



Supplement of

Evolution of firework-related barium aerosols: insights from single-particle analysis and mass concentration monitoring

Xiufeng Lian et al.

Correspondence to: Mei Li (limei@jnu.edu.cn)

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1. Concentration Weighted Trajectory (CWT) model

To identify potential source regions affecting the receptor sites, the concentration-weighted trajectory (CWT) method was applied (Watson et al., 2008). In this approach, each grid cell is assigned a weighted concentration by averaging the measured concentrations of all trajectories that pass through that cell (Hsu et al., 2003) (Equation R1):

$$C_{ij} = \frac{1}{\sum_{l=1}^M \tau_{ijl}} \sum_{l=1}^M C_l \tau_{ijl} \quad (R1)$$

$$W(i,j) = \begin{cases} 1.0 & (3n_{ave} < n_{ij}) \\ 0.7 & (1.5n_{ave} < n_{ij} < 3n_{ave}) \\ 0.4 & (n_{ave} < n_{ij} < 1.5n_{ave}) \\ 0.2 & (n_{ave} < n_{ij}) \end{cases} \quad (R2)$$

where l is the trajectory index, M is the total number of trajectories, C_l is the concentration measured at the receptor upon the arrival of trajectory l , and τ_{ijl} is the residence time of trajectory l in the ij^{th} cell. A weighting function $W(i,j)$ is applied to account for the uncertainty due to low endpoint counts, where n_{ij} is the number of trajectory endpoints in ij cell. The resulting weighted concentration fields reveal spatial gradients of potential source contributions. All back-trajectory analyses were performed using the HYSPLIT4 model (Draxler and Rolph, 2003) developed by the U.S. National Oceanic and Atmospheric Administration (NOAA), with an arrival height of 100 m above ground level and a backward calculation time of 24 h.

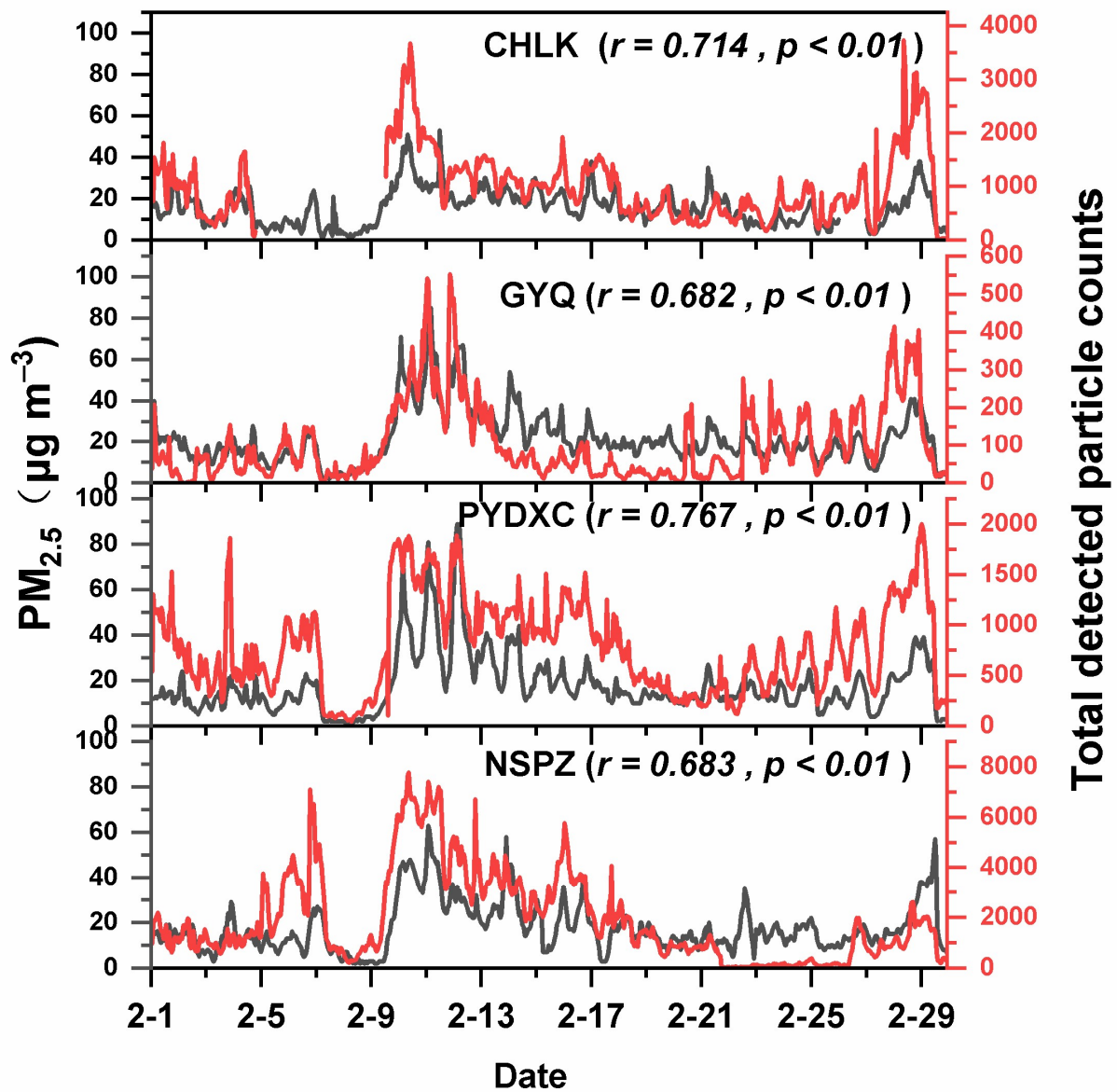


Figure S1. The time series and correlations of total detected particle counts and PM_{2.5} mass concentration at the four sites, respectively.

