Globally analysing spatiotemporal trends of anthropogenic PM$_{2.5}$ concentration and population's PM$_{2.5}$ exposure from 1998 to 2016

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**A R T I C L E  I N F O**

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**A B S T R A C T**

Air pollution in the form of particulate matter (PM) is becoming one of the greatest current threats to human health on a global scale. This paper firstly presents a Bayesian space–time hierarch piecewise regression model (BSTHPRM) which can self-adaptively detect the transitions of local trends, accounting for spatial correlations. The spatiotemporal trends of the approximately anthropogenic PM$_{2.5}$ removed natural dust (PM$_{2.5,\text{No Dust}}$) concentrations and the corresponding population’s PM$_{2.5,\text{No Dust}}$ exposure (PPM$_{2.5,E}$) in the global continent from 1998 to 2016 were investigated by the presented BSTHPRM. The total areas of the high and higher PM$_{2.5,\text{No Dust}}$ polluted regions, whose spatial relative magnitude of PM$_{2.5,\text{No Dust}}$ pollution to the global continental overall level was between 1.89 and 14.68, accounted for about 13.4% of the global land area, and the corresponding exposed populations accounted for 56.0% of the global total population. The spatial heterogeneity of the global PM$_{2.5,\text{No Dust}}$ pollution increased generally from 1998 to 2016. The areas of hot, warm, and cold spots with increasing trends of PM$_{2.5,\text{No Dust}}$ concentration initially contracted and then later expanded. The local trends of the global continental PM$_{2.5,\text{No Dust}}$ concentrations and PPM$_{2.5,E}$ can be parted into three changing stages, early, medium, and later stages, using the BSTHPRM. The area proportions of the regions experiencing a decreasing trend of PM$_{2.5,\text{No Dust}}$ concentrations and PPM$_{2.5,E}$ were greater in the medium stage than in the early and later stages. The local trends of PM$_{2.5,\text{No Dust}}$ concentration and PPM$_{2.5,E}$ in the two higher PM$_{2.5,\text{No Dust}}$ polluted areas, northern India and eastern and southern China, increased in the early stage and then decreased in the medium stage. In the later stage (recent years), northern India displayed a stronger increasing trend; nevertheless, the follow-up decreasing trend still occurred in eastern and southern China. In the first two stages, more than half of the areas in Europe experienced a decreasing trend of PM$_{2.5,\text{No Dust}}$ Concentration and PPM$_{2.5,E}$; later, more than half of areas in Europe exhibited increasing trends in the later stage. North America and South America experienced a similar local trend of PPM$_{2.5,E}$ to Europe. The PPM$_{2.5,E}$ trend in Africa generally increased during the study period.

1. Introduction

Air pollution has become a common global challenge to human health that affects the quality of life. Particulate matter (PM) is widely believed to be the most deadly form of air pollution as well as a major cause of non-communicable disease according to the Global Burden of Disease in 2015 (Ahmad Kiadaliri and Norrving, 2016). PM smaller than 2.5μm (PM$_{2.5}$) constitutes most major components of air pollutants (Delfino et al., 2005). Many studies have revealed that long-term exposure to PM$_{2.5}$ can increase the risk of lung disease (e.g., lung cancer) (Hamra et al., 2014), cardiovascular disease (Puett et al., 2009), and cerebrovascular disease (Stafoggia et al., 2014), among others. PM$_{2.5}$ exposure may also increase mortality rates (Beelen et al., 2014; Hoek et al., 2013; Sacks et al., 2010).

Global estimates of ground-level PM$_{2.5}$ concentrations using multisource, remotely sensing data have become possible based on the technological development of Earth observations (Zhang and Li, 2015). Additionally, many high-precision algorithms retrieving PM$_{2.5}$ concentrations from remote sensing data have been developed (Hsu et al., 2013; Hu et al., 2014; Lee et al., 2011; Levy et al., 2013). Remotely sensed PM$_{2.5}$ concentration data that are collected on a large scale and possess the advantages of spatial continuity (Chen et al., 2006; Peng et al., 2016) are a good supplement to ground monitoring (Ma et al., 2015). Therefore, remotely sensed PM$_{2.5}$ concentration data can
provide an efficient mode with which to study the spatial and temporal
variations of air pollution (Tian and Chen, 2010).

Since 2010, the research team led by van Donkelaar has initiated the
production of spatiotemporal, remotely sensed, and annual mean global
continental PM$_{2.5}$ concentration data. The first version of the series of
PM$_{2.5}$ concentration data from 2001 to 2006 (van Donkelaar et al.,
2010) was integrated with the atmospheric chemical transport model
and the Moderate-Resolution Imaging Spectroradiometer (MODIS)
Aerosol Product. The second version of the data from 2001 to 2010 was
adopted from Aerosol Optical Depth (AOD) products and inverted from
Multiscale Imaging Spectroradiometer (MISR); SeaWiFS satellite
remote sensing was released in 2014 (Boys et al., 2014). In 2016, the
third version of the global continental PM$_{2.5}$ concentration from 1998
to 2014 was generated by van Donkelaar et al. (van Donkelaar et al.,
2016); currently, the time range has been updated for 1998–2016. The
latest dataset included more sourced, remotely sensed products and
clearly improved the inversion accuracy based on on-site monitoring
data (van Donkelaar et al., 2016). The data of the global continental
PM$_{2.5}$ concentration was produced in two formats; total components
included natural dust, and, to a smaller extent, sea salt. PM$_{2.5,\text{Total}}$, as
well as the format that excluded natural dust and sea salt (PM$_{2.5,\text{No}}$
Dust), approximated the anthropogenic components of PM$_{2.5}$ (Evans
et al., 2013), based on Goddard Earth Observing System Chemistry
(GEOS-Chem) simulations (Evans et al., 2013).

