| 1 | Quasi-weekly oscillation of regional PM _{2.5} transport over |
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| 2 | China driven by the synoptic-scale disturbance of East |
| 3 | Asian Winter Monsoon circulation |
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| 21 | Abstract: The regional PM _{2.5} transport is one of the important causes for atmospheric |
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understanding of regional PM_{2.5} transport with source-receptor relationship and the meteorological
 mechanism in atmospheric environment change.

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Key words: regional PM_{2.5} transport, quasi-weekly oscillation, source-receptor relationship,
extended empirical orthogonal function (EEOF)

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41 1 Introduction

PM2.5 pollution has attracted worldwide attention due to its adverse impact on the 42 43 environment and human health (Fan et al., 2016; Geng et al., 2021; Lin et al., 2018). The PM_{2.5} pollution in the cold season has become one of the major atmospheric environmental problems in 44 45 China (An et al, 2019; X-Huang et al, 2020). The high-concentration $PM_{2.5}$ tends to occur with extensive spatiotemporal coverage (Tao et al, 2016; Zhang et al, 2019), and synthetic 46 physical-chemical processes caused such heavy PM_{2.5} pollution events (Ding et al, 2017; Quan et 47 al., 2020), including emissions (Liu et al, 2016; Zheng et al, 2018a), chemical formation (Huang et 48 49 al, 2014; Nie et al, 2014), atmospheric boundary layer processes (Huang et al, 2018; Zhong et al, 50 2019), localized circulation (Miao et al, 2015; Shu et al, 2021; Zheng et al, 2018b), as well as 51 weather and climate (Cai et al, 2017; Wu et al, 2016). The interactions among these physical and 52 chemical processes make it more challenging to comprehend the severe haze formation, which 53 serves as one of the major difficulties in forecasting and controlling atmospheric environment 54 change and heavy air pollution (Zhang et al., 2012; Zhang et al., 2019).

PM2.5 is featured with complex spatiotemporal changes on multiscale (Georgoulias and 55 56 Kourtidis, 2012; Wu et al, 2021). $PM_{2.5}$ oscillates periodically at multi-time scales, and the 57 periodic oscillation of atmospheric circulation is the leading cause of the cyclical variations of 58 $PM_{2.5}$ (Chen et al, 2020; Dong et al, 2021; Fu et al, 2020; Perrone et al, 2018). To be specific, the 1-d periodic change or diurnal variation of near-surface PM2.5 concentrations is mainly attributed 59 60 to the atmospheric boundary layer process and localized circulation (Miao et al, 2019); the 61 periodic change of around 7 days may be controlled by the fluctuation of the long-wave trough in middle and high latitudes (Guo et al, 2014); the oscillating cycle of about 14 days is closely 62 63 related to the quasi-biweekly oscillation of the synoptic circulation (Gao et al, 2020; Zhao et al, 64 2019); and the 30-60-d intra-seasonal oscillation is mainly caused by the impact of monsoon

circulation change (Xu et al, 2014; Zhang et al, 2019). Comprehensively revealing the interaction between $PM_{2.5}$ and meteorology at different time scales is essential for solving air pollution problems more effectively (B äumer and Vogel, 2007; Wang et al, 2020). Previous studies mainly focused on the multiscale periodic variation of atmospheric pollutants in a certain region or local area, <u>have_not</u> yet found on the $PM_{2.5}$ trans-regional and periodic oscillation in the large area of central and eastern China (CEC).

71 East Asian Winter Monsoon (EAWM) is one of the most active atmospheric circulation system in the cold season over the Northern Hemisphere (Ding et al, 2017; Wu and Wang, 2002), 72 73 which is also a critical leading factor for the variation of wintertime air pollution in CEC (Chin, 74 2012; Li et al, 2016). Being the major circulation system of EAWM, the Siberian High dominates 75 the cold seasons, acting as a particular driver of cold airflows, so having an important impact on 76 the wintertime atmospheric environment in CEC (An et al, 2019; Shen et al, 2021, 2022; Wu et al, 77 2016). The rapid southward advance of cold air with strong Siberian High can effectively drive the 78 regional transport of air pollutants with less accumulations across CEC, while the weak Siberian High with the slow southward movement of cold air can particularly favorable for the transport of 79 80 air pollutants from the northern source regions to southern receptor region over CEC (Hou et al., 81 2020; Zhang et al., 2016). When the position of Siberian High is more eastern than normal, the transport of air pollutants from northern China to the south is weakened, and the aggravation of 82 83 pollution is enhanced in northern China (Jia et al., 2015). Regional pollutant transport driven by 84 the southward movement of a cold front with the Siberian High would exacerbate the air quality in 85 the corresponding receptor regions (Kang et al., 2019; Hu et al., 2021; Shen et al, 2022). The 86 characteristics of atmospheric circulation anomalies favoring heavy haze pollution in China have 87 changed in recent years, and the leading formation mechanism of severe haze has been shifting 88 from local accumulation to regional transport processes in eastern China (X-Yang et al, 2021b). Therefore, studying the influence of EAWM circulation system on regional pollutant transport 89 90 over CEC is an important issue in atmospheric environment changes (Bai et al, 2021, 2022; Ge et 91 al, 2018; Merrill and Kim, 2004; Tan et al, 2021; W. Yang, et al, 2021a). 92 Previous studies have primarily focused on the relationship between atmospheric 93 intraseasonal oscillations in the mid-to-high latitudes of the Eurasian region and the persistent

