Supporting Information *for*

Haze episodes before and during the COVID-19 shutdown in Tianjin, China: Contribution of fireworks and residential burning

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Contents of this file

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Text S1. HYSPLIT backward-trajectory calculation.

Air trajectory analysis provides useful information regarding the spatial scale of the airflows that arrived at a given location (here N 38.99°, E 117.34°). 48 h backward trajectories were calculated at heights of 100 m AGL using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) for every hour during the measurement campaign. Clustering analysis was performed for the derived 48 h backward trajectories thus produced four clusters: C1-Southwestly, C2-Northerly, C3-Northeasterly, and C4-Northwesterly (Fig. S6).

Text S2. Evolution of pollution episodes

HE1. The peak values of $PM_{2.5}$ in the studied cities were decreased from the north (Beijing) to the south (Cangzhou) of the BTH region. Before reached its peak value at 11:00 on 4 January (Fig. 1a), the $PM_{2.5}$ concentration in Tianjin elevated under poor dispersion conditions that associated with low VC values (Fig. 2(b, c, d), periods of MLH less than 400 m and weak surface winds, resulting in average hourly increase rate of 2 μg m^{-3} $PM_{2.5}$ per hour. Surface winds in Tianjin were gradually shifted from SW-W to N at heights below 3 km AGL (Fig. 1(c, d)). Since then, the pollutants were carried off by cold high-speed N and NE winds (Fig. 2b). Given that the arrival time of the N winds to each city delayed from north to south in the BTH region, the pollution episode was first ended in Beijing, followed by Lang'fang, Tangshan, Tianjin and Cangzhou (Fig. 1a). The end of HE1 in Tianjin was facilitated by the sharply increased VC (>1000 m^2 s^{-1}) in the late afternoon on 4 January.

HE2. There were four transient periods with pronounced increases in $PM_{2.5}$ during HE2. The $PM_{2.5}$ concentration was sharply elevated from ~ 50 μg m^{-3} to the first peak value of 175 μg m^{-3} when the air masses started shifting from N to SW in the early morning (05:00) on 15 January, together with MLH remained <200 m and surface wind speed as weak as <1 m s^{-1}. During this period enhanced by poor dispersion, the average increase rate of $PM_{2.5}$ was 18.3 μg m^{-3} h^{-1}, with a maximum rate of 29 μg m^{-3} $PM_{2.5}$ per hour. The first peak value in Tianjin was higher than other mentioned surrounding cities. There was significantly elevated number concentration of particles with sizes around 100 nm without notable size shifting in the morning on 15 January (HE2), which was likely

resulted from near-surface regional transport. The second pronounced increase of $PM_{2.5}$ from 127 μg m^{-3} to the second peak value of 285 μg m^{-3} was due to the decrease of MLH from ~700 m to ~200 m, as well as wind speed decreased from 1.5 m s^{-1} to less than 1 m s^{-1}. The average increase rate of $PM_{2.5}$ was 9.2 μg m^{-3} h^{-1}, with a maximum rate of 42 μg m^{-3} $PM_{2.5}$ per hour. In the afternoon of 16 January, a weak humid northerly wind arrived at Tianjin for a short period thus reduced the surface $PM_{2.5}$ concentrations. The third shapely increase of $PM_{2.5}$ from 156 to 285 μg m^{-3} was again partly due to the decrease of MLH, with a maximum increase rate of 22 μg m^{-3} h^{-1}. From the night of 17 January to the next day, it was foggy with rime appeared on a large scale. Note that a persistent strong temperature inversion above the fog (>200 m AGL) was observed in the early morning of 18 January. The low temperature and high humidity within the MLH provided favorable conditions for the formation of sulfate and nitrate through aqueous-phase and/or heterogenous chemistry. Those adverse meteorological effects together resulted in the fourth elevation of $PM_{2.5}$ from 113 to 293 μg m^{-3} within 10 h, with an average increase rate of $PM_{2.5}$ of 20 μg m^{-3} h^{-1} (maximum value of 48 μg m^{-3} h^{-1}). Since then, the $PM_{2.5}$ was gradually decreased along with the surface winds increased to ~2 m s^{-1}. With the arrived air masses shifted from SW (2~3 m s^{-1}) to N to NW (~16 m s^{-1}), $PM_{2.5}$ quickly declined from ~173 to ~20 μg m^{-3}. As shown in Fig. 1b, the end of HE2 in Beijing ahead of those in Langfang, Tangshan, Tianjin and Cangzhou. Overall, the three times drop of $PM_{2.5}$ in HE2 were all occurred during daytime when MLH rose. Whereas every shapely increases of ground $PM_{2.5}$ associated with decreased MLH at nighttime as a likely result of the downmixing of aerosol aloft (Fig. 1b), particularly for the last increase. In that case, the increase of $PM_{2.5}$ in Tianjin was ahead of those in Langfang, Beijing and Tangshan, suggesting the pollution transported from south to north of the BTH.