Few researchers have conducted studies regarding spatiotemporal
trends of global PM$_{2.5}$ pollution. Boys et al. (Boys et al., 2014) and
van Donkelaar et al. (van Donkelaar et al., 2015b) employed the basic linear
regression model to analyse the linear trends of global PM$_{2.5}$ concentration
from 1998 to 2012. Li et al. (Li et al., 2015b) investigated the space–time
complexity of the global continental PM$_{2.5,\text{Total}}$ pollution from 2000 to
2014 by employing a modified Bayesian space-time hierarch model
(BSTMH) based on multiscale statistical units. Yang et al. (Yang et al.,
2018) used a simple, classical linear regression model to analyse the low
frequencies of the global PM$_{2.5,\text{Total}}$ concentration from 1998 to
2015 in each country or region across the globe on the national scale.
Neither study explored the turning situation of the global continental
PM$_{2.5}$ pollution during the study period. Moreover, few studies
investigated the spatiotemporal trends of the population’s PM$_{2.5}$ exposure
in detail. In addition, the topic of the previous two studies was
PM$_{2.5,\text{Total}}$ pollution caused by the mixture of natural and human ac-
tivities, whereas the toxicity and the controlling measures of PM$_{2.5,\text{Total}}$
and PM$_{2.5,\text{No Dust}}$ could be different (Evans et al., 2013). Based on these
perspectives, this paper focuses on PM$_{2.5,\text{No Dust}}$ pollution, anthropo-
genic PM$_{2.5}$ pollution, and the population’s PM$_{2.5,\text{No Dust}}$ exposure
(PM$_{2.5,\text{E}}$).

To explore local trends more thoroughly, this paper firstly presents a
Bayesian space-time model that can self-adaptively detect the turning
points during the study period, considering spatial correlations, and
then apply this to investigate the spatiotemporal trends of global
continental PM$_{2.5,\text{No Dust}}$ concentrations and PM$_{2.5,\text{E}}$ from 1998 to 2016.

2. Data and methodologies

2.1. Data sources and pre-processing

The latest version of the remotely sensed PM$_{2.5}$ concentrations da-
taset (V4.GL.20) with the spatial resolution of 0.1’ × 0.1’
(−10km × 10km) used in this paper was produced by van Donkelaar’s
team (van Donkelaar et al., 2015a; van Donkelaar et al., 2016). The
remotely sensed PM$_{2.5}$ concentrations were produced in two formats:
PM$_{2.5,\text{Total}}$: including natural components, and PM$_{2.5,\text{No Dust}}$:excluding
natural components. The latter was used for this study. The details and
validation of the dataset can be found in the related references (van
Donkelaar et al., 2015a; van Donkelaar et al., 2016). Moreover, a global
population density dataset with the spatial resolution of 2.5’ × 2.5’
(−5km × 5km)(GPWv4) in 2000, 2005, 2010, and 2015 (Center for
International Earth Science Information Network - CIESIN - Columbia
University, 2017) was used in our study. The population density dataset
was adjusted to match the 2015 Revision of the United Nation’s World
Population Prospects (UN WPP) country totals and is consistent with
national censuses and population registers as rasterized data to facil-
itate data integration. The continuous yearly global population density
dataset from 1998 to 2016 was obtained by the linear interpolation
method (van Donkelaar et al., 2015a).

We used the spatial neighbourhood average method to estimate the
missing pixel values of the dataset. In addition, the spatial resolutions
and projected coordinate system of the population density dataset were
adjusted in accordance with the remotely sensed PM$_{2.5,\text{No Dust}}$ annual
concentration dataset (0.1’ × 0.1’,−10km × 10km) when the global
continental PM$_{2.5,\text{E}}$ was calculated.

2.2. Methods

2.2.1. The q-statistic method

The q-statistic method can measure the degree of spatial hetero-
genocity (Wang et al., 2010; Wang et al., 2016) which refers to aniso-
tropic distributions of traits, events, or their relationships across a re-
region (Anselin, 2010; Dutilleul, 2011). The q-statistic index, q, can be
expressed as follows:

\[
q = 1 - \frac{\sum_{i=1}^{N_h} (Y_i - \bar{Y})^2}{\sum_{i=1}^{N_h} (Y_i - \bar{Y})^2} = 1 - \frac{\sum_{h=1}^{N_h} N_h \sigma^2_h}{N \sigma^2}
\]

(1)

where \(N\) is the number of the spatial statistical units of the global
continent which is stratified into \(h = 1, 2, \ldots, L\) stratum, countries and
regions in the world; \(Y_i\) and \(Y_h\) denote the value of unit \(i\) in the population and in
stratum \(h\), separately. \(\sigma^2\) and \(\sigma^2_h\) represent the population mean
and variance. The stratum mean and variance, \(\bar{Y}_h\) and \(\sigma^2_h\), can be expressed as:

\[
\bar{Y}_h = \frac{\sum_{i=1}^{N_h} Y_i}{N_h}, \quad \sigma^2_h = \frac{\sum_{i=1}^{N_h} (Y_i - \bar{Y}_h)^2}{N_h}
\]

(2)

the value of q-statistic index is between 0 and 1. The q-Statistic index
value increases monotonously with the increase of spatial heterogeneity
(Wang et al., 2016). The closer the value of q-statistic index gets to 1,
the higher the degree of spatial heterogeneity is, and vice versa.

2.2.2. Multiscale homogeneous subdivision method

To implement the Bayesian statistical model in remotely sensed
PM$_{2.5,\text{No Dust}}$ concentration data, the multiscale homogeneous subdivi-
sion method presented by Li et al. (Wang et al., 2018a) was employed in
this paper. The procedure of this method involves structuring a set of
nested, multiscale grids through square quadtree subdivision (Samet
et al., 1984) based on local variation or heterogeneity which is mea-
sured by the coefficient of variation (CV); it can be expressed as follows
(Everitt, 2002):

\[
CV = \frac{\sigma}{\bar{X}} \times 100\%
\]

(3)

where \(\sigma\) and \(\bar{X}\) represent standard deviation and mean of the pixel
value, PM$_{2.5,\text{No Dust}}$ concentration, within the subdivided grid for each
year, respectively. In statistics, sample data can generally be regarded
as homogeneous if their CV is < 15% (Snedecor and Waddle, 1980).
Therefore, the CV threshold value \(\lambda_0\) is generally assigned with < 15%.

