

Supplementary Information

Title: Measurement Report: Rapid changes of chemical characteristics and health risks for high time-resolved trace elements in PM_{2.5} in a typical industrial city response to stringent clean air actions

Authors: Rui Li^{a*}, Meng Peng^{b,c*}, Weidong Zhao^{c*}, Gehui Wang^a, Jiming Hao^b

Affiliations: ^a *Key Laboratory of Geographic Information Science of the Ministry of Education, School of Geographic Sciences, East China Normal University, Shanghai, 200241, PR China*

^b *State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, 100084, P.R. China*

^c *Institute of energy conservation and environmental protection, China Electronic Information Industry Development Research Institute, Beijing, 100084, P.R. China*

*** Corresponding author**

Prof. Li (rli@geo.ecnu.edu.cn), Dr. Peng (mvponesky@163.com), and Prof. Zhao (zhaoweidong@ccidthinktank.com)

Number of pages: 16

Number of figures: 9

Number of tables: 6

Figure S1 Monthly average values of WS (m/s), T (°C), relative humidity (RH) (%) and air pressure (P) (hPa) during the sampling period. The blue, green, orange, and pink bobbles denote WS, T, RH, and P, respectively.

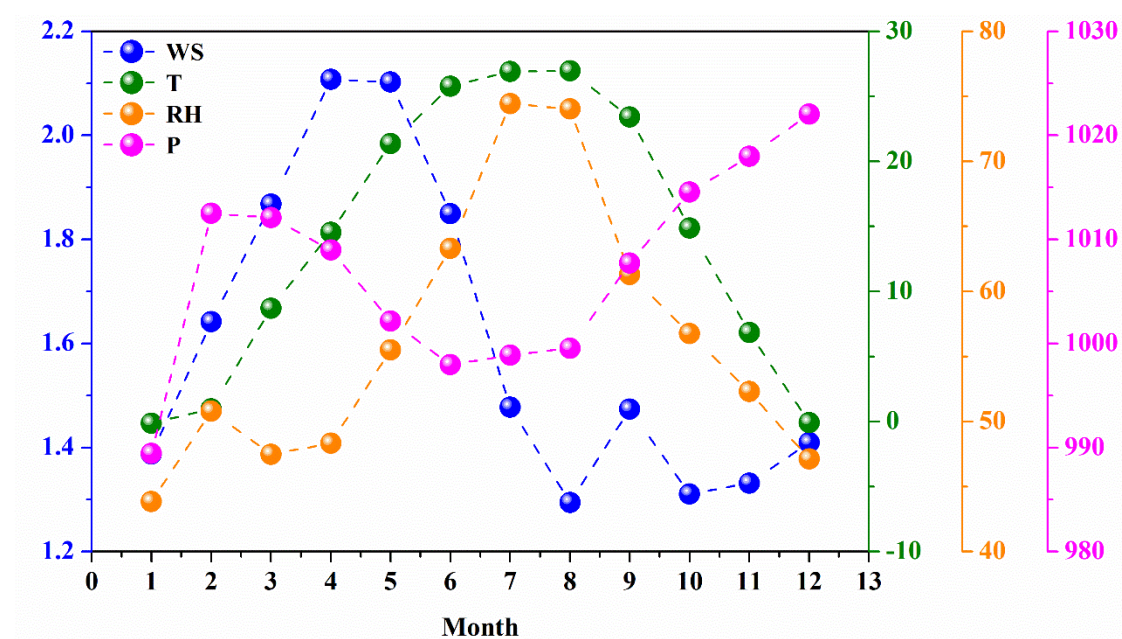


Figure S2 The performance of the random forest model in predicting the hourly concentrations of gaseous pollutants. The model was constructed with 80% original data and the remained data was applied to validate the model. The black solid line denotes the best-fitting curve for all of the points, while the black dashed line represents the diagonal, which means the same observed and simulated values.