94 PM_{2.5} pollution (An et al., 2022; Gao et al., 2020; Li et al., 2021; Liu et al., 2022; Wu et al., 2023;

| 95 | Yang et al., 2024b). PM _{2.5} concentration anomalies in North China exhibit significant lifetimes of |
|-----|-----------------------------------------------------------------------------------------------------------------|
| 96 | 10-30 days, with anticyclonic anomalies and related meteorological conditions (e.g., surface air |
| 97 | temperature, boundary layer height) in Northeast Asia influencing local PM2.5 accumulation and |
| 98 | hygroscopic growth (An et al., 2022; Yang et al., 2024b). These studies have investigated the |
| 99 | quasi-biweekly lifecycle of persistent PM2.5 pollution events in North China through phase |
| 100 | synthesis methods (Gao et al., 2020; Wu et al., 2023; Yang et al., 2024b). However, there remains |
| 101 | a lack of systematic studies on the synoptic-scale oscillation of regional PM _{2.5} transport. |
| 102 | The "harbor" effect on the eastern lee of the Tibetan Plateau's large topography on the |
| 103 | westerlies is possibly an important factor influencing the regional distribution of PM _{2.5} pollution |
| 104 | in CEC with weak horizontal winds and sinking motion in the lower troposphere, which |
| 105 | exacerbates the environmental impacts of local air pollutant emissions establishing a |
| 106 | "susceptibility zone" in this region (Xu et al., 2016; Zhu et al, 2018). Anticyclones and cyclones |
| 107 | alternatively affect the region on a time scale of 3-7 days, resulting in periodic air pollution in |
| 108 | cities (Guo et al., 2014). Thus, the weather system in the CEC is basically characterized by |
| 109 | periodic changes and the cold air in winter with EAWM oscillates in quasi-weekly periods (Wu |
| 110 | and Wang, 2002; Wu et al., 2016). However, the influence of the synoptic-scale disturbance of the |
| 111 | EAWM on regional PM _{2.5} transport over CEC is not yet clear. Responding to this problem, this |
| 112 | study aims to reveal from a new perspective the quasi-weekly oscillation (QWO) of regional $PM_{2.5}$ |
| 113 | transport over CEC affected by EAWM and its underlying mechanism with the synoptic-scale |
| 114 | oscillation of the EAWM circulation. This study could be beneficial to deepen the understanding |
| 115 | of regional PM _{2.5} transport, its source-receptor relationship and meteorological mechanism in the |
| 116 | atmospheric environment changes, and provide scientific evidence for air pollution forecast, early |
| 117 | warning and coordinated control. |
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119 2 Data and methods
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- 120 2.1 Environmental and meteorological data
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The daily dataset of PM_{2.5} concentrations selected for this study was from China National
 Environmental Monitoring Center (http://datacenter.mee.gov.cn/), including daily PM_{2.5}
 concentrations from 1079 air quality monitoring stations in CEC during the winters

125 (December-February) of 2015-2019.

Meteorological data were selected out of the NCEP/NCAR global reanalysis daily data (https://psl.noaa.gov/data/gridded/tables/daily.html) with a grid resolution of 2.5 °×2.5 ° for the large-scale circulation analysis. It is composed of the daily sea level pressure (SLP), air temperature at 1000 hPa, and the U- and V-components of wind at 1000 hPa during the winters of 2015–2019.

131 addition, the ERA5-land In high-resolution reanalysis hourly dataset 132 (https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-land?tab=form) with spatial resolution of 0.1 °×0.1 ° was selected for the calculation of transport flux (TF) of PM_{2.5} in CEC. 133 134 The U- and V-components of the 10-m wind over CEC were obtained at 00, 06, 12, and 18 UTC 135 daily during the winter (December-February) of 2015-2019. In order to match the resolution of 136 PM_{2.5} daily data, the ERA5-Land high-resolution 10-m wind was processed into daily average 137 data.

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139 $2.2 \text{ PM}_{2.5}$ TF and its divergence

141 In order to quantitatively characterize the horizontal transport direction and intensity of PM2.5 142 as well as convergence or divergence during regional PM2.5 transport, we introduced the concepts of PM2.5 TF and divergence of PM2.5 TF. Generally, there are two types of TF: horizontal and 143 vertical. This study only addresses the near-surface horizontal PM2.5 TF. The horizontal PM2.5 TF 144 145 is defined as the PM2.5 mass concentrations passing through the unit area in unit time (unit: µg m⁻² 146 s⁻¹), expressed as the product of wind vector and PM_{2.5} concentration (Liu et al., 2019; Ma et al., 147 2021), and its vector points to the same direction as the horizontal wind. The zonal component (F_u) 148 and meridional component (F_{y}) of PM_{2.5} TF vector (TFV) and the magnitude (TFM) are calculated 149 as follows:

$$F_u = C u \tag{1}$$

$$F_{\nu} = C v$$
 (2)

$$152 TFV = F_u i + F_v j (3)$$

$$153 TFM = \sqrt{F_u^2 + F_v^2} (4)$$

where *C* is the surface $PM_{2.5}$ concentration, *u* and *v* are the zonal and meridional components of the 10-m wind speed, respectively. Firstly, the U- and V-components of ERA5-Land high-resolution 10-m wind are interpolated to 1079 stations of environmental measurements in CEC for calculations of near-surface $PM_{2.5}$ TF in this study. Then, the daily $PM_{2.5}$ TF of the 1079 stations for the winters from 2015 to 2019 are calculated according to the calculation by Formulas (1)–(4).

160 The divergence of $PM_{2.5}$ TF can be an indicator for the $PM_{2.5}$ budget. When positive 161 divergence occurs, the air pollutants were net outflow from the domain region, and vice versa 162 (Wang et al., 2021). The divergence of horizontal $PM_{2.5}$ TF near the surface is calculated as 163 follows (Wang et al., 2021):

164
$$D = \frac{\partial F_u}{\partial x} + \frac{\partial F_v}{\partial y}$$
(5)

where *D* is the horizontal $PM_{2.5}$ TF divergence, unit: $\mu g m^{-3} s^{-1}$. If *D* is positive (negative), it indicates divergence (convergence) of $PM_{2.5}$ TF.

