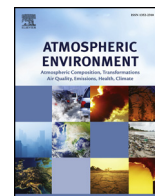




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journal homepage: www.elsevier.com/locate/atmosenvGlobal distribution and evolution of urbanization and PM_{2.5} (1998–2015)Dongyang Yang^{a,b}, Chao Ye^{a,*}, Xiaomin Wang^c, Debin Lu^{a,b,d}, Jianhua Xu^{a,b,**}, Haiqing Yang^{a,b}^a School of Geographic Sciences & Institute of Eco-Chongming, East China Normal University, Shanghai 200241, China^b Research Center for East-West Cooperation in China, East China Normal University, Shanghai 200241, China^c School of Geography, Beijing Normal University, Beijing 100875, China^d Department of Tourism and Geography, Tongren University, Tongren, Guizhou Province 554300, China

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ABSTRACT

PM_{2.5} concentrations increased and have been one of the major social issues along with rapid urbanization in many regions of the world in recent decades. The development of urbanization differed among regions, PM_{2.5} pollution also presented discrepant distribution across the world. Thus, this paper aimed to grasp the profile of global distribution of urbanization and PM_{2.5} and their evolutionary relationships. Based on global data for the proportion of the urban population and PM_{2.5} concentrations in 1998–2015, this paper investigated the spatial distribution, temporal variation, and evolutionary relationships of global urbanization and PM_{2.5}. The results showed PM_{2.5} presented an increasing trend along with urbanization during the study period, but there was a variety of evolutionary relationships in different countries and regions. Most countries in East Asia, Southeast Asia, South Asia, and some African countries developed with the rapid increase in both urbanization and PM_{2.5}. Under the impact of other socioeconomic factors, such as industry and economic growth, the development of urbanization increased PM_{2.5} concentrations in most Asian countries and some African countries, but decreased PM_{2.5} concentrations in most European and American countries. The findings of this study revealed the spatial distributions of global urbanization and PM_{2.5} pollution and provided an interpretation on the evolution of urbanization-PM_{2.5} relationships, which can contribute to urbanization policies making aimed at successful PM_{2.5} pollution control and abatement.

1. Introduction

Many regions of the world have seen a rapidly increased urbanization in the last few decades, simultaneously, PM_{2.5} pollution with its negative health impacts on humans have been one of the major social problems globally (Butt et al., 2017; World Health Organization, 2014). Urbanization transformed the natural and social-economic factors within the rural-urban territorial system and affected local PM_{2.5} pollution, and even the development of megacities and urban agglomerations could produce implications on the distribution of global PM_{2.5} concentration (Gurjar et al., 2016; Seto et al., 2017). The development level and speed of urbanization showed obvious disparities between different countries or regions, along with which was the regional disparities of PM_{2.5} concentration. PM_{2.5} pollution has been an unprepared challenge that many countries faced, while urbanization is projected to continue to present an accelerated growth in the near future (Alhowaish, 2015; World City Report, 2016). Hence, it is urgent to grasp the relationship between urbanization and PM_{2.5} pollution from a global perspective, which can contribute to urbanization policies

making to control and reduce air pollution and achieve the global urban sustainability goals.

Although urbanization theoretically has the potential to transform living conditions, including services, income, and health, toward better, it also concentrated and increased pollution sources. PM_{2.5} pollution is therefore one of the environmental changes caused by the transitions of urban components, including industry, motor vehicles, building yard et al., during the process of urbanization (Timmermans et al., 2017). The absolute numbers of urban dwellers increased 77 million per year between 2010 and 2015, and in 2015, the proportion of the world's urban population has reached to 54 percent (4 billion) according to World Cities Report in 2016 (World City Report, 2016). While, less than 20% of people living in urban areas that the PM_{2.5} concentrations met the WHO Air Quality Guideline (AQG) level (10 µg/m³), and approximately half of the population in monitored cities were exposed to air pollution that was at least 2.5 times higher than the WHO recommended level (World Health Organization, 2016). Inevitably, serious and long-term additional health risks come with the air pollution.

Researchers from multiple disciplines have conducted numerous

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studies on both the issues of PM_{2.5} and the issues of urbanization. Firstly, the development of air monitoring sites and atmospheric remote sensing has had a great progress in recent years, which provided substantial data sources for researches on PM_{2.5} pollution. Relevant studies mainly involved several aspects, including the spatial and spatiotemporal distribution, the source apportionment, and the influencing factors of PM_{2.5} (Gummeneni et al., 2011; Hao and Liu, 2016; Heimann et al., 2015; Lu et al., 2017; Yang et al., 2017). Prior studies contributed to understanding the spatiotemporal variations, chemical compositions, and driving mechanism of PM_{2.5} pollution. Secondly, urbanization was one of the hot topics for many disciplines, such as geography, economics, demography, etc., which absorbed long-term concern from governments and scholars. The basic aspects of urbanization, including increasing population, transformational industrial structure, and expanded urban space, has been widely discussed (Wei and Ye, 2014; Ye et al., 2017). Besides, dramatic transitions of these socio-economic elements within urbanization posed environmental disruption and pollution in many cities all over the world (Bekhet and Othman, 2017; Cao et al., 2014; Hu et al., 2013; Wei and Ye, 2014). Urban development, accompanied by the exacerbated environmental problems, triggered a number of studies on the relationship between urbanization and environment (Cui et al., 2015; Zhao et al., 2016). However, existing researches approved that the relationship between PM_{2.5} pollution and urbanization get less attention except a few researches which have examined the relationship between PM_{2.5} pollution and urbanization in some cities over the last few decades (Cavalcante et al., 2017; Han et al., 2016; Tuo et al., 2013). Especially, few studies were conducted to research the relationship between PM_{2.5} pollution and urbanization from a global perspective.