2.2.3. A developed Bayesian space–time model

Based on the BSTHM presented by Li et al. (Li et al., 2014) and
piecewise regression model (Malash and El-Khiaiery, 2010), this paper
presented a Bayesian space-time hierarchical piecewise regression
model (BSTHPRM). The BSTHPRM can explore non-linear local trends
by capturing self-adaptively turning points of local trends according to
Fig. 1. Map of global annual average PM$_{2.5}$ concentrations (μg/m$^3$) from 1998 to 2016.
the sampled data and considered spatial correlations. The mathematical forms of the presented BSTHPRM can be expressed as follows:

\[ \rho_i \sim \text{Normal}(\mu_{ij}, \sigma^2 \rho) \]  
(4)

\[ \mu_{ij} = \alpha + S_{ij}(t + B M_i) + b_1 M_i(t - a_1 M_i) + G_{a_1, M_i} + b_2 M_i(t - a_2 M_i) + G_{a_2, M_i} + e_{ij} \]  
(5)

\[ G_{a_1, M_i} = \frac{1}{1 + e^{-(t - a_1 M_i)}} \]  
(6)

\[ G_{a_2, M_i} = \frac{1}{1 + e^{-(t - a_2 M_i)}} \]  
(7)

\[ a_1 M_i \sim \text{Uniform}(3, T - 4) \]  
(8)

\[ a_2 M_i \sim \text{Uniform}(a_1 M_i + 2, T - 2) \]  
(9)

\[ k_1 M_i = b_0 + b_1 M_i \]  
(10)

\[ k_2 M_i = b_0 + b_1 M_i + b_2 M_i \]  
(11)

\[ k_3 M_i = b_0 + b_1 M_i + b_2 M_i + b_3 M_i \]  
(12)

where \( \mu_{ij} \) is the mean parameter of the \( M_i \)-th multiscale statistical unit; \( \alpha \) is the corresponding variance, and \( \alpha \) is the overall global mean of PM\(_{2.5, \text{No Dust}} \) concentration from 1998 to 2016. \( S_{ij}(t + B M_i) \) describes overall time trend globally containing a linear \( b_0 \) and non-linear tendency by \( B \) whose prior distribution adapted Gaussian distribution; \( b_1 M_i \) and \( b_2 M_i \) are local piecewise linear regression coefficients; \( a_1 M_i \) and \( a_2 M_i \) are the first and second turning points of the \( M_i \)-th multiscale statistical unit. The spatiotemporal process can be decomposed into three stages of variation. To determine the number of turning points, the root mean square errors (RMSE) and goodness of fitting \( R^2 \) were used to evaluate and calculated under three conditions with no turning point, one turning point, and two turning points. If there are two turning points, i.e. the first (early), second (medium), and third (later) stage. The corresponding linear variation parameters are \( k_1 M_i \) and \( k_2 M_i \). It should be pointed out that the situation with three changing stages includes the situation of two or one changing stages. It is the situation with two stages of variation if \( k_1 M_i \) equals to \( k_2 M_i \) (\( k_1 M_i = k_2 M_i \), or \( k_2 M_i \) equals to \( k_3 M_i \)(\( k_2 M_i = k_3 M_i \)). There is only one changing stage if all the three piecewise linear regression parameters \( k_1 M_i \), \( k_2 M_i \), and \( k_3 M_i \) are equal \( (k_1 M_i = k_2 M_i = k_3 M_i) \). \( G_{a_1, M_i} \) and \( G_{a_2, M_i} \) are the logistic function as described in Eqs. (6) and (7). \( \lambda \) is a shape parameter that is generally assigned to the value \( > 10 \). \( T \) is the number of time nodes \( (T = 19 \) in this paper). Because linear regression is necessary only when there are more than two sample values, namely at least three sample values, both \( a_1 M_i \) and \( a_2 M_i \) are therefore limited between 3 and \( T - 4 \) and between \( a_1 M_i \) and \( a_2 M_i \) are both assigned to the prior of uniform distribution, and can be self-adaptively estimated according to sample data. The term \( e_{ij} \) is a Gaussian noise error whose prior distribution is assigned as normal distribution \( N(0, \sigma^2) \). Through the BSTHPRM, the spatial relative magnitude of PM\(_{2.5, \text{No Dust}} \) pollution in the \( M_i \)-th multiscale statistical unit, denoted as \( S_{RM_i} \), is used to quantify the PM\(_{2.5, \text{No Dust}} \) pollution density relative to the overall level on earth, can be estimated. \( S_{RM_i} \) can be expressed as follows:

\[ S_{RM_i} = \frac{\mu_{ij}(t')}{N \sum_{ij}(t) \mu_{ij}(t')} \]  
(13)

where \( t' \) represents the centre point of the study period from 1998 to 2016. \( N \) is the number of the subdivided multiscale statistical units. The parameters \( b_1, M_i \), \( b_2, M_i \), \( b_3, M_i \), \( a_1, M_i \), and \( a_2, M_i \) are considered simultaneously spatial, structured, and unstructured random effects through the assignment of the BYM model (Besag et al., 1991). Thereinto, the conditional autoregression (CAR)-Normal prior based on the first-order spatial adjacency matrix was assigned to the five regression parameters. The spatial correlation was established based on the topological relation of the constructed, multiscale statistical units. The Bayesian statistical estimate was implemented by WinBUGS (Lunn et al., 2000) based on the Markov chain Monte Carlo (MCMC) algorithm in this paper. The number of iterations was set to 100,000. 90,000 of these iterations were associated with the burn-in period, and 10,000 were associated with the number of iterations of the posterior distribution of parameters. The convergence of the Bayesian inference results was monitored by standard autocorrelation plots and trace plots.