167 In the *i* and *j* grids, the expression of Formula (5) for the differential calculation with grid 168 spacing to be d is

169
$$D = \frac{Fu_{i+1,j} - Fu_{i-1,j} + Fv_{i,j+1} - Fv_{i,j-1}}{2d}$$
(6)

170 When calculating the horizontal divergence of transport $PM_{2.5}$ flux, it is necessary to 171 interpolate the station data of zonal and meridional components (F_u , F_v) of $PM_{2.5}$ TFV to grid 172 spacing with 0.25 by 0.25 degree in longitude and latitude in CEC and then calculate the 173 divergence of $PM_{2.5}$ TF at each grid point according to Formula (6).

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175 <u>2.3 Butterworth filter</u>

177 Atmospheric motion encompasses a variety of temporal and spatial scales. The sequences of 178 meteorological variables often contain complex periodic components and exhibit multi-time-scale 179 variations, including daily, weekly, seasonal, and interannual variations. Numerous observations 180 have found QWO with periods of less than 10 days across various meteorological elements in the 181 EAWM system (Compo et al., 1999; Murakami, 1979; Wu and Wang, 2002). Synoptic-scale atmospheric variations are closely related to atmospheric longwave adjustments, with QWO 182 periods of 4-7 days observed in cold air activities of the EAWM (Bai et al., 2022; Wu and Wang, 183 184 2002). The synoptic-scale disturbance regulates the generation, transport, and removal of PM_{2.5} in air pollution, which is a key mechanism behind the 4-7 day periodic changes in PM25 in CEC 185

186 during the periods of EAWM (Guo et al., 2014; Liu et al., 2018; Quan et al., 2014, 2020). Based 187 on the research objectives, identifying the desired periodic components from the original 188 observational sequences is referred to as sequence filtering. In this study, we employed a 189 Butterworth filter to extract QWO from observational data. The Butterworth filter is commonly used to separate atmospheric periodic variations across 190 191 specific frequency bands. Due to its smooth amplitude response, linear phase characteristics, and 192 ease of implementation, Butterworth filter has been widely applied in climate and meteorological studies (Gouirand et al., 2012; Yang et al., 2024a). The Butterworth filter can be configured as a 193 194 low-pass, high-pass, or band-pass filter, depending on the specific requirements. A band-pass 195 filtering only allows signals within a defined frequency range to pass through with attenuating 196 signals outside the defined frequency range. It is often employed to extract and analyze signals 197 within specific frequency bands, such as particular weather patterns and climate cycles. In this 198 study, to investigate the QWO (8-d) of regional PM25 transport over the CEC under the influence 199 of EAWM circulations in the synoptic scale, we applied Butterworth band-pass filtering to the 200 daily TFM of PM_{2.5} change and daily SLP anomalies during the winters of 2015-2019 for 201 identifying at the quasi-weekly (6-9 days) synoptic-scale component of regional transport of 202 PM2.5 over CEC.

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204 2.4 Extended empirical orthogonal function (EEOF)

The Empirical Orthogonal Function (EOF) analysis is a widely-applied climate statistical method in atmospheric and oceanographic scientific studies (Kim et al., 2015; Li et al., 2019; Schepanski et al., 2016), also used to investigate the variability of atmospheric aerosols at different spatiotemporal scales (Bai et al., 2022; Feng et al., 2020). The mathematical process of EOF analysis is to decompose the variable field $X_{m \times n}$, which consists of observations at *n* times at *m* spatial points, into a linear combination of *p* spatial eigenvectors (modes) with corresponding time-weighting coefficients:

213

$$X_{m \times n} = V_{m \times p} T_{p \times n} \tag{7}$$

where *V* is the spatial eigenvector (load) and *T* represents the time coefficient. The main information of variable field $X_{m \times n}$ is represented by several eigenvectors. Since the method has been maturely applied, the detailed calculation steps of EOF decomposition are omitted here, andour focus is on how to construct the observation matrix.

Firstly, we decompose the daily PM_{2.5} TFM anomalies of 1079 stations in CEC during the
 winters of 2015-2019 by EOF method. Thus, the following observation matrix can be obtained:

221
$$X = \begin{bmatrix} x_{11} \cdots x_{1n} \\ \vdots \\ x_{m1} \cdots x_{mn} \end{bmatrix}$$
(8)

where X represents the PM_{2.5} TFM anomalies, *m* represents the spatial points for 1079
stations, and *n* represents the observation times of 450 days. Then, the variable field X is
decomposed into the sum of the product of space and time functions according to Formula (7).
EOF decomposition of PM_{2.5} TFV anomalies can be performed by employing the complex

226 matrix, hence the following observation matrix is constructed:

227
$$X = \begin{bmatrix} u_{11} \cdots u_{1n} \\ \vdots \\ u_{m1} \cdots u_{mn} \\ v_{11} \cdots v_{1n} \\ \vdots \\ v_{m1} \cdots v_{mn} \end{bmatrix}$$
(9)

where *X* is the $PM_{2.5}$ TFV anomalies, and *u* and *v* refer to the zonal and meridional components of TFV anomalies.

With EOF analysis we can get the spatial distribution structure, which is in a fixed time pattern of climate variables, but we cannot get a temporally moving spatial distribution structure. EEOF is an extension of the EOF to analyze the autocorrelations of the variable field over time. By selecting a lag time, the original observational matrix is expanded into multiple continuous time matrices, diagnosing the temporal changes in the spatial structure of variable fields. This method has found-widespread applications in the analysis and prediction of marine and atmospheric motions (Dey et al., 2018; Qian et al., 2019; H.-Wang et al., 2019).

