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Supporting Information for

Evidence of haze-driven secondary production of supermicrometer aerosol nitrate and sulfate in size distribution data in South Korea

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Contents of this file

Supplemental section S1, Tables S1, Figures S1 – S9

Section S1. Inha University Ionic Charge Balance Results

A charge balance was conducted between the measured anions and cations based on individual stages of each DLPI^{+} filter set collected at Inha University (Fig. S3; anions/cation on y/x axes, respectively). A slope and y-intercept of one and zero, respectively, would signify perfect balance between anions and cations. The cumulative (polluted, transition, and clean periods) and polluted period data yielded similar slopes of 0.76 and 0.77, respectively, and similar y-intercepts of -0.04 and -0.02, respectively. The transition and clean period sets exhibited decreasing slopes (0.59 and 0.37, respectively), with the clean period's best fit line exhibiting the lowest correlation coefficient ($r = 0.72$). Therefore, there were undetected anions for the entire study period, which became more significant as PM levels decreased. Sources of undetected anions include potentially a wide variety of organic species that are difficult to speciate with IC. That the charge balance was better in more polluted conditions suggests that secondary inorganic anions such as NO_3^- and SO_4^{2-} (i.e., easier to detect via IC), were relatively more abundant as compared to other species such as organics.

Table S1. Limits of detection (LOD) for species analyzed from DLPI$^+$ size-resolved filters collected at Inha University during the study period and the percentage of samples for each species that had concentrations below the LOD and thus replaced with LOD/2. Bromide was excluded from calculations as it was always below its LOD.

Parameter	LOD (ppb)	% of samples below LOD
Adipate	22.66	85
Br^-	25.09	100
Ca^{2+}	45.23	5
Cl^-	2.144	7
K^+	26.24	14
Maleate	6.970	95
Mg^{2+}	36.93	49
MSA	12.32	98
Na^+	43.48	9
NH_4^+	42.43	10
NO_3^-	8.917	7
Oxalate	12.31	7
Phthalate	20.69	95
SO_4^{2-}	11.98	4