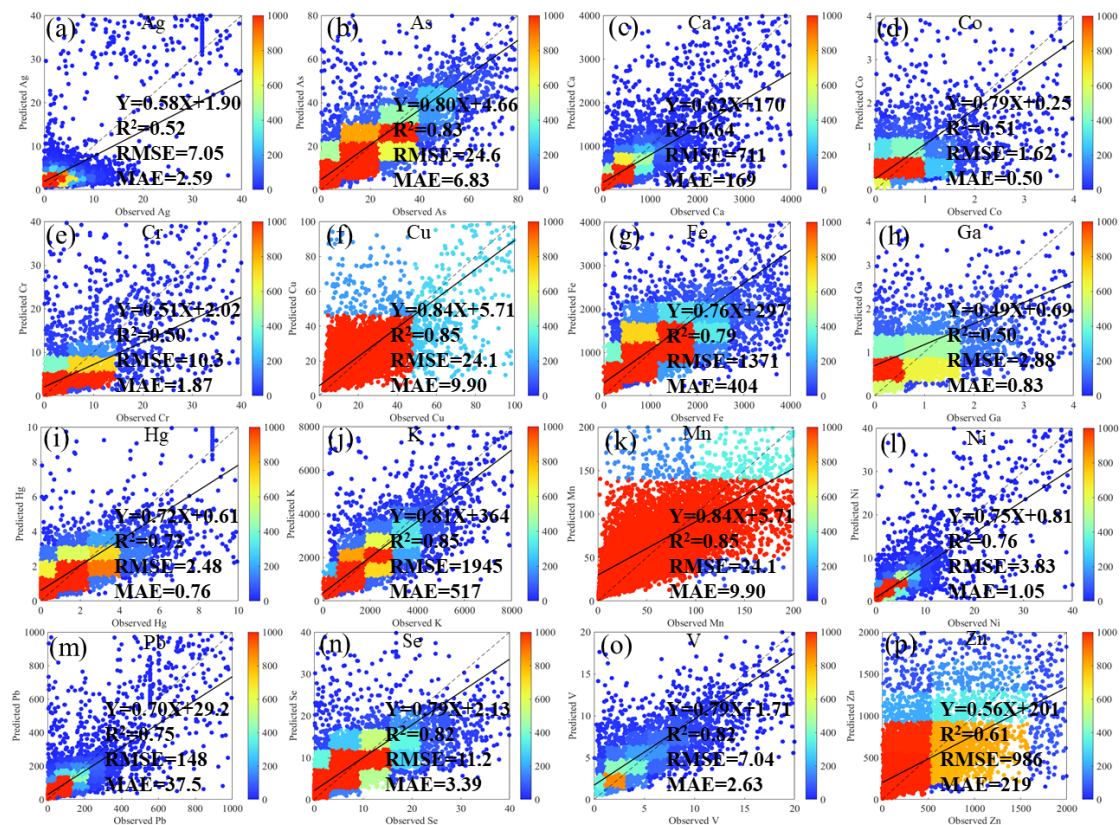


Figure S3 The changes in observed concentrations of trace elements from 2017 to 2020 against the changes derived from the emission and meteorological changes. The blue, orange, and green columns denote the original concentrations, emission-induced concentrations, and meteorology-induced ones, respectively.

