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Long-term $PM_{2.5}$ pollution over China: Identification of $PM_{2.5}$ pollution hotspots and source contributions

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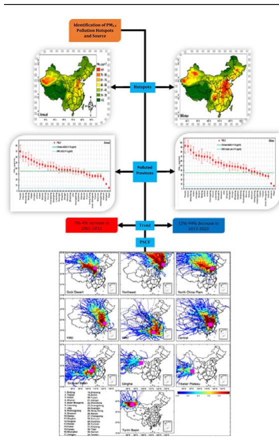
HIGHLIGHTS

- $PM_{2.5}$ was highest in the North China Plain (annual mean > 61 μg/m^3).
- $PM_{2.5}$ in 33 provinces was 1.11 to 13.99 times higher than the WHO AQG (≤ 5 μg/m^3).
- $PM_{2.5}$ increased significantly (3–43 %) from 2001 to 2012.
- $PM_{2.5}$ reduced significantly (12–94 %) from 2013 to 2020.
- China's air quality is mainly affected by local $PM_{2.5}$ sources.

GRAPHICAL ABSTRACT



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ABSTRACT

Fine particulate matter, with an aerodynamic diameter ≤ 2.5 μm ($PM_{2.5}$), is a severe problem in China. The lack of ground-based measurements and its sparse distribution obstruct long-term air pollution impact studies over China. Therefore, the present study used newly updated Global Estimates (V5. GL.02) of monthly $PM_{2.5}$ data from 2001 to 2020 based on Geographically Weighted Regression (GWR) by Washington University. The GWR $PM_{2.5}$ data were validated against ground-based measurements from 2014 to 2020, and the validation results demonstrated a good agreement between GWR and ground-based $PM_{2.5}$ with a higher correlation ($r = 0.95$), lower error (8.14), and lower bias (−3.10 %). The long-term (2001 − 2020) $PM_{2.5}$ data were used to identify pollution hotspots and sources across China using the potential source contribution function (PSCF). The results showed highly significant $PM_{2.5}$ pollution hotspots

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in central (Henan, Hubei), North China Plain (NCP), northwest (Taklimakan), and Sichuan Basin (Chongqing, Sichuan) in China, with the most severe pollution occurring in winter compared to other seasons. During the winter, $PM_{2.5}$ was in the range from 6.08 to 93.05 μg/m^3 in 33 provinces, which is 1.22 to 18.61 times higher than the World Health Organization (WHO) Air Quality Guidelines (AQG-2021; annual mean: 5 μg/m^3). In 26 provinces, the reported $PM_{2.5}$ was 1.07 to 2.66 times higher than the Chinese Ambient Air Quality Standard (AAQS; annual mean: 35 μg/m^3). Furthermore, provincial-level trend analysis shows that in most Chinese provinces, $PM_{2.5}$ increased significantly (3–43 %) from 2001 to 2012, whereas it decreased by 12–94 % from 2013 to 2020 due to the implementation of air pollution control policies. Finally, the PSCF analysis demonstrates that China's air quality is mainly affected by local $PM_{2.5}$ sources rather than by pollutants imported from outside China.

1. Introduction

China has been experiencing severe air pollution since 2000 due to a steep rise in energy consumption due to urbanization and industrial development (Chan and Yao, 2008). Air pollution in China has become a severe threat and challenge, especially for $PM_{2.5,}$ which reached 59 μg/m^3 in 2010 in most Chinese cities (Apte et al., 2015), where >80 % of the people live. The annual $PM_{2.5}$ concentrations in most Chinese cities were about 12 times higher than the WHO AQG (annual average: 5 μg/m^3) (World Health Organization, 2021). Over the years, $PM_{2.5}$ pollution increased significantly in most Chinese cities (Ma et al., 2019). In 2013, the annual average $PM_{2.5}$ concentrations were 106 μg/m^3 in Beijing-Tianjin-Hebei (BTH), 67 μg/m^3 in the Yangtze River Delta (YRD), and 47 μg/m^3 in the Pearl River Delta (PRD) regions (Zheng et al., 2018) and these concentrations were 9 to 21 times higher than the WHO AQG-2021. During a severe haze day in January 2013, hourly $PM_{2.5}$ concentrations in Beijing reached about 600 μg/m^3 (Fu and Chen, 2017). Moreover, the Chinese Ministry of Environmental Protection reported that in most Chinese cities, the $PM_{2.5}$ pollutants were >80.3 % higher than the standards of WHO AQG during heavy or severe pollution days (MEP, 2017). Numerous studies have demonstrated that exposure to elevated $PM_{2.5}$ contributes to several health problems (Lu et al., 2015), including weakened lungs and immune function (Hao et al., 2017), cardiovascular (Dockery, 2001; Guo et al., 2009) and respiratory diseases (Duan et al., 2016; Gordian et al., 1996), asthma (Ding et al., 2017), and increased mortality rate (Cao et al., 2011; Chen et al., 2017a, b; Pope et al., 2002). The impact of outdoor pollution is estimated to cause three million deaths annually. $PM_{2.5}$, in particular, has risen in the world's top five lethal risks. In 2016, $PM_{2.5}$ caused 4.2 million deaths worldwide, including 7.6 % in eastern and southern Asia, especially in China (Cohen et al., 2017; Song et al., 2019). Moreover, $PM_{2.5}$ pollution negatively affects social and economic activity; for example, it can cause haze which may delay or cancel flights and even close transportation services (An et al., 2019; Li and Zhang, 2014). Thus, $PM_{2.5}$ pollution has now become a top concern for the public. To better understand the impacts of $PM_{2.5}$ on human health, there must be a better understanding of its formation, spatiotemporal distribution, and variability (Islam et al., 2023; Li et al., 2014; Liu et al., 2019; Mhawish et al., 2020; Zhang et al., 2013) as well as its source identification.

Since the start of the 21st century, the Chinese government has implemented air pollution control policies to promote the economy and improve air quality. China has implemented the Five-Year Plan (FYP) every five years since 2000 for economic and social development. The country has four recent FYPs: the current (13th) FYP (2016–2020), the 10th FYP (2001–2005), the 11th FYP (2006–2010), and the 12th FYP (2011–2015). The Beijing government substantially measured environmental management and protection for the Olympic games in 2008 (Du and Mendelsohn, 2011) and continued implementing its air pollution control measures (Jin et al., 2016). Since 2013, high-pollution episodes involving $PM_{2.5}$ and the resultant haze in eastern China (BTH, YRD, PRD) have attracted growing attention from the scientific community and government. Consequently, the Chinese government developed an extensive air quality monitoring network, including $PM_{2.5}$ sensors (Fan et al., 2021). $PM_{2.5}$ concentrations have been monitored for the first time in the National Ambient Air Quality Standards (NAAQS: GB 3095–2012). Monitoring and publishing air quality data was launched by the National Ministry of Environmental Protection (MEP) and the National Environmental Monitoring Center (NEMC) in 2013. The public has been able to access these data in real-time since January 2013, and the number of monitoring stations for $PM_{2.5}$ has increased over the years. Scientists and decision-makers can find adequate information using these data by evaluating and analyzing China's spatiotemporal distribution and air quality variations. In 2013, the Chinese national government implemented the five-year "Action Plan on Air Pollution Prevention and Control," with the goal was to make the skies blue again by reducing concentrations of $PM_{2.5}$ and other pollutants (e.g., PM_{10}, NO_2, CO, O_3, and SO_2) from 2013 to 2017 in China (Li et al., 2020). Consequently, various state-specific policies and control measures have led to noticeable PM2.5 prevention and control results, improving air quality. But, China's $PM_{2.5}$ pollution remains exceptionally high (Wang et al., 2022). For example, a recent global study (2000 − 2020) also reported high $PM_{2.5}$ concentrations in 23 cities in China, ranging from 75 μg/m^3 to 100 μg/m^3 (Li et al., 2022a, b), which do not meet the Chinese AAQS and WHO AQG. The national government again launched a new action plan in 2018 (i.e., a three-year Blue-sky Defense Battle Plan: 2018–2020) to reduce total emissions of major air pollutants and increase citizens' quality of life (Wei et al., 2021).

To effectively control and decrease air pollution, it is necessary to understand the spatiotemporal distribution and variations of $PM_{2.5}$ (Hu et al., 2013). Previous studies validated and investigated spatiotemporal distribution and variations of $PM_{2.5}$ over China using reanalysis data from MERRA-2 (Ali et al., 2022; He et al., 2019; Ma et al., 2020) and CAMS (Wu et al., 2020). The results demonstrated a significant difference compared to ground-based $PM_{2.5}$ measurements. Several earlier studies also investigated the spatiotemporal characteristics of $PM_{2.5}$ in different parts of China based on Geographically Weighted Regression (GWR) models (Boys et al., 2014; van Donkelaar et al., 2016), standard deviation ellipse analysis (Peng et al., 2016), meta-analysis (Fontes et al., 2017), spatial auto-correlation method (Xu et al., 2017), and the spatial interpolation method (Wang et al., 2017a, b) derived $PM_{2.5}$ concentrations. Although performed in different parts of China, these studies used old versions and short data periods from 1999 to 2014. As an alternative, newly developed GWR Global Estimates (van Donkelaar et al., 2021) will be used in this study after validation, identifying $PM_{2.5}$ pollution hotspots across China by analyzing the long-term (2001–2020) spatiotemporal distribution, variations, and trends, as well as potential source contribution function (PSCF) analysis is used to identify the potential sources of pollution.

$PM_{2.5}$ can be considered an effective source of information for explaining and assessing emission reduction measures based on spatiotemporal distribution and variations. Long-term $PM_{2.5}$ with a high spatial and temporal resolution is needed to study the physical and chemical processes that affect air quality and the corresponding health risk (Wang et al., 2014b). Most foregoing studies on $PM_{2.5}$ pollution have focused on the spatiotemporal distributions (Seltenrich, 2016) and its chemical composition (Zhang et al., 2016), health effects (Guo et al., 2020), and source identification (Wang et al., 2015; Zhang et al., 2015; Zheng et al., 2005; Zíková et al., 2016). However, these studies mainly focus on specific regions, like the North China Plain (Si et al., 2019), the BTH region (Yan et al., 2018), the YRD region (Yang et al., 2020), Beijing (Yang et al., 2016; Zhang et al., 2020), Lanzhou (Filonchyk and Yan, 2018), and Shanghai (Wang et al., 2016; Wang et al., 2013), with a small data range. Apart from these, few studies examined air quality in the background or rural areas of China, where $PM_{2.5}$ is also elevated (Lai et al., 2016; Yao et al., 2016; Zhao et al., 2009). Some recent studies examined only multi-temporal variations of $PM_{2.5}$ using ground-based $PM_{2.5}$ from 2013 to 2019 (Jiang et al., 2020; Shen et al., 2020; Zhao et al., 2020). These studies did not focus on the long-term (2001–2020) spatiotemporal distributions and variations of $PM_{2.5}$, trends, and potential source identification over the entire China. Therefore, to formulate realistic pollution control goals, quantify the urban and regional contribution to pollution levels, and oversee air quality management, it is necessary to investigate the long-term spatiotemporal distributions and variations of $PM_{2.5}$, including trends and source identification in both urban and rural areas in China. The present study is designed based on the following two main objectives: 1) to validate GWR Global Estimates of $PM_{2.5}$ against ground-based measurements, and 2) to identify the long-term (2001–2020) $PM_{2.5}$ pollution hotspots and sources by analyzing spatiotemporal distribution, variations, and trends, as well as identification of potential sources using PSCF analysis.

