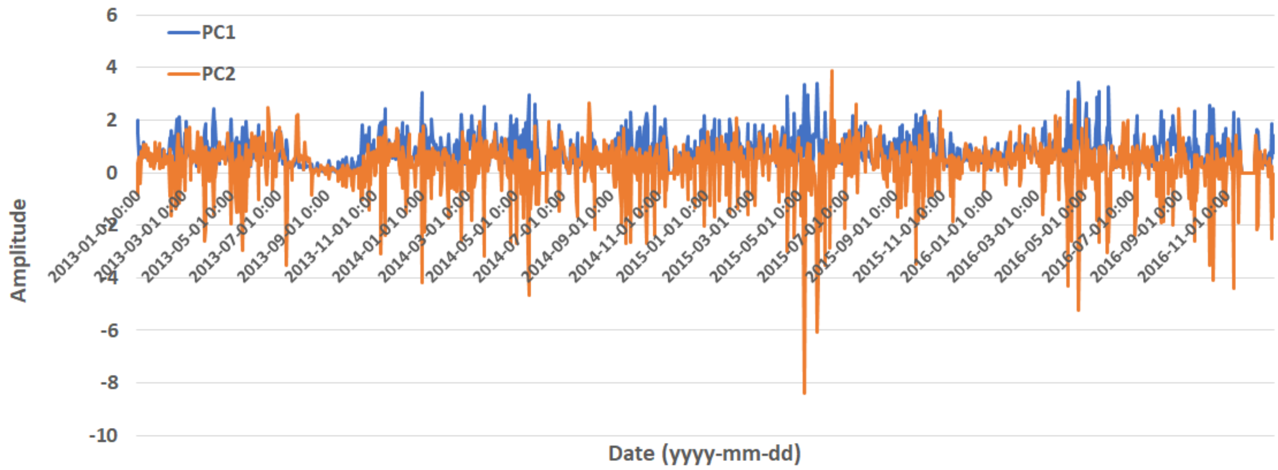


Figure S1. Map of the study region. Red points denote measurement sites: Jeju Air Quality Research Center in Aewol (33.21° N, 126.23° E, 600 m asl) and Gosan Climate Observatory (GCO; 33.17° N, 126.10° E) in Jeju island, and Seoul (37.61° N, 126.93° E), Ansan (33.17° N, 126.10° E, 70 m asl), and Icheon (37.27° N, 127.43° E) in Seoul metropolitan area (SMA).

a)



b)

