

12 **Data and Methods**

13 **Data**

14 Designed as a mix of nine urban sites and one background sites for the point layout of ambient air
15 quality monitoring, presently there are ten national automated monitoring sites in Hefei. Monitoring
16 systems were installed and used to measure the pollutant concentrations according to China
17 Environmental Protection Standards (HJ655-2013). The mass concentrations of PM_{2.5} and PM₁₀ were
18 measured using the Tapered Element Oscillating Microbalance analyzers, and levels of SO₂, NO₂, O₃
19 and CO were measured using the using of the ultraviolet fluorescence method, the chemiluminescence
20 method, the UV-spectrophotometry method, and the non-dispersive infrared absorption method (or the
21 gas filter correlation infrared absorption method), respectively. The diurnal concentrations of each
22 pollutant were calculated only when there were more than 16 h valid data. To better study the temporal
23 changes of pollutants, the corresponding data are divided into four seasons: spring (March–May),
24 summer (June–August), autumn (September–November) and winter (December–February).

25 **Methodology**

26 *Hybrid Single Particle Lagrange Integrated Trajectory (HYSPLIT) model*

27 HYSPLIT-4 model was developed in the Air Resources Laboratory (ARL) of the National Oceanic
28 and Atmospheric Administration (NOAA). Corresponding meteorological data with a 1° × 1° grid used
29 in the HYSPLIT-4 model was acquired from the Global Data Assimilation System (GDAS)
30 (<ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/>). The backward trajectories were run four times per day
31 at a starting times of 0:00, 06:00, 12:00, and 18:00 UTC (8:00, 14:00, 20:00, and 02:00 LT, respectively)
32 with starting height of 500 m above ground level (agl). In order to divide the seasonal trajectories into
33 distinct transport groups, cluster analysis was carried out on the basis of the hierarchical clustering
34 method [1].

35 *Potential source contribution function (PSCF)*

36 The zone of concern is divided into small equal grid cells ($i \times j$). The PSCF value in the ij^{th} cell
37 represents the probability of air parcel residence time and is expressed as:

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$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \quad (1)$$

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where i and j denote the latitude and longitude, n_{ij} is on behalf of the total number of endpoints that fall in the ij^{th} cell, and m_{ij} represents the number of endpoints in the same cell associated with samples for which the monitored mass concentration of pollutant exceeds the criterion value in the ij^{th} cell. The GB3095-2012 PM_{2.5} guideline value for daily average of 75 $\mu\text{g}/\text{m}^3$ is used as the criterion-value. To better reflect the uncertainty in cells, the PSCF values should be multiplied by an arbitrary weighting function $W(n_{ij})$ when n_{ij} is lower than three times of average number of trajectory endpoints (n_{ave}) in each cell [2,3], and the weight PSCF (WPSCF) is described as follows:

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$$WPSCF_{ij} = \frac{m_{ij}}{n_{ij}} \times W(n_{ij}) \quad (2)$$

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$$W(n_{ij}) = \begin{cases} 1.00, & 3n_{ave} < n_{ij} \\ 0.70, & 1.5n_{ave} < n_{ij} \leq 3n_{ave} \\ 0.40, & n_{ave} < n_{ij} \leq 1.5n_{ave} \\ 0.20, & n_{ij} \leq n_{ave} \end{cases} \quad (3)$$

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Concentration-weighted trajectory (CWT) model

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Since PSCF just reflects the proportion of polluted trajectories in a cell grid, which is impossible to distinguish the contribution to the pollution levels of the target object for those who have the same PSCF value, CWT model is applied to weight trajectories with related PM_{2.5} concentrations for better unraveling above limitation and calculating the relative contribution of different sources-areas. The geographical field is divided into grid cells representing an area of $0.5^\circ \times 0.5^\circ$. The CWT is defined as follows:

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$$C_{ij} = \frac{\sum_{h=1}^M C_h \times \tau_{ijh}}{\sum_{h=1}^M \tau_{ijh}} \times W(n_{ij}) \quad (4)$$

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Where C_{ij} is designed as the average weight concentration of the trajectory h in the ij^{th} cell, h represents the index of the trajectory, M represents the total number of trajectories, C_h denotes PM_{2.5} concentrations in the trajectory h through ij^{th} cell, τ_{ijh} is the time that trajectory h resides in the ij^{th} cell (i ,

60 *j*). The $W(n_{ij})$ used in PSCF also is utilized to CWT method for reducing the uncertainty in cells.

61 The study domain is in the range of 20–60 °N and 90–130 °E, which includes more than 95% of
62 areas covered by all the paths. The total number of endpoints in each season is about 51840, and the
63 area covered by the trajectories is divided into 6400 grid cells (0.5° × 0.5°). Hence, each cell owns an
64 average of 9 trajectory endpoints, that is, the n_{ave} is equal to 9. Both PSCF and CWT analysis are run by
65 the MeteInfo software-TrajStat Plugin.