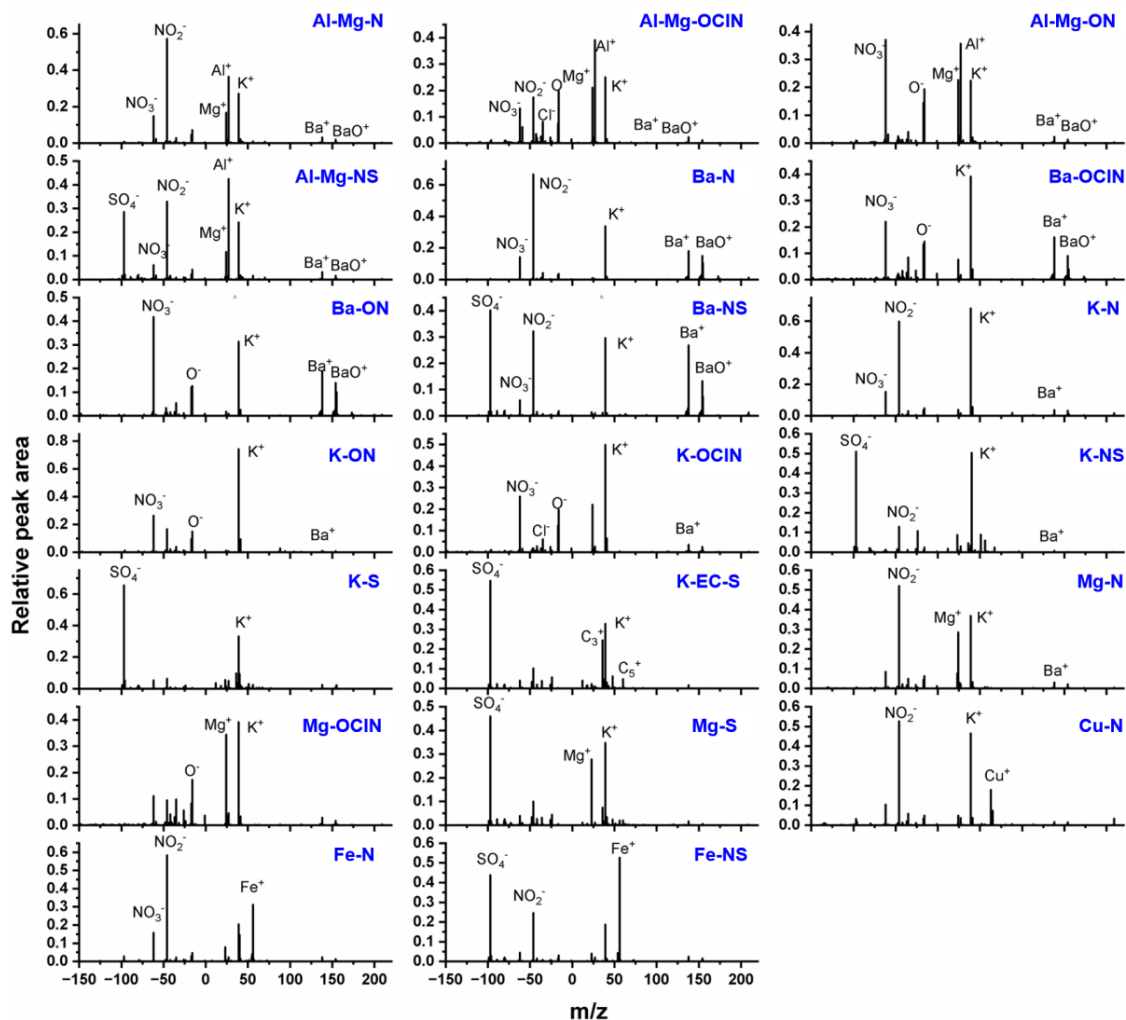


Figure S2. Average mass spectra for the main Ba-containing particle types.