2. Data and method

2.1. Ground-based $PM_{2.5}$ measurements in China

China has 22 provinces, including Anhui, Fujian, Gansu, Guangdong, Guizhou, Hainan, Hebei, Heilongjiang, Henan, Hubei, Hunan, Jiangsu, Jiangxi, Jilin, Liaoning, Qinghai, Shaanxi, Shandong, Shanxi, Sichuan, Yunnan, and Zhejiang with four municipalities (Beijing, Chongqing, Shanghai, and Tianjin), five autonomous regions (Guangxi, Inner Mongolia, Ningxia, Xinjiang, and Tibet), and two special administrative regions (Hong Kong and Macao). However, from the point of view of air quality studies, the four municipalities and five autonomous regions are equivalent to provinces; therefore, hereafter, they are all referred to as provinces (33 in total). These provinces belong to seven specific regions such as central (including the provinces of Henan, Hubei, and Hunan), eastern (Anhui, Jiangsu, Jiangxi, Fujian, Shanghai, Shandong, Zhejiang), north (Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia), northeast (Liaoning, Jilin, Heilongjiang), northwestern (Xinjiang, Shaanxi, Gansu, Qinghai, Ningxia), south (Guangdong, Guangxi, Hong Kong, Macau, Hainan), and southwest (Chongqing, Sichuan, Guizhou, Yunnan, Tibet) regions of China (Fig. 1). The country has a total of 1675 sites across 33 provinces that provide hourly $PM_{2.5}$ data, which is obtained from the China National Environmental Monitoring Center (CNEMC: http://www.cnemc.cn/en/;accessed date: 15th January 2021) from 13th May 2014 to 31st December 2020. In this network, PM measurements follow the China Environmental Protection Standards, using a micro oscillating balance and the β absorption method, providing the data with an uncertainty of <5 μg/m^3 (Kong et al., 2021; Miao and Liu, 2019). The monitoring sites for $PM_{2.5}$ are mainly located in the metropolitan areas of China (Fig. 1c).

2.2. GWR global estimates of $PM_{2.5}$

In the present study, newly updated Geographically Weighted Regression (GWR) Global Estimates of monthly $PM_{2.5}$ are used, developed by the atmospheric composition analysis group at Washington University

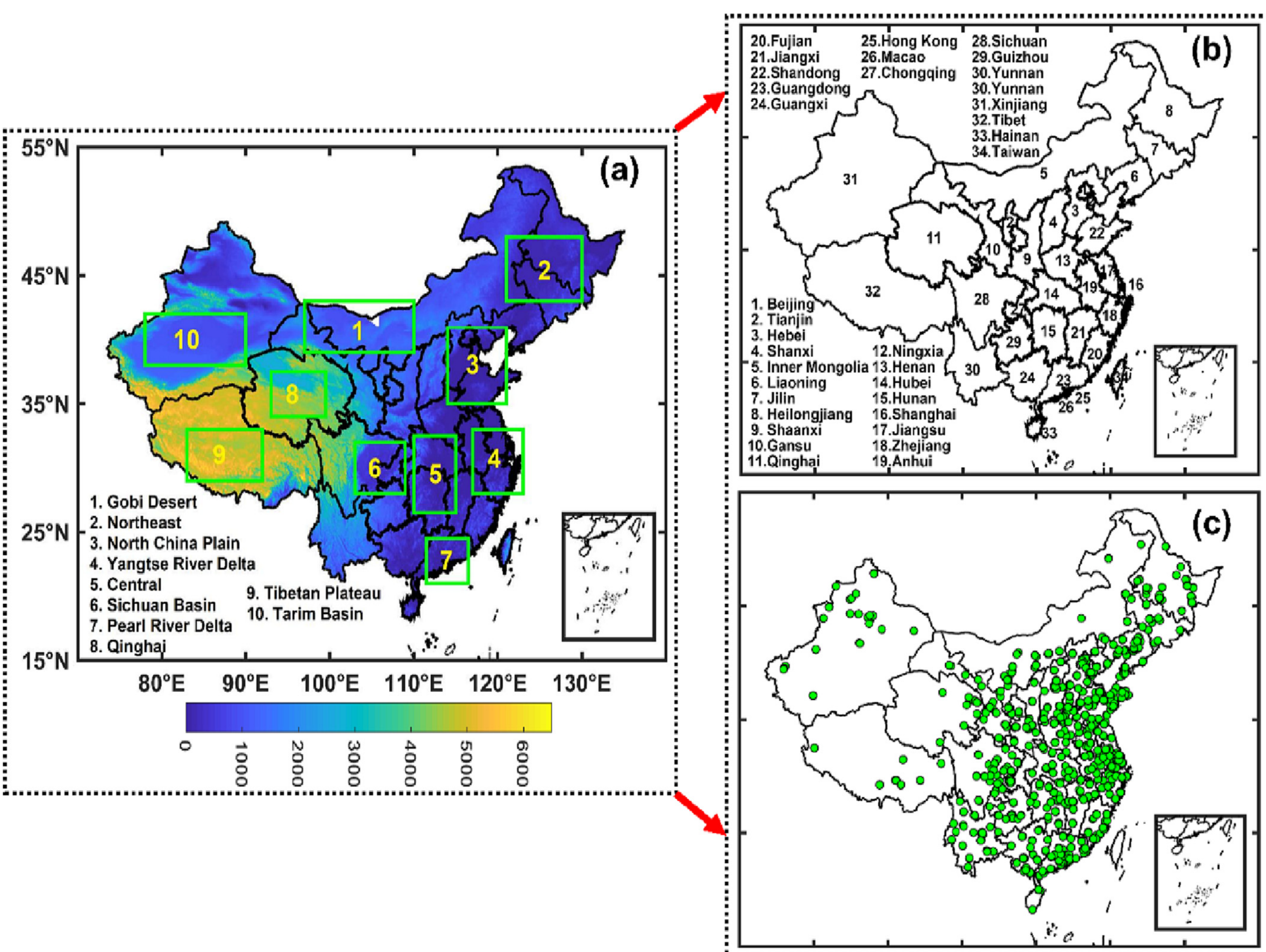

Fig. 1. Geographical maps of China showing (a) elevation and selected study areas (green rectangles) for PSCF analysis, (b) 22 provinces, four municipalities, and five autonomous regions, and (c) in-situ $PM_{2.5}$ monitoring sites (green circles).

(van Donkelaar et al., 2021). This data is created by combining AOD retrievals from multiple sources (i.e., ground measurements, satellite retrievals, GEOS-Chem chemical transport model, meteorological fields, and land data) through the Geographically Weighted Regression (GWR) technique. Data from 1998 to 2020 can be obtained using the link https://sites.wustl.edu/acag/datasets/surface-pm2-5/(last access: 7th May 2022).

2.3. Methodology for analyzing $PM_{2.5}$

The following step-by-step methods were applied in this study:

- For validation, the study extracted point data from monthly GWR Global Estimates of $PM_{2.5}$ using the spatial window of 10 km × 10 km surrounding the ground-based measurement sites. For ground-based $PM_{2.5}$ measurement, a micro-oscillating balance and the absorption method are used, providing data with a level of uncertainty of 5 μg/m^3, according to the China Environmental Protection Standards (Kong et al., 2021; Miao and Liu, 2019). The performance of the GWR Global Estimates dataset was examined using several statistical metrics such as slope and intercept using the Reduced Major Axis (RMA) regression method Pearson's correlation (r), root means squared error (RMSE), and relative mean bias (RMB) (Ali et al., 2021; Ali and Assiri, 2019; Bilal et al., 2022; Bilal et al., 2019; Bilal et al., 2016; Mhawish et al., 2021; Mhawish et al., 2020).

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(GWR\ based\ PM_{2.5} - Ground\ based\ PM_{2.5}\right)^2} \quad (1)$$

$$RMB = \frac{GWR\ based\ PM_{2.5} - Ground\ based\ PM_{2.5}}{Ground\ based\ PM_{2.5}} \times 100 \quad (2)$$

- Long-term annual and seasonal averages of $PM_{2.5}$ (μg/m^3) were calculated over the study domain from 2001 to 2020. In addition, for annual and seasonal analysis, area-averaged $PM_{2.5}$ was extracted for each province using the shapefile.
- The study has used MAC (mean annual cycle) and SSA (annual cycle using singular spectrum analysis) methods to calculate trends on seasonal-adjusted time series (i.e., removed seasonality). The following Eq. (3) is used to calculate the seasonal-adjusted time series (a):

$$A = Y–S \quad (3)$$

Here, Y defines the original time series of $PM_{2.5,}$ and S indicates the seasonal cycle. However, the ordinary least square (OLS) regression method is applied to calculate the slope (α_2) and pivot points from the seasonal-adjusted time series (Eq. (4)).

$$A = \alpha_1 + \alpha_2\ t + \varepsilon_t \quad (4)$$

Here, α_1 indicates the intercept, and ε_t defines the residual error (Verbesselt et al., 2012). More details about pivot point and slope calculation can be found in (Forkel et al., 2013).