Therefore, this study examined the spatiotemporal variation of urbanization and PM_{2.5} pollution and their evolution relationship using global data on urbanization and PM_{2.5} concentrations from 1998 to 2015. The findings in this study would be useful to identify the relationship between urbanization and PM_{2.5} globally, assist in the urbanization policy-making to improve air quality and the realization of development goals of urban sustainability.

2. Context and literature review

As one of major environmental and social issues, PM_{2.5} pollution was seen increasing concern from multidisciplinary researchers. Researches on source apportionment of PM_{2.5} have proved that coal and biomass combustion, transport, and industry were the general sources (Chowdhury et al., 2007; Huang et al., 2014; Marcazzan et al., 2003; Timmermans et al., 2017; Wang et al., 2015). And researches have also documented that social-economic factors, including economic growth, industrial production, thermal power generation, building construction et al., drove increases in PM_{2.5} concentrations in many cities in the world (de Miranda et al., 2012; Guan et al., 2014; Hao and Liu, 2016; Li et al., 2016; Rao et al., 2017). Urban was the concentrated space of these social-economic factors and characterized by these factors, urbanization which transformed the socio-economic factors, would inevitably affect PM_{2.5} pollution. While, urbanization presented different modes, levels, and speeds in different countries and regions, and not all the urbanization deteriorated PM_{2.5} pollution. The power from environmental regulation, technical improvement, and capital could promote the improvement of air pollution (Liu et al., 2015; Rao et al., 2017; Walsh, 2014; Wang et al., 2010; Wang and Hao, 2012).

Hence, PM_{2.5} pollution should be related to the level and development speed of urbanization, as they were respectively the reflection of the development state of urban components and their transforming speed. In this aspect, Northman first proposed an S-curve to depict the general process of urbanization and urban development mode in mid-1970s (Northam, 1975), which provided a theoretical basis for understanding the transitions on population, industry, and development speed within the urbanization process (Antrop, 2000; Chen et al., 2014;

Pannell, 2002). Northman's S-curve interpreted the process of urbanization as a triple transitional process of initial stage (urbanization rate < 30%), acceleration stage (30% < urbanization rate < 70%), and terminal stage (urbanization rate > 70%). In the initial stage of urbanization, the industrial productivity was low, industry could just provide limited employment opportunity and transferred the surplus labor force slowly. Urbanization rate could reach 30 percent after several decades, even a hundred years of development. In the accelerating stage, urbanization rate rose from 30% to 70%, the industrial productivity enhanced and transferred a large rural population into urban population. Urbanization rate could pass 50% in a short time, and then went up to 70%. In the third stage, the increase in the urban population tends to be slow and even stagnant, industry transferred to tertiary industry.

It was worth noting that the Environmental Kuznets Curve (EKC) hypothesis, early verified by Panayotou (1993), Selden and Song (1994), Grossman and Krueger (1995), and others, contributed to understanding the relationship between economic growth and environmental pollution. EKC described that environmental pollution would experience a process of pollution increase, reaching a “turning point”, and pollution reduction along with the economic growth. Given that urbanization was partly a comprehensive representation of economic development, EKC has also been widely used to examine the relationship between urbanization and the issues of environmental pollution and emissions in recent decades (Bekhet and Othman, 2017; He et al., 2017; Li et al., 2012; Zhao et al., 2016).

Northman's S-curve is a generalization of urbanization process. Given the diversity of urbanization across the world and the potentially drastic changes in the accelerating stage of urbanization, we redivided the urbanization process into four sub-processes: initial stage (urbanization rate < 30%), transition stage (30% < urbanization rate < 50%), climbing stage (50% < urbanization rate < 65%), and terminal stage (urbanization rate > 65%) according to the S-curve. This paper would examine the spatiotemporal profiles of PM_{2.5} in these different urbanization stages, and inspect the evolution of urbanization-PM_{2.5} relationships according to the EKC.

3. Data and methods

3.1. Data

PM_{2.5} data is a published global PM_{2.5} concentration dataset obtained from Atmospheric Composition Analysis Group at Dalhousie University (http://fizz.phys.dal.ca/~atmos/martin/?page_id=140). This dataset provides global PM_{2.5} concentrations from 1998 to 2015 with a geographical range of from 54.995°S to 69.995°N and from 179.995°W to 179.995°E. The spatial resolution is 0.1° * 0.1°. It is the ground-level PM_{2.5} estimated by combining Aerosol Optical Depth (AOD) with the GEOS-Chem chemical transport model and subsequently calibrated by using the geographically weighted regression method based on monitoring data of PM_{2.5} (Boys et al., 2014; Van Donkelaar et al., 2016). This dataset has good accuracy ($R^2 = 0.81$) with the largest coverage and longest time span and has been used in many researches at national or regional scale (Lu et al., 2017; Luo et al., 2017; Ma et al., 2016; Pinault et al., 2016).