In this study, we utilized the EEOF analysis to reveal the evolution of $PM_{2.5}$ TF to reveal the spatiotemporal variations of regional $PM_{2.5}$ transport. On the basis of Formula (8), a new extension matrix of $PM_{2.5}$ TFM is constructed. Due to the study on the synoptic scale, 5 lag times are

selected, and each lag time is 1 day in length. The constructed observation matrix is as follows:

$$X = \begin{bmatrix} x_{1,1} \cdots x_{1,n-5} \\ \vdots \\ x_{m,1} \cdots x_{m,n-5} \\ x_{1,2} \cdots x_{1,n-4} \\ \vdots \\ x_{m,2} \cdots x_{m,n-4} \\ x_{1,3} \cdots x_{1,n-3} \\ \vdots \\ x_{m,3} \cdots x_{m,n-3} \\ x_{1,4} \cdots x_{1,n-2} \\ \vdots \\ x_{m,4} \cdots x_{m,n-2} \\ x_{1,5} \cdots x_{1,n-1} \\ \vdots \\ x_{m,5} \cdots x_{m,n-1} \\ x_{1,6} \cdots x_{1,n} \\ \vdots \\ x_{m,6} \cdots x_{m,n} \end{bmatrix}$$
(10)

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Seen from Formula (10), the new extended matrix is composed of $X_{6m,n-5}$, where X is the PM_{2.5} TFM anomalies, *m* is the spatial points of observation station, and *n* is the observation times of 450 days. When EEOF decomposition is performed on PM_{2.5} TFV, the complex matrix is still used for the extension, and the same lag scheme is adopted to construct a new extended matrix of PM_{2.5} TFV based on Formula (9). After constructing the initial data matrix, the EEOF decomposition method is in line with the classical EOF decomposition method.

Additionally, existing studies have utilized wavelet analysis, power spectrum analysis, and
band-pass filtering methods to extract intraseasonal oscillation sequences of regional PM_{2.5}
concentrations (An et al., 2022; Gao et al., 2020; Li et al., 2021; Liu et al., 2022; Wu et al., 2023;
Yang et al., 2024b). Such approaches may serve as alternative methods to EEOF analysis for
establishing the quasi-weekly lifecycle of regional PM_{2.5} transport.

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254 3 Results and discussion

- 256 3.1 QWO of regional PM_{2.5} transport over CEC
- 257

The EOF decomposition is carried out on the daily anomalies of $PM_{2.5}$ TFM and TFV in the winters of 2015-2019 over CEC. The first two EOFs explain 26.6% and 14.2% (29.1% and 11.8%) of the total anomalous variations of $PM_{2.5}$ TFM (TFV), which is very helpful for better characterizing regional $PM_{2.5}$ transport variations.

262 Two principal modes govern the variations of PM2.5 TF anomalies over CEC: the first leading 263 mode of monopole (EOF1) and the second mode of meridional dipole (EOF2) (Fig. 1). EOF1 264 indicates the enhanced PM2.5 TF over CEC (Fig. 1a). The large value center of TF mainly occurs 265 in central China, and the transport vector direction is abnormally by north. The horizontal PM_{2.5} 266 transport is unusually strong in central China affected by the EAWM, presenting a typical channel 267 for regional PM2.5 transport over CEC (Yang W. et al., 2021a). The dipole mode of PM2.5 TF 268 anomalies displays a south-north out-of-phase pattern, with the flux large value centers located in 269 the North China Plain (NCP) and the Twain-Hu Basin (THB) respectively, and the vector 270 directions are opposite (Fig. 1b). This mode indicates that the air pollutants from NCP in the 271 upwind are transported to THB in the downwind driven by the prevailing northerlies of EAWM (Hu et al., 2021; Shen et al., 2022), and the PM_{2.5} flux in NCP decreases while that in THB 272 273 increases in the regional PM2.5 transport process.

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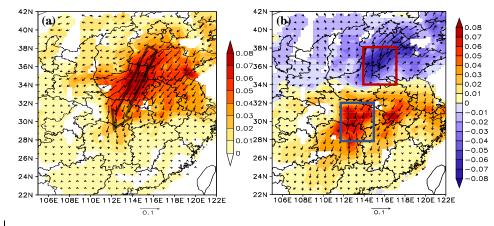




Figure 1. Spatial pattern of the (a) EOF1 and (b) EOF2 loads in the daily change of PM_{2.5} TFV anomalies (vectors_a
unitless) and TFM anomalies (color contours<u>unitless</u>) over CEC in the winters of 2015-2019. The red and blue
boxes indicate NCP and THB, respectively<u>The grid cells in white represent "missing values".</u>

Through EOF decomposition, the $PM_{2.5}$ TF could be understood from the perspective of a fixed time pattern of climate, but the <u>temporal changes in the moving spatial structure</u> of $PM_{2.5}$ TF over CEC failed to be obtained. However, EEOF decomposition can be used to analyze the continuous structural evolution of the main modes of regional $PM_{2.5}$ TF over CEC.

284 The EEOF decomposition was carried out for the daily variations of PM2.5 TFM anomalies 285 and TFV anomalies respectively over CEC during the winters of 2015-2019. Figure 2 and Figure 286 S1 show the spatial distribution of different lag times for the main modes of EEOFs, which account for about 20% of the total variation. According to the analysis, the PM2.5 TFM anomalies 287 288 for EEOF2 and EEOF3, as well as TFV anomalies for EEOF1 and EEOF2, all show the structural 289 evolutions in the different phases of regional $PM_{2.5}$ transport in one cycle. As it can be seen, 290 Figures. 2a-d, S1a-d, and 2e-h respectively describe the evolution of the first and second four phases in a cycle and the first four phases in the next cycle (one phase represents 1day). 291

292 Figures 2a-d illustrate the positive anomalies of PM2.5 TF shifting from NCP to THB in the 293 first four phases under the effect of the EAWM, causing the upwind PM2.5 TF to decrease and the 294 downwind PM2.5 TF to increase, which is in line with the spatial pattern of the EOF modes in 295 Figure 1. The last four phases show the out-of-phase pattern of the first half cycle (Figs. S1a-d). It 296 is noted that when anomalies of $PM_{2.5}$ TFV in the NCP turn to the northerly direction (Fig. S1d and Fig. 2e), it is a strong signal initiating the regional $PM_{2.5}$ transport. Then, the transport is 297 298 repeated in the next periodic cycle (Figs. 2e-h). Therefore, the regional PM_{2.5} transport over CEC 299 enjoys a quasi-weekly (8-d) oscillation pattern.