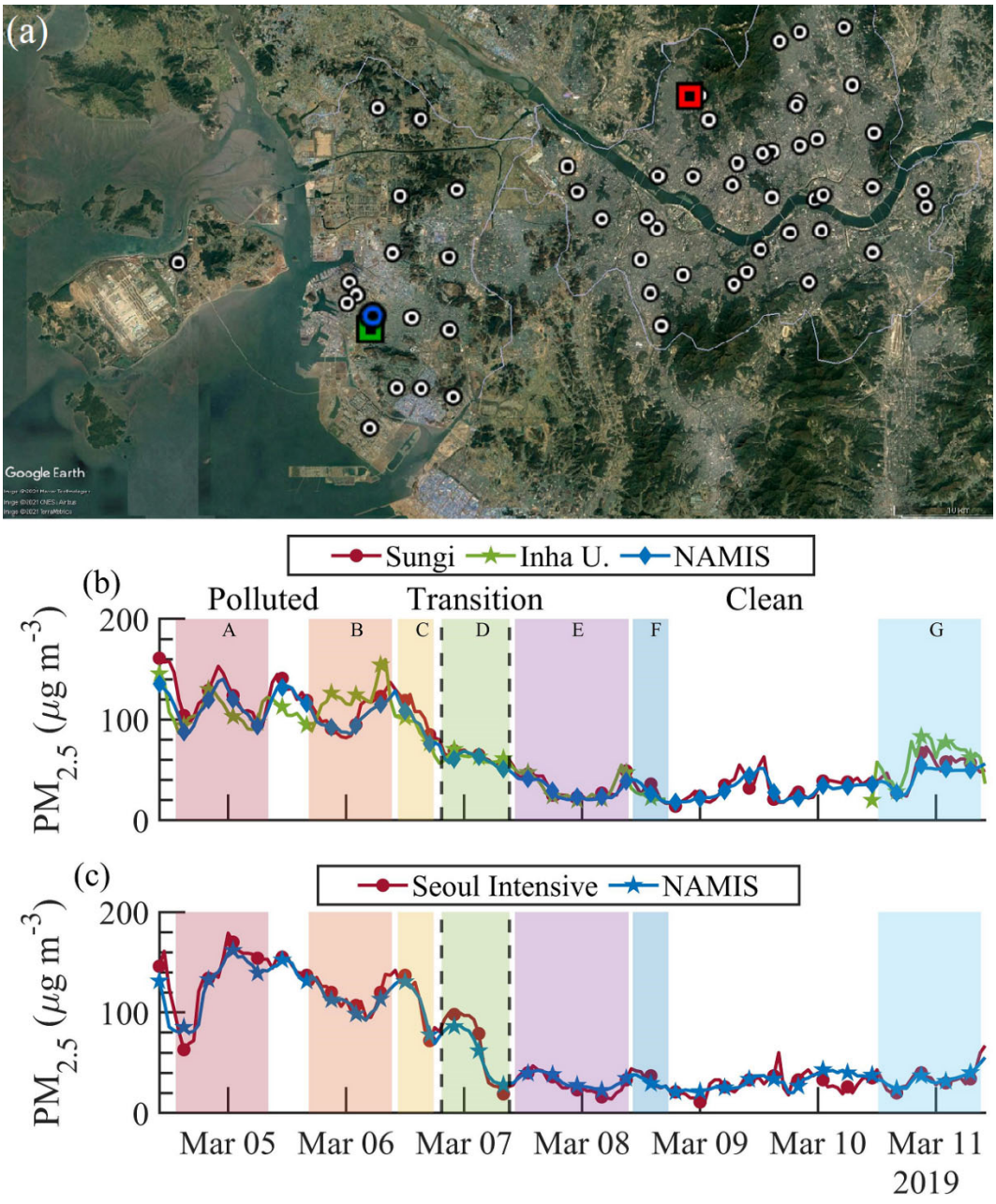


Figure S1. (a) Spatial map showing the 17 and 40 National Ambient air quality Monitoring Information System (NAMIS) stations in Incheon and Seoul, respectively, along with the three main surface sites relied on for this study (green = Inha University, blue = Sungi [also a NAMIS station], red = Seoul Intensive Monitoring Station). $PM_{2.5}$ comparison between (b) city-wide Incheon mean values and those for Sungi and Inha University, and between (c) city-wide Seoul mean values and those for Seoul Intensive Monitoring Station. Coefficients of determination (R^2) between the data points: (b) (Inha University) $R^2 = 0.82$ and (Sungi) $R^2 = 0.98$; (c) $R^2 = 0.96$. Shaded regions of panels b-c are labeled with individual DLPI+ sets overlapping in time. All times are reported in Korea standard time (KST), where KST is UTC + 9 hrs. Image Data: ©2021 Google Earth, Maxar Technologies, CNES/Airbus, TerraMetrics.

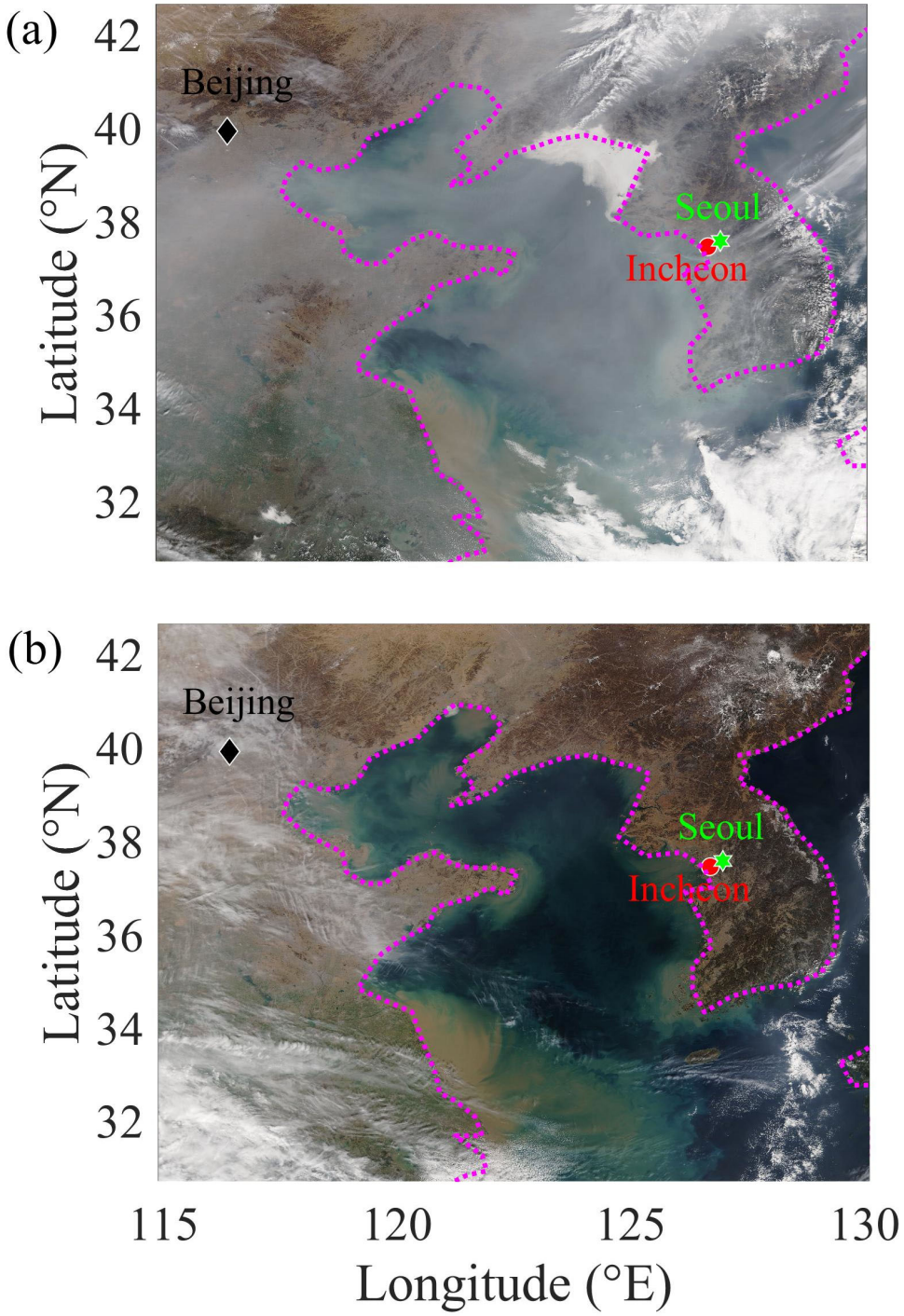


Figure S2. Satellite view of the study region comparing (a) a polluted day on 4 March 2019 and (b) a relatively cleaner day on 8 March 2019. Markers are shown for Seoul and Incheon. These are representative days during the sampling period for polluted and clean conditions without significant cloud interference. MODIS Terra image retrieved from https://worldview.earthdata.nasa.gov/.