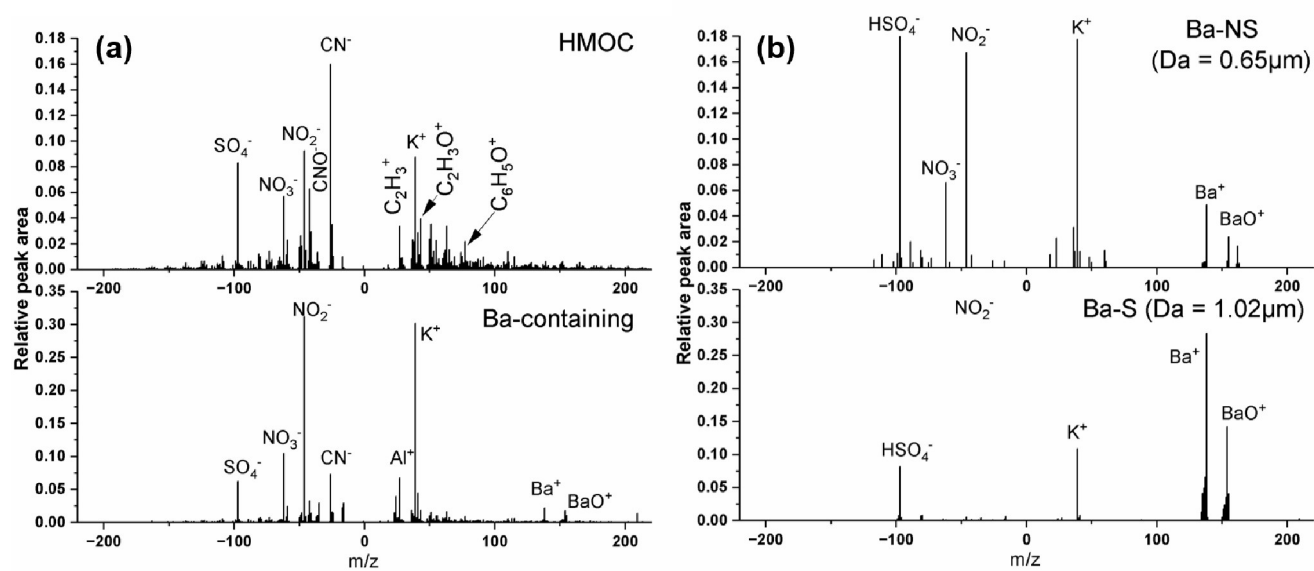
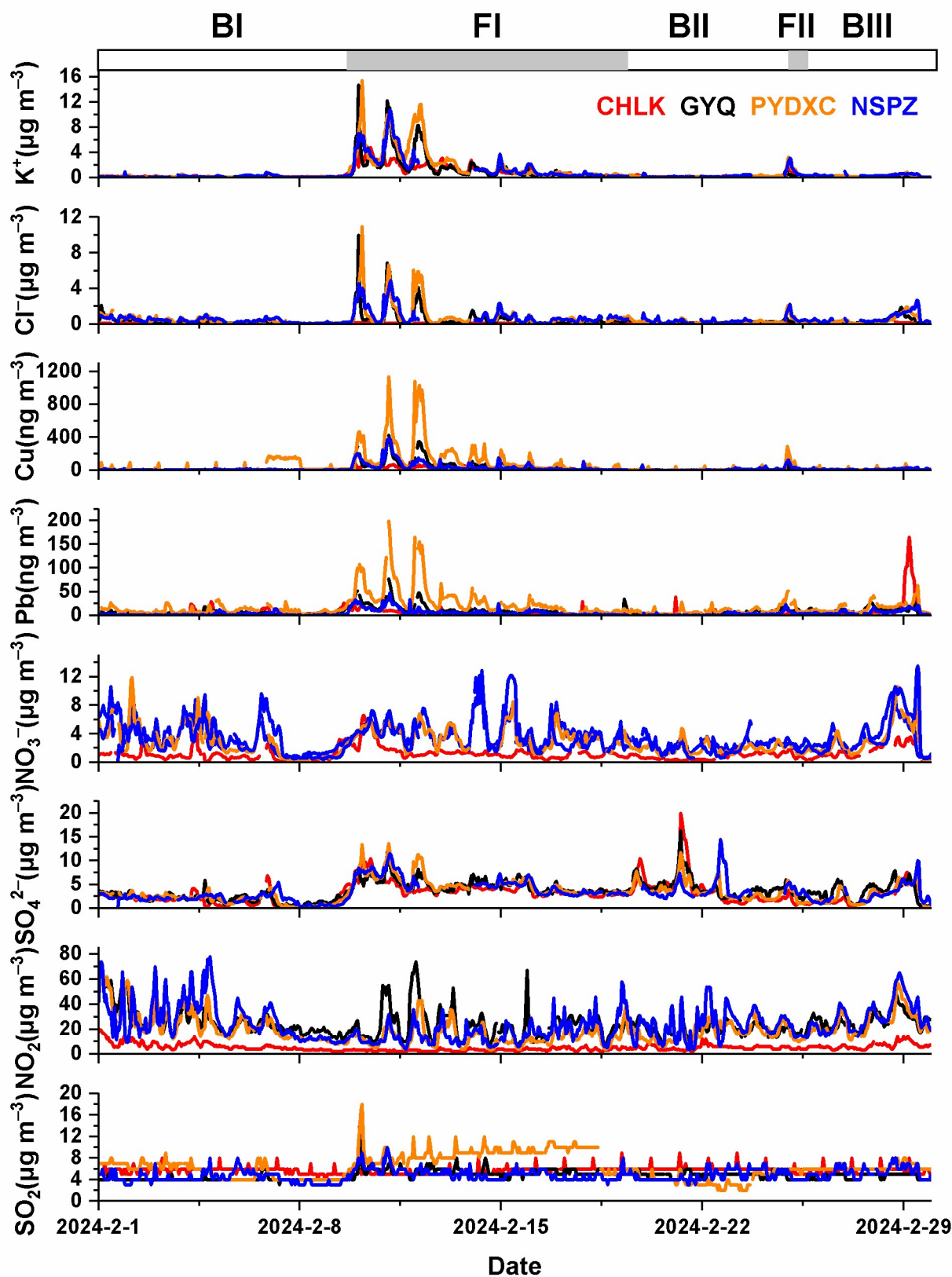


Figure S3. (a) The average mass spectrum of the high-molecular-weight organic (HMOc) type particles and Ba-containing particles at the CHLK site; (b) Representative property spectra of Ba-NS and Ba-S particle types during Firework Episode I (FI).