3. Results

3.1. Descriptive statistical result

The spatial pattern of global continental PM\(_{2.5, \text{No Dust}} \) pollution remained roughly stable (Fig. 1) from 1998 to 2016, although some distinctions occurred during different years; specifically, a distinct spatiotemporal trend occurred during the study period. The maximum global mean of PM\(_{2.5, \text{No Dust}} \) concentration from 1998 to 2016 was 97 \( \mu \)g/m\(^3\), and the corresponding area located in northern India. In addition, a polarizing phenomenon became generally increasingly severe from 1998 to 2016. To quantificationally measure the spatial heterogeneity of global annual mean PM\(_{2.5} \) concentration during 1998–2016, this paper calculated the annual q-Statistic index value with the stratum of 196 countries and regions. The q-statistic index values from 1999 to 2016 were all > 0.50 and reached the maximum of 0.6450 in 2014. This indicated that the global PM\(_{2.5} \) pollution exhibited stronger discrepancy between countries. Moreover, the q-statistic index value showed an overall increasing trend from 1998 to 2016 (Fig. 2 (A)). Considering that the CV index was often used to assess variation of sample data in statistics, this paper has also calculated the CV of the global PM\(_{2.5, \text{No Dust}} \) annual concentrations from 1998 to 2016. The result showed that the variation of global PM\(_{2.5} \) pollution usually increased during the study period (Fig. 2 (B)). The trend of CV was similar to that of the q-statistic index. Although it can be concluded that the heterogeneity of global PM\(_{2.5, \text{No Dust}} \) pollution across various regions generally increased from 1998 to 2016, the turning feature in the trends of the q-statistic and CV values appeared in the study period. This implied that the local trends of PM\(_{2.5, \text{No Dust}} \) pollution have the turning character that will be demonstrated by the following Bayesian statistical results.

3.2. Multiscale statistical unit construction

A set of nested, multiscale statistical units need to be constructed before conducting the presented BSTHPRM. The multiscale statistical units applicable to the dataset used in this study were structured by the abovementioned multiscale homogeneous subdivision method. Three-cycles-subdivision was implemented, and a set of three-scales nested statistical grids were structured for the spatiotemporal remotely sensed PM\(_{2.5, \text{No Dust}} \) Concentration from 1998 to 2016 (Fig. 3). The threshold of CV, denoted as \( \lambda_0 \), was assigned at 12% so that the variance of the multiscale statistical units maintained > 99.5% compliance with that of the original data. Consequently, the global, multiscale statistical grids/units contained a total of 16,756 grids—5675 grids with a scale of \( 1.6 \times 1.6 \), 2308 grids with a scale of \( 0.8 \times 0.8 \), and 8773 grids with a scale of \( 0.4 \times 0.4 \). In other words, the CVs of PM\(_{2.5, \text{No Dust}} \) concentration value of pixel within the 16,756 grids were all less than \( \lambda_0 \) during each year of the study period (1998–2016).
3.3. Bayesian statistical results

3.3.1. Overall spatial trends

The presented BSTHPRM in this paper estimated the overall spatial relative magnitude of PM$_{2.5,\text{No Dust}}$ concentration, namely the overall spatial patterns, based on the simultaneous consideration of the space–time interaction from 1998 to 2016. The overall spatial patterns may be quantified by the posterior medians of the coefficient SR$_{M(i)}$, described by formula (13), whose value indicates the magnitude of the PM$_{2.5,\text{No Dust}}$ concentration in the $M[i]$-th multiscale grid relative to the overall global continental mean level. Namely, the posterior median of SR$_{M(i)}$ directly indicates that the PM$_{2.5,\text{No Dust}}$ level of the $M[i]$-th multiscale grid is SR$_{M(i)}$ times the overall global continental level.

Fig. 4 illustrates the common spatial patterns of global continental PM$_{2.5,\text{No Dust}}$ pollution. The peak value of the posterior median of SR$_{M(i)}$ is 14.68 (14.28, 15.08), and the corresponding area is located in northern India. According to the BSTHPRM’s estimates of SR$_{M(i)}$, based on the geometrical interval method (Conolly and Lake, 2006), the global continent was classified into five grades (or levels) (Fig. 5): lower (SR$_{M(i)}$: 0.01–0.86), low (SR$_{M(i)}$: 0.86–1.12), middle (SR$_{M(i)}$: 1.12–1.98), high (SR$_{M(i)}$: 1.98–4.89), and higher (SR$_{M(i)}$: 4.89–14.68).

The most severely PM$_{2.5,\text{No Dust}}$-polluted area (dark red-coloured in Fig. 5), where the posterior median of SR$_{M(i)}$ was 4.89–14.68, was located mainly in three areas: eastern and southwestern China (193.8 × 10$^4$km$^2$), northern India (139.9 × 10$^4$km$^2$), and central areas of the Indo-China Peninsula (0.6 × 10$^4$km$^2$). The PM$_{2.5,\text{No Dust}}$-polluted exposed population reached up to 1945 million (about 27% of the world’s population) in the three higher PM$_{2.5,\text{No Dust}}$-polluted regions. The PM$_{2.5,\text{No Dust}}$ concentrations in the three areas were 40.2 (26.8, 53.6) μg/m$^3$, 47.1 (32.7, 61.53) μg/m$^3$, and 38.1 (30.1, 45.5) μg/m$^3$ from 1998 to 2016. The high-level PM$_{2.5,\text{No Dust}}$-polluted areas (orange-red-coloured in Fig. 5), whose posterior median of SR$_{M(i)}$ ranged from 1.98 to 4.89, were located mainly in three regions: southern Asia (732.4 × 10$^4$km$^2$), eastern Europe (283.3 × 10$^4$km$^2$), and southern Africa (641.8 × 10$^4$km$^2$). The corresponding exposed populations were 1409 million (southern Asia), 307 million (eastern Europe), and 370 million (southern Africa), respectively. The total areas of the high and higher PM$_{2.5,\text{No Dust}}$-polluted regions were 1991.8 × 10$^4$km$^2$, accounting for about 13.4% of the global land area; however, the corresponding exposed populations were up to 4031 million and comprised nearly 56.0% of the total global population.