HE3. The $PM_{2.5}$ concentrations were elevated to pollution level (>75 μg m^{-3}) at the late night of 21 January when the air masses shifted from N to SW with speed decreased from >10 m s^{-1} to <2 m s^{-1}, concurrent with MLH decreased from ~800 m to <200 m. The $PM_{2.5}$ concentrations increased again around next midday, during the time when MLH and surface wind speed rose. Meanwhile, there was clearly sinking of aerosol at late night of 21 January (Fig. 3). Before $PM_{2.5}$ reached its maximum value, the MLHs were stay below 200 m, surface wind speeds were <1.0 m s^{-1}, and the average air masses

speed approximate 1.5 m s^{-1}. In this stagnant environment, the ground temperature was below 0°C and RH reached about 90%. $PM_{2.5}$ gradually increased to the maximum value (239 μg m^{-3}) from the afternoon of 22 January to the next morning, accompanied by strong sinking of aerosol from >1.0 km AGL down to the ground (Fig. 1b). Since then, $PM_{2.5}$ concentrations were declined to 89 μg m^{-3} then increased again to 113 μg m^{-3}, as the MLH elevated from 150 to 1000 m and surface winds increased from ~1.0 to ~5.0 m s^{-1}. The last increase of $PM_{2.5}$ was possibly caused by winds convergence in Tianjin. The SE winds run into the N winds for a short period (Fig. 1(c, d)). Pollutants were aggregated in Tianjin before the strong dry northerly winds arrived at 19:00 on 23 January with moving speeds great than 13.0 m s^{-1}. The wind profiler radar also observed strong convection flows after the MLH increased (Fig. 1b). The last increase of PM before the end of the pollution episode was featured enhanced particle number concentrations as shown in Fig. 3d, which was caused by regional transport since the airflows were able to carry pollutants from upwind polluted areas, thus exacerbating local pollution. It should be noted that the northerly airflows have passed through Tangshan before arrival in Tianjin (Fig. 2b). Fig. 1a also clearly demonstrates that HE3 was ended first in Beijing, then in Langfang, Tangshan and Tianjin, and Cangzou. The shape of $PM_{2.5}$ concentrations of HE3 is more similar to HE1 than other HEs.

HE4. HE4 was gradually formed since the midnight of 24 January (Chinese New Year Eve) when the direction of air masses shifted from N-NE to SW. The speeds of air masses were decreased from ~10 to 3~4 m s^{-1} and the MLH remained <200 m until $PM_{2.5}$ reached to its highest value of 280 μg m^{-3} at 15:00 on 26 January, with average hourly increase rate of 7.0 μg m^{-3} h^{-1}. The fireworks tracer species (SO_2, Cl^-, K^+ and Mg^{2+}) concurrently rose with $PM_{2.5}$ while NO_2 and CO unchanged. The above results suggest that the increase of $PM_{2.5}$ may be primarily from fireworks emissions. Fireworks display is a traditional event over China to celebrate the Chinese New Year since a long time ago. Note that fireworks displays were banned throughout Tianjin but this was not the case for other cities in the BTH area particularly in the suburban/rural areas, where fireworks were extensively used. Before $PM_{2.5}$ reached its peak value, and the air masses were shifted from SW to NE with speed of ~3.0-4.0 m s^{-1}, and the northeasterly surface winds also increased. Thus, the fireworks emissions were transported from the rural areas