- Furthermore, Theil-Sens Slope (Sen, 1968; Theil, 1992) and bootstrapping methods with the Mann-Kendall (MK) test (Kendall, 1975; Mann, 1945) are used to evaluate the robustness of the OLS regression-based trend since the method is sensitive to data outliers. The significance of the $PM_{2.5}$ trend was calculated using a two-tailed

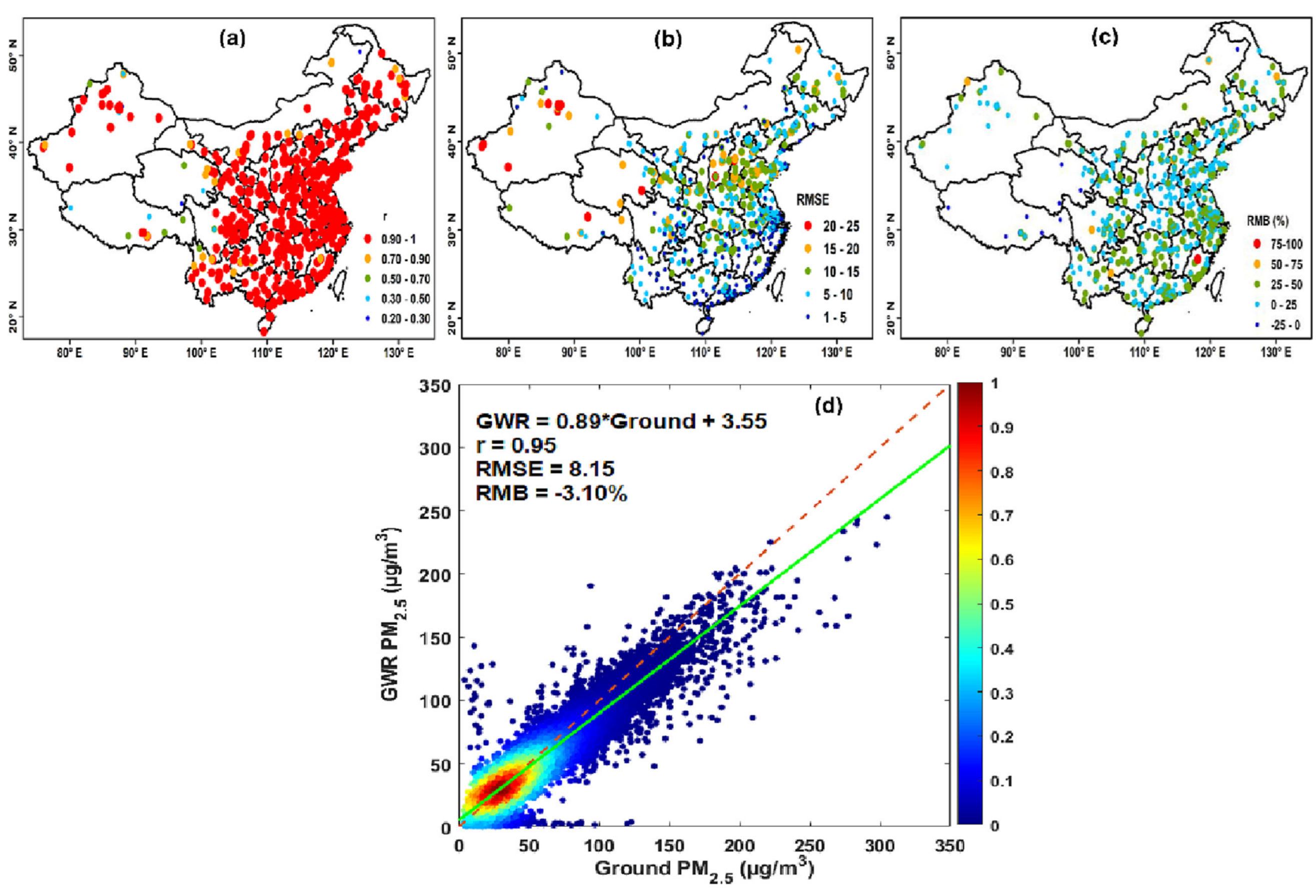

Fig. 2. Validation of GWR Global Estimates of monthly $PM_{2.5}$ (μg/m^3) versus ground-based measurements at 1675 sites across China from 2014 to 2020 for a) spatial map of correlation coefficients (r), b) RMSE, c) RMB, and d) density scatter plot. The data density is shown in color. The red dashed line is the identity line (1:1), and the green solid line is the regression line. Statistical metrics are presented in the legend in the left-top corner. The values of r, RMSE, and RMB are color-coded in the legends.

test at a 95 % confidence level. More details about the methods can be found in (Wang et al., 2021a, 2021b). Due to the variability of $PM_{2.5}$ across China, we calculated the percent change in $PM_{2.5}$ that makes the trend across 33 provinces comparable. The percent change in $PM_{2.5}$ was calculated by Eq. (5), also used in several earlier studies (Mhawish et al., 2021; Sogacheva et al., 2018; Yue and Hashino, 2003).

$$Percent\ Change\ of\ \mathrm{PM}_{2.5}(\%) = \frac{Trend \times total\ numbers\ of\ year}{Mean\ of\ \mathrm{PM}_{2.5}} \times 100 \qquad (5)$$

- To identify the source of air masses, the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model (Stein et al., 2015) from the NOAA (National Oceanic and Atmospheric Administration) (Fleming et al., 2012) was used. The model demonstrates complete chemical transformation, dispersion, and transportation. Combining the NOAA HYSPLIT model with a potential source contribution function (PSCF) was used to identify the potential sources of $PM_{2.5}$ that influence China's air quality. In this study, 72-h HYSPLIT backward trajectories at 500 m above ground level (AGL) were calculated for every hour, at seasonal scales from 2014 to 2020, using the Global Data Assimilation System (GDAS) based meteorological data with a spatial resolution of $1^\circ \times 1^\circ$ (link: ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1; last accessed: 15th July 2021). The 500 m height is very suitable for representing pollution for the mixed layer height (Begum et al., 2005). The PSCF analysis used hourly ground-based $PM_{2.5}$ concentrations over a grid size of 0.5°, for the days that exceeded the Chinese AAQS ($PM_{2.5}$ = 35 μg/m^3). More details about the method can be found in Bilal et al. (2021) and Wang et al. (2021a).

3. Results and discussion

3.1. Accuracy assessment of $PM_{2.5}$

The GWR monthly $PM_{2.5}$ data validation against ground-based measurements at 1675 sites across China from 2014 to 2020 is presented in Fig. 2. Fig. 2c shows that the GWR monthly $PM_{2.5}$ is in good agreement with ground-based measurements, with a correlation (r) of 0.95 and low RMSE (8.14) and RMB (−3.10 %). The correlation coefficient (r) map in Fig. 2a shows that correlations are high (> 0.90) over most areas in China, except for a few sites located in eastern (Fujian), north-northeast (Hebei, Inner Mongolia, and Heilongjiang), northwest (Xinjiang, Qinghai, Gansu), and southwestern (Guizhou, Tibet, Yunnan), where r is lower. Over the areas with high correlation, however, RMB and RMSE are generally also small. At the same time, these metrics are higher over areas with lower correlation (Fig. 2 a-c). This overall good agreement justifies using GWR monthly $PM_{2.5}$ data for identifying the long-term hotspots for $PM_{2.5}$ throughout China.

3.2. Spatial distributions of annual and seasonal $PM_{2.5}$

The long-term spatial distributions of the annually and seasonally averaged $PM_{2.5}$ obtained from GWR Global Estimates over China during the period 2001–2020 are shown in Fig. 3. Generally, the spatial patterns of annually and seasonally averaged $PM_{2.5}$ are consistent, with the highest $PM_{2.5}$ over the largest polluted areas like central China, North China Plain (NCP) in eastern China, north Taklimakan desert in northwestern, and Sichuan Basin in southwest regions of China. However, most regions of China include the central (Henan, Hubei), NCP, northwestern (Xinjiang), and Sichuan Basin. The high $PM_{2.5}$ concentrations in these areas (annual mean > 61 μg/m^3) (Fig. 3) are mainly due to the rapid growth of industrialization and urbanization and dense population with maximum energy consumption (Ali et al., 2021; de Leeuw et al., 2018; He et al., 2021; Ma et al., 2019), together with meteorological conditions such as those conducive to the formation of haze (e.g., Cai et al., 2017) or long-range transport (e.g., Hou et al., 2020). The socioeconomic factors (e.g., economic expansion, end-of-pipe control impulses, energy-climate policies, economical system) (Geng et al., 2021) and meteorological conditions (Chen et al., 2020) significantly affect the spatiotemporal distributions of $PM_{2.5}$ in China. A recent study (Xu et al., 2020) reported that the impacts of meteorological conditions on $PM_{2.5}$ concentrations in southern and southeast coastal regions of China were more noticeable than in areas of central China. According to an earlier study (Cai et al., 2017), unfavorable meteorological conditions and higher anthropogenic emissions (e.g., coal combustion, industrial exhaust, value-added from the heavy pollution industry, and heavy-polluting vehicles) may cause elevated $PM_{2.5}$ over the North China Plain (NCP) (e.g., the BTH, Shandong, Henan). High $PM_{2.5}$ over the NCP topography is further enhanced by frequent air stagnation caused by the interactions between meteorological conditions and terrain (Wang et al., 2018a, b). In contrast, Xinjiang province also experienced significant levels of $PM_{2.5}$ from a natural source, namely the Taklimakan Desert, where dust events are more frequent (Ying et al., 2018; Zhao et al., 2015). Additionally, urban areas in several provinces, including Anhui, Hunan, Jiangsu, parts of Inner Mongolia, Liaoning, Jilin, and Zhejiang were experiencing the annually second-highest $PM_{2.5}$ concentrations (46–60 μg/m^3) (Fig. 3). Lower $PM_{2.5}$ concentrations (<30 μg/m^3) were observed across the Tibetan Plateau, Qinghai, the northeastern part of Inner Mongolia, Heilongjiang, Hainan, Fujian, and Yunnan, mainly due to the lower primary emissions and good atmospheric ventilation (Chen et al., 2019; Yin et al., 2017).

In conjunction with the annual spatial distributions, $PM_{2.5}$ in the highly polluted regions of China was higher during the winter than in spring, autumn, and summer (Fig. 3). In winter, $PM_{2.5}$ concentrations are often high across different regions of China, including the central, eastern, north, northwestern, and southwest because of increased anthropogenic activities and stable atmospheric conditions (shallower boundary layer and stagnant conditions) (Ding et al., 2019; Li et al., 2017a, b). The use of coal as the primary energy source for winter heating has been shown to increase the levels of $PM_{2.5}$ pollution in Chinese cities by 20 %–30 % (Cao et al., 2012; He et al., 2001; Ye, 2003; Zhang et al., 2013). During the El Nino winters (2015, 2018), $PM_{2.5}$ was higher in the Fenwei Plain (e.g., Beijing-Tianjin-Hebei, Shanxi-Shaanxi-Henan) due to unusual southerly winds favoring particle accumulation (Xie et al., 2022). The springtime dust storms in the Taklimakan Desert in the northwest result in higher levels of $PM_{2.5}$ (over 91 μg/m^3) and transportation to other parts of China (Ge et al., 2014; Zhang and Cao, 2015a; Proestakis et al., 2018). A large amount of open biomass burning (e.g., rice or wheat straw field burning) occurs from south to north China in late May and June, contributing to enhanced $PM_{2.5}$ pollution during spring and summer (Chen et al., 2017a, b). In China, plenty of precipitation (i.e., wet deposition) may result in lower levels of $PM_{2.5}$ pollution (Wang et al., 2017a, b) in summer. The burning of corn residue in northern China and rice straw in south China occurs in October, contributing to high autumn $PM_{2.5}$ pollution (Chen et al., 2017a, b; Zhang and Cao, 2015b). The largest city in the PRD region, Guangzhou, reports up to 19 % of its total $PM_{2.5}$ emissions are from biomass burning (Wang et al., 2007).