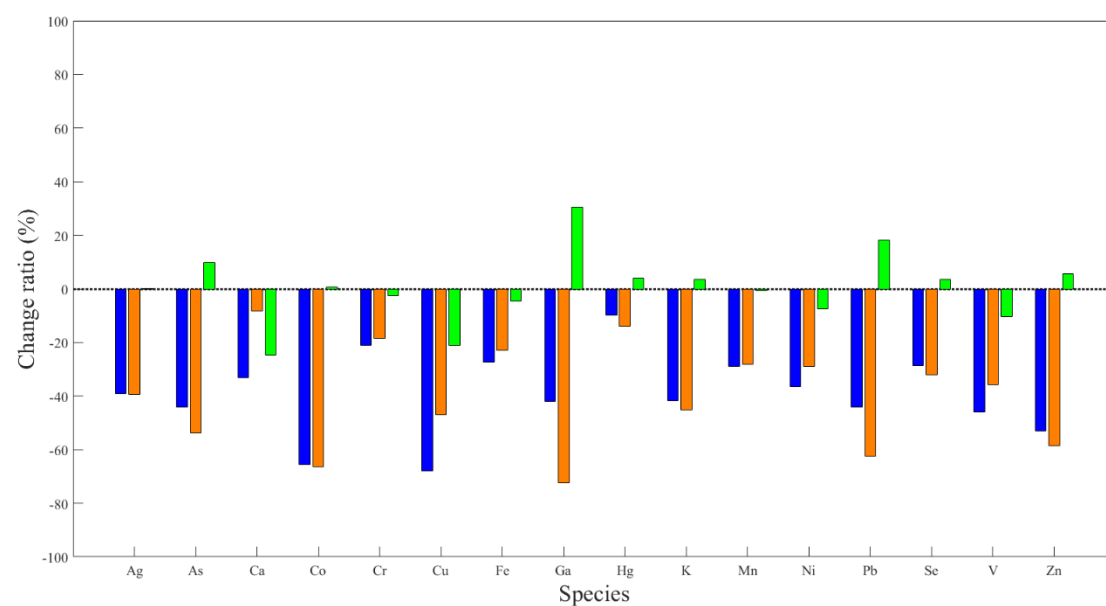


Figure S4 The source apportionment of trace elements in PM_{2.5} during 2017-2018. Red, orange, green, blue, pink, and dark yellow bars denote the sources of biomass burning, non-ferrous metal smelting, coal combustion, ferrous metal smelting, heavy oil combustion, and traffic-related dust, respectively.

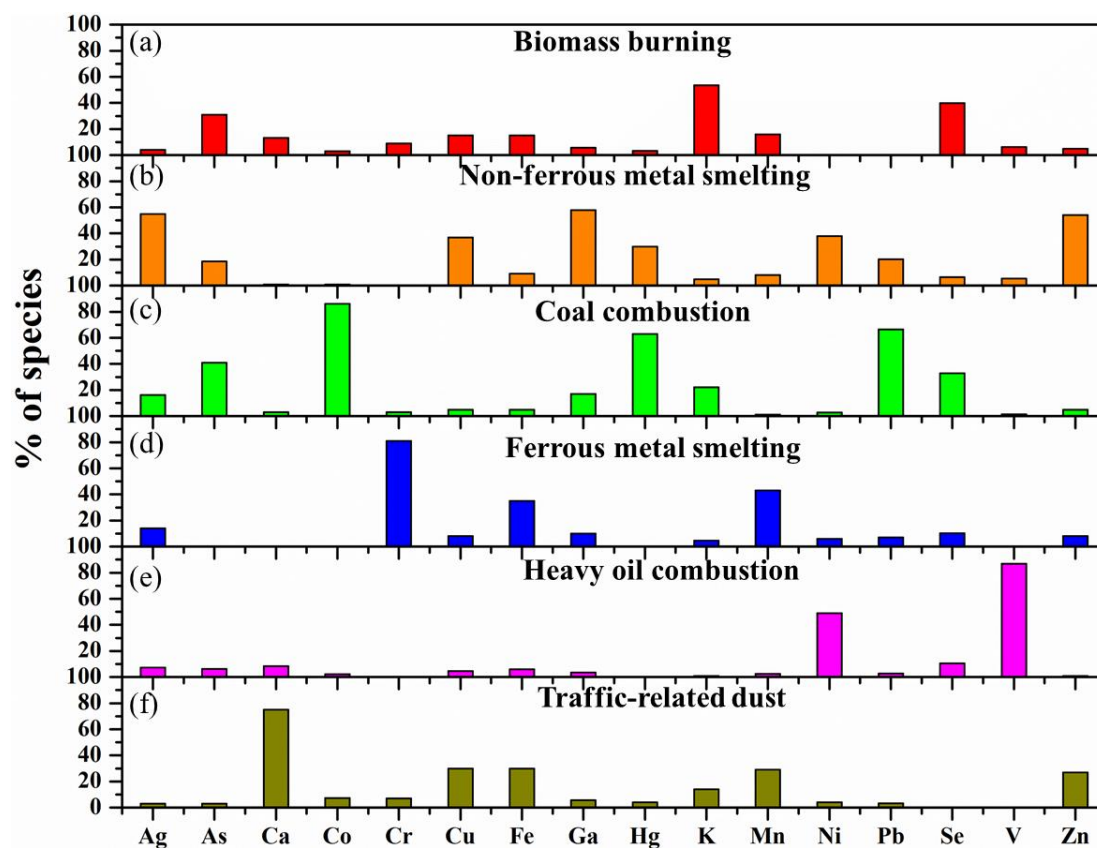


Figure S5 The source apportionment of trace elements in PM_{2.5} during 2018-2019. Red, orange, green, blue, pink, and dark yellow bars denote the sources of biomass burning, non-ferrous metal smelting, coal combustion, ferrous metal smelting, heavy oil combustion, and traffic-related dust, respectively.