surrounding Tianjin, particularly from the northeast of Tianjin where the severest pollution occurred, as shown in the pollution map (Fig. S4). $PM_{2.5}$ was shapely decreased in Tangshan and elevated in Tianjin and Cangzhou (Fig. 1a). During the midnight of 27 January, the air masses were shift to SW again for a short period. Since then, $PM_{2.5}$ fell from 265 to 181 μg m^{-3}, with an average decrease rate of 9 μg m^{-3} h^{-1}. The last increase of $PM_{2.5}$ in the morning of 29 January was likely due to the downmixing of transported aerosol aloft when the MLH began to rise (Fig. 1b). Though the VC were extremely low during the period of HE4, both the northeast of China and the whole BTH areas experienced serious heavy haze pollution after the Chinese New Year Eve. HE4 was ended by the strong northerly winds that passed through Tangshan, which is similar to HE3. During the daytime of 25 and 26, January, the mass concentrations of both PM_1 and $PM_{2.5}$ transiently increased as the increases of particle number concentration (100-200 nm). The sink of regional transport aerosol was responsible for such increases in PM, as confirmed by the aerosol lidar measurement (Fig. 3). In the subsequent developing stage, strong sinking airflows have been frequently recorded (Fig. 1b). The elevated surface secondary inorganic aerosol (SIA) was partly attributed to the sinking airflows.

HE5. The whole period of HE5 was consisted of two coterminous stages. HE5 was formed when air masses shifted from NE and SW, coupled with decreased MLH. During the first stage, $PM_{2.5}$ was increased from ~20 to 171 μg m^{-3} within 42 h, with a average increase rate of 3.5 μg m^{-3} h^{-1}. As is obviously shown in Fig. 1(a, c, d), the shortly alleviation of $PM_{2.5}$ pollution on 8 February was due to the arrival of relatively clean northerly air. From the midnight of 6 February to the early morning of 8 February, the mass concentrations of both PM_1 and $PM_{2.5}$ transiently increased along with the increase of particle number concentration (100-200 nm). The increase in particulate mass concentrations was caused by the sinking of aerosols transported from the southwest (Fig. 2b), as confirmed by the aerosol lidar measurement (Fig. 3). Before $PM_{2.5}$ reached its first peak value, a short period of inversion was formed in the morning due to the arrival of the dry and relatively warmer northerly winds in the urban canopy layer (Fig. 1(e, f)). Given that the northerly winds were not strong enough to sweep away all pollutants, the local dispersion condition has backed to stagnant again after it disappeared. After the air masses shifted to SW again, the second stage was formed. During this period,

temperature inversion has been observed again and lasted for several days, with the maximum intensity of >4℃ though the ground temperature dramatically rose (Fig. 1f). The increase of $PM_{2.5}$ after the midnight of 8 February was associated with elevated K^+, Cl^- and SO_4^{2-}, indicating enhanced emissions from fireworks because it is also popular to play fireworks during the Lantern festival as well. Winds from SW tend to move pollutants toward north as $PM_{2.5}$ in Cangzhou and Tianjin were gradually decreased and elevated in Langfang and Cangzhou (Fig. 1a). The impact of transported firework-related emissions on HE5 was likely less strong than that in HE4. The pollutants were finally removed by the strong northeasterly winds initially originated from the north.