3.3. Annual and seasonal variations in provincial level $PM_{2.5}$

This section presents the 20-year annual and seasonal average $PM_{2.5}$ variations over 33 Chinese provinces in Fig. 4 and S2. In 33 Chinese provinces (Fig. 4), the annual $PM_{2.5}$ concentrations averaged over 2001–2020 varied from 5.57 μg/m^3 to 69.97 μg/m^3, with the Tianjin province identified as the most polluted and Tibet as relatively clean (Fig. 4). $PM_{2.5}$ in 33 provinces is 1.11 to 13.99 times higher than the WHO AQG (≤ 5 μg/m^3, annual mean), indicating polluted air that is hazardous to human health (World Health Organization, 2021). Of these 33 provinces, in 22 provinces (e.g., Tianjin, Henan, Shandong, Beijing, Hebei, Jiangsu, Anhui, Hubei, Shanxi, Xinjiang, Hunan, Chongqing, Shanghai, Liaoning, Guangxi, Ningxia, Jiangxi, Shaanxi, Zhejiang, Jilin, Guangdong) $PM_{2.5}$ is 1.01 to 2.00 times higher than the Chinese AAQS for Grade-II (GB 3095–2012; ≤ 35 μg/m^3 annual mean) (Li et al., 2017a, b), signifying

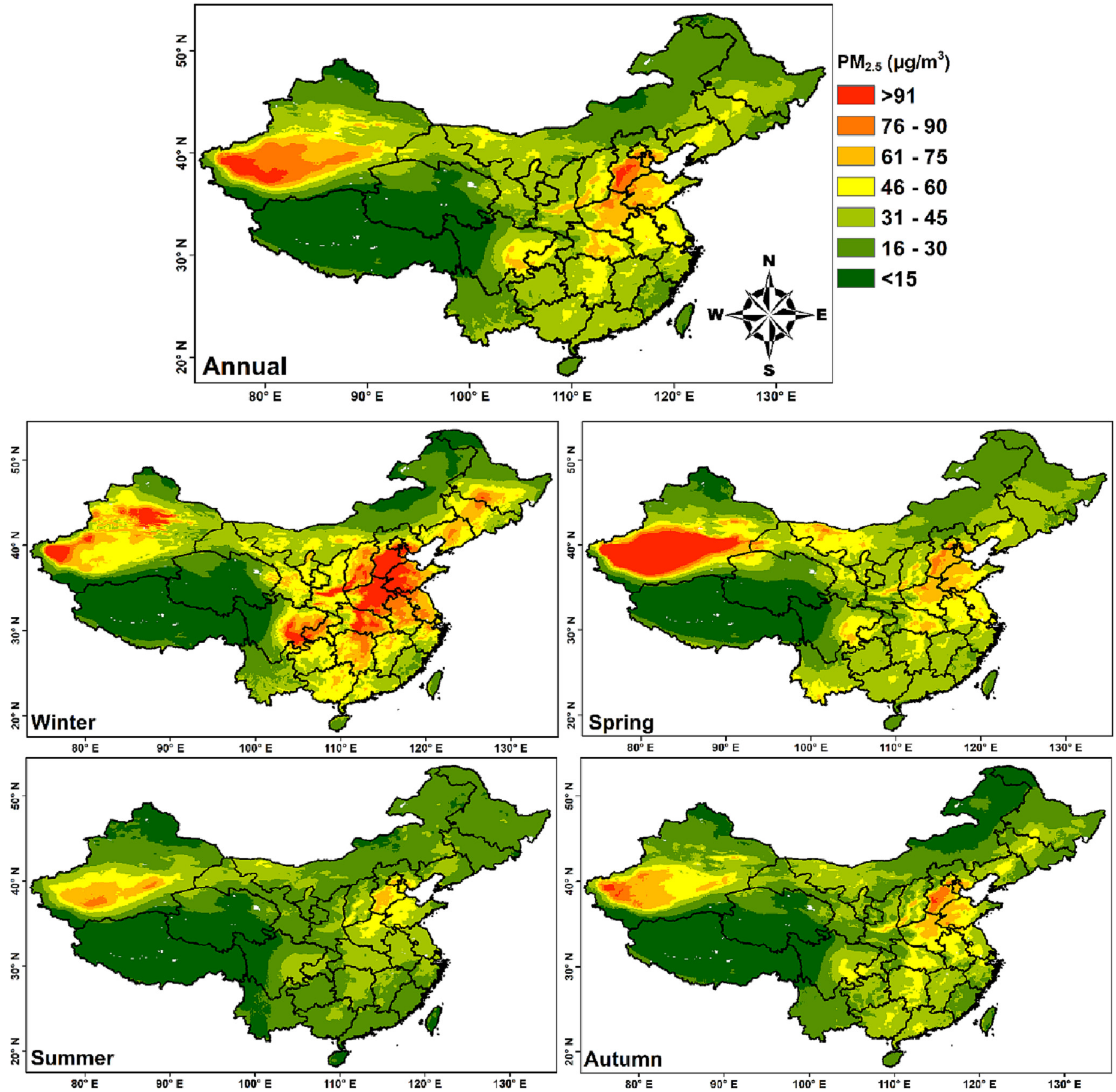

Fig. 3. Spatial distributions of annual and seasonal mean $PM_{2.5}$ (μg/m^3) concentrations were obtained from GWR Global Estimates over China from 2001 to 2020. The figures show the average over these years.

these provinces as polluted (Fig. 4). An earlier study (Pope et al., 2002) reported that mortality, lung cancer, and cardiopulmonary diseases could rise by 8 %, 6 %, and 4 %, respectively, due to the long-term exposure to $PM_{2.5}$. Over 30.8 million adult premature deaths were attributed to long-term exposure to $PM_{2.5}$ in China from 2000 to 2016 (Liang et al., 2020). Across China, about 1.55 million premature deaths were accounted for in 2016, mainly due to long-term exposure to $PM_{2.5}$ (Zheng et al., 2021). Cao et al. (2018) also reported that long-term exposure to $PM_{2.5}$ (41.36 μg/m^3 in 2008) is associated with 0.53 million lung cancer deaths in China. A recent study from northern China reported that long-term exposure to $PM_{2.5}$ increased stroke mortality (Yang et al., 2021). Apart from these, Chen et al. (2022) reported that long-term exposure to $PM_{2.5}$ (e.g., 13.2 μg/m^3 to 72.1 μg/m^3) in China is linked to higher obesity risk and abdominal obesity.

China's AAQS shows a large number of polluted provinces by season, with the majority being polluted in winter (26 provinces out of 33), followed by spring (22), autumn (18), and summer (7) (Fig. 4). The 20-year averaged wintertime $PM_{2.5}$ concentrations vary from 6.08 μg/m^3 to 93.05 μg/m^3 across 33 provinces, with the Tianjin province identified as the most polluted and Tibet as relatively clean (Fig. 4). In 26 of these 33 provinces (including Tianjin, Henan, Shandong, Hebei, Jiangsu, Shanxi, Beijing, Anhui, Hubei, Chongqing, Hunan, Liaoning, Shanghai, Shaanxi, Jilin, Ningxia, Guangxi, Jiangxi, Zhejiang, Guizhou, Xinjiang, Guangdong, Macao, Gansu, Sichuan, Hong Kong) $PM_{2.5}$ was 1.07 to 2.66 times higher than the Chinese AAQS (Fig. 4). In spring, $PM_{2.5}$ ranged from 7.48 μg/m^3 to 64.96 μg/m^3 across 33 provinces and in 22 of them (including Tianjin, Xinjiang, Shandong, Henan, Beijing, Jiangsu, Hebei, Anhui, Shanghai, Hubei, Ningxia, Liaoning, Shanxi, Hunan, Chongqing, Gansu, Guangxi, Zhejiang, Jiangxi, Yunnan, Shaanxi, Jilin) $PM_{2.5}$ was 1.01 to 1.86 times higher than the Chinese AAQS (Fig. 4). $PM_{2.5}$ was lowest in the summer and ranged from 4 μg/m^3 to 56.15 μg/m^3 across 33 provinces (Fig. 4).