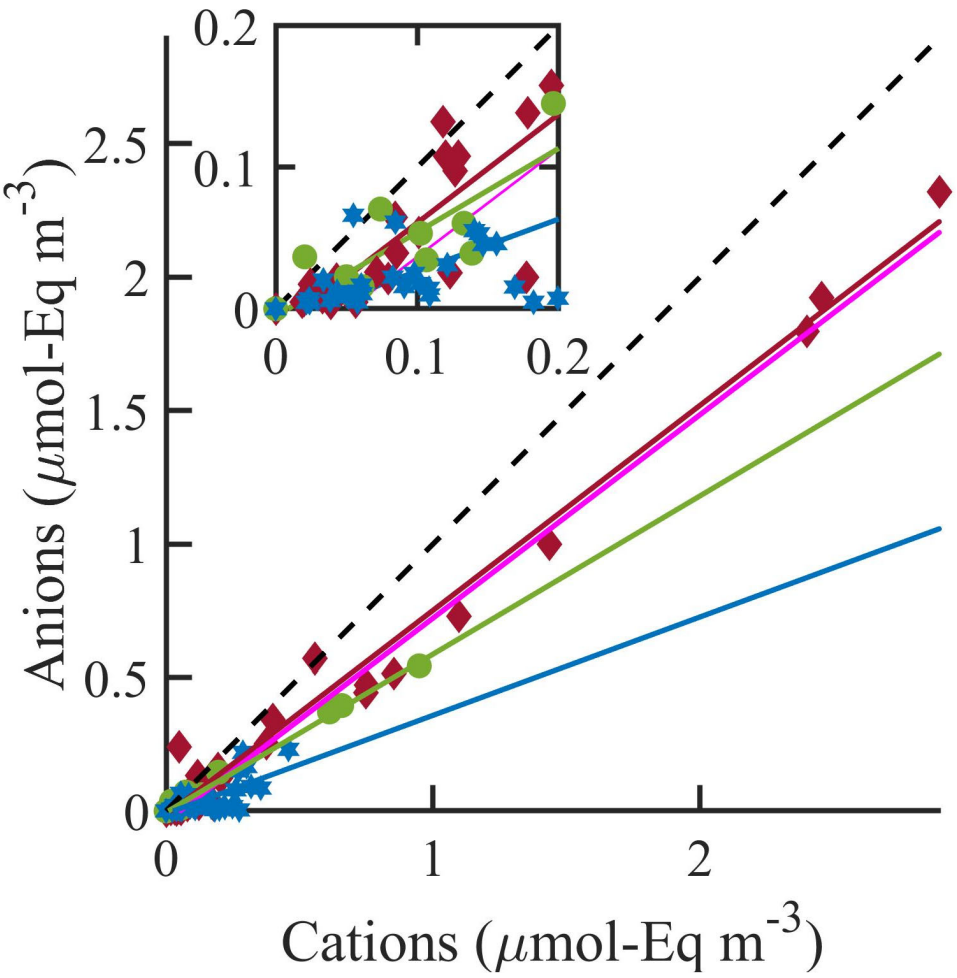
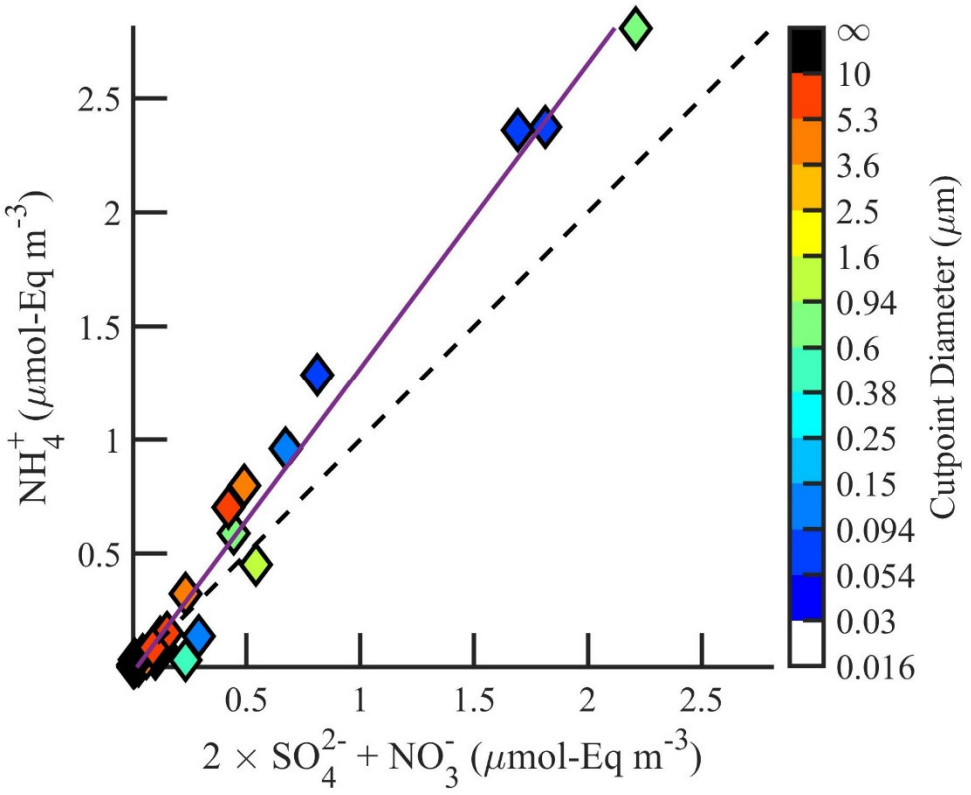