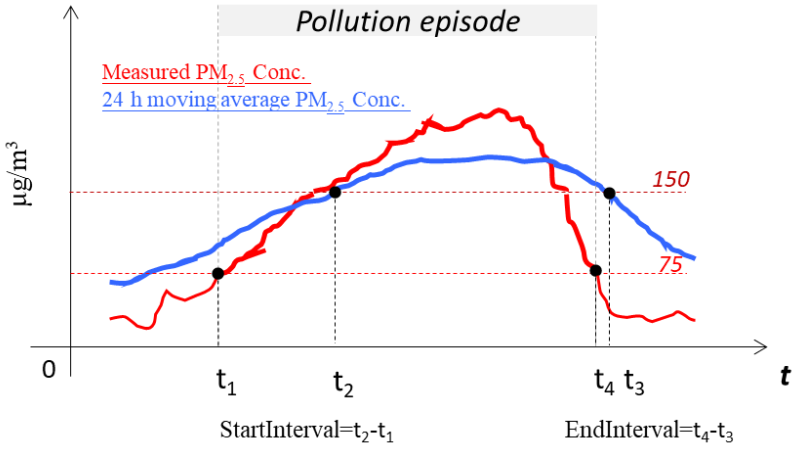
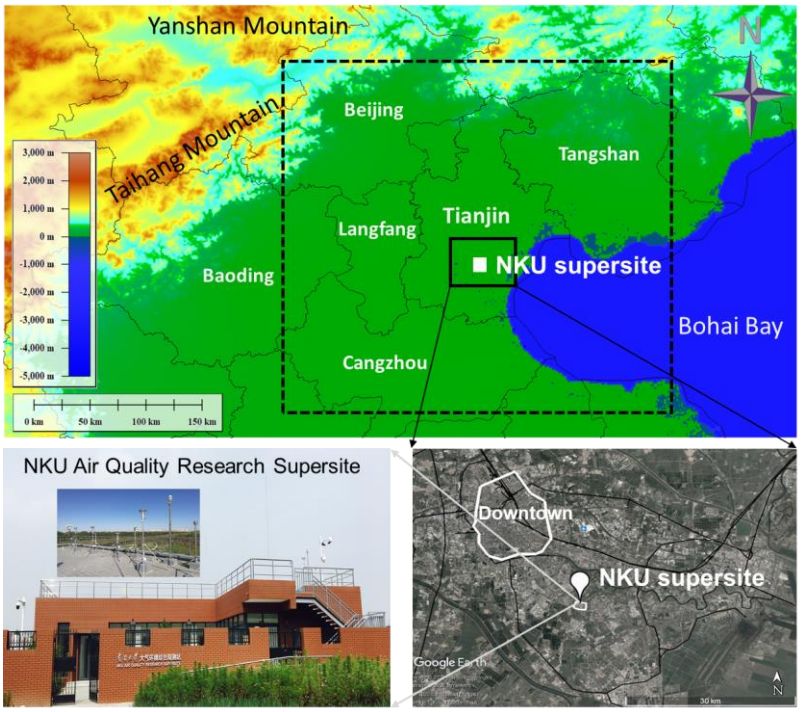


Figure S1. Schematic diagram of starting and ending nodes and duration of haze pollution process. The starting time t_1 is the time when $PM_{2.5}$ concentration begins to reach 75 μg m^{-3}, while the ending time t_4 is the time when $PM_{2.5}$ concentration begins to decrease below 75 μg m^{-3}. The duration is t_4-t_1. This definition was revised based on Zheng et al. (2016).



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174 **Figure S2.** Location of the measurement site in Tianjin.

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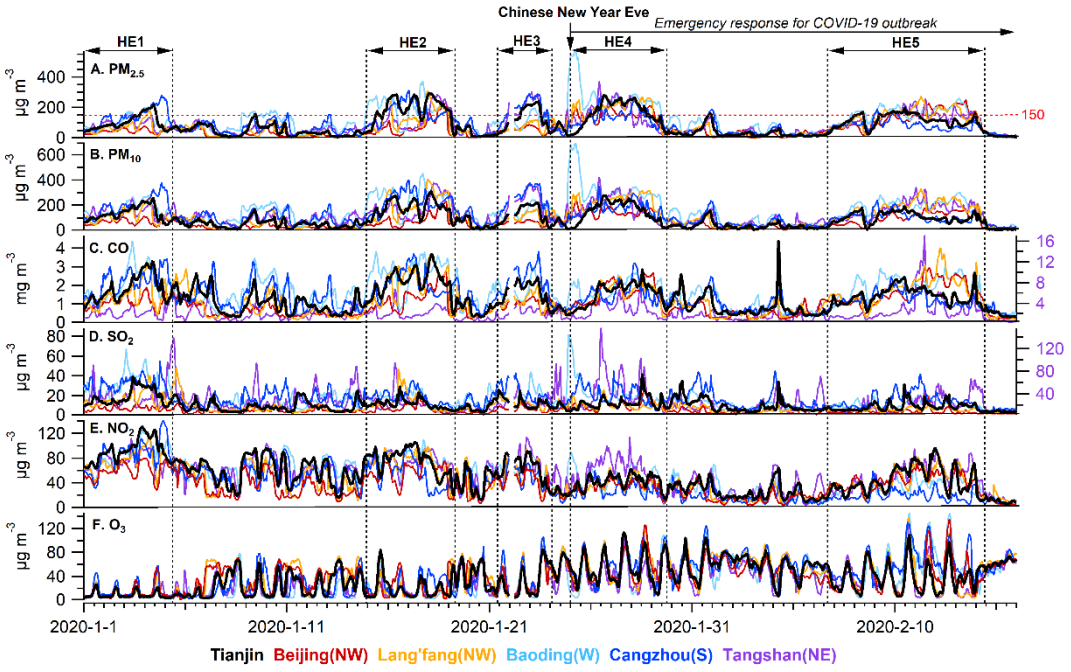