66 ***Integrated exposure–response (IER) model***

67 This model is based on the assumption that the cause-specific mortality rates are independent of
68 PM_{2.5} concentrations. The relative risk (RR) for a specific disease due to PM_{2.5} exposure is the ratio of
69 the probability of an event occurring in an exposed group to the probability of the event occurring in a
70 comparison, nonexposed group [4,5]. Adult STR (stroke, including ischemic and hemorrhagic stroke),
71 Ischemic heart disease (IHD), lung cancer (LC) and chronic obstructive pulmonary disease (COPD),
72 ranking top ten in the international classification, are selected as the causes of mortality. The mortality
73 burdens attributable to ambient PM_{2.5} exposure in 2014 and 2015 are calculated using the
74 disease-specific mortality and Hefei population data. The RR of several causes of premature mortality
75 (Δ Mort) among adults including STR, IHD, LC, and COPD, are estimated according to the IER
76 functions.

77 The RR for each disease is calculated based on equation (5):

$$78 \quad RR(C) = \begin{cases} 1 + \alpha (1 - e^{-\gamma(C-C_0)^\delta}), & C \geq C_0 \\ 1, & C < C_0 \end{cases} \quad (5)$$

79 where C is the annual PM_{2.5} exposure concentration ($\mu\text{g}/\text{m}^3$); C_0 is the endpoint-specific theoretical
80 minimum-risk concentration of PM_{2.5}; α , δ and γ are parameters used to describe the shape of C–RR
81 curves. The population attributable fraction (AF), representing the fraction of mortality for a specific
82 disease due to PM_{2.5} exposure, is calculated by RR factors through the formula (6):

$$83 \quad AF = (RR - 1)/RR \quad (6)$$

84 The AFs are estimated with the annual average PM_{2.5} concentrations in 2014 and 2015 for four
85 diseases. Formula (7) is used to calculate the Δ Mort for a specific disease attributable to PM_{2.5} exposure

86 [6,7]. y_0 is the baseline mortality rate caused by a specific disease, and Pop is the total population
87 exposed to PM_{2.5} in Hefei. The population data of Hefei in 2014 and 2015 are obtained from Hefei
88 Statistics Yearbook.

$$89 \quad \Delta \text{Mort} = y_0 \times \text{AF} \times \text{Pop} \quad (7)$$

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91 **References**

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93 deposition. *Aust. Meteorol. Mag.* 47, 295–308.
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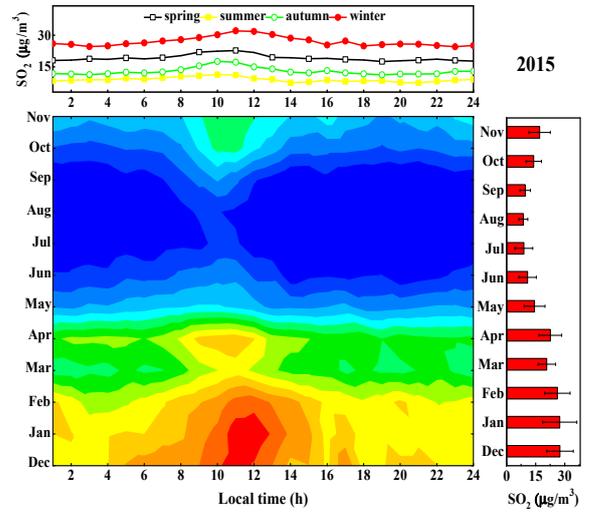
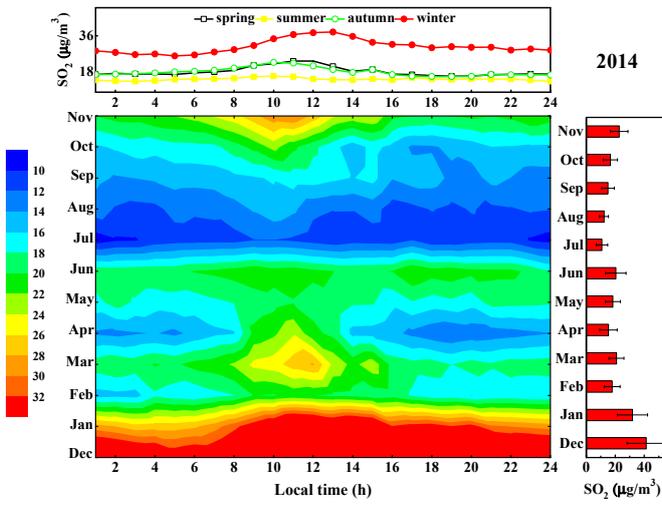
110 **Table S1** The attributable fractions due to PM_{2.5} for STR, IHD, LC and COPD.