Figure S3. Charge balance analysis for each DLPI^{+} filter stage among the seven sets collected during the study period at Inha University (Incheon). Information for the four best fit lines is as follows (Slope, y-intercept, correlation coefficient (r), number of points): All (magenta) = 0.76, -0.04, 0.97, 98; Polluted (red) = 0.77, -0.02, 0.99, 42; Transition (green) = 0.59, -0.01, 0.99, 14; Clean (blue) = 0.37, -0.01, 0.72, 42. The dashed-black line represents the 1-to-1 line. An inset panel shows a zoomed version of the lowest concentration points.



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75 **Figure S4. Charge balance between ammonium and its association inorganic anions (sulfate and nitrate) for**
76 **the three polluted sets collected at Inha University (Incheon). Markers are color-coded by the cutpoint diameter**
77 **of specific DLPI$^+$ filter samples. The best-fit line is the solid curve (slope = 1.34, y-int = -0.02, r = 0.99, n = 20),**
78 **and the dashed line is the 1:1 line.**

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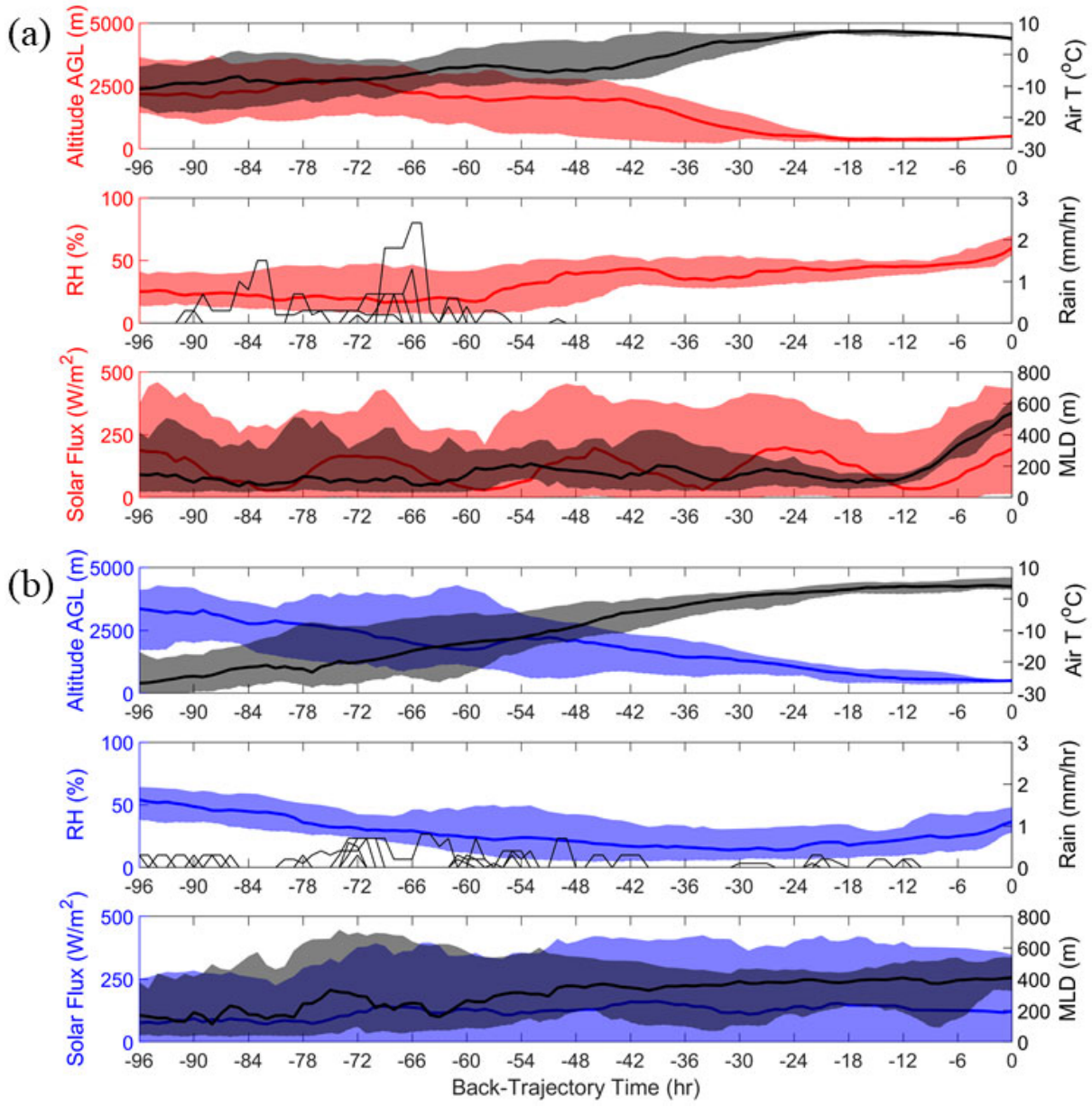