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177 **Figure S3.** Time series of hourly averaged $PM_{2.5}$, PM_{10}, CO, SO_2, NO_2 and O_3 concentrations in

178 Beijing, Tianjin, Lang'fang, Baoding, Tangshan and Cangzhou during the measurement

campaign. Beijing and Lang'fang are located at the northwest direction of Tianjin, while Baoding, Tangshan and Cangzhou are seated at west, northeast and south of Tianjin.

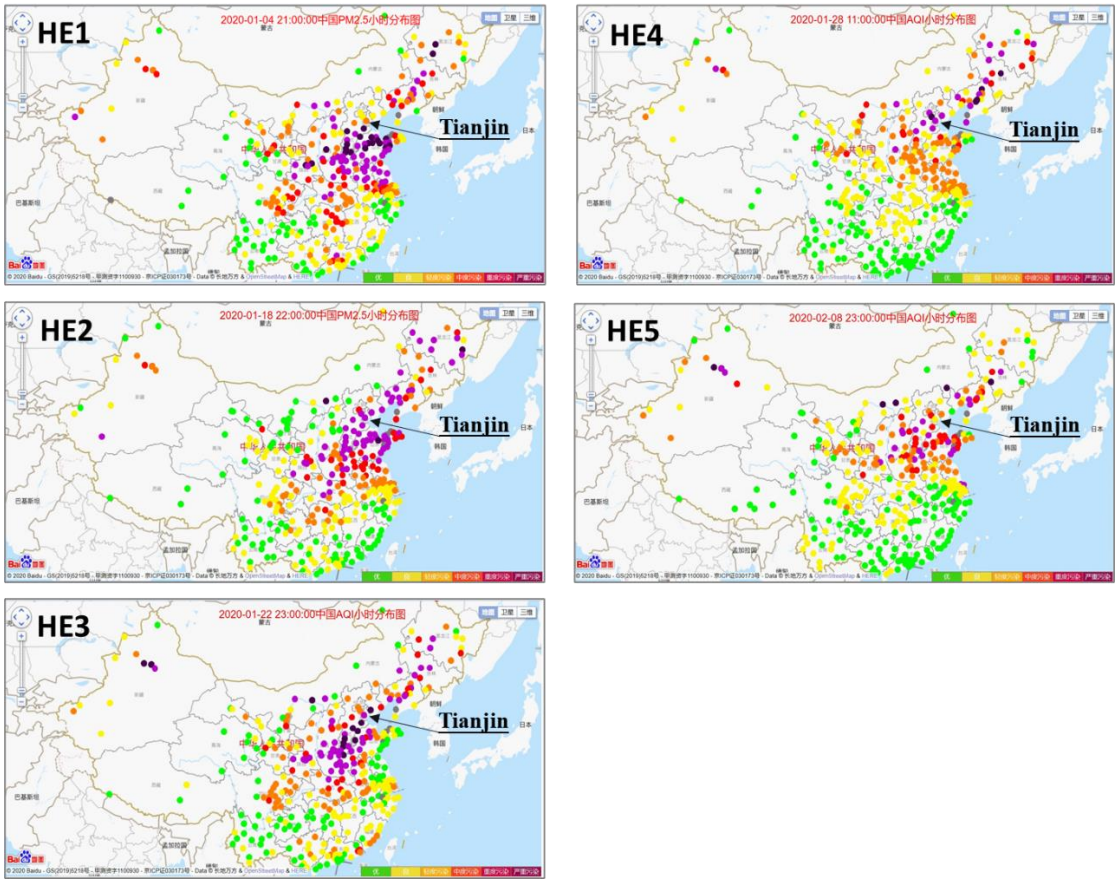

Figure S4. Screenshots of each HE reaching its maximum spatial scales (Data source: https://www.aqistudy.cn/).