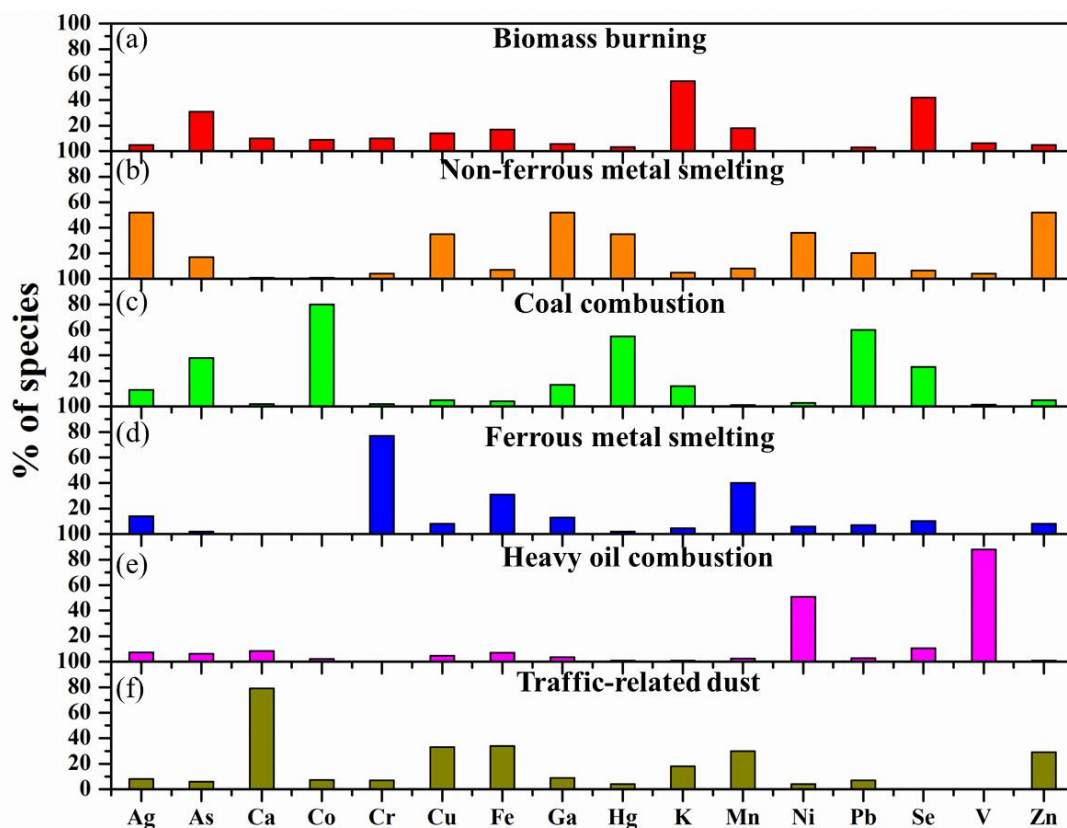


Figure S6 The source apportionment of trace elements in PM_{2.5} during 2019-2020. Red, orange, green, blue, pink, and dark yellow bars denote the sources of biomass burning, non-ferrous metal smelting, coal combustion, ferrous metal smelting, heavy oil combustion, and traffic-related dust, respectively.