Figure S5. Time series of various parameters (altitude, air temperature, relative humidity, rain fall, downward solar radiation flux, mixing layer depth) along the back-trajectories in the (a) polluted (4 March 10:00 KST – 6 March 19:00 KST) and (b) clean (7 March 09:15 KST – 11 March 10:00 KST) periods. Solid lines represent the median, shaded regions represent the 25th and 75th percentiles, and for rain, data shown represented by the black solid lines correspond to individual trajectories.

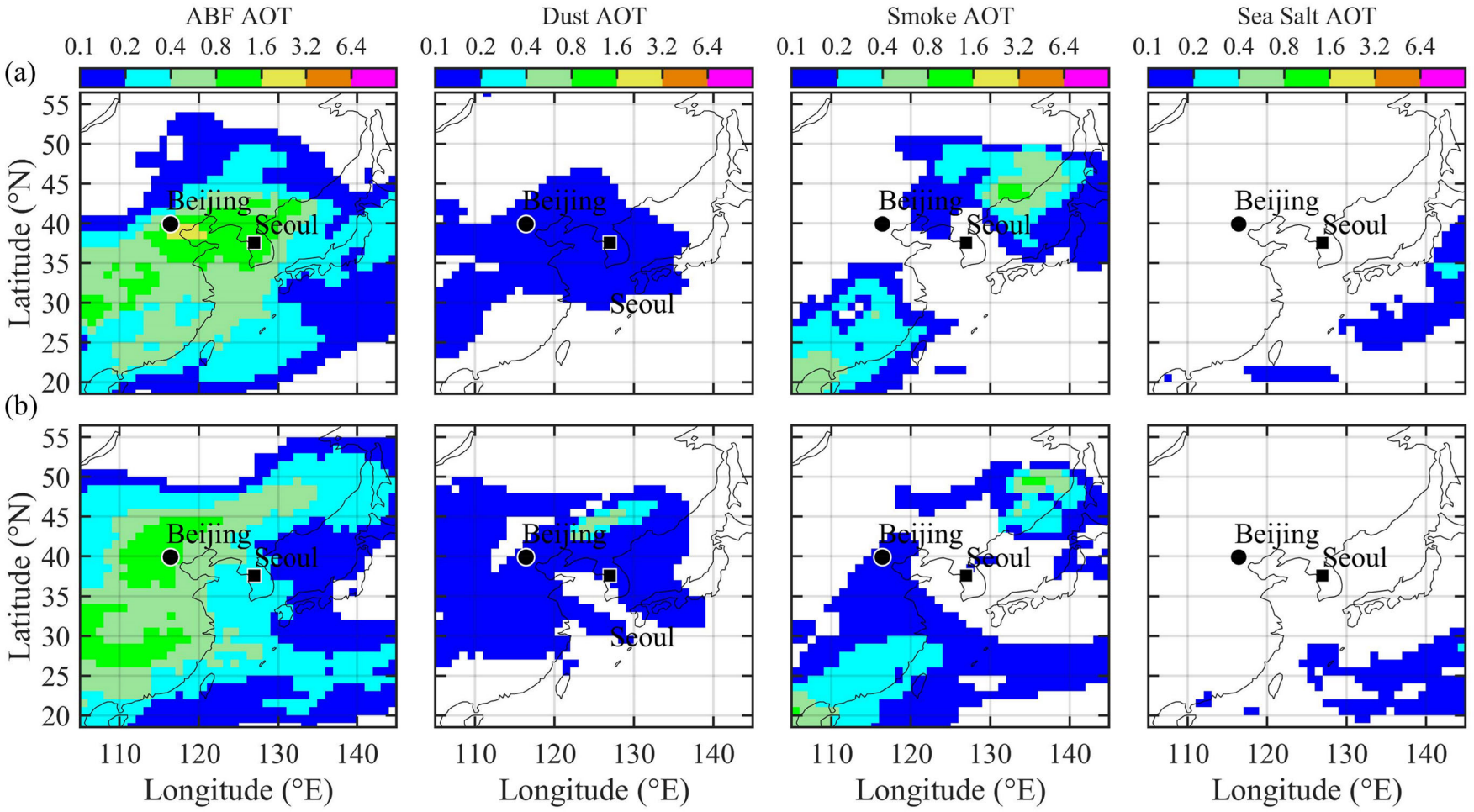


Figure S6. Spatial maps of NAAPS speciated 555 nm AOT for days representative of (a) polluted (5 March 2019 at 03:00 KST) and (b) clean (9 March 2019 at 03:00 KST) conditions. Figure S2 shows the visual satellite imagery around the time of these two days.