3.3.2. Local trends

The multiscale statistical units whose 19-year (1998–2016) averages of PM$_{2.5,\text{No Dust}}$ annual mean concentration were greater than 25μg/m$^3$, the WHO air guidelines of PM$_{2.5}$ annual mean concentrations of the interim target-2 (IT-2) (WHO, 2006) were selected. Fig. 6 (left) shows the corresponding time series polylines of PM$_{2.5}$ annual concentration from 1998 to 2016. The highlighted red, orange, and blue polylines in Fig. 6 belong to the high (38.62–93.09μg/m$^3$), medium
(29.92–38.61μg/m³), and low (25.00–29.91μg/m³) level of PM$_{2.5\text{,No Dust}}$ pollution, and the corresponding 19-year averages of PM$_{2.5}$ annual mean concentration were 71.96μg/m³, 45.79μg/m³, and 25.03μg/m³, respectively. Fig. 6 (right) shows the regression lines of the selected three-time series under the conditions with no turning point, one turning point, and two turning points. It can be seen that three changing stages, namely two turning points, may properly describe the changing process. The RMSE and $R^2$ were calculated (Table 1) under the three conditions with no turning point, one turning point, and two turning points. The results show that the RMSE and $R^2$ of the selected three time series are minimum and maximum under the condition with two turning points. Of course, it can also be parted into four or more stages of change. Yet, according to Ockham’s Razor principle (Gauch, 2003; Hoffmann et al., 1997), simpler models are preferable to more complex...
ones because they are more testable (Baker, 2004) and explicable. From the above, the three stages and two turning points are determined in the BSTHPRM.

Fig. 7(A) and (B) illustrated the estimated results of the first and second turning points of the local trend of the PM$_{2.5}$ concentration in each multiscale statistical unit, namely the parameters $a_{1, M[i]}$ and $a_{2, M[i]}$ of the BSTHPRM. The turning year equals to 1997 plus the integer of $a_{1, M[i]}$ and $a_{2, M[i]}$, e.g. if $a_{1, M[i]}$ is 4.5, then the turning year is 2001. The first local trend’s transition occurred mainly between 2006 and 2009 in many areas (orange in Fig. 7(A)) and 2003–2006 in other areas (yellow in Fig. 7(A)). A few areas, e.g. western Alaska of USA, central regions in Canada, south central Brazil, and some areas of western China also arose local turning trends before 2003 (light green)

Fig. 5. Five grades of PM$_{2.5}$ polluted, spatial, distributed areas according to the classifications of the posterior median of the parameter SRM[$i$] based on the geometrical interval method. (For interpretation of the references to colour in this figure, the reader is referred to the web version of this article.)

Table 1

<table>
<thead>
<tr>
<th>Three selected polylines</th>
<th>Regression indicator</th>
<th>No turning point</th>
<th>One turning point</th>
<th>Two turning points</th>
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<tr>
<td></td>
<td></td>
<td>RMSE</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 (71.96)</td>
<td>Root Mean Square</td>
<td>7.7238</td>
<td>7.2428</td>
<td>4.7463</td>
</tr>
<tr>
<td>2 (45.79)</td>
<td>Error (RMSE)</td>
<td>7.2332</td>
<td>2.5370</td>
<td>2.4632</td>
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<tr>
<td>3 (25.03)</td>
<td></td>
<td>1.8367</td>
<td>1.7947</td>
<td>1.4366</td>
</tr>
<tr>
<td></td>
<td>$R^2$</td>
<td>0.4236</td>
<td>0.9224</td>
<td>0.7823</td>
</tr>
<tr>
<td>1 (71.96)</td>
<td></td>
<td>0.4369</td>
<td>0.9224</td>
<td>0.9268</td>
</tr>
<tr>
<td>2 (45.79)</td>
<td></td>
<td>0.8776</td>
<td>0.8831</td>
<td>0.9251</td>
</tr>
</tbody>
</table>

Fig. 6. The time series polylines of PM$_{2.5}$ annual concentration from 1998 to 2016 whose 19-year averages of PM$_{2.5}$ annual concentration is greater than 25μg/ m$^3$ (left), and the regression lines of the selected three three-time series under the conditions with no turning point, one turning point, and two turning points (right). (For interpretation of the references to colour in this figure, the reader is referred to the web version of this article.)
in Fig. 7(A)) or after 2009 (orange red in Fig. 7(A)). The second local trend’s transition occurred predominately during 2010–2012 (brilliant yellow in Fig. 7(B)) in most areas, or between 2012 and 2014 in some areas, e.g. western regions in Africa, China, southern Argentina, etc.

Additionally, the areas where the first turning point is before 2003 generally experienced the second turning point before 2010 (emerald green and light green in Fig. 7(B)).

The presented BSTHPRM has estimated three-piecewise local linear change rates, \( k_1, M[i] \), \( k_2, M[i] \), and \( k_3, M[i] \), of each multiscale statistical unit corresponding to the two turning points, \( a_1, M[i] \) and \( a_2, M[i] \). We defined the three piecewise local trends as early, medium, and later-change local trends, which can be measured quantitatively by the parameters \( k_1, M[i] \), \( k_2, M[i] \), and \( k_3, M[i] \), respectively.

Figs. 8–10 illustrate the spatial patterns of the three stages’ yearly local linear rates of the global continental PM\(_{2.5, \text{No Dust}}\) concentration from 1998 to 2016. In the early stages before the first turning point, \( a_1, M[i] \), the higher-polluted areas located in Asia, northern India, eastern and southwestern China, western Indonesia, and southern Malaysia.
experienced increased local trends. The highest yearly increases in the four areas were up to 3.24 (2.82, 3.65) μg/m³ per year, 5.35 (4.91, 5.77) μg/m³ per year, 3.88 (3.44, 4.26) μg/m³ per year, and 2.15 (1.72, 5.57) μg/m³ per year, respectively. Although western Europe had the high level of PM2.5 pollution, many regions in Europe possessed a decreased local trend, the highest decrease was −2.45 (−2.87, −2.03) μg/m³ per year. Except for southern Brazil, the majority of the regions in South America experienced an increased trend. Moreover, some lower-polluted regions in western Canada, Alaska (North America), western North America, and northern Africa also experienced increased local trends.