The percentage of urban population in the total population was regarded as urbanization rate. Data on the percentage of urban population in total population from 1998 to 2015 for all countries were obtained from the World Bank (<https://data.worldbank.org/indicator/SP.URB.TOTL.IN.ZS?view=chart>). This dataset included the urban population (% of total) in 217 countries (or regions and cities), but it was null in individual countries (or regions) or in individual years. Given the data integrality and to match the vector maps of global countries, a total number of 197 countries or regions were extracted and used to analyze in this study. Data on industrial added values (million US dollars) and annual percentage growth rate of GDP (%)

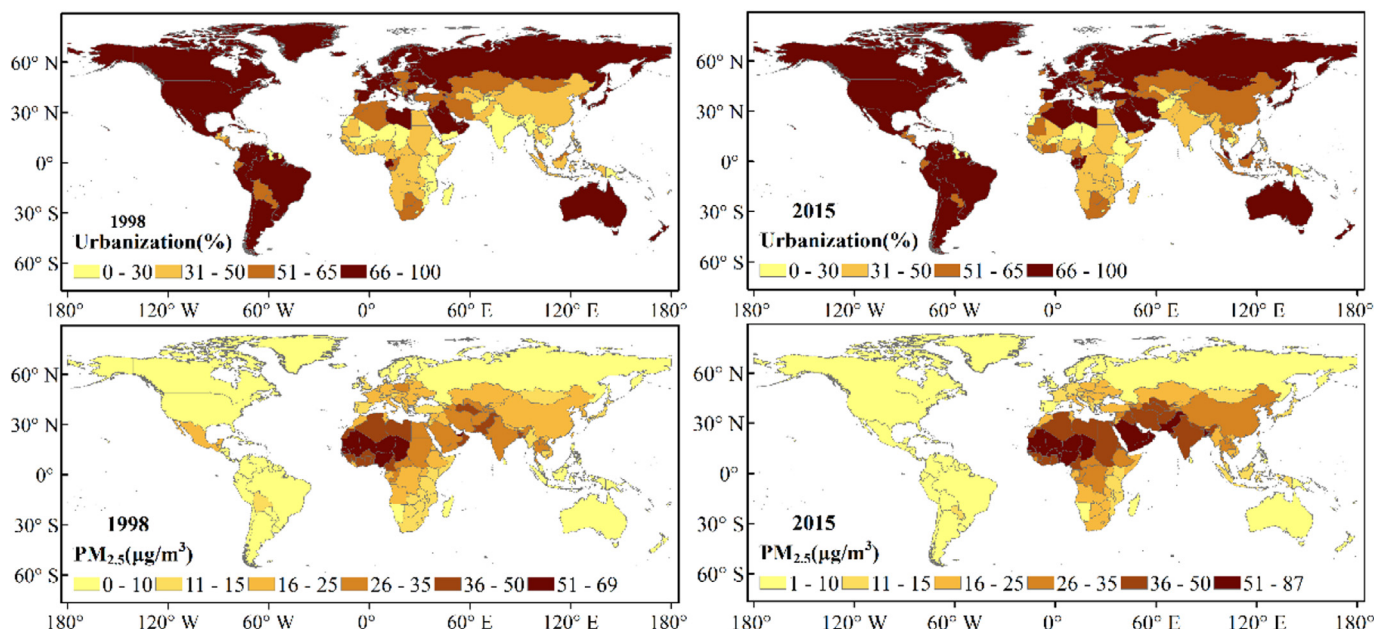


Fig. 1. Global urbanization and PM_{2.5} concentrations in 1998 and 2015.

during the study period were also gathered from the World Bank.

3.2. Methods

3.2.1. Temporal trend (slope)

The slope of urbanization rates and PM_{2.5} concentrations can be calculated to analyze their temporal trend using the unitary linear regression model. The slope can reflect the variation trend by fitting the linear relation of the attribute in each unit (country or region) and time. The formula is as follows:

$$Slope = \frac{\sum_{i=1}^n i \cdot Y_i - \frac{1}{n} (\sum_{i=1}^n i) (\sum_{i=1}^n Y)}{\sum_{i=1}^n i^2 - \frac{1}{n} (\sum_{i=1}^n i)^2}$$

where, Y donates the attribute (urbanization rate or PM_{2.5} concentration) in each research unit, n is the time span, and i is the time unit (year). If $Slope > 0$, it means the attribute increase and presents a rising trend over time. If $Slope < 0$, it means the attribute present a downward trend over time. The size of the absolute value of $Slope$ reflects the increased or decreased speed.