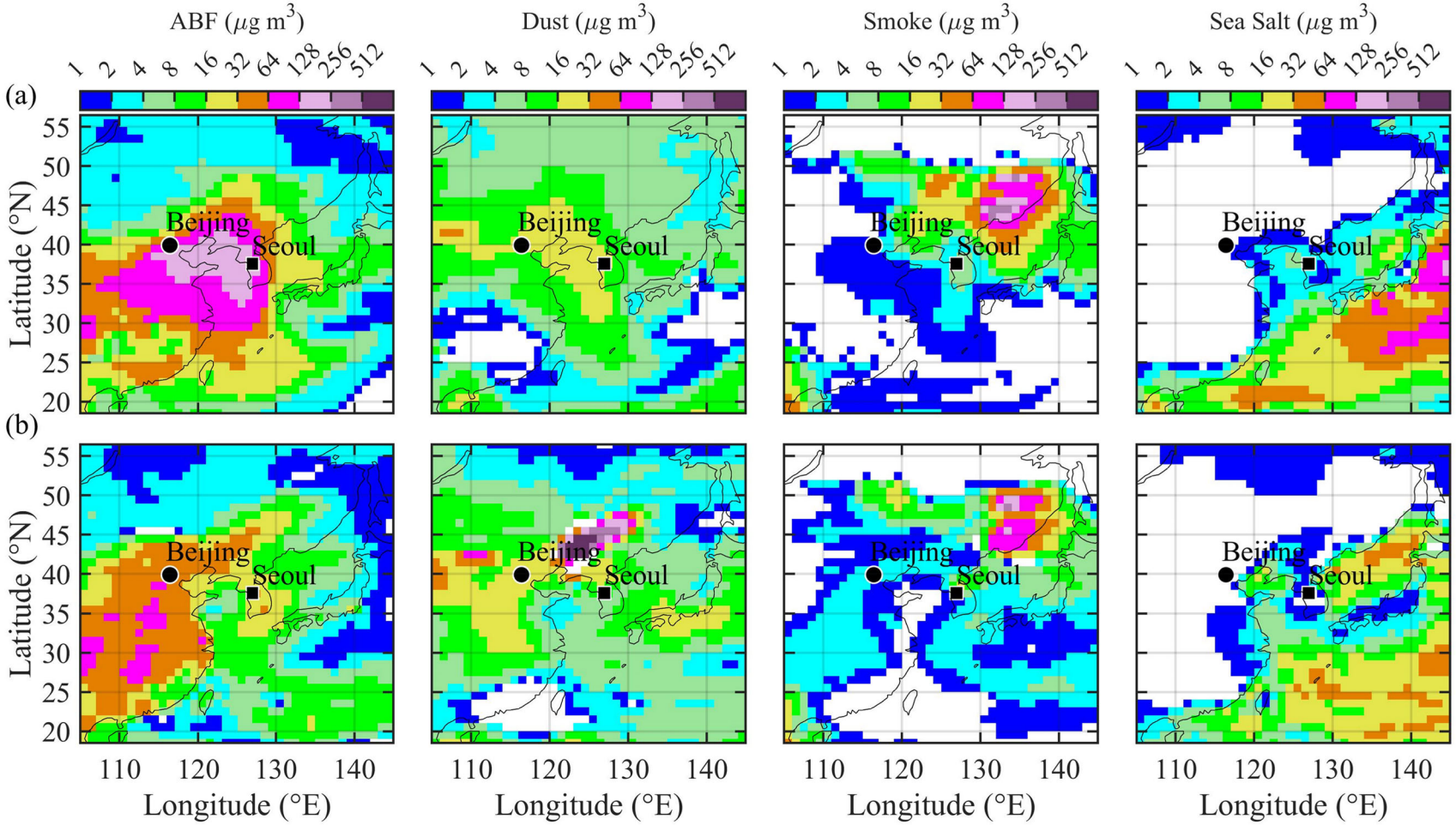


Figure S7. Spatial maps of NAAPS speciated surface mass concentrations for days representative of (a) polluted (5 March 2019 at 03:00 KST [UTC + 9 hrs]) and (b) clean (9 March 2019 at 03:00 KST) conditions. Figure S2 shows the visual satellite imagery around the time of these two days.