In the medium stage, many regions exhibited a decreased local trend (Fig. 9). Four areas, Indochina peninsula, Alaska (North America), central regions in Russia, and the mid-southern regions of...
Africa possessed increasing trends; the corresponding highest yearly increases were 5.27 (4.86,5.68) μg/m³ per year, 6.68 (6.26,7.10) μg/m³ per year, 1.93 (1.51,2.35) μg/m³ per year, and 1.34 (0.94,1.76) μg/m³ per year, respectively. Except for northwest Italy and south Sweden, the majority of Europe still possessed a stronger decreased local trend in the medium stage. The local trends of the higher-polluted areas, northern India, eastern and southwestern China, western Indonesia, and southern Malaysia have transformed from increasing to decreasing. The corresponding strongest yearly decreases of the four areas were −7.89 (−8.35,−7.41) μg/m³ per year, −3.13 (−3.62,−2.68) μg/m³ per year, −5.91 (−6.32,−5.50) μg/m³ per year, and −2.37 (−2.80,−1.91) μg/m³ per year, respectively. Additionally, most regions in Canada and Australia experienced a weak increasing trend.

Within the later stage, the local trends have changed considerably. A strong increasing trend occurred in many regions of global continents (Fig. 10). The local trends of eastern Europe transformed from decreasing to increasing with the highest value of 2.74 (2.04,3.54) μg/m³ per year. The strongest increase occurred in Kalimantan Island and Sumatra Island of Indonesia in Asia. The higher PM2.5 polluted areas, northern India and surrounding areas including Pakistan, Nepal, and Bangladesh, also experienced a stronger increase in the later stage; the highest yearly increase was up to 9.10 (8.34,9.86) μg/m³ per year. The local trends of China exerted a distinct spatial difference. An obvious decrease appeared in Central and southern China in the later phase; the local trends of eastern Europe transformed from decreasing to increasing with the highest value of 2.74 (2.04,3.54) μg/m³ per year. The local trends of the higher-polluted areas, northern India, eastern and southwestern China, western Indonesia, and southern Malaysia, the decreasing trends appeared only in the second stage; the stronger increasing trends arose in the first and third stage. Almost all of the areas throughout China experienced the increasing trends in the first stage, then the decreasing trends in the second stage. In the third stage, the decreasing trends occurred continuously in the northern, central, and southern regions within China, but the local trends in western, northeast, and a few areas of eastern China switched to an increasing trend. Table 2 listed the parameters' estimates of local trends of the representative 21 multiscale statistical units whose $SR_{M[i]}$ are > 4.89, belonging to the higher PM$_{2.5}$NoDust polluted areas (deep red in Fig. 5). The results can provide representative and specific statistical information.

The overall spatial pattern may be clustered into three categories: hot, warm, and cold spots—where the PM$_{2.5}$NoDust levels are high, medium, and low. Furthermore, the local trend of the three types of areas can be investigated by the following two-stage classification rule (Li et al., 2014). First, the spatial units were identified as hot, warm, and cold spots when the posterior probability of $SR_{M[i]}$ $P(SR_{M[i]} > 1.0 | \rho_i)$ was > 0.80, between 0.20 and 0.80, and < 0.20, respectively (Richardson et al., 2004). Second, based on the posterior probability of the local trend parameters $P(k_i, M[i] > 0 | \rho_i)(r = 1,2,3)$, the spatial units could also be classified into three categories: > 0.80, between 0.20 and 0.80, and < 0.20. Table 3 listed the regional areas ($\times10^4$km$^2$) and the corresponding percentages of the three types of local trends (increasing, stable, and decreasing) in the hot, warm, and cold spot regions. The areas of hot, warm, and cold spots with increasing trends all firstly contracted from 2698.9 $\times10^4$ km$^2$ (50.0%) in the first stage to 1488.3 $\times10^4$ km$^2$ (27.6%) in the second stage, then expanded from 1488.3 $\times10^4$ km$^2$ (27.6%) in the second stage to...
3277.6 \times 10^4 \text{ km}^2 (60.7\%) in the third stage, the areas of hot, warm, and cold spots with decreasing trends exhibiting an inverse trait. The areas with stable trends in hot and cold spots shrank continuously. The areas of warm spots with stable local trends initially expanded slightly but later shrank.

4. Spatiotemporal variability of PPM2.5E

4.1. Descriptively statistical results

PM2.5 concentration constantly serves as a risk indicator for air pollution exposure (Hystad et al., 2011; Zhong et al., 2013). However, the indicator considering only PM2.5 concentration ignores the heterogeneity of the population's spatial distribution. Kouza et al. (Kouza et al., 2002) presented an assessment model for the population's exposure (Hystad et al., 2011; Zhong et al., 2013). However, the indicator considering only PM2.5 concentration ignores the heterogeneity of the population's spatial distribution. Kouza et al. (Kouza et al., 2002) presented an assessment model for the population's exposure...

This paper calculated the differences of global continental PPM2.5E between 2000 and 2015 (Fig. 11), between 2005 and 2010 (Fig. 12), and between 2010 and 2015 (Fig. 13) in northern India, there always existed many regions where the PPM2.5E in 2000 was greater than in 2005, that in 2005 was greater than in 2010, that in 2010 was greater than in 2015. From 2000 to 2005, nearly all densely populated regions in India and Indonesia experienced a greater increase of PPM2.5E (Fig. 11). Meanwhile, the decreasing trends in many regions throughout China (except for the Beijing-Tianjin-Hebei region and Yangtze River Delta, but including Shanghai City and Jiangsu province) have been occurring from 2005 to 2010 (Fig. 12) and 2010–2015 (Fig. 13). This result revealed the positive effects of the continuously tightened emission-control policies implemented by China’s government since 2005. It should be mentioned that the maximum increased value of PPM2.5E in India was greater than that in China between 2005 and 2010 (+86.66 × 10^4 vs +40.46 × 10^4 persons/km^2-m^3) and between 2010 and 2015 (+193.64 × 10^4 vs +136.91 × 10^4 persons/km^2-m^3), with the exception of 2000–2005 (+86.98 × 10^4 vs +151.28 × 10^4 persons/km^2-m^3). Eastern Europe and southern Nigeria also experienced a slight increase of PPM2.5E from 2000 to 2015, while other regions experienced a general decrease during the period from 2005 to 2015. Java Island, a populated (about 14.5 million inhabitants) region in Indonesia, experienced a continuous growth of PPM2.5E from 2000 to 2015. Other regions, including North and South America, Oceania, and other sparsely populated regions, experienced a stable, even declining trend of PPM2.5E.