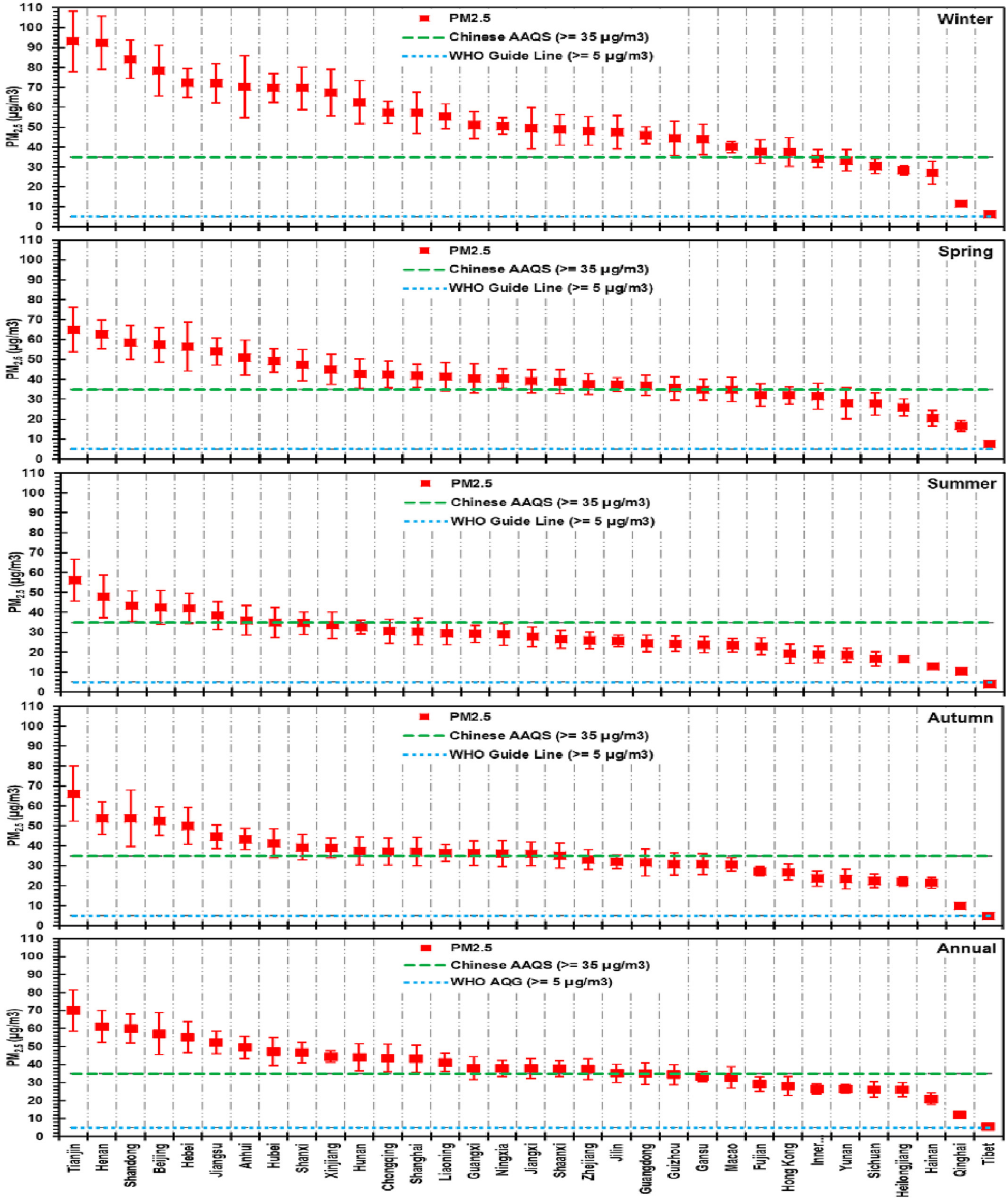

Fig. 4. Variation per province in annual and seasonal mean $PM_{2.5}$ ($\mu g/m^3$) obtained from GWR Global Estimates over China and averaged from 2001 to 2020. The Green dashed line = the Chinese Ambient Air Quality Standard (AAQS), and the cyan dashed line = the WHO Air Quality Guideline (AQG). The error bar indicates the standard deviation of $PM_{2.5}$.

This result is in line with a few earlier studies on China (Wang et al., 2017a, b; Yan et al., 2018; Zhang and Cao, 2015a). The summertime $PM_{2.5}$ from 26 provinces met the Chinese AAQS. In comparison, in 7 provinces (including Tianjin, Beijing, Shandong, Henan, Hebei, Jiangsu, and Anhui), $PM_{2.5}$ was 1.01 to 1.60 times higher than the Chinese AAQS (Fig. 4). In summer, Zhang et al. (2013) estimated that motor vehicles contribute 63 % to the total carbonaceous fraction of the $PM_{2.5}$ mass in Beijing and 30.3 % is contributed by coal combustion. In autumn, $PM_{2.5}$ was found between 4.75 μg/m^3 to 66.06 μg/m^3 across 33 provinces (Fig. 4). In 18 provinces (including Tianjin, Shandong, Beijing, Henan, Hebei, Jiangsu, Anhui, Hunan, Hubei, Shanxi, Guangxi, Guangdong, Macao, Xinjiang, Jiangxi, Chongqing, Liaoning, Shanghai) the $PM_{2.5}$ concentrations in the autumn are 1.01 to 1.89 times higher than the Chinese AAQS (Fig. 4). With respect to the WHO AQG, $PM_{2.5}$ in 33 provinces is higher than the standard by 1.22 to 18.61 times in winter and by 1.50 to 12.99 times in spring (Fig. 4). Moreover, $PM_{2.5}$ in 32 provinces is 2.11 to 11.23 times higher in summer and 1.99 to 13.21 times higher in autumn. Only Tibet is relatively clean in the summer and autumn seasons.

3.4. $PM_{2.5}$ trend

To better understand the variability in $PM_{2.5}$ across 33 provinces in China, we calculate the $PM_{2.5}$ trend using the ordinarily least square (OLS) regression method, and the significance of the trend is determined using the Man-Kendall trend test applied to seasonally-adjusted time series. $PM_{2.5}$ trend analysis was performed by segmenting the data into two categories: 1st segment (before the pivot point) and 2nd segment (after the pivot point), as shown in Table 1. The year 2014 was calculated as a pivot point for Anhui, Guangdong, Hong Kong, Jiangsu, Jiangxi, and Tianjin, where $PM_{2.5}$ ranged from 27.39 μg/m^3 to 74.85 μg/m^3 during the 1st segment (2001–2014) and progressively declined during the 2nd segment (2015–2020: 21.39–58.57 μg/m^3) (Table 1). During the 1st segment, the significant ($p < 0.05$) highest increase in $PM_{2.5}$ was in Jiangsu (39 %), followed by Tianjin (32 %), Guangdong (27 %), Hong Kong, Anhui (19 %), and Jiangxi (17 %). During the 2nd segment, the significant largest decrease in $PM_{2.5}$ was in Tianjin (−52 %), followed by Hong Kong (−43 %), Jiangxi (−41 %), Jiangsu (−38 %), and Anhui (−35 %) (Table 1). In Beijing, Hebei, and Shanghai, the pivot point occurred in 2012; $PM_{2.5}$ ranged from 44.46 μg/m^3 to 61.52 μg/m^3 during the 1st segment (2001 − 2012) and gradually decreased during the 2nd segment (2013–2020: 41.37–51.78 μg/m^3) (Table 1). As shown in the 1st segment, the significant highest increase in $PM_{2.5}$ was in Beijing (30 %) and Hebei (24 %). However, for Shanghai, the trend was a statistically insignificant negative trend (−4 %). During the 2nd segment, the significant largest decrease in $PM_{2.5}$ was in Beijing (−94 %), followed by Hebei (−72 %) and Shanghai (−69 %). In Chongqing, Hubei, Shaanxi, and Shanxi, the pivot point was identified in 2010. In these provinces, $PM_{2.5}$ ranged from 38.66 μg/m^3 to 49.11 μg/m^3 during the 1st segment (2001 − 2010), and these concentrations decreased during the 2nd segment (2011–2020: 36.64–45.76 μg/m^3) (Table 1). During the 1st segment, the significant highest increase in $PM_{2.5}$ was in Hubei (32 %), followed by Hebei (24 %), Chongqing (28 %), Shaanxi (16 %), and Shanxi (7 %). In the 2nd segment, the significant largest decrease in $PM_{2.5}$ occurred in Chongqing (−73 %), followed by Hebei (−69 %), Shaanxi (−49 %), and Shanxi (−48 %). In Fujian province, the pivot point occurred in 2007; here, $PM_{2.5}$ was 30.86 μg/m^3 during the 1st segment (2001–2007) and decreased during the 2nd segment (2008–2020: 28.11 μg/m^3) (Table 1). However, a 23 % significant increase in $PM_{2.5}$ was noticed in Fujian during the 1st segment, while a 50 % decrease during the 2nd segment. In Gansu, Heilongjiang, Henan, Hunan, and Zhejiang provinces, the pivot point was identified in 2013, where $PM_{2.5}$ ranged from 25.82 μg/m^3 to 64.94 μg/m^3 during the 1st segment (2001 − 2013) and these concentrations were decreased during the 2nd segment (2014–2020: 26.67–54.07 μg/m^3) (Table 1). During the 1st segment, Hunan (43 %) showed the significant highest increase in $PM_{2.5}$, compared to Henan (33 %), Heilongjiang (12 %), Gansu (10 %), and Zhejiang (5 %), respectively. In the 2nd segment, Zhejiang (−72 %)

Table 1
Estimation of Provincial trends of $PM_{2.5}$ (μg/m^3) using the OLS regression method with the Mann-Kendall test using monthly data over China from 2001 to 2020. Here, the 1st segment = before the pivot point, and the 2nd segment = after the pivot point. Trend values in Bold = significant (at the 95 % confident interval) and non-bold = insignificant trend.

Province	pivot point	Mean $PM_{2.5}$ (1st segment)	Trend (1st Segment) (% change)	Mean $PM_{2.5}$ (2nd segment)	Trend (2nd segment) (% change)
Anhui	2014	52.46	**0.72 (19 %)**	43.02	**−2.54 (−35 %)**
Beijing	2012	61.52	**1.55 (30 %)**	50.48	**−5.94 (−94 %)**
Chongqing	2010	46.55	**1.31 (28 %)**	40.77	**−2.99 (−73 %)**
Fujian	2007	30.86	**1.02 (23 %)**	28.11	**−1.08 (−50 %)**
Gansu	2013	34.42	**0.27 (10 %)**	31.22	**−1.66 (−37 %)**
Guangdong	2014	38.14	**0.74 (27 %)**	27.94	**−2.06 (−44 %)**
Guangxi	2015	40.83	**0.91 (33 %)**	29.72	**−1.90 (−32 %)**
Guizhou	2011	36.04	**1.00 (30 %)**	32.43	**−2.49 (−69 %)**
Hainan	2008	22.63	**1.15 (41 %)**	19.99	**−0.65 (−39 %)**
Hebei	2012	57.51	**1.17 (24 %)**	51.78	**−4.69 (−72 %)**
Heilongjiang	2013	25.82	**0.23 (12 %)**	26.67	**−2.06 (−54 %)**
Henan	2013	64.94	**1.63 (33 %)**	54.07	**−3.25 (−42 %)**
Hong Kong	2014	30.98	**0.43 (19 %)**	21.60	**−1.56 (−43 %)**
Hubei	2010	48.67	**1.55 (32 %)**	45.76	**−3.18 (−69 %)**
Hunan	2013	47.01	**1.54 (43 %)**	38.41	**−3.07 (−56 %)**
Jiangsu	2014	27.39	**0.77 (39 %)**	46.34	**−2.96 (−38 %)**
Jiangxi	2014	54.83	**0.68 (17 %)**	31.44	**−2.15 (−41 %)**
Jilin	2015	36.87	**0.72 (29 %)**	30.20	**−2.00 (−33 %)**
Liaoning	2015	43.27	**0.48 (17 %)**	35.18	**−1.56 (−22 %)**
Macao	2008	35.97	**1.57 (35 %)**	30.80	**−1.49 (−58 %)**
Inner-Mongolia	2017	27.25	0.04 (3 %)	22.03	**−0.89 (−12 %)**
Ningxia	2005	38.22	**−1.21 (−16 %)**	37.83	**−0.99 (−39 %)**
Qinghai	No break	12.21	**−0.05 (−9 %)**		
Shaanxi	2010	38.66	**0.61 (17 %)**	36.64	**−1.80 (−49 %)**
Shandong	2006	58.56	**3.69 (38 %)**	60.56	**−1.67 (−39 %)**
Shanghai	2012	44.46	−0.14 (−4 %)	41.37	**−3.58 (−69 %)**
Shanxi	2010	49.11	**0.35 (7 %)**	44.41	**−2.14 (−48 %)**
Sichuan	2009	27.93	**0.90 (29 %)**	24.67	**−1.42 (−63 %)**
Tianjin	2014	74.85	**1.71 (32 %)**	58.57	**−5.07 (−52 %)**
Xinjiang	No break	44.39	**0.16 (7 %)**		
Tibet	2011	5.80	**−0.20 (−39 %)**	5.29	**−0.16 (−27 %)**
Yunnan	2006	26.49	**1.26 (29 %)**	26.44	**−0.61 (−32 %)**
Zhejiang	2013	39.95	**0.16 (5 %)**	32.64	**−3.35 (−72 %)**