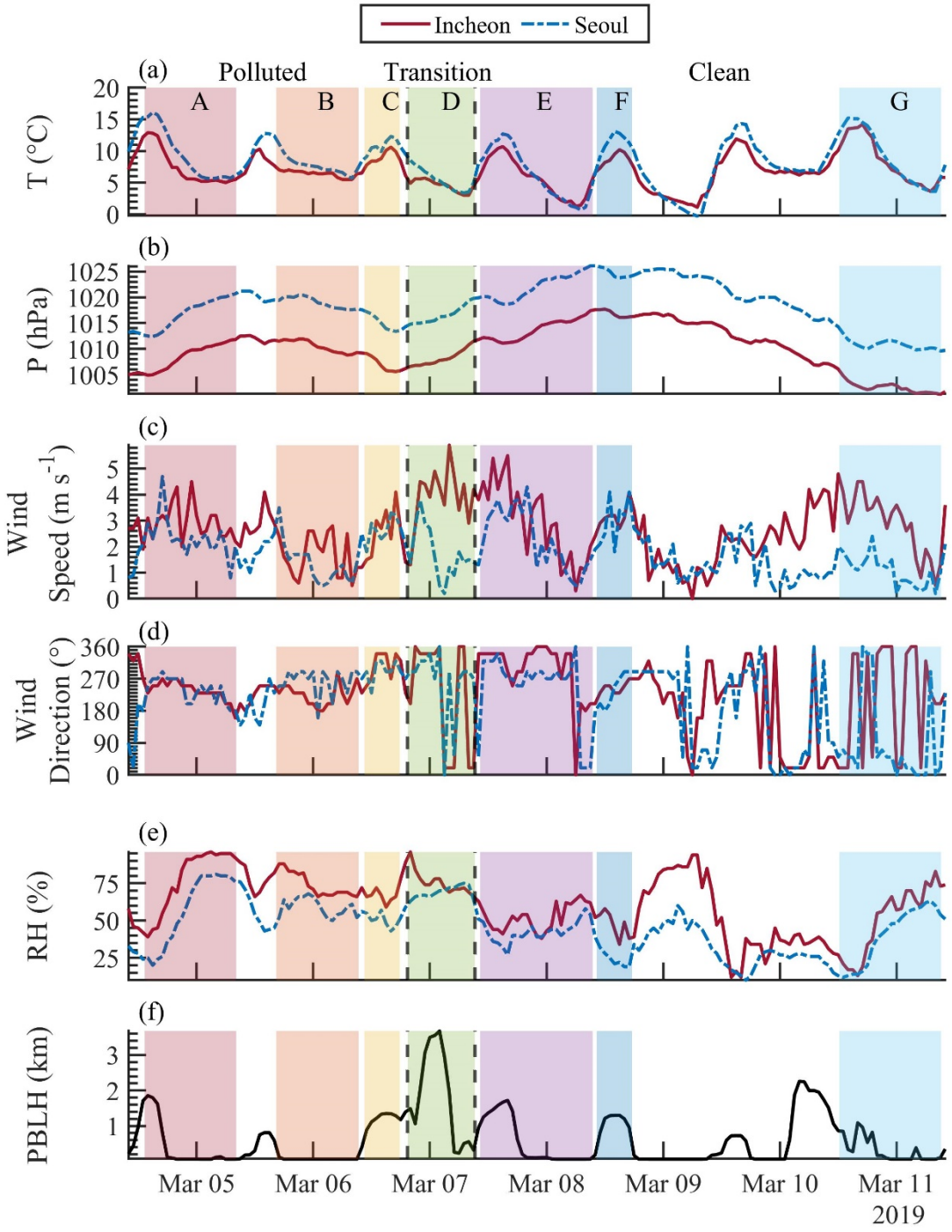
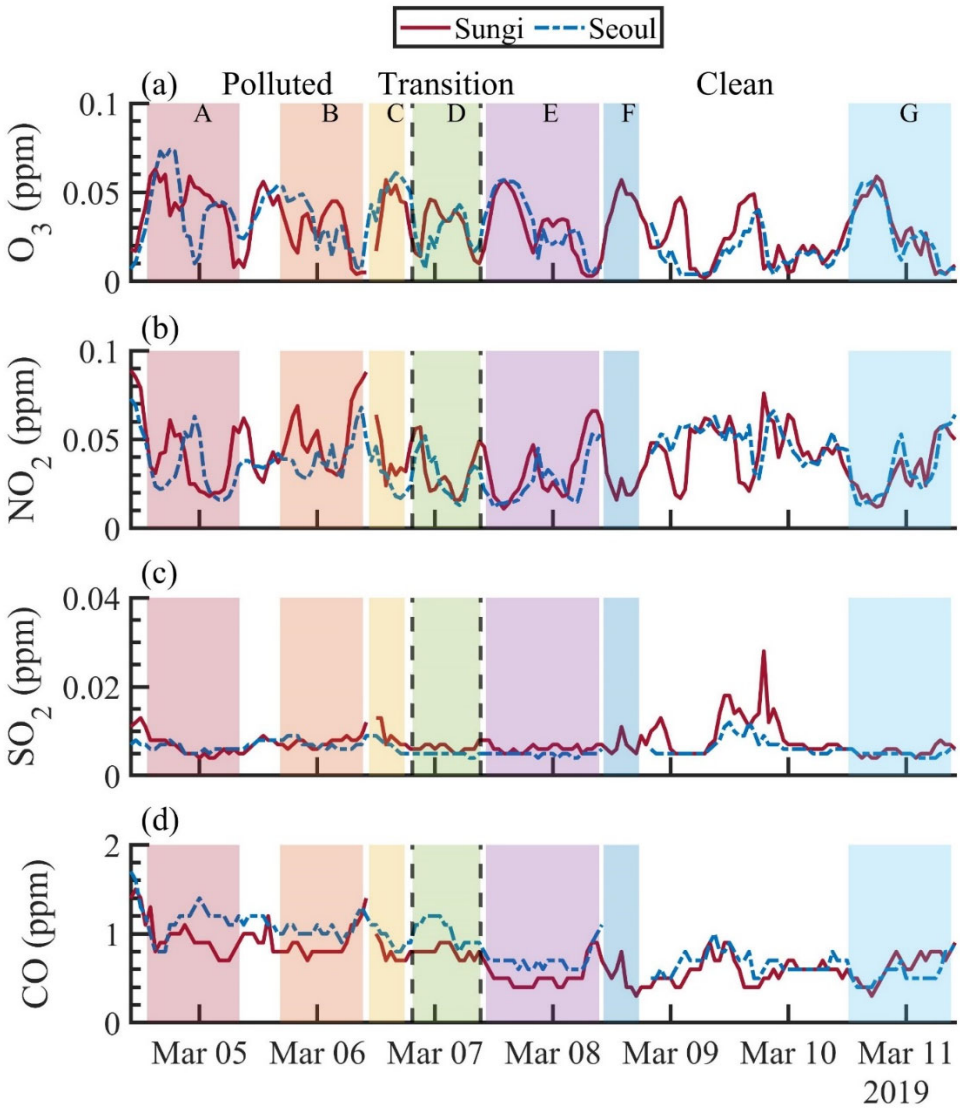