3.2.2. Bivariate spatial association

The bivariate spatial association is an extension of the spatial correlation analysis, which can be used to examine the spatial association of bivariate observations (Lee, 2001). This paper used the bivariate local indicators of spatial association (LISA) to identify the local spatial association type of the variation trends of PM_{2.5} and urbanization. The bivariate LISA is defined as:

$$I_{kl}^i = Z_k^i \sum_{j=1}^n W_{ij} Z_l^j$$

where, w_{ij} is the spatial weight matrix, $Z_k^i = [x_k^i - \bar{x}_k] / \sigma_k$, $Z_l^j = [x_l^j - \bar{x}_l] / \sigma_l$, x_k^i is the observation k (the Slope of PM_{2.5}) at location i , x_l^j is the observation l (the Slope of urbanization) at location j , σ_k and σ_l is the variance of x_k and x_l , respectively.

The results of bivariate LISA can be visualized using the Moran's I scatter plot (Anselin et al., 2002), which categorized the nature of spatial correlation into four groups (four quadrants divided by the vertical axis and the horizontal axis). The first quadrant and the third quadrant meant the bivariate variables in these spatial units are

positive spatial correlation, or spatial clusters (high-high and low-low). The second quadrant and the fourth quadrant meant the bivariate variables in these spatial units are negative spatial correlation, or spatial outliers (high-low and low-high) (Matkan et al., 2013). The spatial clusters or spatial outliers are the values of the bivariate variables in some spatial units where there are significant positive or negative spatial correlation. The GeoDa software was implemented to conduct the bivariate LISA analysis in this paper.

3.2.3. EKC model

Given that other socioeconomic factors, such as industrial level and economic growth, may affect the urbanization-PM_{2.5} relationship. To further examine the relationship between urbanization and PM_{2.5} under socioeconomic impact, we included the industrial added values (million US dollars), urbanization rates (%), and annual percentage growth rate of GDP (%) into the following EKC model:

$$Y_{it} = \alpha + \beta_1 x_{it} + \beta_2 x_{it}^2 + \beta_3 U_{it} + \beta_4 G_{it} + \varepsilon_{it}$$

where, Y_{it} is the PM_{2.5} concentrations in country i in year t ; x_{it} , U_{it} , and G_{it} is respectively the industrial added values, urbanization rates, and annual percentage growth rate of GDP in country i in year t . Thus, β_3 is the expected coefficient to reveal the correlations between urbanization and PM_{2.5} with considering socioeconomic impact.

4. Results

4.1. The spatial distribution of global urbanization and PM_{2.5}

Based on the four sub-stages of urbanization mentioned above, global countries were classified and mapped according to their urbanization stages (Fig. 1). From Fig. 1 (a), we could find that the countries in the terminal stage of urbanization were mainly distributed in Europe and America in 1998. Some countries in other continents, such as Gabon and Libya in Africa, Australia and New Zealand in Oceania, South Korea and Japan in East Asia, Singapore and Brunei in Southeast Asia, and several countries in Arabian Peninsula were also found in the terminal stage. While, most countries in Southeast Asia, South Asia, Africa, and China, were found with lower urbanization rate. In 2015, obvious changes mainly occurred in countries with lower urbanization rate formerly in Asia and Africa, some of which strode into a higher

Table 1
Urbanization rate and PM_{2.5} in 1998 and 2015 and the variation trend in countries in the four urbanization stages.

| Urbanization stage | PM _{2.5} (μg/m ³) | | Urbanization (%) | | Slope | |
|--------------------|--|-------|------------------|-------|-------------------|--------------|
| | 1998 | 2015 | 1998 | 2015 | PM _{2.5} | Urbanization |
| Initial stage | 17.97 | 23.35 | 20.32 | 25.32 | 0.23 | 0.29 |
| Transition stage | 21.23 | 24.15 | 39.00 | 45.24 | 0.03 | 0.37 |
| Climbing stage | 15.17 | 16.24 | 56.80 | 63.07 | 0.02 | 0.37 |
| Terminal stage | 14.18 | 17.75 | 79.68 | 82.75 | 0.13 | 0.18 |
| Total | 16.91 | 20.17 | 53.59 | 58.46 | 0.10 | 0.29 |

urbanization stage from a lower urbanization stage, such as India entered into the transition stage from the initial stage and China entered the climbing stage from the transition stage (Fig. 1b). The distributions of PM_{2.5} were opposite to the distribution of global urbanization both in 1998 and 2015 generally, except several countries partly or mostly covered by tropical or temperate desert. PM_{2.5} concentrations increased obviously in most Asian countries and some countries in Sub-Saharan Africa in 2015, compared with that in 1998.

4.2. The temporal variation of urbanization and PM_{2.5}

4.2.1. The temporal variation of global urbanization and PM_{2.5}

The statistical results of urbanization rate and PM_{2.5} in 1998 and 2015, and the variation trend (the fitted slope) during the study period were shown in Table 1. Global mean urbanization rate was 53.59% in 1998, and reached to 58.46% in 2015. The mean PM_{2.5} concentrations was 16.91 μg/m³ in 1998, and reached to 20.17 μg/m³ in 2015. The fitted slope of global urbanization was 0.29, higher than the slope of PM_{2.5} concentrations (Slope = 0.10). It indicated that both global urbanization and PM_{2.5} generally increased in the study period, while the increased trend of urbanization was rapid and the increased trend of PM_{2.5} was relatively slow. Their specific temporal variations were shown in Fig. 2. It could be found that global mean urbanization rate increased progressively during the study period, the mean PM_{2.5} concentrations presented an upward trend generally but appeared drastic fluctuations from 2008. It indicated that PM_{2.5} generally increased along with urbanization in the study period, especially the increased trend was significant before 2008.