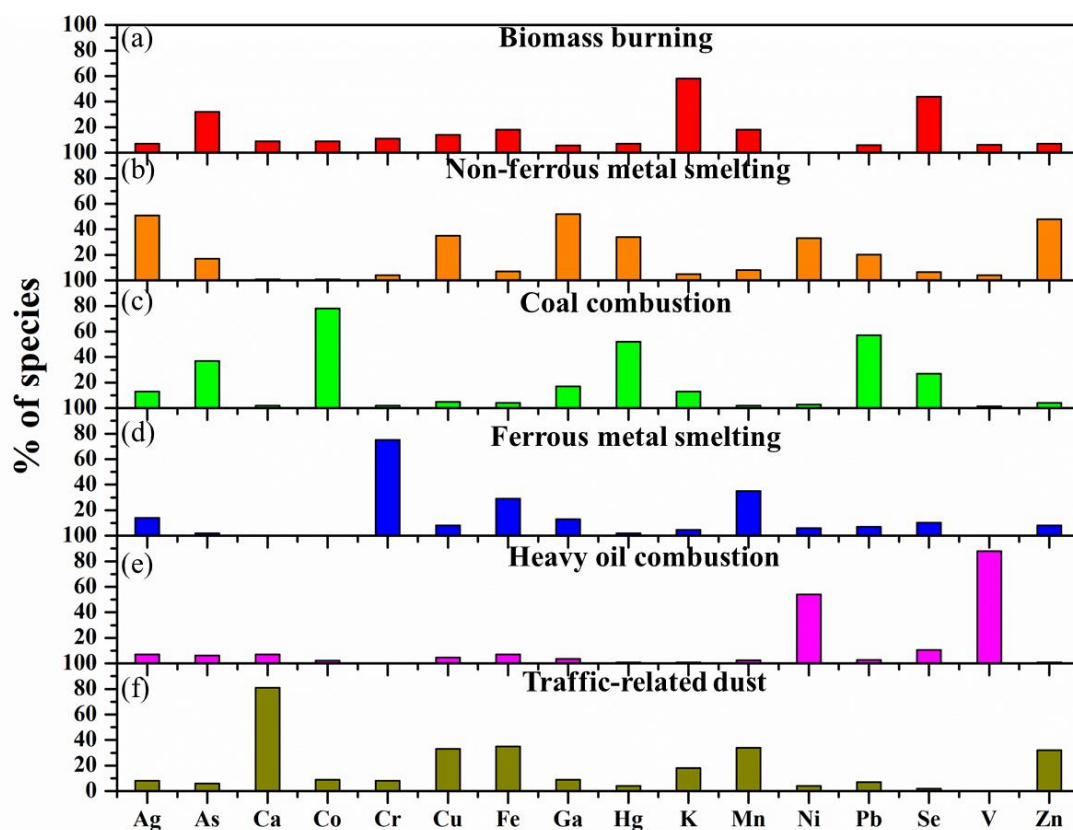


Figure S7 The 72-h backward trajectories arriving at Tangshan during biomass burning episodes in harvesting season.