	PM _{2.5} (µg/m ³)	Attributable Fractions (AFs) (%)			
		STR	IHD	LC	COPD
2014	89	48	32	32	25
2015	61	44	29	26	21
CAAQS grade II	35	34	23	18	15
WHO IT-2	25	25	20	14	11
WHO IT-3	15	11	15	8	7
WHO AQG	10	4	10	4	3

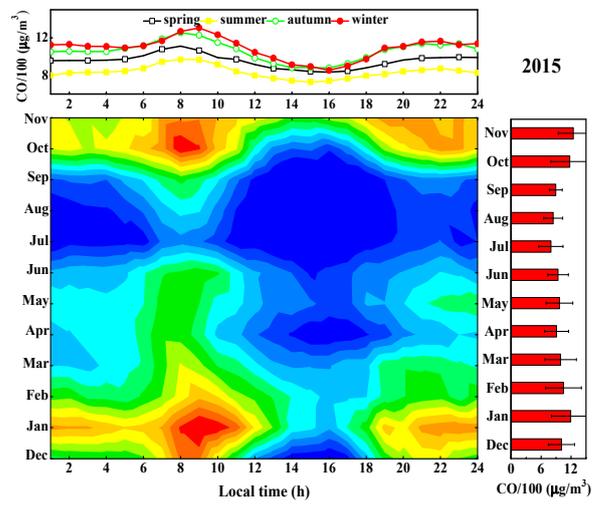
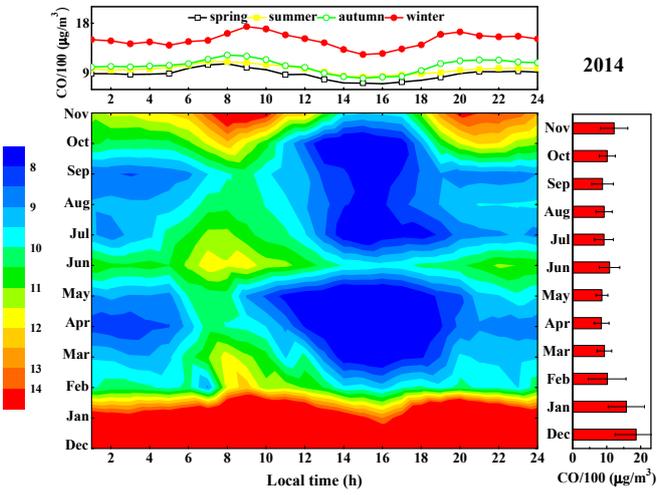
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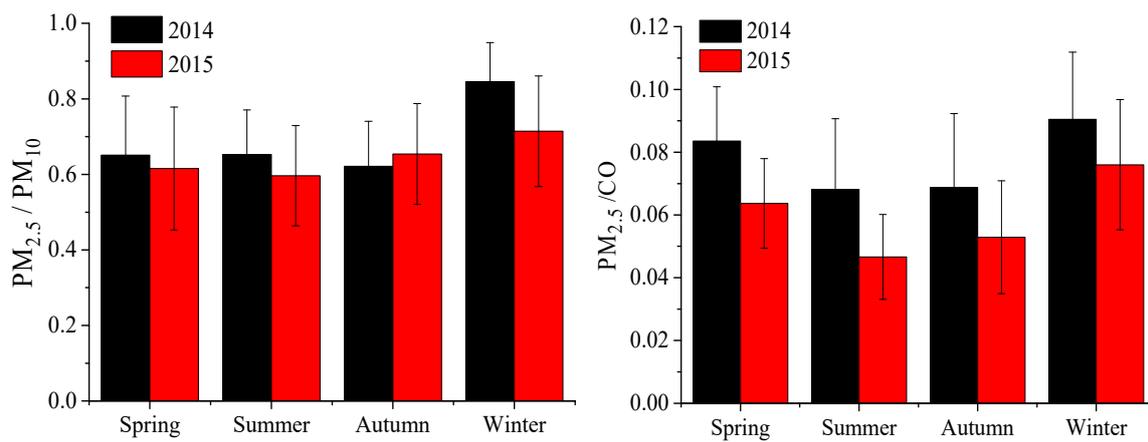
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116 **Fig. S1.** Diurnal variations of SO₂ (upper) and CO (down).