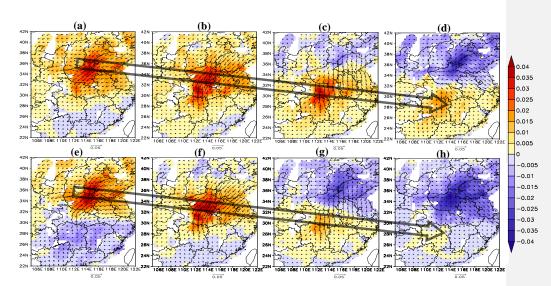
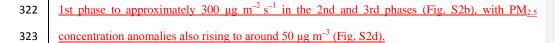


Figure 2. (a)-(d) The first four phases (days) of QWO (8-d) during the regional $PM_{2.5}$ transport over CEC; (e)-(h) the first four phases (days) of the next cycle. The Loads of $PM_{2.5}$ TFM anomalies (color contours, <u>unitless</u>) for EEOF2 and TFV anomalies (vectors, <u>unitless</u>) for EEOF1 with lag time (a) 0 d, (b)1 d, (c) 2 d and (d) 3 d, and loads of TFM anomalies (color contours, <u>unitless</u>) for EEOF3 and TFV anomalies (vectors, <u>unitless</u>) for EEOF2 with lag time (e) 2 d, (f) 3 d, (j) 4 d and (h) 5 d over CEC in the winters of 2015-2019.

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To further study the variations of regional $PM_{2.5}$ transport over CEC, we have screened out 23 typical events with greater than 1.5 times standard deviations based on the standardized time coefficient of EEOF, and then used the 8 consecutive days of each event as the 8 phases of QWO in the composite analysis on the 23 typical events of regional $PM_{2.5}$ transport over CEC.

312 Figure 3 shows the composited PM2.5 TF, divergence of PM2.5 TF, and PM2.5 concentration 313 anomalies in the first four phases of QWO. The high fluxes of PM2.5 transport from north to south 314 persists for 3-4 days over CEC and decline in the THB (Fig. 3a-d). The regional PM_{2.5} transport 315 lifetime corresponding to synoptic systems is about 3-5 days (H-Huang et al., 2020a). Abnormal 316 northerly winds drive the heavy PM_{2.5} pollution from the upwind NCP to the downwind regions, 317 aggravating PM_{2.5} pollution in the downwind THB (Figs. 3e-h). Under the context of QWO, the average PM_{2.5} TFM in NCP decreases from approximately 400 µg m⁻² s⁻¹ in the 1st and 2nd phases 318 to 200 and 100 µg m⁻² s⁻¹ in the 3rd and 4th phases, respectively (Fig. S2a). Correspondingly, the 319 $PM_{2.5}$ concentration anomalies decline from around 100 µg m⁻³ to approximately -50 µg m⁻³ (Fig. 320 321 <u>S2c). In the downwind THB, the average PM_{2.5} TFM increases from about 200 μ g m⁻² s⁻¹ in the</u>



324 It is noteworthy that the regions PM_{2.5} TF convergence zone (negative value of divergence) 325 matches spatially the centers positive anomaly centers of PM2.5 concentrations, which is confirmed 326 with a significantly negative correlation of the PM2.5 concentrations with divergences of PM2.5 TF 327 in the 23 typical events (Fig. $\frac{\$2\$3}{1}$). The PM_{2.5} transport is accompanied by flux convergence, 328 which is beneficial to the $PM_{2.5}$ accumulation. In addition, the $PM_{2.5}$ TF in the upwind NCP changes from convergence to divergence, and the divergence of the $PM_{2.5}$ TF in the downwind 329 330 THB alters to convergence in the meantime (Figs. 3i-1), indicating that the PM_{2.5} over THB is 331 transported from the upwind NCP. The source-receptor relationship between NCP and THB during 332 the regional PM_{2.5} transport over CEC is discussed in detail in the next section.



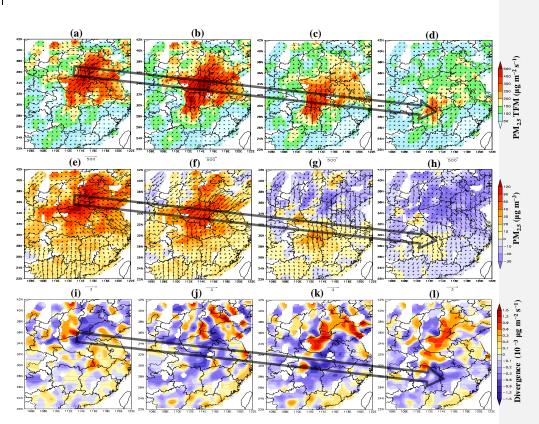




Figure 3. Spatial distributions of the composited (a-d) $PM_{2.5}$ TFM (color contours, unit: $\mu g m^{-2} s^{-1}$) and TFV (vectors, unit: $\mu g m^{-2} s^{-1}$), (e-h) anomalies of $PM_{2.5}$ concentrations (color contours, unit: $\mu g m^{-3}$) and 10-m wind vectors (unit: $m s^{-1}$), (i-l) divergence of $PM_{2.5}$ flux (color contours, unit: $10^{-3} \mu g m^{-3} s^{-1}$) in the first four phases of