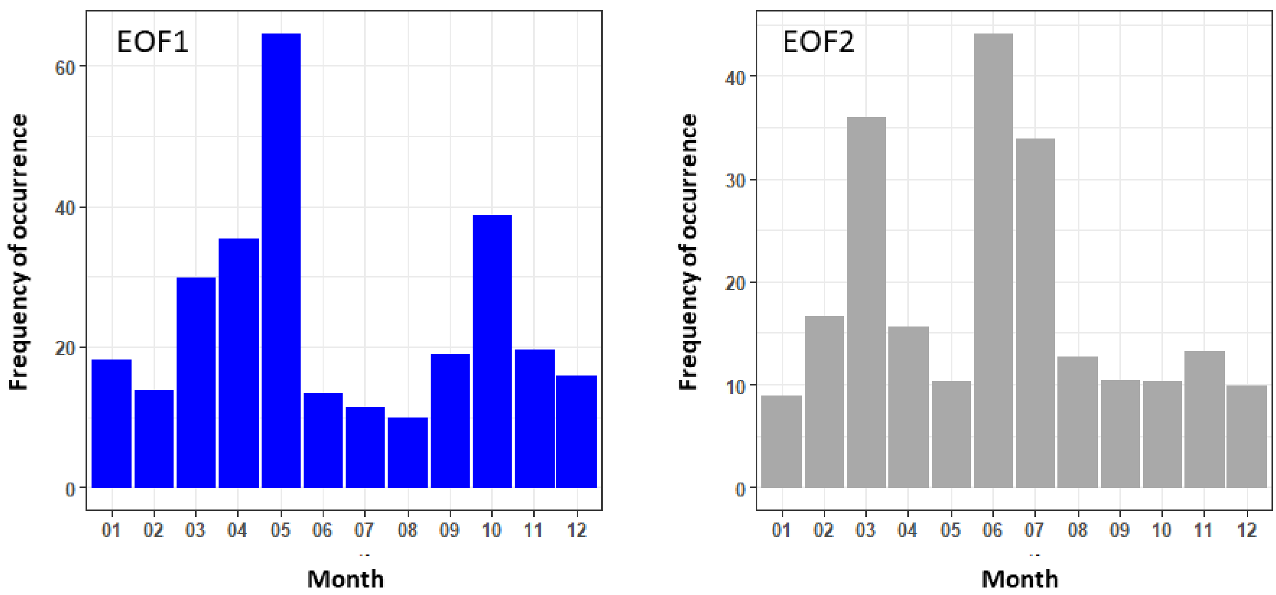


Figure S2. Summary of two major PC time series. (a) Amplitude of PC1 and PC2. (b) Monthly frequency of occurrence for days with top 10% amplitudes of PC1 and PC2, referred to as “EOF1” and “EOF2”.