showed the significant largest decrease in $PM_{2.5}$, compared to Hunan (−56 %), Heilongjiang (−54 %), Henan (−42 %), and Gansu (−37 %), respectively. In Guangxi, Jilin, and Liaoning provinces, the pivot point was identified in 2015, where $PM_{2.5}$ ranged from 21.24 μg/m^3 to 43.27 μg/m^3 during the 1st segment (2001–2015), and these concentrations were decreased during the 2nd segment (2016–2020: 17.05–35.18 μg/m^3) (Table 1). During the 1st segment, the significant highest increase in $PM_{2.5}$ occurred in Guangxi (33 %), followed by Jilin (29 %) and Liaoning (17 %). In the 2nd segment, the significant largest decrease in $PM_{2.5}$ occurred in Jilin (−33 %), followed by Guangxi (−32 %) and Liaoning (−22 %). In Guizhou and Tibet, the pivot point occurred in 2011, and in these provinces, $PM_{2.5}$ ranged from 5.80 μg/m^3 to 36.04 μg/m^3 during the 1st segment (2001−2011) and decreased during the 2nd segment (2012–2020: 5.29–32.43 μg/m^3). There was a greater decrease in $PM_{2.5}$ in Tibet during the 1st segment (−39 %) than in the 2nd segment (−27 %), while in Guizhou, an increase of 30 % occurred during the 1st segment, followed by a 69 % decrease during the 2nd segment. In Hainan and Macao, the pivot point occurred in 2008; $PM_{2.5}$ increased from 22.63 μg/m^3 to 35.97 μg/m^3 during the 1st segment (2001–2008) and decreased during the 2nd segment (2009–2020: 19.99–30.80 μg/m^3). In Hainan, $PM_{2.5}$ increased by 41 % during the 1st segment and by 35 % in Macao, while during the 2nd segment, it decreased by 58 % in Macao and 39 % in Hainan. In Inner Mongolia, the pivot point occurred in 2017; here, $PM_{2.5}$ was 27.25 μg/m^3 during the 1st segment (2001–2017) and decreased during the 2nd segment (2018–2020: 22.03 μg/m^3) (Table 1). In Inner Mongolia, during the 1st segment, an insignificant increase in $PM_{2.5}$ was noticed (3 %), while during the 2nd segment,

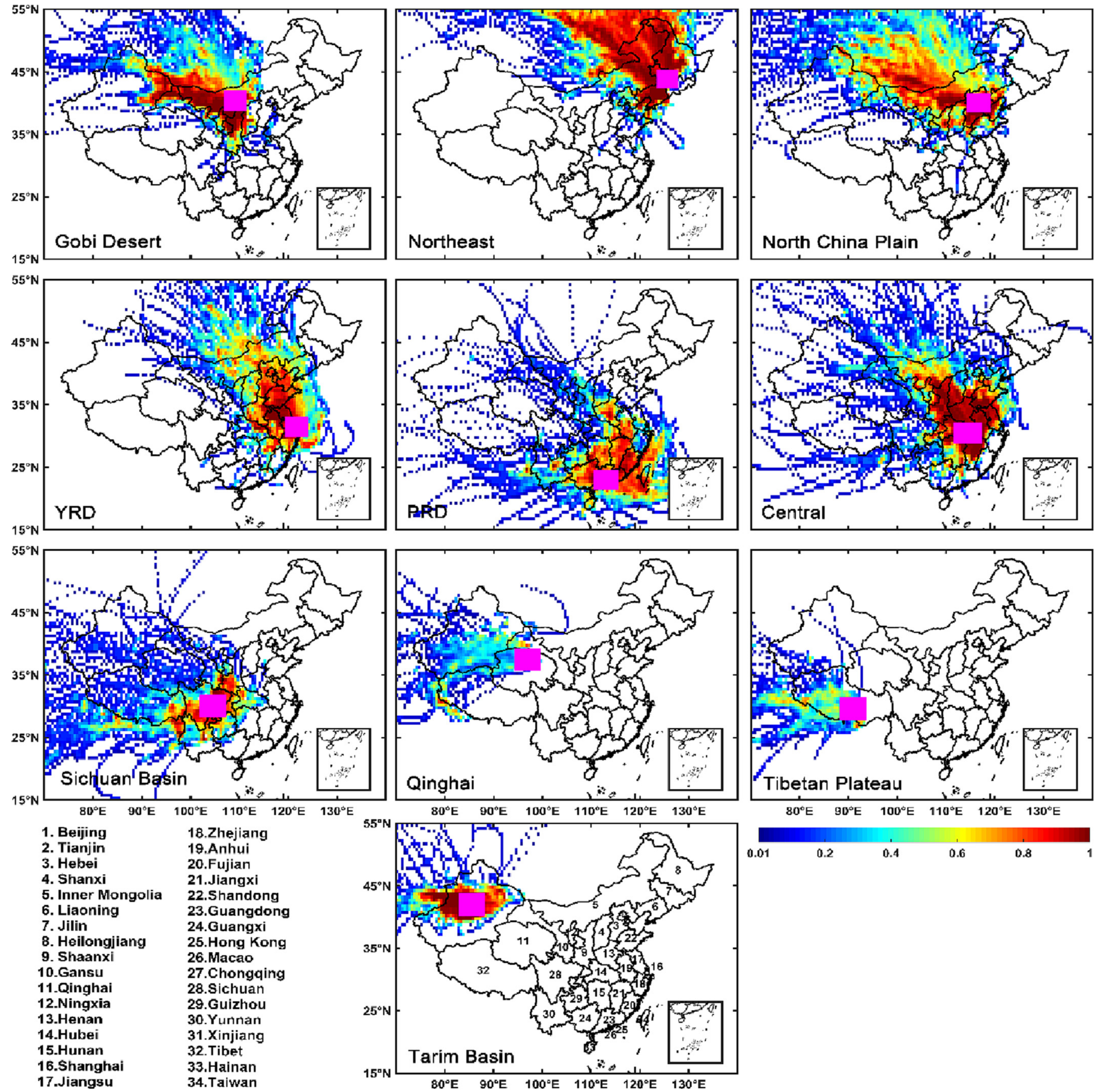

Fig. 5. Source identification for $PM_{2.5}$ over 10 selected study regions in China using PSCF analysis during the winter season from 2014 to 2020. Source regions are identified by province or autonomous area, indicated in the Tarim Basin's bottom figure. The pink-color box inside the figure indicates the 10 receptor sites.

$PM_{2.5}$ decreased by 12 %. In Ningxia, the pivot point occurred in 2005; during the 1st segment (2001–2005), $PM_{2.5}$ was 38.22 μg/m^3 and slightly dropped during the 2nd segment (2006–2020: 37.83 μg/m^3). The 2nd segment saw a significant decrease in $PM_{2.5}$ (−39 %) than the 1st segment (−16 %). No pivot point was detected in the PM2.5 time series in Qinghai and Xinjiang. From 2001 to 2020, $PM_{2.5}$ decreased by −9 % in Qinghai and increased by 7 % in Xinjiang. In Shandong and Yunnan, the pivot point occurred in 2006; during the 1st segment, $PM_{2.5}$ significantly increased by 38 % in Shandong and by 29 % in Yunnan, while during the 2nd segment, $PM_{2.5}$ decreased by 39 % in Shandong and by 32 % in Yunnan. In Sichuan, the pivot point occurred in 2009; during the 1st segment (2001–2009), $PM_{2.5}$ significantly increased by 29 %, while it decreased by 63 % during the 2nd segment.

Theil-Sen's Slope and bootstrapping methods were applied to the same $PM_{2.5}$ time series to test the results' robustness using the OLS regression. The results are presented in Table S1. It is worth mentioning that the trends estimated using all two methods are comparable regarding significance and magnitude. However, several factors could contribute to the increase or decrease of $PM_{2.5}$ concentrations across China from 2001 to 2020. Most Chinese provinces showed an increase in $PM_{2.5}$ from 2001 to 2012, during which three FYPs were implemented (10th, 11th, and 12th FYP periods: 2001–2005; 2006–2010; 2011–2015). However, although China's economy and industrial development were rapidly growing, adequate controls on $PM_{2.5}$ were not included in the 10th and 11th FYPs (Li et al., 2022a, b; Wang et al., 2017a, b). For example, the national government targeted the reduction of SO_2 by 10 %, with industrial dust with soot (Jiang, 2014;