40 Figure S4. Synchronous time curves of potassium ion (K^+), chloride ion (Cl^-), copper (Cu), lead(Pb), nitrate (NO_3^-), sulfate (SO_4^{2-}), NO_2 , and SO_2 mass concentrations at the four sites, respectively. The time series of Ba mass concentrations is presented in Figure 1a of the main text.

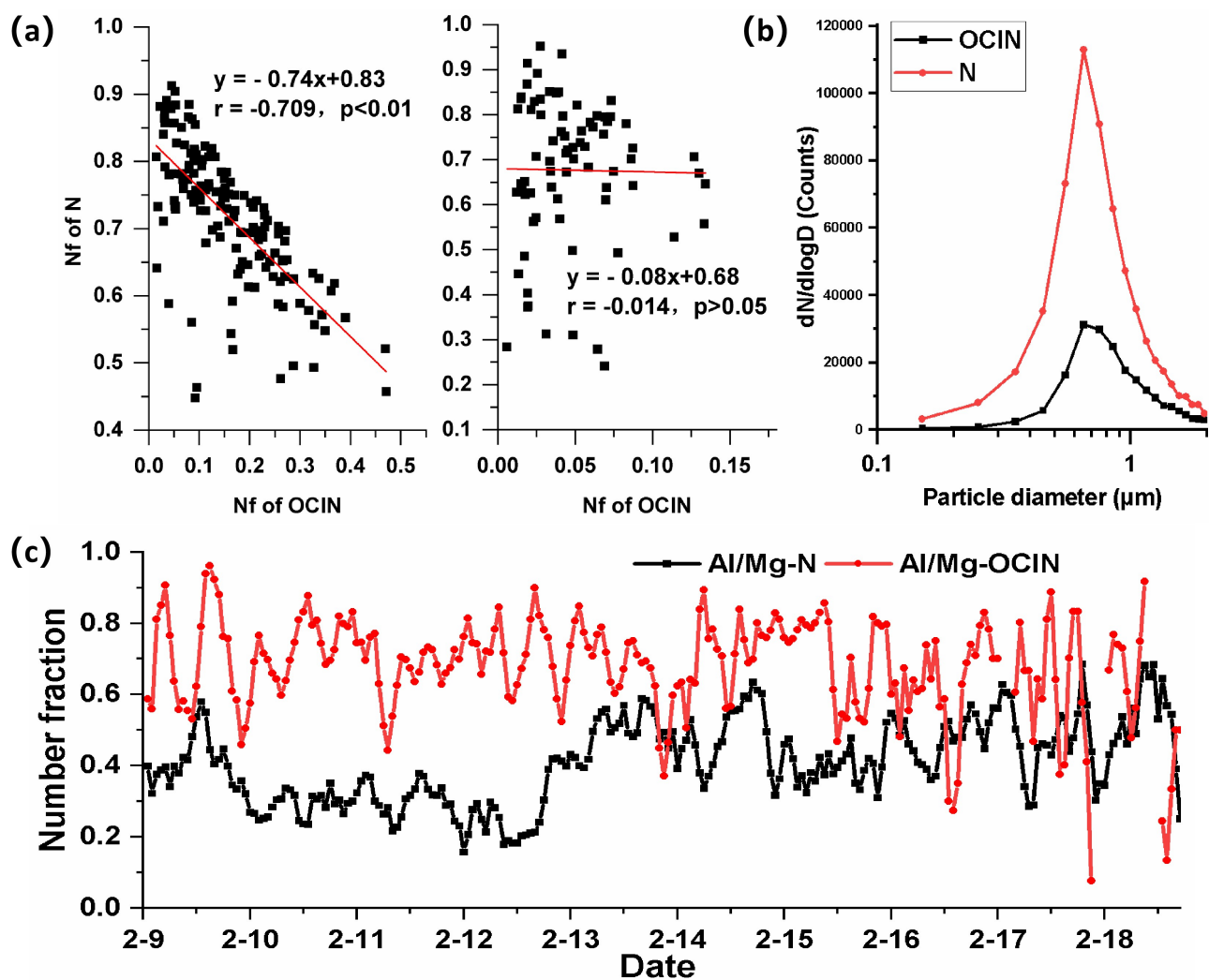


Figure S5. N and OCIN particle characteristics at the four sites during FI. (a) Correlation between the relative number fractions (N_f) during 9–15 February (left) and 16–19 February (right); (b) Particle size distributions of N and OCIN particles (plotted as $dN/d\log D$); (c) Time series of Al- or Mg-containing fractions in N and OCIN particles (3-hour moving average applied).