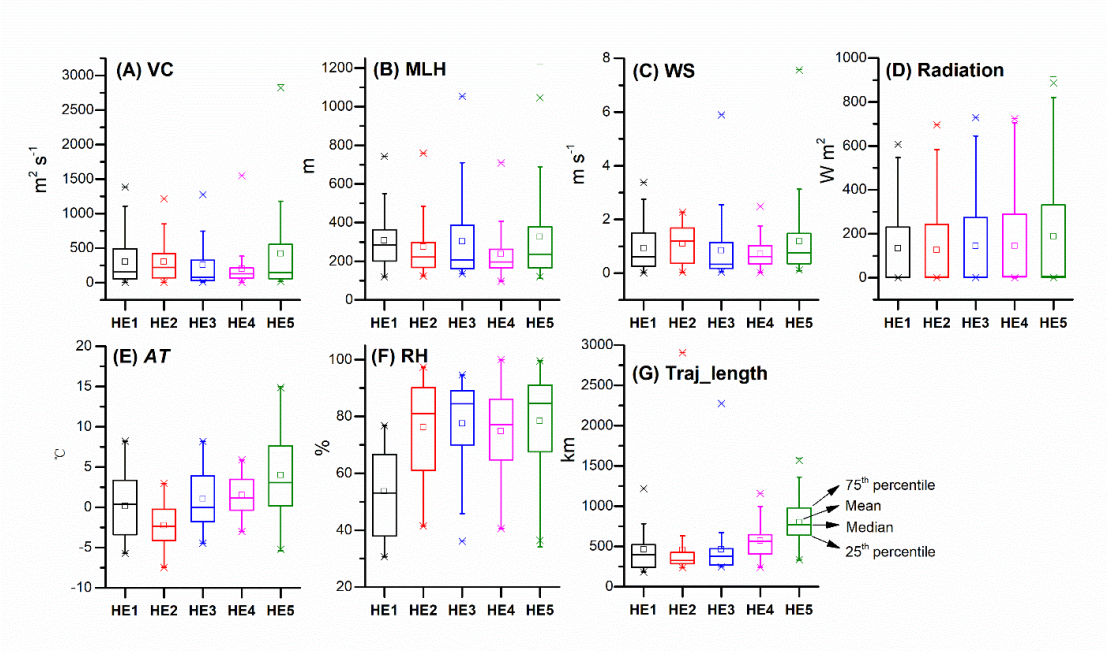

Figure S5. Boxplots for VC, MLH, WS, radiation, temperature, RH and the length of air mass backward trajectory during each HE.

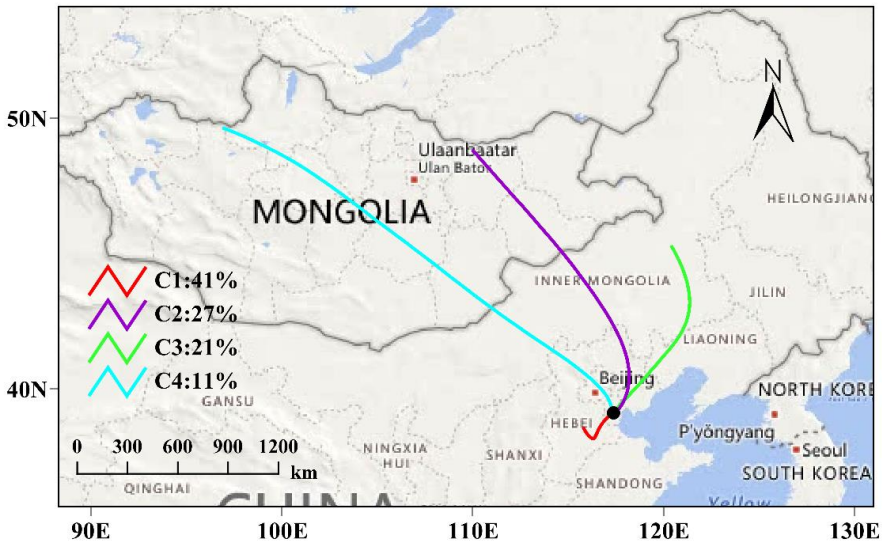

Figure S6. Clusters of the 48h air mass backward trajectories arrived in Tianjin at the height of 100 m AGL during the measurement campaign (C1: Southwesterly, C2: Northerly, C3: Northeasterly, C4: Northwesterly).