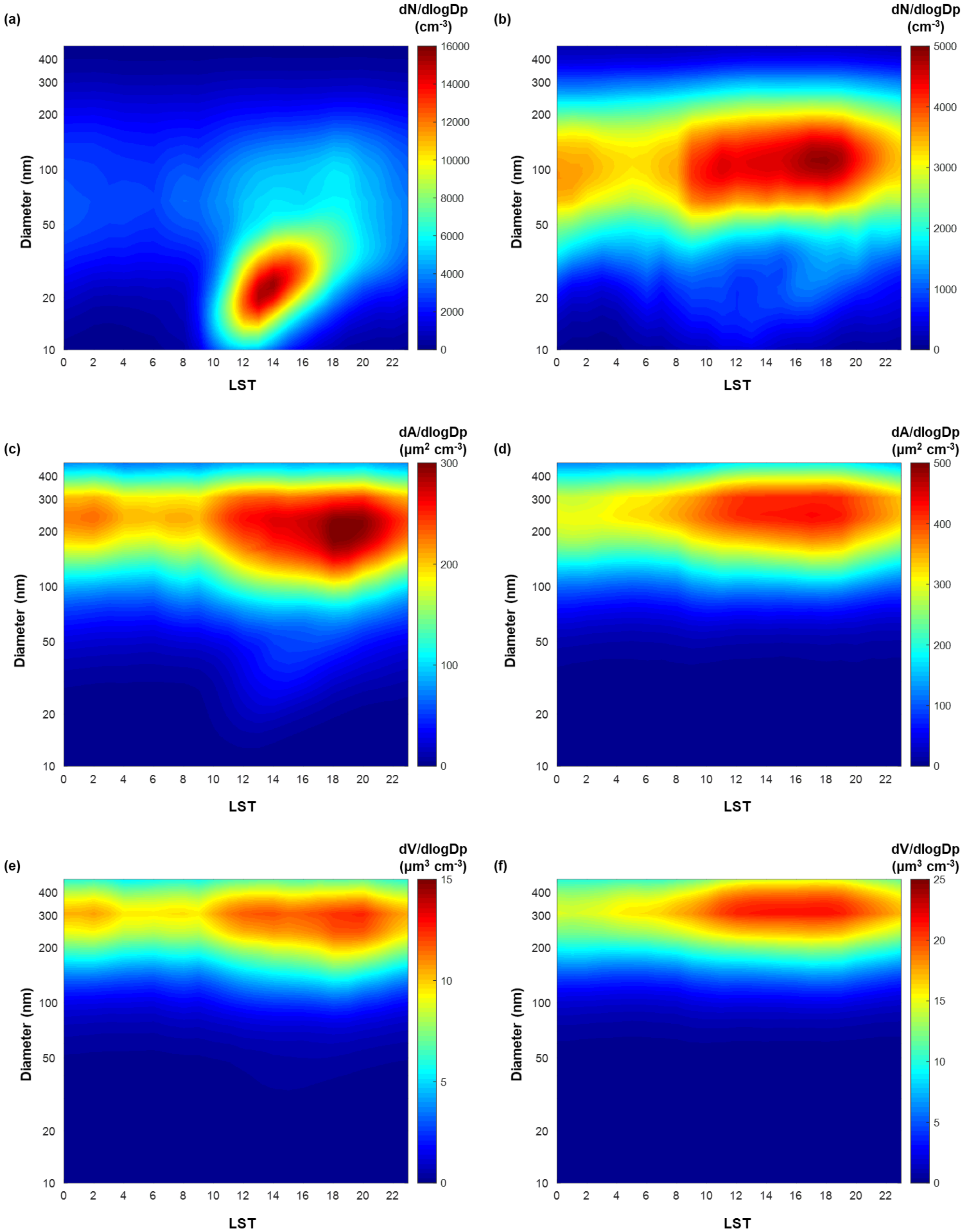
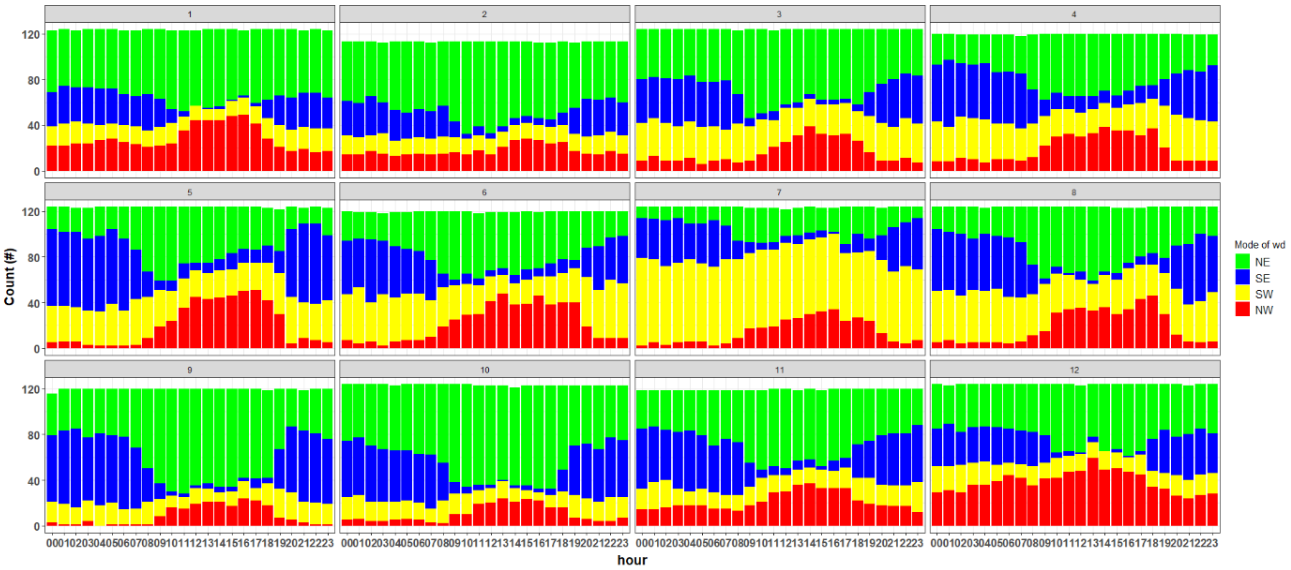
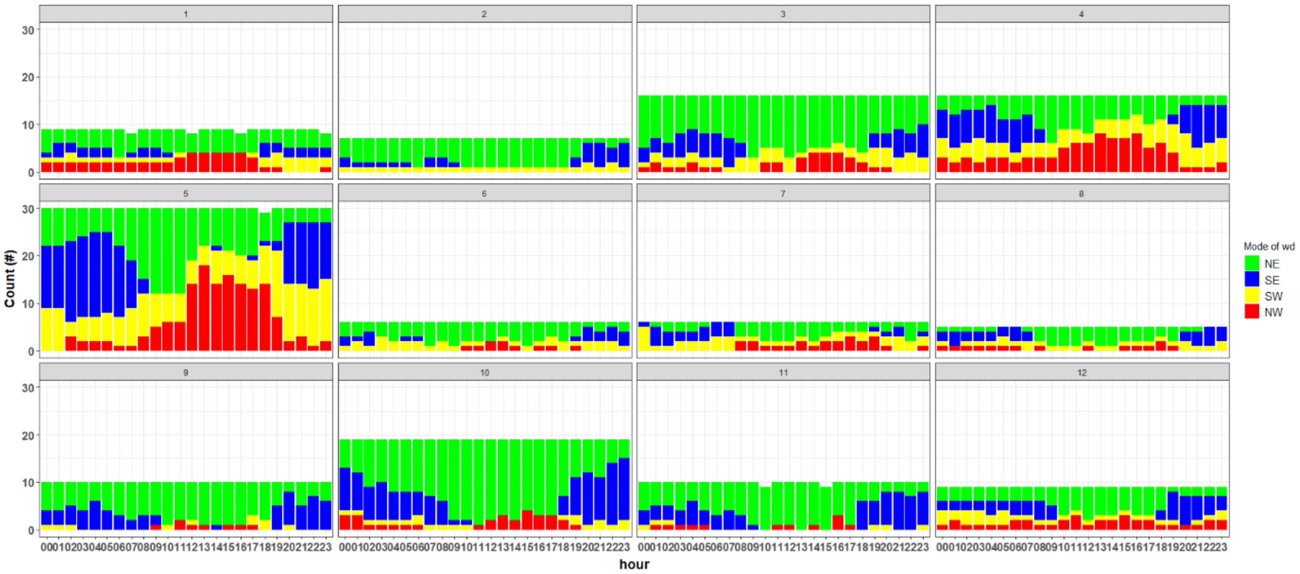