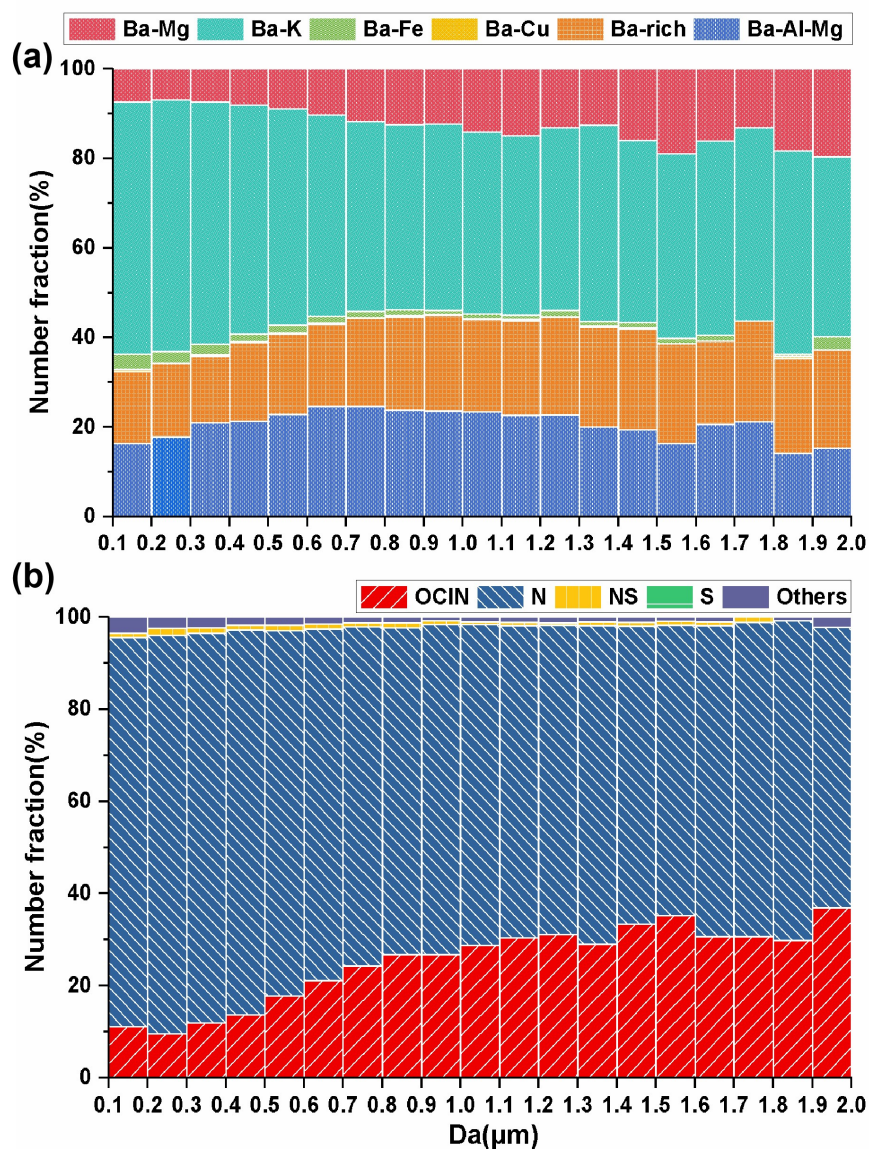


Figure S6. Relative number fractions of the main types of Ba-containing particles by particle size at the NSPZ site. (a) Classification based on positive ion mass spectra; (b) Classification based on negative ion mass spectra.