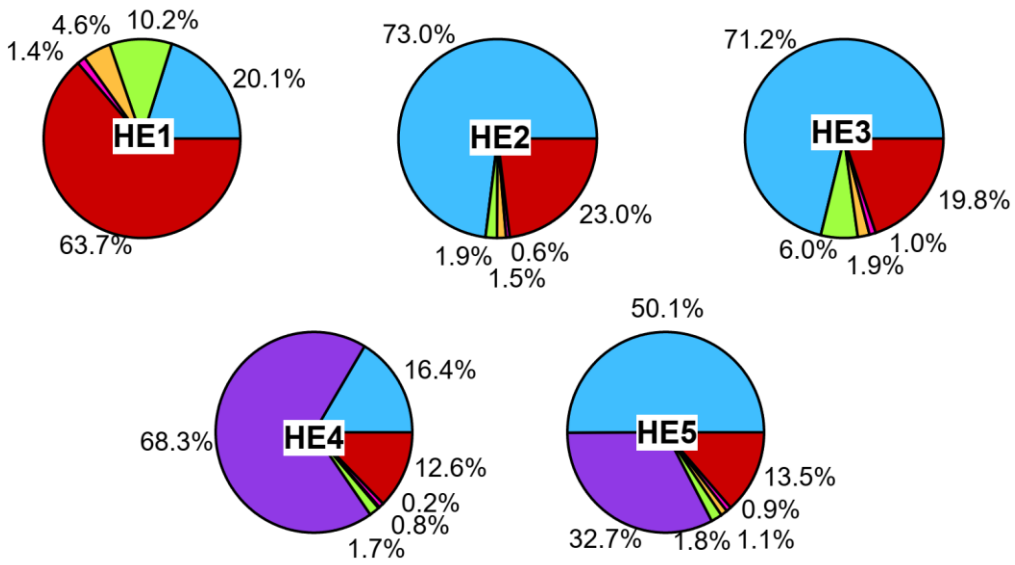

Figure S7. Average source contributions to $PM_{2.5}$ during each HE.

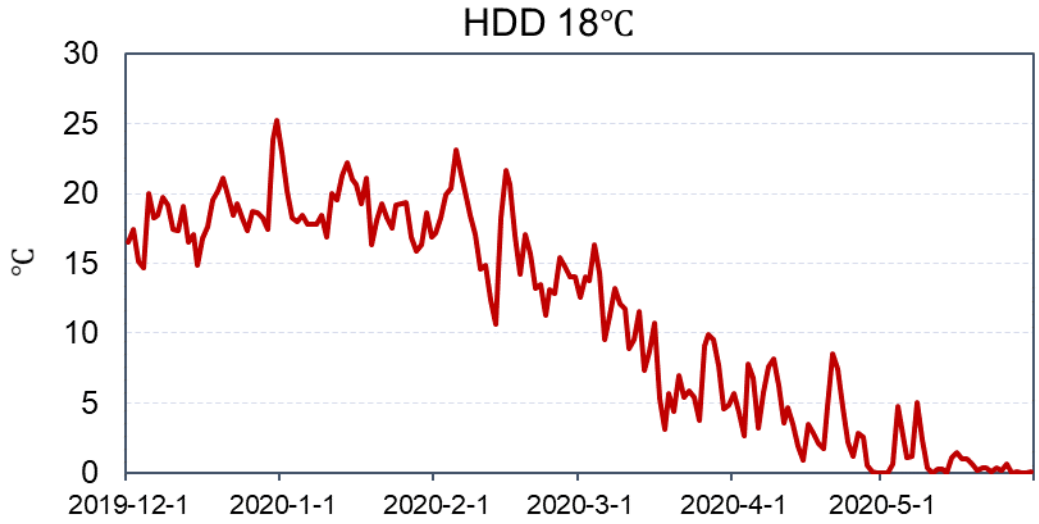

Figure S8. Celsius-based heating degree days with a base temperature of 18℃ from December 2019 to May 2020 in Tianjin. Data source: www.degreedays.net.