Figure S3. Aerosol size distributions of EOF1 (left panels) and EOF2 (right panels). (a) and (b) aerosol number size distributions, (c) and (d) aerosol surface size distributions, and (e) and (f) aerosol volume size distributions.

a)



b)



c)

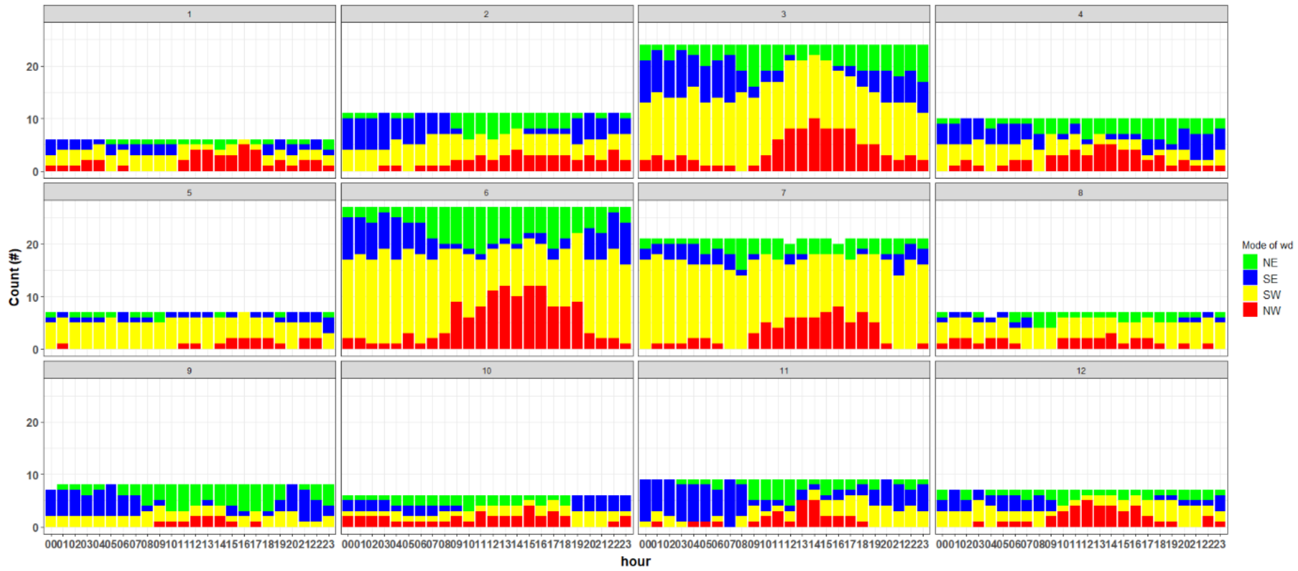


Figure S4. Monthly count of hourly wind sector. (a) Entire period of years 2013–2016, (b) EOF1 period, (c) EOF2 period. Wind directions were categorized into “NE,” “SE,” “SW,” and “NW” directions.