QWO during the 23 typical events of regional $\ensuremath{\text{PM}_{2.5}}$ transport over CEC.

| 339 340 341 | 3.2 Source-receptor relationship in regional PM _{2.5} transport from NCP to THB |
|-------------------|------------------------------------------------------------------------------------------------------------------------------|
| 342 | The regional pollutant transport governed by emissions and meteorology leads to a complex |
| 343 | source-receptor relationship of air pollution changes (Yu et al., 2020). Band-pass filtering is |
| 344 | performed on the daily $PM_{2.5}$ TFM anomalies at a quasi-weekly (6-9 days) synoptic scale in the |
| 345 | winters of 2015-2019. In Figure 4a, we composite the filter components of $PM_{2.5}$ TFM in the 8 |
| 346 | phases of QWO during the 23 typical events of regional $PM_{2.5}$ transport over the NCP and THB, |
| 347 | respectively. The $PM_{2.5}\ TF$ exhibits an obvious QWO on the synoptic scale (Fig. 4a). The $PM_{2.5}$ |
| 348 | TF over the NCP continues to decline in the first four phases, while that of THB first rises and |
| 349 | then falls in the last four phases, the $\ensuremath{\text{PM}_{2.5}}$ TF over the NCP increases continuously, while that of |
| 350 | THB falls first and then rises. We can see that the QWO of $\text{PM}_{2.5}\text{TF}$ over THB lags behind the |
| 351 | NCP by 2 phases (Fig. 4a). The high TFM of $PM_{2.5}$ from NCP in the first phase spread to THB, |
| 352 | resulting in the peak of $PM_{2.5}$ TF over THB in the third phase. |
| 353 | In addition, the distribution of the differences in $PM_{2.5}$ TF and the vectors between phase 3 |
| 354 | and phase 1 of the QWO, and the $PM_{2.5}\ TF$ decrease and increase from phase 1 to phase 3 |
| 355 | respectively over the upwind NCP and the downwind THB, which is in accordance with the |
| 356 | spatial pattern of the EOF mode (Figs.1b and 4b), indicating that the source-receptor relationship |
| 357 | over CEC exist the regions NCP and THB of regional PM _{2.5} transport over CEC. |

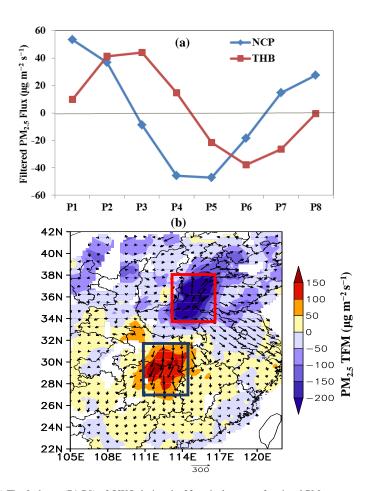


Figure 4. (a) The 8 phases (P1-P8) of QWO during the 23 typical events of regional $PM_{2.5}$ transport over the NCP and THB with composited 6-9 d band-pass filtering of $PM_{2.5}$ TFM; (b) spatial distribution of the differences in PM_{2.5} TFM (color contours, unit: $\mu g m^{-2} s^{-1}$) and TFV (vectors, unit: $\mu g m^{-2} s^{-1}$) between the 3rd phase and the 1st phase of QWO. The red and black boxes represent NCP and THB.

364

The statistical analysis based on long-term observation also shows that there is a significant 2-day lag relationship of positive correlation between NCP and THB in $PM_{2.5}$ TF in the QWO (Fig. 5a). This discloses that the air pollutants are transported from the upwind NCP to the downwind THB in 2 days, confirming a quasi-2-d lag in the regional $PM_{2.5}$ transport from NCP to THB (Hu et al., 2021; Shen et al., 2021). Additionally, in the long-term change of air pollution, the divergences of $PM_{2.5}$ TF in the NCP are significantly negatively correlated to that of THB (Fig. 5b), that is, the $PM_{2.5}$ TF convergences in the downwind THB fits well with the $PM_{2.5}$ TF

372 divergence in the upwind NCP. It can be reflected that the changes in the synoptic scale of EAWM

373 atmospheric circulation impel the regional $PM_{2.5}$ transport to build the source-receptor relationship

of atmospheric pollutants between the NCP and THB.

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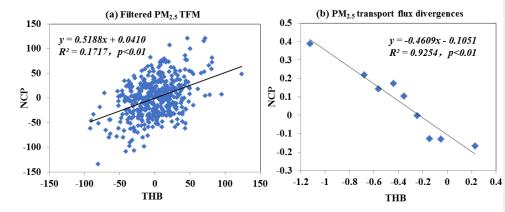


Figure 5. (a) Scatter plot of 6-9-d filtering components of $PM_{2.5}$ TFM ($10^{-3} \mu g m^{-2} s^{-1}$) over THB in 2-day lag and NCP during the winters of 2015-2019; (b) scatter plot of $PM_{2.5}$ TF divergences ($10^{-3} \mu g m^{-3} s^{-1}$) between THB and NCP, and the $PM_{2.5}$ TF divergences are averaged over the value interval of 0.1.

| 381 | Driven by prevailing winds of EAWM, the THB became the main receptor for regional |
|-----|------------------------------------------------------------------------------------------------------------|
| 382 | transport of air pollutants over CEC (Bai et al., 2022; Shen et al., 2021). During 2015-2019, |
| 383 | approximately 65.2% of the total PM2.5 heavy pollution events in the THB were triggered by |
| 384 | regional transport of air pollutants over CEC (Hu et al., 2022; Shen et al., 2021). Such PM _{2.5} |
| 385 | transport from upstream source regions in CEC contributes 51%-85.7% of the PM _{2.5} pollution |
| 386 | over the THB receptor region (Hu et al., 2021; Lu et al., 2017; Shen et al., 2022; Yu et al., 2020), |
| 387 | revealing the dominance of regional transport of air pollutants from CEC to the THB with the |
| 388 | meteorological drivers. Our research emphasizes the QWO of regional PM2.5 transport over CEC |
| 389 | with the driver of the synoptic-scale disturbances of EAWM circulation, confirming the |
| 390 | source-receptor relationships with their 2-day lagging effects in the regional PM2.5 transport |
| 391 | between the upstream NCP source region and the THB receptor region. |
| 392 | |
| 393 | 3.3 Effect of synoptic-scale disturbance of EAWM circulation on QWO of regional PM2.5 transport |