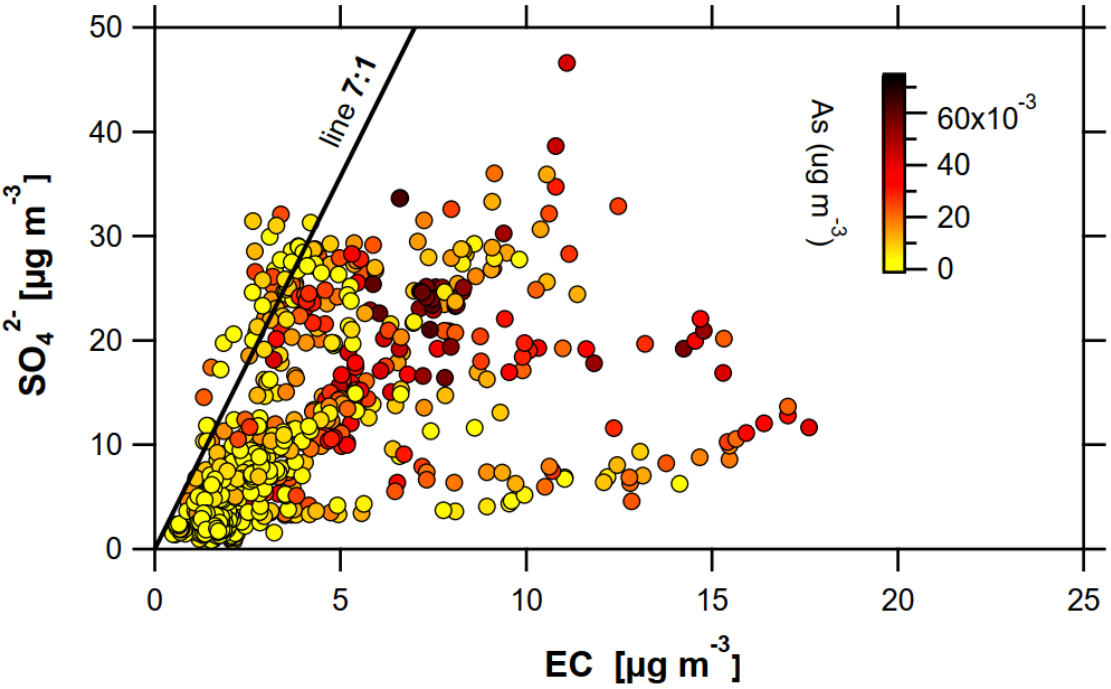

Figure S9. Scatter plot of SO_4^{2-} versus EC measured during the sampling campaign. Data points were colored by arsenic concentrations. Dash line denotes the 95th percentile line for the ratios of SO_4^{2-} to EC.

Table S1. Duration and the maximum hourly $PM_{2.5}$ concentrations of the five HEs in Tianjin and surrounding cities.

	Event	Tianjin	Beijing	Tangshan	Lang'fang	Cangzhou	Baoding
Duration (hr)	HE1	73	-	-	-	75	63
	HE2	93	27	42	81	102	88
	HE3	48	-	49	-	51	41
	HE4	88	89	89	89	85	89
	HE5	174	130	126	129	-	171
Maximum $PM_{2.5}$ ($\mu g\ m^{-3}$)	HE1	217	-	-	-	279	202
	HE2	293	234	286	310	311	373
	HE3	239	-	303	-	289	198
	HE4	280	210	370	270	263	571
	HE5	199	249	256	272	-	261

Table S2. Squared Person correlation coefficient (r^2) of $PM_{2.5}$ versus primary gaseous pollutants measured in Tianjin during the five HEs.

	SO_2	NO_2	CO	PM_{10}
E1	0.68	0.77	0.89	0.84
E2	0.00	0.18	0.87	0.89
E3	0.00	0.47	0.65	0.91
E4	0.01	0.09	0.43	0.85
E5	0.01	0.21	0.68	0.89