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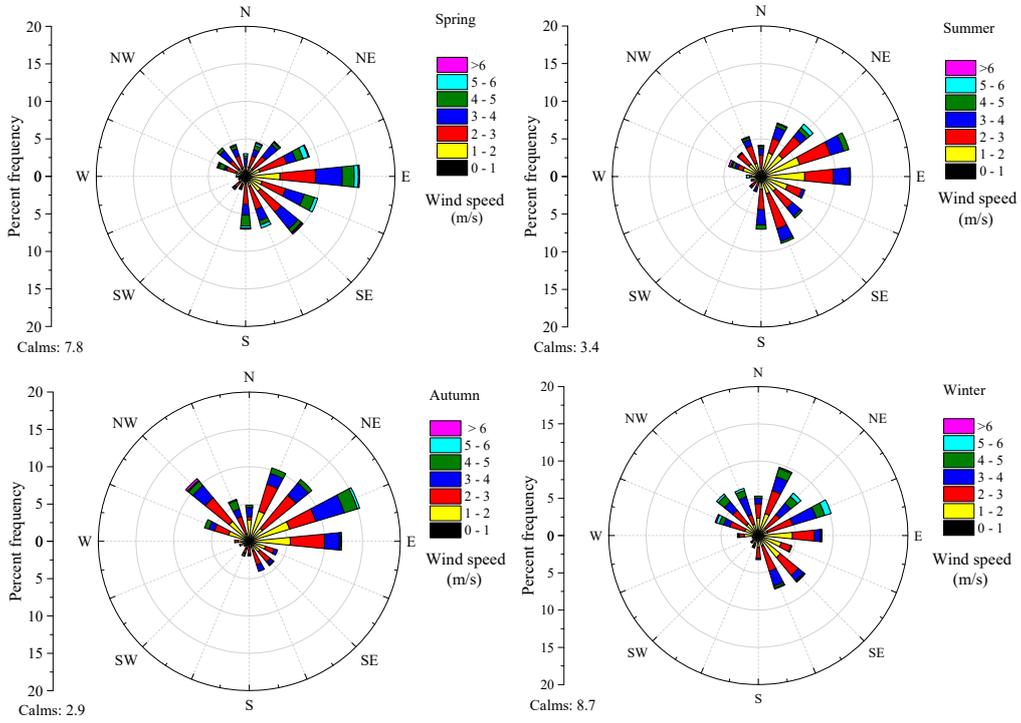


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119 **Fig. S2.** The seasonal variations of PM_{2.5}/PM₁₀ ratios and PM_{2.5}/CO ratios.

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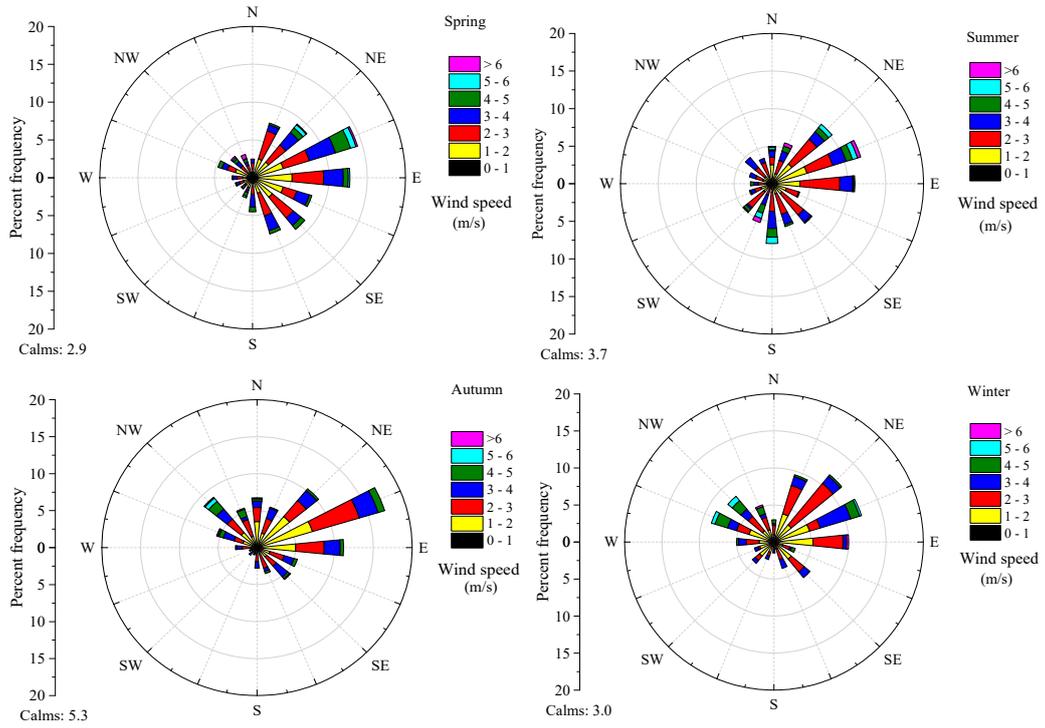
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124 **Fig. S3.** The relationship between percent frequency and wind speed, direction for seasonal distribution in 2014 at
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130 **Fig. S4.** The relationship between percent frequency and wind speed, direction for seasonal distribution in 2015 at
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