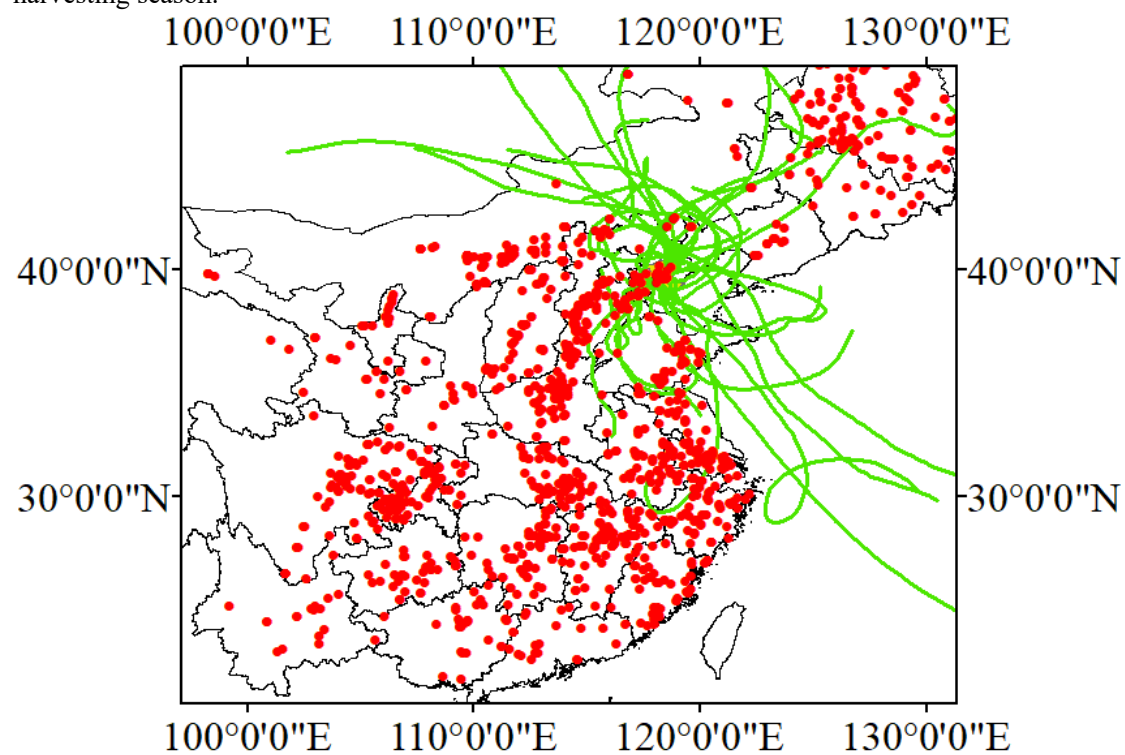


Figure S8 The 72-h air masses trajectories (a) and cluster result (b) in Tangshan during spring, 2018. The red, green, and blue solid line account for 16%, 55%, and 29% of total air masses, respectively.

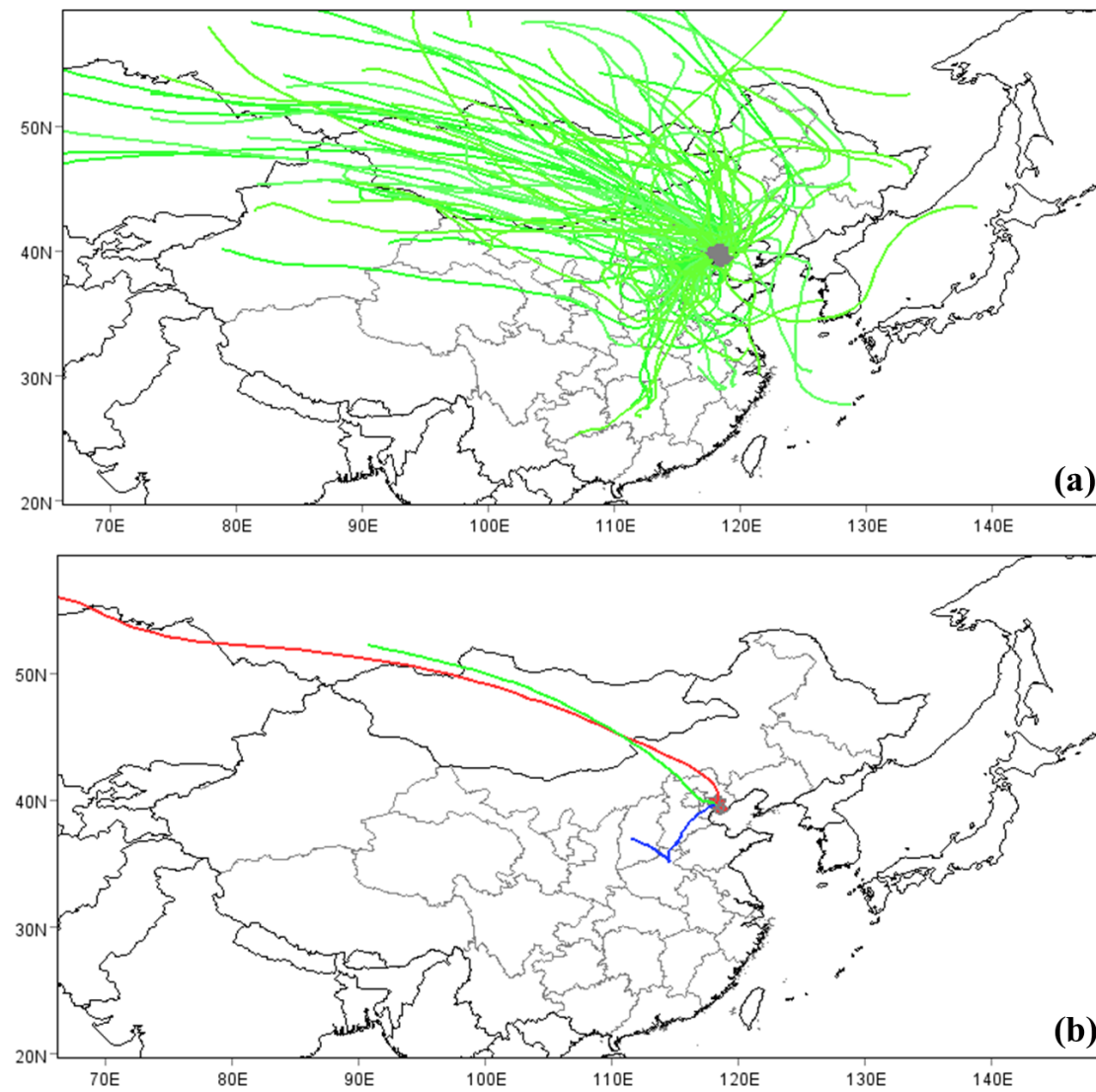


Figure S9 The dominant wind direction of the sampling site in summer and autumn.