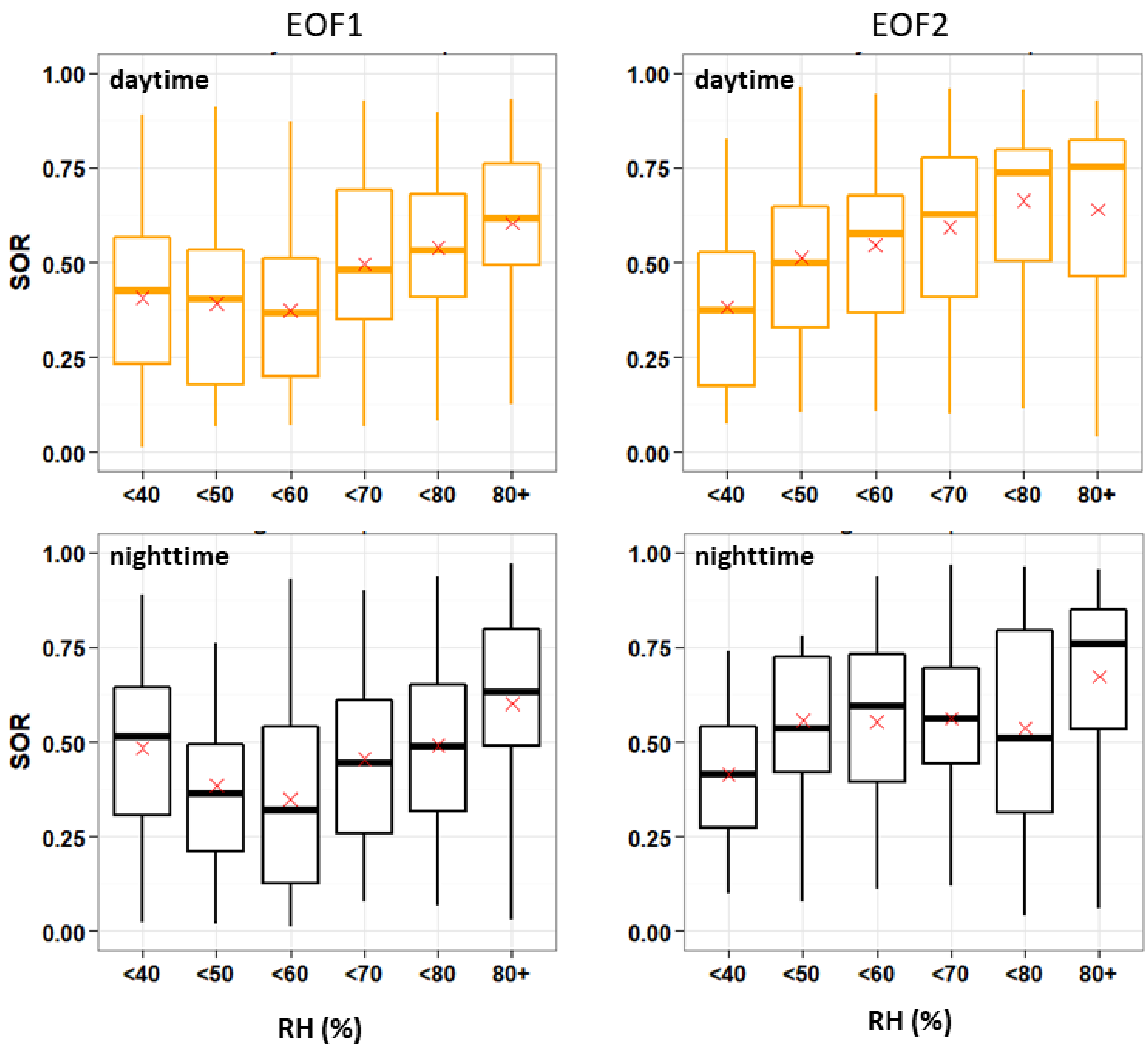


Figure S5. Variations of sulfur oxidation ratio (SOR) as a function of RH for EOF1 (left panels) and EOF2 (right panels) during daytime (8 a.m.–7 p.m. in local time; top panels) and nighttime (8 p.m.–7 a.m. in local time; bottom panels).

References

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