4.2.2. The temporal variation of urbanization and PM_{2.5} in countries in different urbanization stages

The means of urbanization rate and PM_{2.5}, and their slopes in countries in the four urbanization stages were calculated to investigate the profiles of urbanization and PM_{2.5} pollution and their variation trends in different urbanization stages. In 1998, the mean PM_{2.5} concentration in transition stage was the highest (21.23 μg/m³), followed by 17.97 μg/m³ in the initial stage. In 2015, the mean PM_{2.5} concentration in transition stage was 24.15 μg/m³, followed by 23.35 μg/m³ in the initial stage, far higher than PM_{2.5} in the other two urbanization stage (Table 1). Urbanization rate presented the slowest growth trend in the terminal stage and presented the fastest growing trend in both the transition stage and climbing stage. While, PM_{2.5} increased fastest in countries in the initial stage and presented slower growth trends in transition stage and climbing stage.

The specific temporal variations of average urbanization rate and PM_{2.5} in the four urbanization stages were shown in Fig. 3. It could be found that the average urbanization rate of countries in the four urbanization stages all presented an increasing trend. While, PM_{2.5} presented a complex temporal variation in different urbanization stages. PM_{2.5} presented fluctuation according to time but presented the overall trend of growth in the initial stage and terminal stage. In the transition stage, the variation of PM_{2.5} was relatively stable, but it increased greatly in 2015. In the climbing stage, it showed an obvious fluctuation and increased year by year after 2011.

4.2.3. The spatial distribution of temporal trends for urbanization and PM_{2.5} in different countries

The slope fitted using unary linear regression method in different countries were mapped (Fig. 4) to show their regional differences. From Fig. 4a, we can see that the countries with slow growth in urbanization were mainly concentrated in several areas including Europe, Central Asia, North America, both the north and the south of South America, and some countries in West Asia, North Africa, and Oceania. Mongolia, China, Japan, and parts of countries in Indo-China Peninsula have seen the fastest growth in urbanization, the growth in urbanization was also faster in some countries, such as archipelagic countries in Southeast Asia, some countries in West Africa, Iran, and Tanzania. The countries with a faster increase in PM_{2.5} were mainly concentrated in Asia except some Central Asian countries. Several countries in east and south Africa have also seen a fast increase in PM_{2.5}. The countries with slow or negative PM_{2.5} growth were mainly concentrated in some countries in Europe, North America, and some countries near the Gulf of Guinea

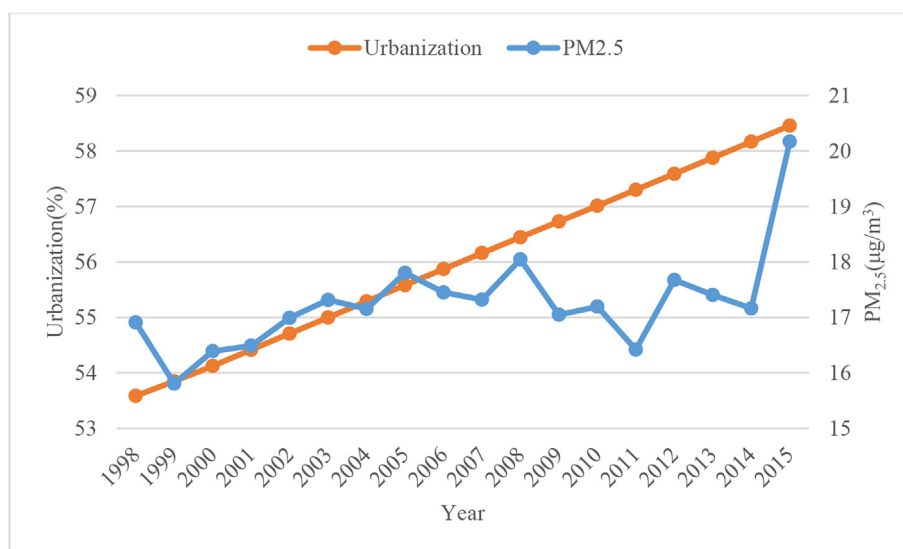


Fig. 2. Temporal variation of global urbanization rate and PM_{2.5} concentrations from 1998 to 2015.