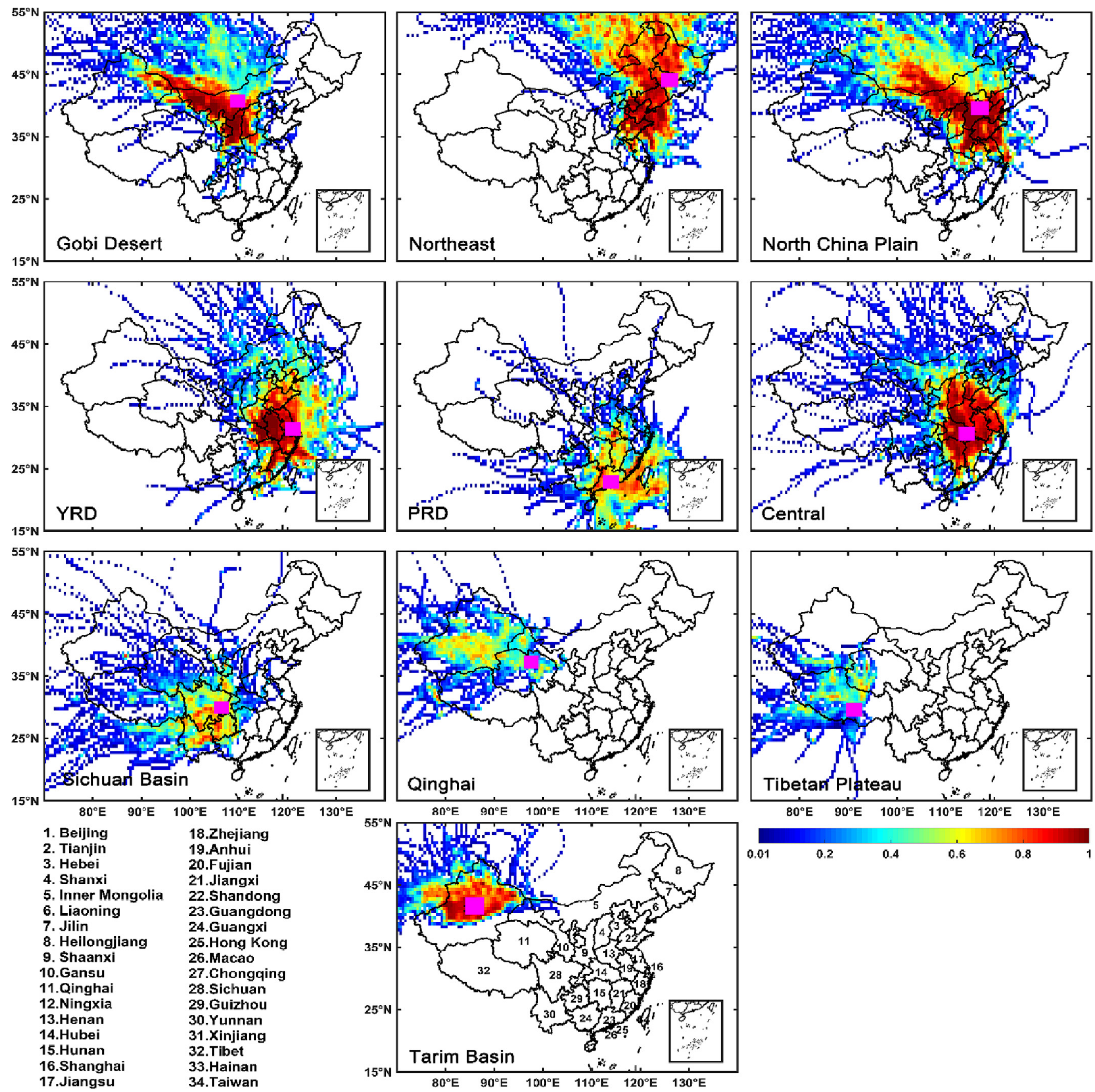

Fig. 6. Source identification for $PM_{2.5}$ over 10 major hotspot areas in China using PSCF analysis during the spring season from 2014 to 2020.

Schreifels et al., 2012). It is important to mention that the 12th FYP policy did include the reduction of $PM_{2.5}$ by 5 % over key regions (Jingjinji, YRD, and PRD), but the reduction of $PM_{2.5}$ was not noticeable in 2011 and 2012 (Wang et al., 2017a, b). Consequently, a significant increase in $PM_{2.5}$ was evident across China, as is also apparent in our study (see Table 1 and Table S1). Several earlier studies reported that emissions grew rapidly from 1986 to 2006, which accelerated the buildup of $PM_{2.5}$ across China, as evidenced by visibility data (Che et al., 2007; Fu et al., 2014; Han et al., 2016; Wang and Chen, 2016) and satellite observations of AOD (e.g., de Leeuw et al., 2018; de Leeuw et al., 2021; and references cited therein). Furthermore, transportation, emissions, and chemical production may also increase $PM_{2.5}$ in China due to meteorological factors (wind speed, relative humidity, temperature) (Leung et al., 2018; Wang et al., 2014a). In contrast, $PM_{2.5}$ concentrations were substantially reduced in different parts of China during the period from 2013 to 2020 (see Table 1 and Table S1), mainly because of the implementation of three policies: Action Plan of Air Pollution Prevention and Control (APPC-AP: 2013–2017), the 13th FYP (2016–2020), and a three-year Blue Sky Defense Battle Plan (2018–2020) (Li et al., 2022a, b; Wang et al., 2018a, b; Zheng et al., 2018). China's five largest regions (including BTH, Sichuan basin, PRD, and Fenwei Plain: Xian) experienced significant declines in $PM_{2.5}$ (34–49 %) between 2013 and 2018 due to controls of anthropogenic emissions under the influence of meteorology (Zhai et al., 2019).

3.5. Source contribution using PSCF analysis

We used PSCF analysis, based on 72-h back trajectories obtained from the HYSPLIT model and ground-based measurements, to identify the

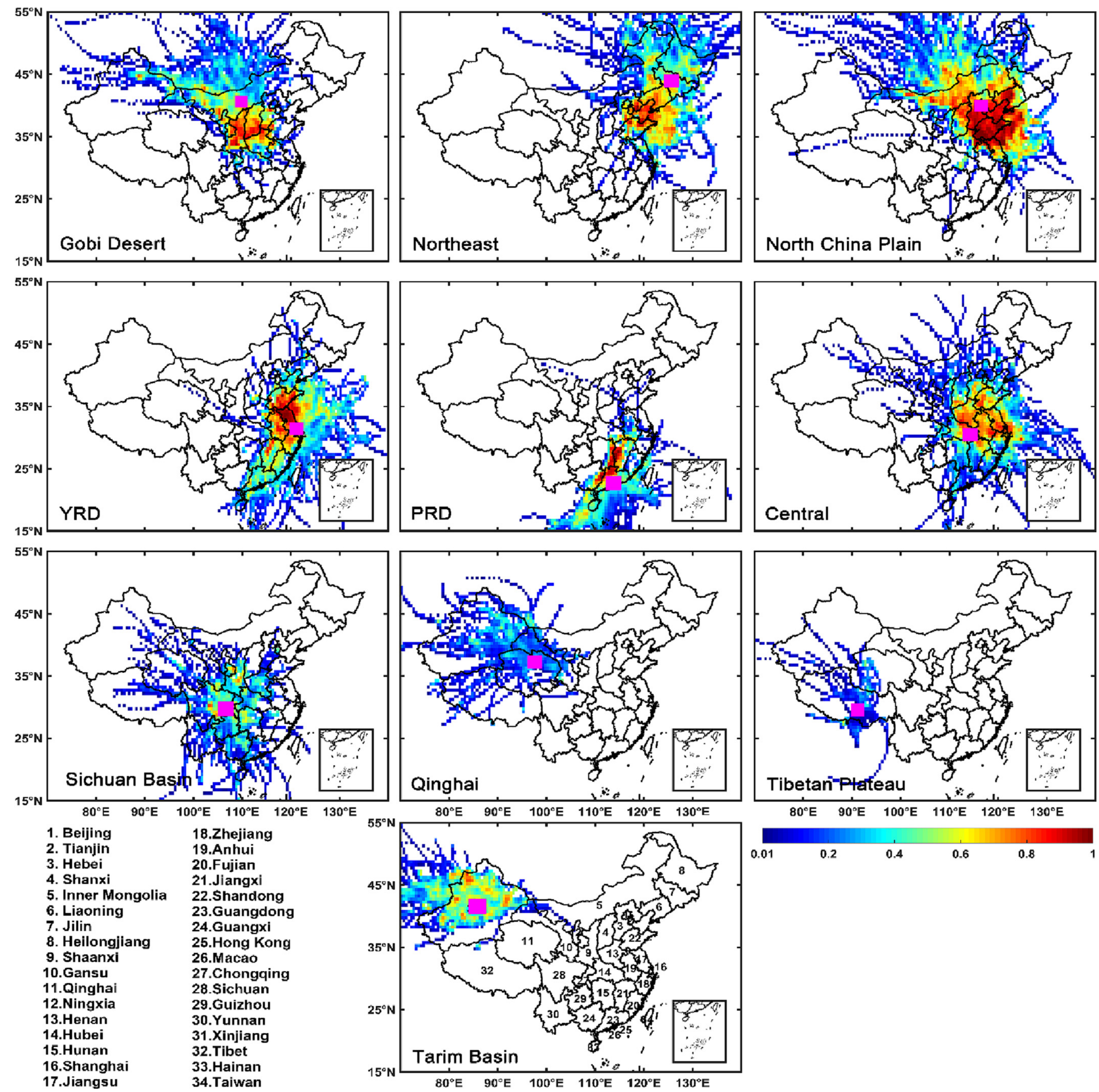

Fig. 7. Source identification for $PM_{2.5}$ over 10 major hotspot areas in China using PSCF analysis during the summer season from 2014 to 2020.

potential source areas of $PM_{2.5}$ for the 10-receptor areas indicated with the pink-color box in Fig. 5: the Gobi Desert, Northeast, NCP, YRD, PRD, Central, Sichuan Basin, Qinghai, Tibetan Plateau, and Tarim Basin, at seasonal timescales from 2014 to 2020 (Figs. 5–8). To this end, 72-h HYSPLIT backward trajectories were computed for each area, arriving every hour at the height of 500 m (AGL). The results from the PSCF analysis in Figs. 5–8 show substantial differences between areas and seasons. Air quality in the Gobi Desert is affected more by local sources (Xinjiang and Gansu, Inner Mongolia, Ningxia, Shaanxi, and Shanxi) than by pollutants transported from abroad (Mongolia and Russia), which were stronger in winter than in spring, summer, or autumn. Additionally, like the Gobi Desert, the pollutants from local sources (Hebei, Heilongjiang, Inner Mongolia, Liaoning, and Jilin) significantly affect the air quality of Northeast China more than pollutants transported from abroad (Northeast of Mongolia and Russia), which typically more intense in winter than in spring, autumn, and summer. In the North China Plain, the air quality is more seriously affected by local sources (Anhui, Beijing, Hebei, Henan, Hubei, Inner Mongolia, Jiangsu, Liaoning, Shandong, Shanxi, Shaanxi, Tianjin, and Xinjiang) rather than imported from abroad (Southern Mongolia and Russia), which were typically more intense in winter than in autumn, spring, and summer. In the YRD, the air quality is much more affected by contributions from local sources (Anhui, Beijing, Hebei, Henan, Hubei, Inner Mongolia, Jiangsu, Jiangxi, Liaoning, Shandong, Shanxi, Shaanxi, Tianjin, and Zhejiang) than from sources outside China (Southern Mongolia), which