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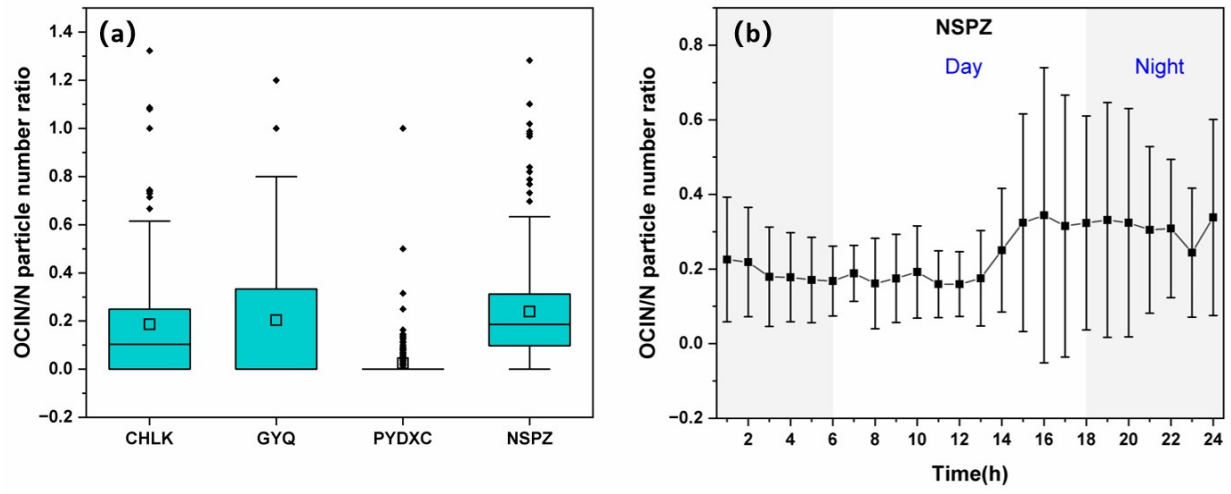
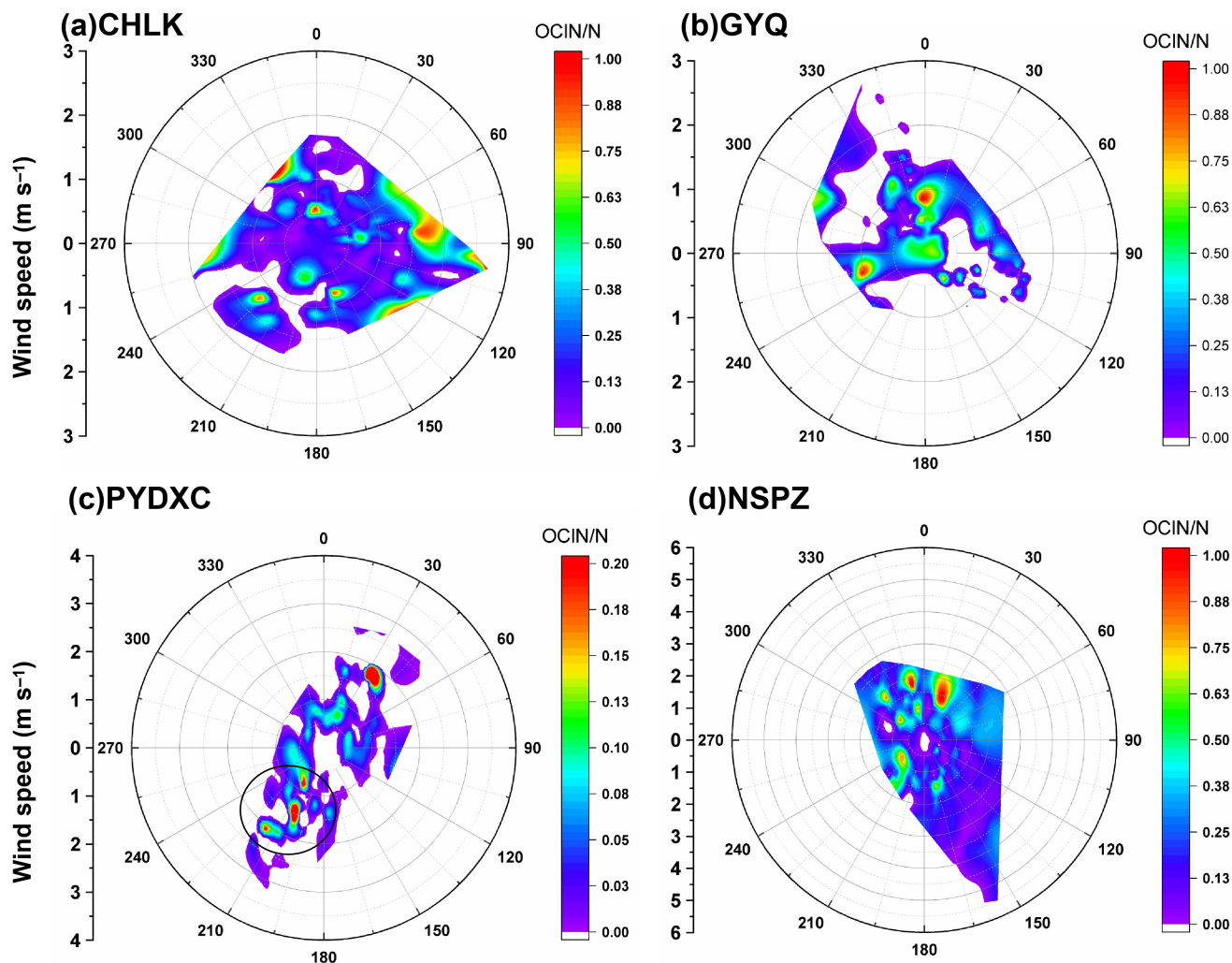
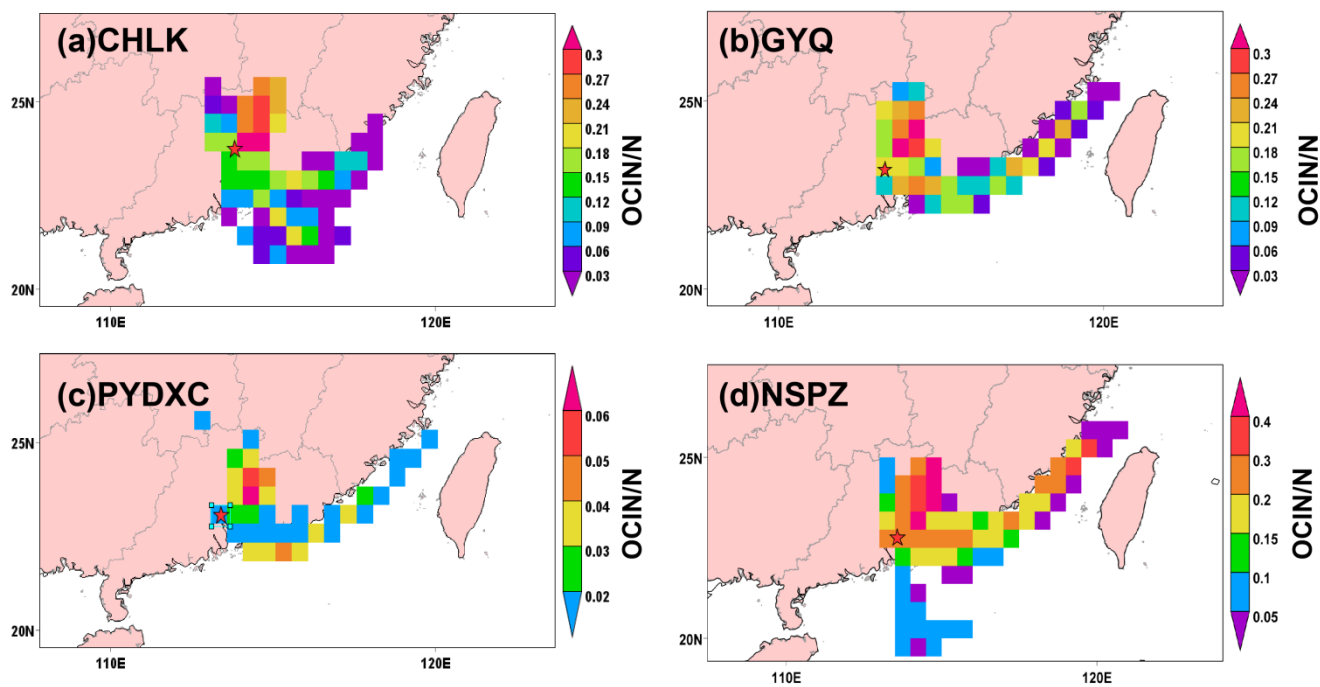