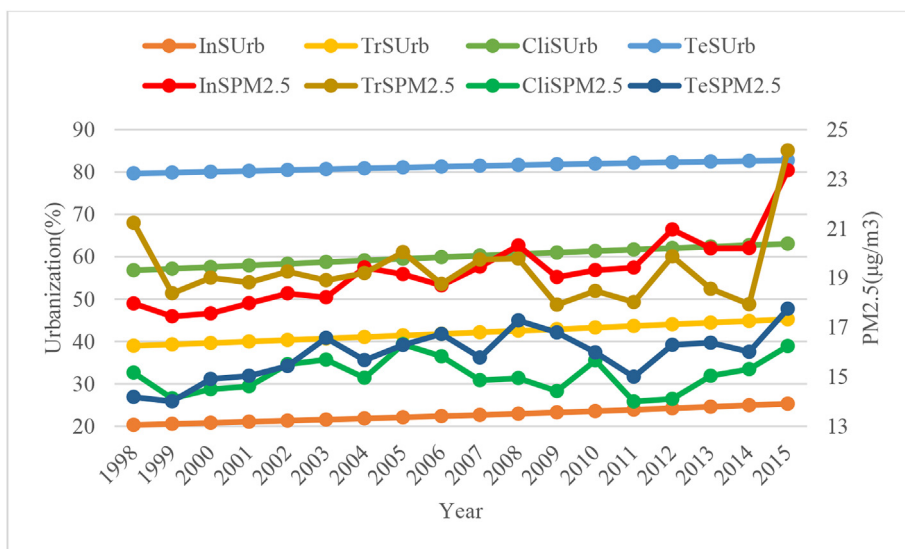


Fig. 3. The time variation of urbanization and PM_{2.5} for countries in the four urbanization stages: (a) initial stage, (b) transition stage, (c) climbing stage, (d) terminal stage.

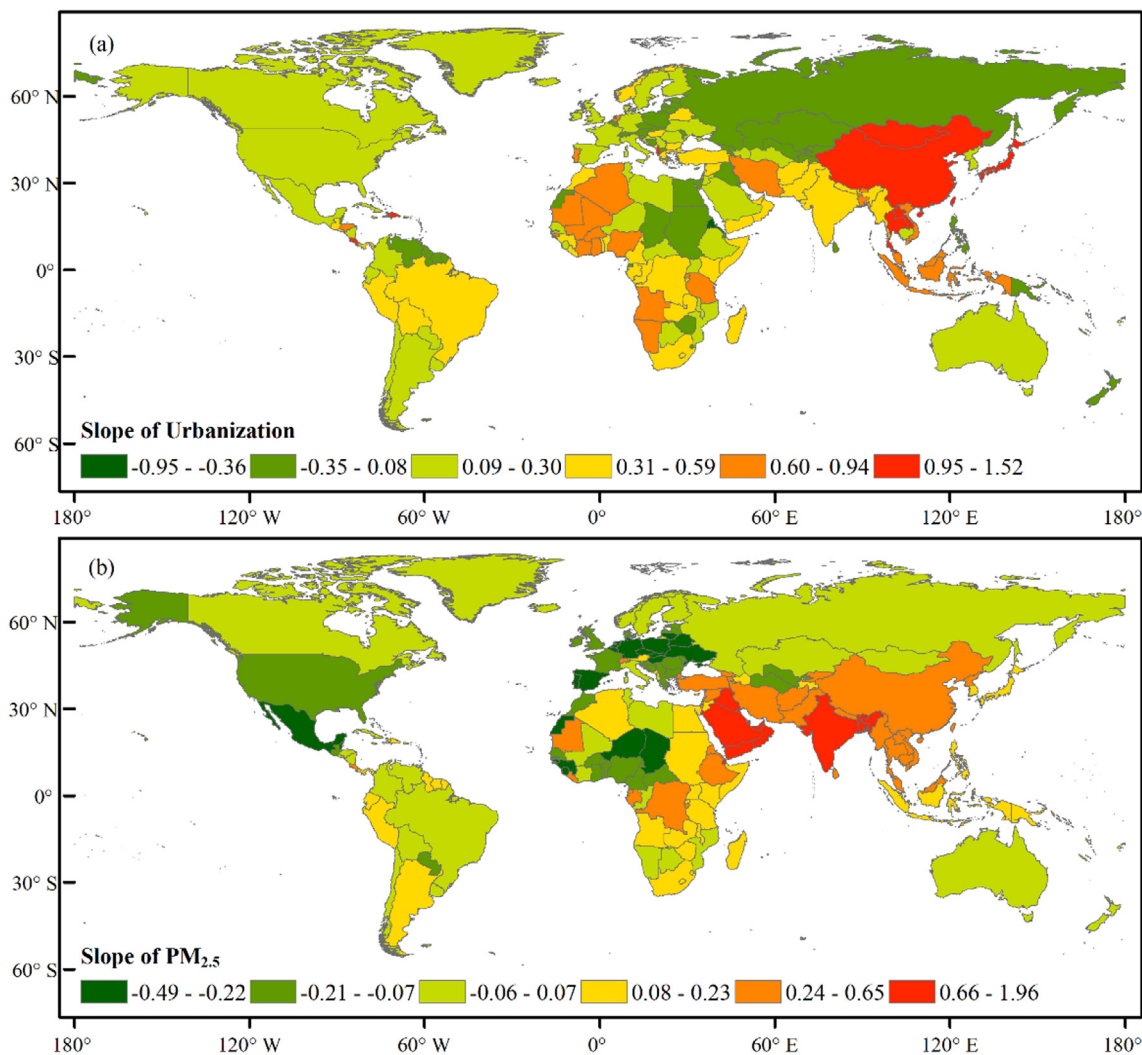


Fig. 4. The slope of global urbanization rates (a) and PM_{2.5} concentrations (b) from 1998 to 2015.