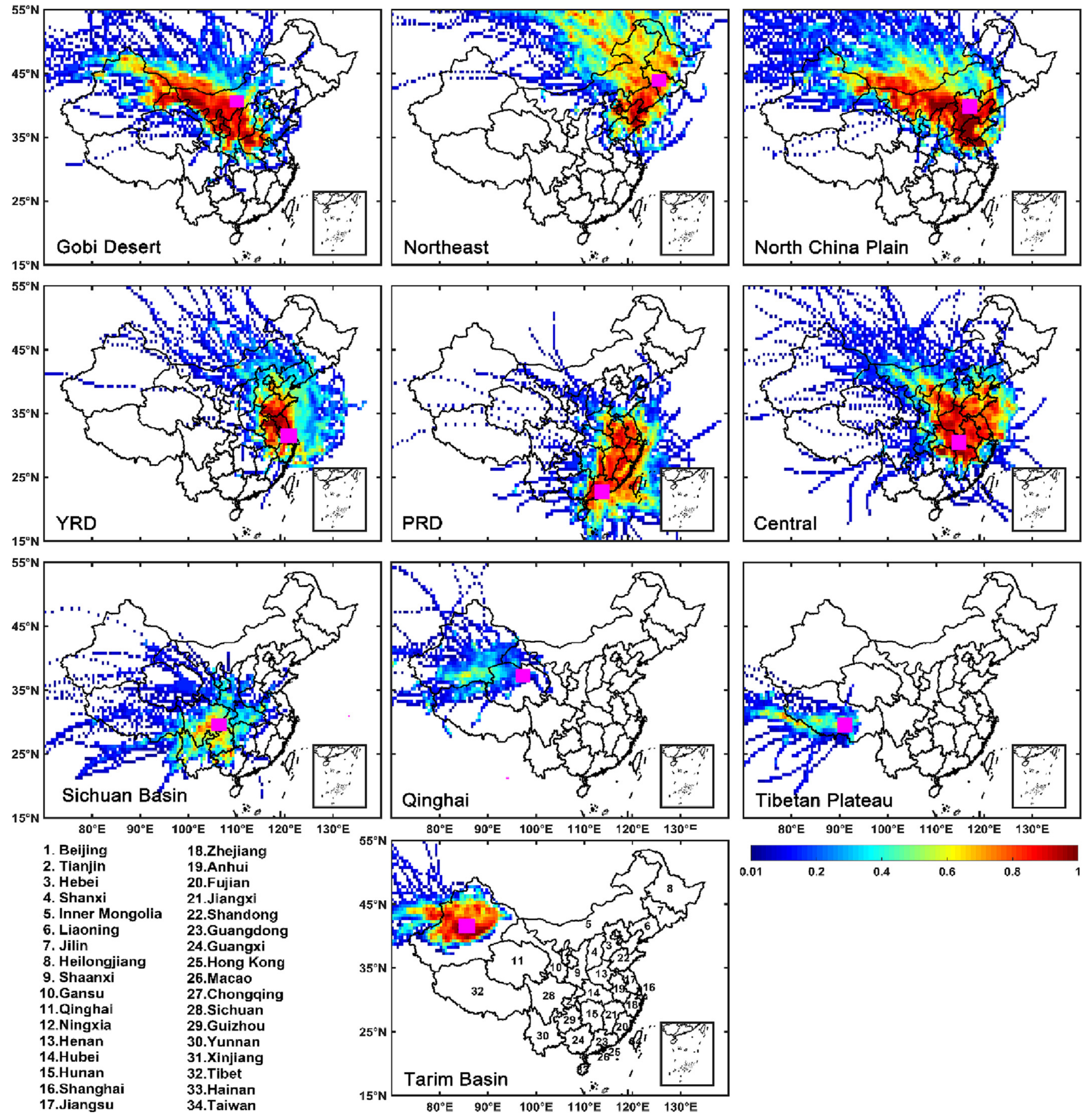

Fig. 8. Source identification for $PM_{2.5}$ over 10 major hotspot areas in China using PSCF analysis during the autumn season from 2014 to 2020.

was stronger in winter than in autumn, spring, and summer. In Central China, the air quality is also much more affected by the contributions from local sources (Anhui, Beijing, Chongqing, Gansu, Guangdong, Guangxi, Hebei, Henan, Hubei, Hunan, Inner Mongolia, Jiangsu, Jiangxi, Ningxia, Shanxi, Shaanxi, Sichuan Basin, Shandong, and Tianjin) than from sources outside China (Southern Mongolia and Burma), which was stronger in winter than in spring, autumn, and summer. In Central China, the air quality of the Sichuan Basin is also significantly affected by local sources (Chongqing, Gansu, Guangxi, Guizhou, Henan, Hubei, Hunan, Inner Mongolia, Ningxia, Qinghai, Shaanxi, Tibetan Plateau, and Yunnan) than from sources outside China (Burma and India), which was stronger in winter than in spring, autumn, and summer. In the PRD, the air quality is much more affected by the contributions from local sources (Anhui, Fujian, Guangdong, Guangxi, Guizhou Hainan, Henan, Hunan, Hubei, Inner Mongolia, Jiangsu, Jiangxi, Shaanxi, Shanxi, and Yunnan) than from sources outside China (Burma and Taiwan), which was stronger in winter than autumn, spring, and summer. In Qinghai, the air quality is much more affected by contributions from local sources (Gansu, Inner Mongolia, Tibetan Plateau, and Xinjiang) than from imported sources (India, Kirgizstan, Nepal, and Tajikistan), which was stronger in spring than in winter, summer, and autumn. In the Tibetan Plateau, the wintertime and autumn air quality are much more affected by contributions from outside sources (India, Ladakh, and Nepal) than local sources (Xinjiang). In contrast, the spring- and summertime air quality is much more affected by local sources (Qinghai and Xinjiang) contributions than sources outside China (Nepal). Finally, the air quality of the Tarim Basin is mainly affected by the Taklimakan Desert than by transport from outside, which was stronger in spring than in winter, summer, and autumn.

4. Conclusion

In the current study, using GWR Global Estimates of $PM_{2.5}$, justified by the validation of these data versus ground-based measurements in China, long-term (2001–2020) $PM_{2.5}$ pollution hotspots and sources were identified by analyzing spatiotemporal distributions, variations, trends, and contributions from potential sources. Our major findings are as follows:

- The validation study shows a good agreement between GWR $PM_{2.5}$ and ground-based measurements, as shown by the high correlation ($r = 0.95$), small error (RMSE = 8.14), and small bias (RMB = −3.10 %). However, this study shows that GWR Global $PM_{2.5}$ data can be effectively used as an indicator to identify local $PM_{2.5}$ pollution hotspots and their impacts on human health in China.
- China's central (Henan, Hubei), eastern (Shandong), northern (Beijing, Tianjin, Hebei, Shanxi), northwestern (Xinjiang), and southwest (Chongqing, Sichuan) regions were identified as hotspots of $PM_{2.5}$ pollution that occur more frequently in winter than in spring, autumn, or summer.
- $PM_{2.5}$ is higher than the Chinese AAQS in 32 provinces (including Tianjin, Henan, Shandong, Hebei, Jiangsu, Shanxi, Beijing, Anhui, Hubei, Chongqing, Hunan, Liaoning, Shanghai, Shaanxi, Jilin, Ningxia, Guangxi, Jiangxi, Zhejiang, Guizhou, Xinjiang, Guangdong, Macao, Gansu, Sichuan, Hong Kong) by 1.07 to 2.66 times in winter. The springtime $PM_{2.5}$ is higher than the AAQS in 22 provinces (including Tianjin, Xinjiang, Shandong, Henan, Beijing, Jiangsu, Hebei, Anhui, Shanghai, Hubei, Ningxia, Liaoning, Shanxi, Hunan, Chongqing, Gansu, Guangxi, Zhejiang, Jiangxi, Yunnan, Shaanxi, Jilin) by 1.01 to 1.86 times. The summertime $PM_{2.5}$ is higher than the AAQS in 7 provinces (Tianjin, Beijing, Shandong, Henan, Hebei, Jiangsu, and Anhui) by 1.03 to 1.60 times. The autumn $PM_{2.5}$ is higher than the AAQS in 18 provinces (including Tianjin, Shandong, Beijing, Henan, Hebei, Jiangsu, Anhui, Hunan, Hubei, Shanxi, Guangxi, Guangdong, Macao, Xinjiang, Jiangxi, Chongqing, Liaoning, Shanghai) by 1.01 to 1.89 times.
- Furthermore, $PM_{2.5}$ is higher than the WHO AQG standard in 33 provinces by 1.22 to 18.61 times in the winter and by 1.50 to 12.99 times in the spring, while $PM_{2.5}$ higher than the WHO AQG standard in 32 provinces by 2.11 to 11.23 times in the summer and by 1.99 to 13.21 times in the autumn. Only the Tibet Province is relatively clean in the summer and autumn seasons.
- In general, $PM_{2.5}$ increased significantly in most Chinese provinces (3–43 %) due to the rapid economic development and industrialization from 2001 to 2012. After that, $PM_{2.5}$ decreased by 12–94 % from 2013 to 2020 because of the implementation of air pollution control policies.
- The PSCF shows that the air quality in China is mainly affected by pollution originating domestically rather than imported from abroad. For example, the air quality of the Gobi Desert (GD), Northeast (NE), North China Plain (NCP), YRD (Yangtze River Delta), Central China, and PRD (Pearl River Delta) regions are mainly affected by the local sources, and winter is the most polluted season.

The present study concluded that the air pollution control policies had the co-benefit of reducing $PM_{2.5}$, resulting in improved air quality over China. As a result, this study strongly recommends that these policies be kept or extended in the future to improve a greater degree of air quality in China, which will benefit the citizens. Furthermore, city-level studies are needed to identify $PM_{2.5}$ hotspots and significant local and urban sources.

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CRediT authorship contribution statement

Md. Arfan Ali: Conceptualization, Data curation, Methodology, Formal analysis, Investigation, Validation, Visualization, Writing – original draft. **Zhongwei Huang:** Supervision, Writing – review & editing. **Muhammad Bilal:** Conceptualization, Investigation, Visualization, Writing – review & editing. **Mazen E. Assiri:** Writing – review & editing. **Alaa Mhawish:** Writing – review & editing. **Janet E. Nichol:** Writing – review & editing. **Gerrit de Leeuw:** Writing – review & editing. **Mansour Almazroui:** Writing – review & editing. **Yu Wang:** Writing – review & editing. **Yazeed Alsubhi:** Writing – review & editing.

Data availability

The data that has been used is confidential.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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