Figure S8. Time series of (a) ambient temperature (T), (b) ambient pressure (P), (c-d) wind speed and direction, (e) relative humidity (RH), and (f) planetary boundary layer height (PBLH) for Incheon and Seoul. The dashed black vertical lines separate the (left) polluted, (middle) transition, and (right) clean periods. The grid size of the MERRA-2 PBLH product encompasses both Incheon and Seoul, hence there is only one curve in panel f. Shaded regions are labeled with individual DLPI$^+$ sets overlapping in time.



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99 **Figure S9. Time series of the following gas species measured at the sites denoted by the colors in the legend: (a)**
100 **ozone (O_3); (b) nitrogen dioxide (NO_2); (c) sulfur dioxide (SO_2); and (d) carbon monoxide (CO). The dashed**
101 **black vertical lines separate the (left) polluted, (middle) transition, and (right) clean periods. Shaded regions**
102 **are labeled with individual DLPI$^+$ sets overlapping in time.**

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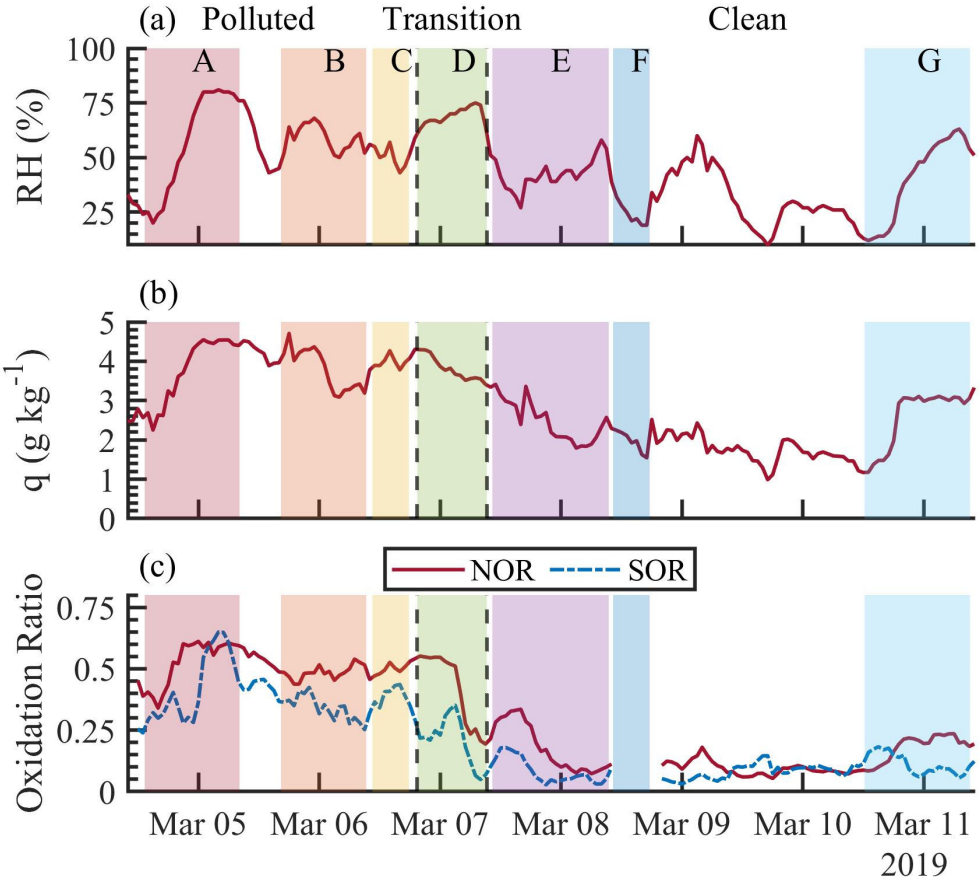

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105 **Figure S10. Time series of (a) relative humidity (RH), (b) specific humidity (q), and (c) nitrogen and sulfur**
106 **oxidation ratios (NOR and SOR, respectively) based on hourly Seoul data. The dashed black vertical lines**
107 **separate the (left) polluted, (middle) transition, and (right) clean periods.**

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