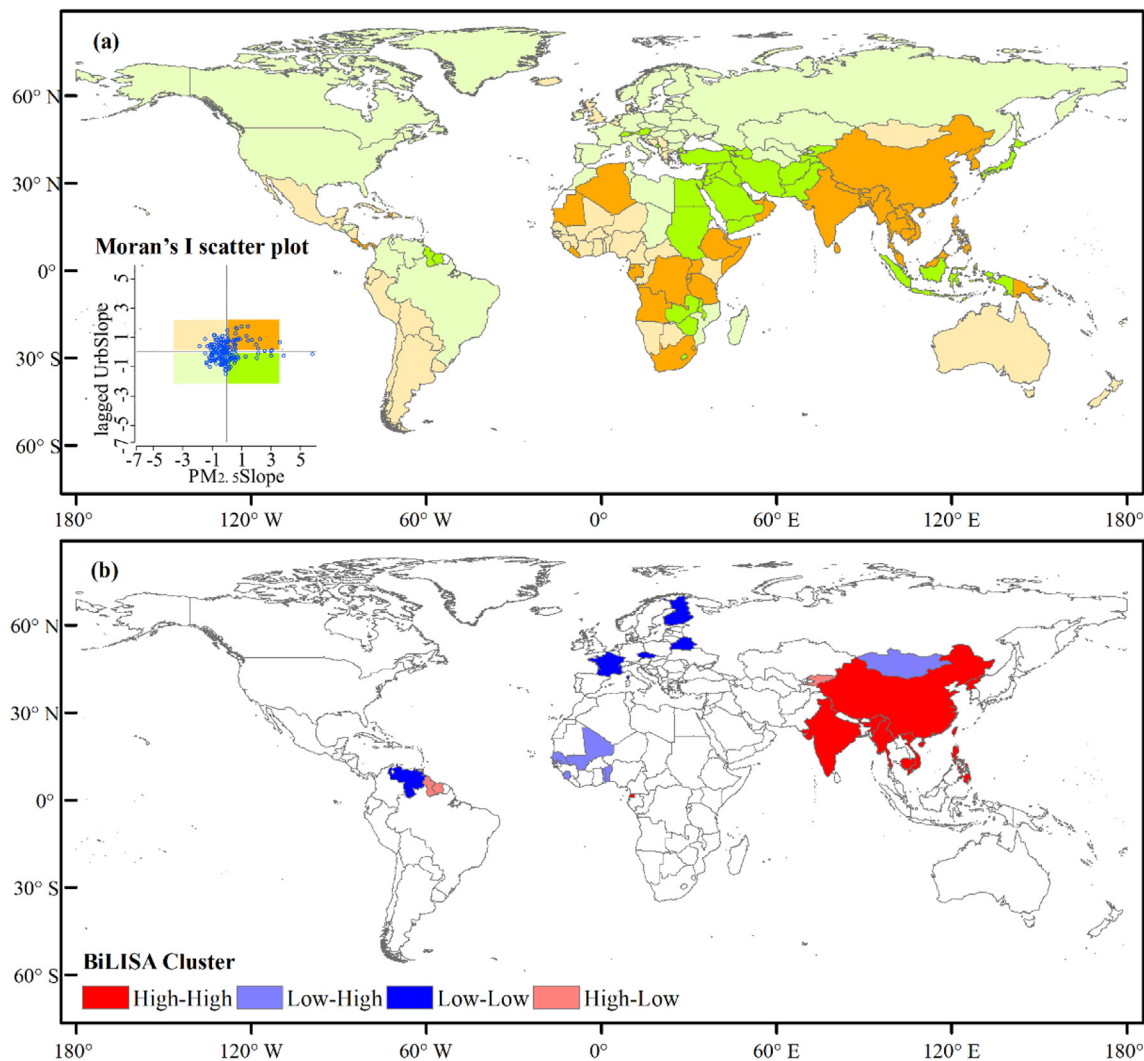


Fig. 5. The map of Moran's I scatter plot and the LISA.

(Fig. 4b).

4.3. The evolutionary relationships between urbanization and PM_{2.5}

4.3.1. Bivariate local spatial association

The bivariate local spatial association analysis was conducted based on the slope of PM_{2.5} and the slope of urbanization rate. According to results of the Moran's I scatter plot, the countries in the four quadrants were mapped in Fig. 5a. Most countries in South Asia and East Asia and some African countries were located in the first quadrant, showing the association of high growth in PM_{2.5} and high growth in urbanization. While most European and American countries were located in the third quadrant, showing the association of low growth in PM_{2.5} and low growth in urbanization. Fig. 5b showed the bivariate spatial clusters and the countries where the spatial association of the bivariate was significant. The spatial cluster of high growth in both urbanization and PM_{2.5} covered China, Korea, most countries in Southeast Asia and South Asia, Aruba, and Equatorial Guinea. The spatial cluster of low growth in both urbanization and PM_{2.5} covered Central Pacific countries including American Samoa, French Polynesia, and Tonga, Central America countries including Venezuela, Antigua and Barbuda, St. Kitts and Nevis, Barbados, Dominica, Grenada, St. Vincent and Grenadines in central America, and European countries including France, Czech, Belarus, and Finland. Mongolia, Sierra Leone, Senegal, Mali, Benin, and Togo in West Africa, Bahamas, and Turks and Caicos Islands, were the

spatial outliers with the evolutionary relationship of low growth in PM_{2.5} and high growth in urbanization. And Guyana, Trinidad and Tobago, British Virgin Islands, Suriname, and Kyrgyz were the spatial outliers with the evolutionary relationship of high growth in PM_{2.5} and low growth in urbanization.