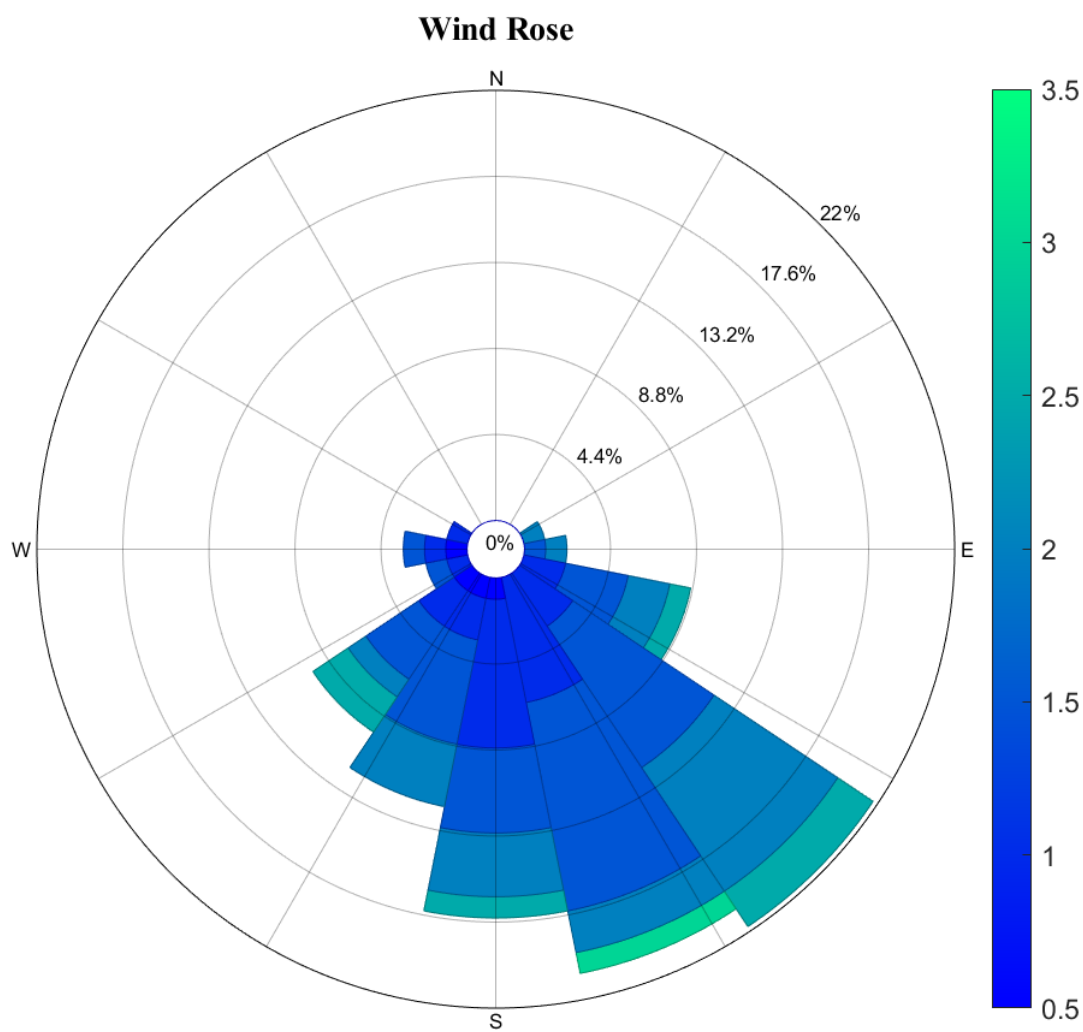


Table S1 LOD (ng/m³) of each element in PM_{2.5} in our study determined by Model Xact 625.