4.2. Bayesian statistical results

This paper also estimated the spatiotemporal trends of global continental PPM2.5E from 1998 to 2016 by employing the presented BTHPRM. Considering the computational burden, the locations, namely pixels, whose population density in 2000 was equal or > 10 persons/km^2 were included in this calculation. In addition, the population does not generally fluctuate in a relative short period for demographic transition (Demeny et al., 2003). The variation process of the global continental annual PPM2.5E may also be decomposed into...
Fig. 11. The difference (increase or decrease) of the PPM$_{2.5}$ over the global continent between 2000 and 2005.

Fig. 12. The difference (increase or decrease) of the PPM$_{2.5}$ over the global continent between 2005 and 2010.
three stages as well as PM$_{2.5,NoDust}$, namely two turning points.

Fig. 14(A) and (B) showed the estimation of the first and second turning points of global continental annual PPM$_{2.5,E}$. The first turning year of the local trends of the PPM$_{2.5,E}$ in America was mainly during 2001–2004 or 2004–2007, the second turning year was in 2008 and beyond. In Europe, the two turning year periods were generally 2004–2007 and 2008–2011. The first turning point in Africa was various, but the second turning point was relatively concentrated in 2011–2014. The first turning year in India concentrated majorly in 2007–2009, the second turning in northern and eastern India occurred during 2011–2013, and other areas of India occurred between 2008 and 2011. The first turning year in China appeared in a wide range between 2001 and 2009. The second turning year in northeast China, Indo-China Peninsula region, Indonesia, and Malaysia, was in 2011–2013, that in other areas of China was predominately between 2008 and 2011.

The local yearly changes, annual increases or decreases, of the global continental annual mean PPM$_{2.5,E}$ in the three stages have been estimated by the presented BSTHPRM; the corresponding spatial distributions were showed by Figs. 15–17. In the early and medium stages, more than half of the areas (60% and 86.8%) in Europe generally experienced a decreasing trend, and a weak increase occurred in the other areas in Europe. The corresponding median annual decrease and increase in the two stages were 1) $-11.62$ and $7.19$ persons/km$^2$$\cdot$μg/m$^3$ per year and 2) $-17.47$ and $5.10$ persons/km$^2$$\cdot$μg/m$^3$ per year. In the later stage, more than half of areas (59.3%) in Europe exhibited increases, and the others decreases; the corresponding median annual decreases and increases were $10.66$ and $-4.95$ persons/km$^2$$\cdot$μg/m$^3$ per year. North America and South America had parallel experiences with Europe. The local trend of PPM$_{2.5,E}$ in Africa showed, in general, an increasing trend, although a declining trend occurred in some areas of western Africa in the medium stage (Fig. 16). In the three stages, the median annual increases in Africa were 8.57, 11.62, and 18.23 persons/km$^2$$\cdot$μg/m$^3$ per year, respectively, and the area percentages with increasing trends were 86.0%, 71.1%, and 86.7%.

The local trends of PPM$_{2.5,E}$ in the higher polluted and densely populated areas of Asia, especially India and China, showed distinct characteristics. In the early stage, the stronger increasing trend occurred in India and southern and eastern China (Fig. 15); the corresponding median annual increases of the two areas reached up to 333.7 and 132.9 persons/km$^2$$\cdot$μg/m$^3$ per year. Moreover, the areas in northern India and eastern China (dark red in Fig. 15) where the PM$_{2.5,NoDust}$ pollution and population density were all at the top level experienced an extremely high PPM$_{2.5,E}$ annual increase of $>1100$ persons/km$^2$$\cdot$μg/m$^3$ per year. The local trend of PPM$_{2.5,E}$ in Africa showed, in general, an increasing tendency, although a declining trend occurred in some areas of western Indonesia.

5. Discussion

It is quite commonly known that PM$_{2.5}$ pollution is a synthetic result of natural and anthropogenic activities. Although natural dust components also pose health risks for humans (Cook et al., 2005), pollution
control policies would be inherently different for natural and anthropogenic compositions of PM$_{2.5}$ (Evans et al., 2013), and the toxicities of natural dust and anthropogenic components of PM$_{2.5}$ could be not identical due to the distinguishing chemical compositions varying between the two components of PM$_{2.5}$. This is also the consideration and starting point as to why this paper focuses on the PM$_{2.5}$ pollution in regards to the removal of natural dust and, to a small extent, sea salt (i.e., PM$_{2.5}$, No Dust pollution). PM$_{2.5}$ pollution is neither a regional nor a national, but rather a global, systematic phenomenon. According to previous reports, global PM$_{2.5}$ pollution was associated with international trade (Lin et al., 2016; Zhang et al., 2017). In order to more thoroughly understand and ultimately control air pollution, which is threatening to human health, it is essential that the phenomenon of PM$_{2.5}$ pollution’s status be studied on a global scale. We previously studied the spatiotemporal evolution of global continental PM$_{2.5}$ concentrations, including natural dust and sea salt from 2000 to 2014 (Li et al., 2018b), but the turning point of local trends was not detectable in the previous study, and the spatiotemporal variability of the global PPM$_{2.5}$E was not sufficiently analysed. The presented BSTHPRM in this paper possesses more advantages than the methods used in our previous research. Because our research subject PM$_{2.5}$, No Dust is primarily caused by human activities, the research results can provide more practical

Fig. 14. Spatial distribution of the first (A) and second (B) turning point of the local trend of the global continental PPM$_{2.5}$E from 1998 to 2016; the turning year equals 1997 plus the integer of the turning point value.
references for the global, joint policies controlling air pollution.