Figure S7. OCIN/N particle number ratios during FL. (a) OCIN/N ratios at the four sites; (b) Diurnal variations of OCIN/N ratios at the NSPZ site.



55 **Figure S8.** Wind-direction dependence of OCIN/N particle number ratios at the four sites during FI. Colors indicate OCIN/N ratio values (see legend). Higher OCIN/N ratios indicate a larger proportion of early-aged OCIN particles. The solid black circle in panel (c) highlights the direction corresponding to the Shiqi Town area, a designated restricted firework zone in Panyu.



60 **Figure S9. CWT plots of potential source areas of OCIN/N particle number ratios at the four sites during FI, respectively. Trajectories were calculated using HYSPLIT4 and visualized with MeteoInfo. © OpenStreetMap contributors.**

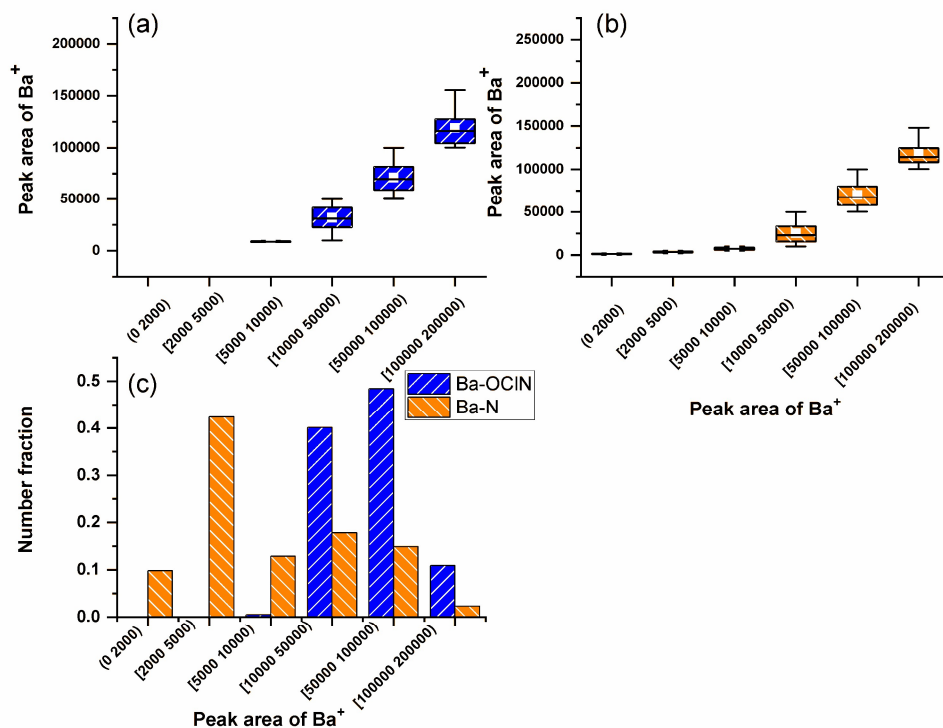


Figure S10. (a, b) Box plots of single-particle Ba peak areas by interval for Ba-OCIN and Ba-N particles at the NSPZ site; (c) Number fraction of particles in each interval at the same site.