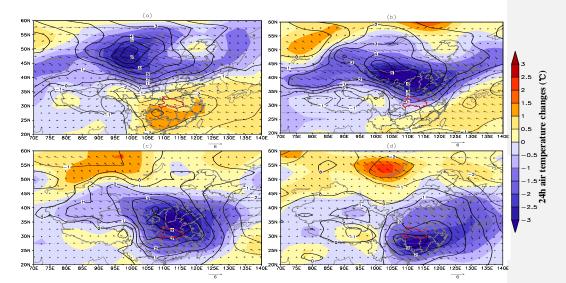
394 over CEC

395

396 Meteorological change is the essential factor in regulating the occurrence and development of 16

397 PM2.5 pollution on synoptic scales. To investigate the QWO of EAWM circulation in the synoptic 398 scale disturbance, this study performs the 6-9-d band-pass filtering of the daily SLP anomalies 399 (denoted as SLP_{OWO}) in East Asia during the winters of 2015-2019. The SLP and SLP_{OWO} fields 400 (Figs. 6 and 7) as well as $PM_{2.5}$ concentrations and 10-m winds (Fig. S3S4) in the 8 phases of 401 QWO during the 23 typical events were composited, respectively. The QWO of regional PM2.5 transport is connected with the "weekly-cycle" synoptic process of PM2.5 transport and 402 403 accumulation over CEC (Fig. S3S4), and it is powered mainly by the Siberian High circulation 404 with the synoptic-scale disturbance of EAWM circulation (Figs. 6 and 7).





406

407 Figure 6. The composited differences between the current day and the previous day of SLP (black contour lines,
408 unit: hPa), 1000 hPa air temperature (color contours, unit: °C) and wind vectors (unit: m s⁻¹) in the first four
409 phases (a-d) of QWO during the 23 typical events.

410

The condition of uniform pressure in the front of Siberian High could favor the $PM_{2.5}$ accumulation over the NCP for triggering regional $PM_{2.5}$ transport over CEC (Fig. 7a). The regional heavy pollution of $PM_{2.5} > 150 \ \mu g \ m^{-3}$ lasts for 1-2 days (Figs. <u>S3a-S4a</u> and <u>S3bS4b</u>). With the development of the Siberian High, the extension of the high pressure guides the cold air to advance southward (Park et al., 2014). As the result of the increasing air pressure gradients, the strong northerly winds in the EAWM circulation system, deliver high-level $PM_{2.5}$ air mass from NCP to THB (Figs. 7a-d, Figs. <u>S3aS4a</u>-d). In addition, the cold and high air pressure system with 17 418 the abnormal northerly airflows moves from the Siberia-Mongolia region to CEC in the first four 419 phases (Fig. 6), providing beneficial synoptic circulation patterns for regional PM_{2.5} transport. 420 Thus, the periodic extension of the Siberian High with the associated strong cold air intrusion is an 421 important driver in the regional PM_{2.5} transport over CEC.

422 Notably, we can see that in the first four phases, the SLP_{QWO} positive anomalies occur, 423 develop, and expand southward from the Siberia-Mongolia region to CEC (Figs. 7a-d). The 424 synoptic-scale disturbance with the extension of Siberian High and the southward movement of 425 cold air could drive the regional PM2.5 transport over CEC (Figs. 7a-d). The situation of the last 426 four phases is opposite to the SLP_{OWO} negative anomalies in Siberia-Mongolia region, inhibiting 427 the Siberian High and cold air intrusion (Figs. 7e-h). The low and uniform pressure is beneficial to 428 the accumulation of PM2.5. Therefore, the periodic changes in the synoptic-scale disturbance of the 429 EAWM circulation impel the QWO of regional PM2.5 transport over CEC.

Corr = 0.81, p < 0.01Corr = 0.86, p < 0.01(ь) 60N 60N 55N 55N 50N 50N 45N 451 40N 40N 351 35N 301 30N 25N 25N 20N 108E 114E 120E 126E 132E 138 20N 72E 725 78E 9ÓE 78E 84E 9ÓE 114E 120E 126E 132E 138E 84E 96E 102E 102E 108E Corr = 0.86, p < 0.01Corr = 0.68, p < 0.01601 601 55N 1037.5 55N 501 501 45N 45N 40N 40N 35N 35N 301 30N 25N 25N 20N



20N 72E 78E 72E 78E 84E 90E 102E 108E 114E 120E 126E 132E 138E 84E 102E 108E 114E 120E 126E 132E 138E 96F 90E 96F

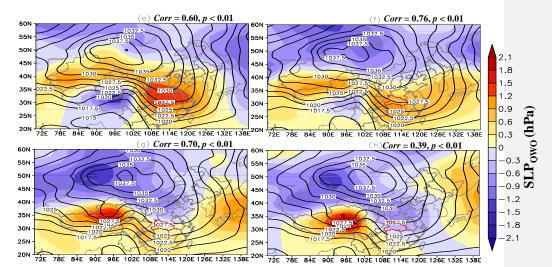


Figure 7. Composited SLP (black contour lines, unit: hPa) and its synoptic-scale filter component SLP_{QWO} (color
contours, unit: hPa) in the 8 phases (a-h) of QWO during the 23 typical events. *Coor* represents the spatial
correlation coefficients between SLP_{QWO} and the load of SLP_{QWO} decomposed by EEOF in Fig. S4.