| Element | LOD |
|---------|------|
| K | 1.18 |
| Ca | 0.56 |
| V | 0.15 |
| Cr | 0.14 |
| Mn | 0.13 |
| Fe | 0.73 |
| Ni | 0.20 |
| Cu | 0.19 |
| Zn | 0.23 |
| As | 0.11 |
| Se | 0.14 |
| Ag | 0.20 |
| Ga | 0.05 |
| Co | 0.05 |
| Hg | 0.19 |
| Pb | 0.22 |

Table S2 Summary of PMF error diagnostics based on DISP.

| | | | | | | |
|----------------------|---------|----------------|-----------------|---------------|------------|------------------|
| Error Code: | 0 | | | | | |
| The largest decrease | 0 | | | | | |
| in Q: | | | | | | |
| %dQ | 0 | | | | | |
| Swaps by Factor: | Biomass | Non-ferrous | Coal combustion | Ferrous metal | Heavy oil | Traffic- related |
| | burning | metal smelting | | smelting | combustion | dust |
| dQmax=4 | 0 | 0 | 0 | 0 | 0 | 0 |
| dQmax=8 | 0 | 0 | 0 | 0 | 0 | 0 |
| dQmax=15 | 0 | 0 | 0 | 0 | 0 | 0 |
| dQmax=25 | 0 | 0 | 0 | 0 | 0 | 0 |

Table S3 Summary of PMF error diagnostics based on BS.

| BS Mapping ($r \geq 0.6$) | Biomass burning | Non-ferrous metal smelting | Coal combustion | Ferrous metal smelting | Heavy oil combustion | Traffic- related dust | Unmapped |
|---|--------------------|----------------------------------|--------------------|------------------------------|-------------------------|-----------------------------|----------|
| Biomass burning and waste incineration | 100 | 0 | 0 | 0 | 0 | 0 | 0 |
| Non-ferrous metal smelting | 0 | 100 | 0 | 0 | 0 | 0 | 0 |
| Coal combustion | 0 | 0 | 100 | 0 | 0 | 0 | 0 |
| Ferrous metal smelting | 0 | 0 | 1 | 98 | 1 | 0 | 0 |
| Heavy oil combustion | 0 | 0 | 0 | 0 | 100 | 0 | 0 |
| Traffic- related dust | 0 | 0 | 0 | 0 | 0 | 100 | 0 |

Table S4 Summary of PMF error estimation diagnostics from BS-DISP.

| | | | | | |
|----------------------|--------------------|--------------------------------------|-----------------|---------------------------|----------------------|
| % of Cases Accepted: | 98% | | | | |
| Largest Decrease in | -22.5 | | | | |
| Q: | | | | | |
| %dQ | -0.0094 | | | | |
| # of Decreases in Q: | 1 | | | | |
| # of Swaps in Best | 0 | | | | |
| Fit: | | | | | |
| # of Swaps in DISP: | 1 | | | | |
| Swaps by Factor: | Biomass burning | Non- ferrous metal smelting | Coal combustion | Ferrous metal smelting | Heavy oil combustion |
| $dQ^{\max}=0.5$ | 0 | 0 | 1 | 0 | 0 |
| $dQ^{\max}=1$ | 0 | 0 | 1 | 0 | 0 |
| $dQ^{\max}=2$ | 0 | 0 | 1 | 0 | 0 |
| $dQ^{\max}=4$ | 0 | 0 | 1 | 0 | 0 |

Table S5 Recommended values of the parameters for health risk assessment of trace metals.

| Parameter | Adult | Child | Unit |
|---------------|--------|--------|-------------------|
| InhR | 16.5 | 8.6 | m ³ /d |
| EF | 365 | 365 | d/a |
| ED | 24 | 6 | a |
| BW | 70 | 15 | kg |
| Cancer AT | 70*365 | 70*365 | d |
| Non-cancer AT | ED*365 | ED*365 | d |

Table S6 Reference dose (RfD) and cancer slope factor (CSF) of selected trace elements in references.

| Element | RfD | CSF |
|---------|----------------------|--------------------|
| Cr | 3×10^{-3} | 5×10^{-1} |
| Mn | 1.4×10^{-1} | |
| Fe | 7×10^{-1} | |
| Co | 3×10^{-4} | |
| Ni | 2×10^{-2} | |
| Cu | 4×10^{-2} | |
| Zn | 3×10^{-1} | |
| As | 3×10^{-4} | 1.5 |
| Pb | 2×10^{-2} | 5×10^{-1} |