The spatiotemporal trends of PM$_{2.5}$ pollution reflect the
preferable, global anthropogenic impact on air pollution. The foundation
tional goal is effectively controlling air pollution by adjusting human
activities: Such as altering industrial structures and reducing emissions. This paper found that the PM$_{2.5}$ pollution and PPM$_{2.5}$ in central
and southern regions of China exhibited a continuously decreased local
trend from the first turning year to 2016, namely in the medium and
later stages. This may be attributed in-part to China’s governmental
implementation of continuously tightened emission-control policies
since 2005. On the other hand, the PM$_{2.5}$ pollution and PPM$_{2.5}$ in India showed a stronger increasing trend in the later stage, namely in
the recent years, although a decreasing trend occurred in the medium
stage. One possible reason for this result is that India’s government has

Fig. 15. Spatial pattern of the early local trends and the yearly change ($\times 10^3$ persons/km$^2$ $\cdot$ μg/m$^3$ per year) of the global continental PPM$_{2.5}$ from 1998 to the first
turning year. (For interpretation of the references to colour in this figure, the reader is referred to the web version of this article.)

Fig. 16. Spatial pattern of the medium local trends, yearly change ($\times 10^3$ persons/km$^2$ $\cdot$ μg/m$^3$ per year) of the global continental PPM$_{2.5}$ from the first to the second
turning year.
not made relevant policies for preventing air pollution. Our study also found that, except for Alaska (USA) and Canada, the area proportion of the regions with a decreasing trend of PM$_{2.5,\text{NoDust}}$ pollution and PPM$_{2.5\text{E}}$ was greater in the medium stage than in the early and later stage. We think that the mitigation of PM$_{2.5,\text{NoDust}}$ pollution and PPM$_{2.5\text{E}}$ in the medium stage possibly arose from the global economic downturn caused by the North American subprime mortgage crisis in 2007. The global electricity production from coal sources (percent of total) declined from the maximum of 41.14% in 2007 to 40.11% in 2010, and the global manufacturing value added (percent of total) decreased markedly from 2006 (17.12%) to 2009 (15.60%) (https://data.worldbank.org/indicator/EG.ELC.COAL.ZS?end=2015&start=1998).

India and China, the areas with the high levels of PM$_{2.5,\text{NoDust}}$ pollution and PPM$_{2.5\text{E}}$, are developing, middle-income countries constituting approximately 36% of the world’s population (https://data.worldbank.org/indicator/). According to the statistical results of our study, PM$_{2.5,\text{NoDust}}$ pollution and PPM$_{2.5\text{E}}$ throughout the northern, central, and southern regions in China appeared to continuously be decreasing in medium and later stages. These results indicate that China’s continuously tightened governmental emission-control policies played a vital role in controlling the PM$_{2.5,\text{NoDust}}$ pollution of China. Nevertheless, to our knowledge, India’s government has not yet proposed any effective emission-control policies during the study period. Although India’s PM$_{2.5,\text{NoDust}}$ concentrations and PPM$_{2.5\text{E}}$ appeared to decreased in the medium stage, this should be considered as a result from the global economic recession. Based on the World Bank’s open data, India’s electricity production from coal sources (percent of total) exhibited a decreasing trend from 2008 to 2011; later, an intensifying increasing trend occurred in the global economic recovery period from 2011 to 2015. However, a decreasing trend of electricity production from coal sources still emerged in northern, central, and southern regions of China from 2011 to 2015.

This paper contributes towards providing not only the concrete statistical results of the spatiotemporal trends of global continental anthropogenic PM$_{2.5}$ pollution and PPM$_{2.5\text{E}}$ from 1998 to 2016, but also a method for analysing spatiotemporal trends of large, remotely-sensed datasets based on the Bayesian statistics paradigm with some advantages. However, our study also possesses some limitations. First, the multiscale, homogeneous subdivision method must be developed in order to enable the construction of a set of more realistic, multiscale statistical units. Second, the presented BSTHPRM can only detect the turning points of local trends during the study period on the basis of the determined number of turning points.

6. Conclusions

This paper presented a BSTHPRM which can self-adaptively detect local temporal turning points, consider spatial correlations, and then apply those to investigate the spatiotemporal trends of the global continental PM$_{2.5,\text{NoDust}}$ pollution and PPM$_{2.5\text{E}}$ from 1998 to 2016. We have drawn the primary, following conclusions.

Generally, a polarization phenomenon has gradually become more severe during the period from 1998 to 2016. The most severely PM$_{2.5,\text{NoDust}}$-polluted areas were northern India, southern and eastern China, and the central areas of the Indo-China Peninsula. The total areas of the high and higher PM$_{2.5,\text{NoDust}}$-polluted regions accounted for about 13.4% of the global land area, and the corresponding exposed populations were up to 4031 million, comprising nearly 56.0% of the total global population.

The areas of hot, warm, and cold spots with increasing trends of PM$_{2.5,\text{NoDust}}$ concentration were firstly contracted from the first to second stage and then expanded from the second to third stage. The two higher polluted areas, northern India and eastern and southern China, have the parallel local trends of PM$_{2.5,\text{NoDust}}$ concentration and PPM$_{2.5\text{E}}$ in the first two stages: Increasing during the early stage, then decreasing during the medium stage. In the later stage (i.e. in the recent years), northern India arose a stronger increasing trend; nevertheless, the follow-up decreasing trend still occurred in the eastern and southern China. In the early and medium stages, more than half of the areas in Europe experienced a gradual decreasing trend of PM$_{2.5,\text{NoDust}}$ concentration and PPM$_{2.5\text{E}}$. Later, more than half of the areas in Europe...
exhibited increasing trends in the recent years. North America and South America experienced similar local trends of PM2.5–8 when compared with Europe. The PM2.5–8 trend in Africa generally increased during the study period.

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