432

437 In addition, the EEOF decomposition is carried out on the SLP_{OWO} field in the winters of 438 2015-2019 to recognize the periodic activities in the synoptic scale of the EAWM circulation. The cold air activity of EAWM presents QWO (Wu and Wang, 2002). The positive (negative) 439 440 synoptic-scale disturbance occurs in the Siberia-Mongolia region, and then spreads to CEC along 441 the northwest-southeast path, contributing to the 8-d cycle of QWO (Fig. 5455). Notably, the 442 spatial correlation coefficients between the load of SLP_{QWO} decomposed by EEOF (Fig. <u>\$4</u><u>\$5</u>) 443 and the SLP_{OWO} composited during 23 typical events (Fig.7) are highly positively correlated in the 8 phases, respectively. Therefore, the QWO in the synoptic-scale activities of the Siberian high is 444 445 an important factor for driving the QWO of regional PM2.5 transport over CEC.

446

447 4 Conclusions

Exploring the periodical oscillations of $PM_{2.5}$ pollution over CEC and the meteorological effect is crucial for understanding the change in the atmospheric environment and improving regional air quality forecasts. In this study with constructing a dataset of the daily $PM_{2.5}$ TF, the EEOF and statistical methods are used to identify the QWO of regional $PM_{2.5}$ transport with the spatiotemporal variations over CEC in winters from 2015 to 2019. The source-receptor relationship is recognized between NCP and THB with the QWO of regional $PM_{2.5}$ transport over CEC with the typical EAWM climate. Furthermore, it is revealed that the driving effect of synoptic-scale disturbance of EAWM circulations on the QWO of regional $PM_{2.5}$ transport over China.

The variations of PM2.5 TF over CEC are dominated by the first leading monopole mode and 457 458 the second meridional dipole mode. The monopole mode indicates the high PM2.5 flux along the channel of regional PM2.5 transport from NCP to THB under the governs of the EAWM 459 circulations, and the dipole mode exhibits a pattern of south-north out-phase with two centers 460 461 existing respectively in the upwind NCP and the downwind THB in regional transport of PM25 over CEC. In terms of the long-term changes in air pollution of 2015-2019, the regional PM_{2.5} 462 463 transport over CEC is featured with the QWO, verifying a source-receptor relationship for the regional PM_{2.5} transport from NCP to THB in 2 days. Such changes are incurred by the QWO in 464 465 the activities of the Siberian High, and this synoptic-scale disturbance of the EAWM circulations 466 is generated in the Siberia-Mongolia region, and then develops, marching into CEC, regulating the 467 QWO of regional PM2.5 transport.

468 The EEOF analysis with the temporal lag of the spatial fields is able to better characterize the 469 spatial and temporal evolution of perturbations, especially propagating waves in the atmosphere 470 (Weare and Nasstrom, 1982; Qian et al., 2019; Yang et al., 2024b). Due to its technical advantages, 471 the EEOF method is commonly employed to extract atmospheric oscillation patterns to reveal the 472 impacts and mechanisms of atmospheric fluctuations and monsoon circulation on regional weather, 473 climate, and atmospheric environments (Dey et al., 2018; Qian et al., 2019; Yang et al., 2024b). In 474 this study, we employed the EEOF method to identify regional $PM_{2.5}$ transport modes in synoptic 475 scale, by constructing PM_{2.5} transport flux vectors (TFV) and the magnitude (TFM) with the 476 product of near-surface PM2.5 concentrations and wind components at 1079 stations across China 477 during the winters of 2015-2019. We performed EEOF analysis on $PM_{2.5}$ TFV and TFM, resulting 478 in the spatial structure of PM2.5 transport flux under the temporal disturbances at the synoptic scale, 479 and revealing the connection between synoptic-scale disturbances in the EAWM and QWO in 480 regional PM_{2.5} transport in CEC. Our study focuses on the driving effects of synoptic-scale 481 disturbances associated with cold air activity with the anomalous northerly winds in EAWM on 482 QWO of regional PM2.5 transport over CEC, exacerbating PM2.5 pollution in the downwind THB. 20

483 Differently from the studies on stagnant meteorological conditions associated with PM_{2.5}
 484 accumulations (Gao et al., 2020; Wu et al., 2023; Yang et al., 2024b), this study provides new
 485 insights into the understanding of regional PM_{2.5} transport with source-receptor relationship with
 486 the meteorological mechanism in atmospheric environment change.

487 Based on the 5-winter (2015-2019) observations of PM2.5 concentrations and the 488 corresponding meteorological reanalysis data, this study with the climate statistical and diagnostic 489 methods investigates the QWO of regional PM2.5 transport in China with the influence of 490 synoptic-scale disturbance of EAWM circulation, providing a new insight into the understanding 491 of regional air pollutant transport with meteorological drivers in atmospheric environment changes. 492 Besides the EEOF method used in this study, the alternative methods of wavelet analysis, power 493 spectrum analysis, and band-pass filtering could be used in further study. Future studies with utilizing long-term observations of air pollutants and meteorology over CEC could more 494 495 comprehensively understand the variations in the regional transport of particles and the gaseous precursors with their contributions to air pollution, through the integration of artificial intelligence 496 497 and physical-chemical process analyses.

498

499 Data availability. All data used in this paper can be provided upon request from Yongqing Bai500 (2007byq@163.com)

- 501 Author contributions. YB and TZ conceived the study. YB designed the graphics and wrote the
- 502 manuscript with help from TZ, KM, YZ, JX, XS, LS, YY, YZ, WH and JY were involved in the
- 503 scientific discussion. All authors commented on the paper.
- 504 *Competing interests.* The authors declare that they have no conflict of interest.
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