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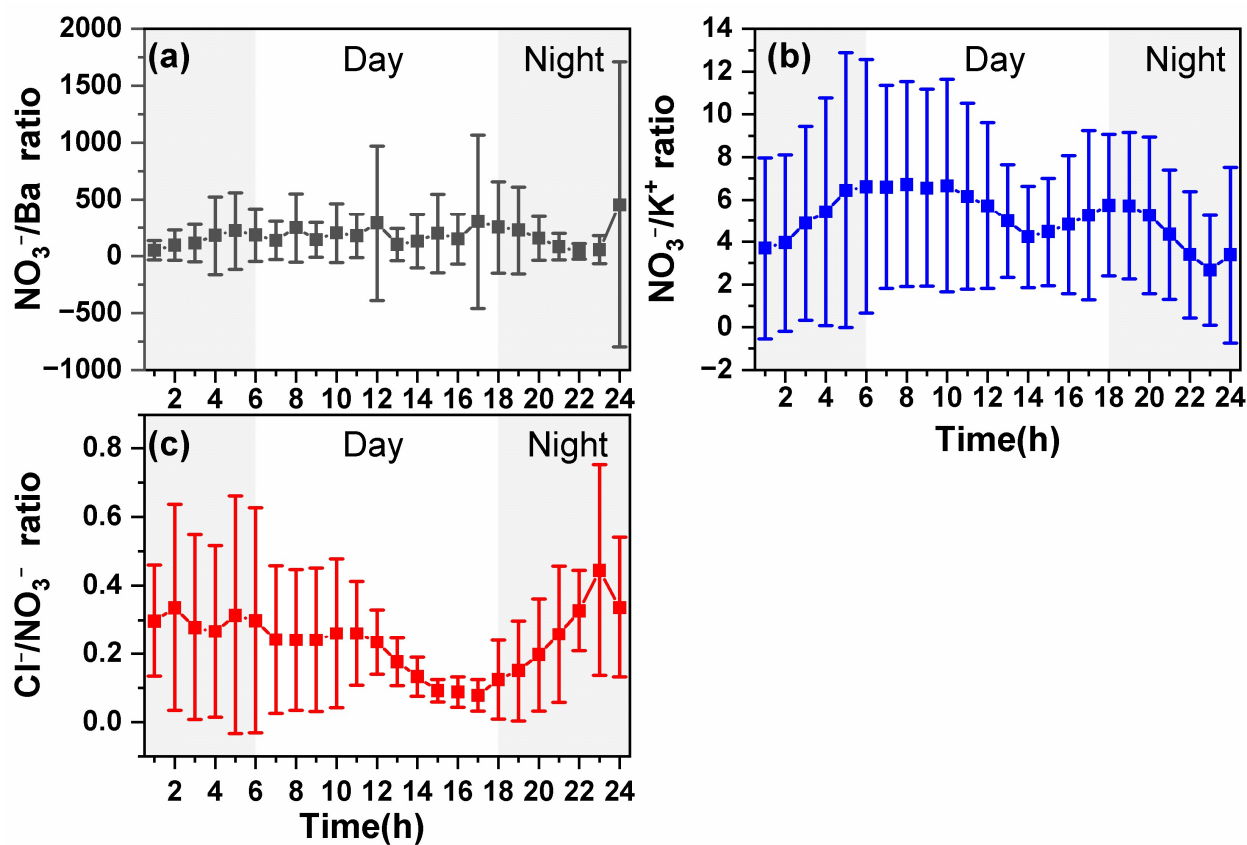
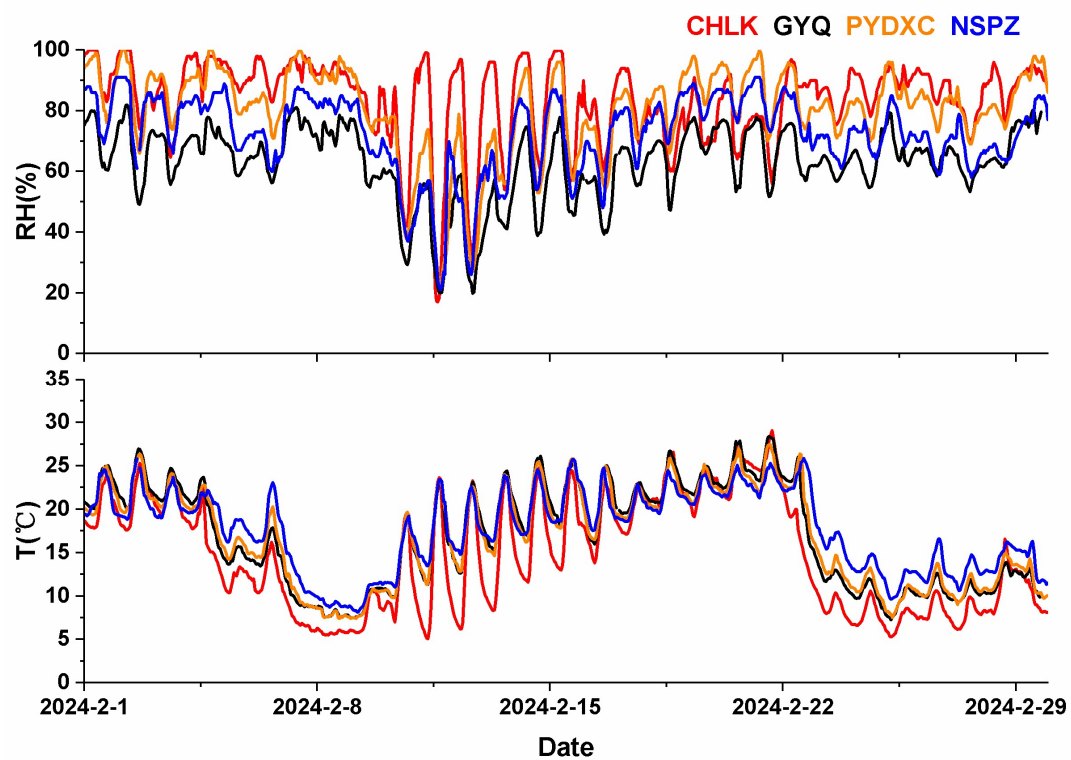


Figure S11. Diurnal variations of NO_3^-/Ba , NO_3^-/K^+ and $\text{Cl}^-/\text{NO}_3^-$ mass concentration ratios at the NSPZ site during FI.



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Figure S12. The time series of relative humidity (RH) and temperature(T) at the four sites during the sampling period, respectively.

References

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