4.3.2. Further verification of the urbanization-PM_{2.5} evolutionary relationships

Based on the above urbanization-PM_{2.5} spatial association identified by Moran's I scatter plot, we put the countries which were located in the same quadrant in one panel and estimate the EKC model to further verify the urbanization-PM_{2.5} relationship. Two kinds of estimation methods, including fixed effects and random effects, were used to estimate the coefficients using the “plm” function in the R statistical software. The estimated coefficient for urbanization rates were

Table 2
Results of estimated coefficient.

| Coefficient | | First quadrant | Second quadrant | Third quadrant | Fourth quadrant |
|----------------|-----------|----------------|-----------------|----------------|-----------------|
| Fixed effects | β_3 | 0.4583 | -0.1327 | -0.0958 | 0.3068 |
| | <i>p</i> | < 0.001 | 0.003 | 0.002 | < 0.001 |
| Random effects | β_3 | 0.3901 | -0.1931 | -0.0847 | 0.1904 |
| | <i>p</i> | < 0.001 | < 0.001 | 0.002 | 0.002 |

extracted and shown in Table 2. The coefficients were significantly positive ($p < 0.01$) in the first quadrant and fourth quadrant countries, indicating that the development of urbanization increased $PM_{2.5}$ concentrations in these countries; the coefficients were significantly negative ($p < 0.01$) in the second quadrant and third quadrant countries, indicating that the increasing urbanization reduced $PM_{2.5}$ concentrations in these countries.

5. Discussion and conclusion

This study examined global distribution and evolution of urbanization and $PM_{2.5}$ during 1998–2015 based on the data for the proportion of the urban population and $PM_{2.5}$ concentrations. The spatial distribution of global urbanization and $PM_{2.5}$ pollution showed that $PM_{2.5}$ concentrations were low in countries with high urbanization rates in Europe and America, and were high in most countries in Southeast Asia, South Asia, Africa, and China with low urbanization rates. The temporal variations showed that global mean $PM_{2.5}$ generally presented a rising trend along with the development of urbanization, $PM_{2.5}$ concentration was the highest in countries in the transition stage of urbanization, but the rising trend was the highest in countries in the initial stage of urbanization. Most Asian countries and some countries in Northwestern Africa and South Africa showed a high increase in both urbanization and $PM_{2.5}$, while most European and American countries showed a low increase in both urbanization and $PM_{2.5}$. Urbanization increased $PM_{2.5}$ concentrations in Asian and African countries with low urbanization rates, but decreased $PM_{2.5}$ concentrations in European and American countries with high urbanization rates.

The findings of the spatial distribution and temporal variation of global urbanization and $PM_{2.5}$ were consistent with previous studies (World City Report, 2016; World Health Organization, 2016). The general distribution of urbanization and $PM_{2.5}$ approved the EKC hypothesis, as $PM_{2.5}$ concentrations were high in most low urbanization countries and were low in most high urbanization countries. While, it was worth noting that urbanization rates were high in some countries in the tropical deserts, such as Libya and Saudi Arabia, where $PM_{2.5}$ were also high. The high $PM_{2.5}$ concentrations should be mainly caused by the source of desert pollution in this area. As Northman's S-curve described that the transforming speed of urban components, including industry and population, was slow in the early stage of urbanization (Northam, 1975). The sources of $PM_{2.5}$ pollution were less and the $PM_{2.5}$ concentrations were low when urbanization level was low, which was also approved by the EKC (Grossman and Krueger, 1995; Panayotou, 1993; Selden and Song, 1994). Hence, it was not difficult to understand that $PM_{2.5}$ concentrations were low and increased slowly in some southeast African countries, where the urbanization rates were lower than 25%. However, the mean $PM_{2.5}$ concentrations were still low in countries in the two higher stages of urbanization and were high in countries in the two lower stages of urbanization. The higher initial value of $PM_{2.5}$ may affect or limit the rising range of $PM_{2.5}$ in the transition stage and the increasing trend of $PM_{2.5}$ may change in the climbing stage, so as that the increasing trend were low in the two urbanization stages (Table 1).

The bivariate spatial association analysis captured the local association between the variation trends of urbanization and $PM_{2.5}$ during the study period. The slow, even negative increasing trends in both $PM_{2.5}$ and urbanization in most countries with high urbanization levels and the rapidly increasing trends in both $PM_{2.5}$ and urbanization in most countries with low urbanization levels were disclosed. As $PM_{2.5}$ presented rapid increase trend in some countries with high urbanization where were most or partly covered by tropical desert or subtropical desert, and other countries, such as Japan, Indonesia, and several countries in the southeast Africa, $PM_{2.5}$ increased faster than urbanization, these countries were identified as countries with high growth in $PM_{2.5}$ and low growth in urbanization. While, $PM_{2.5}$ increased slower than urbanization in Mongolia, Australia, New Zealand, and some West

African countries, these countries were identified as countries with low growth in $PM_{2.5}$ and high growth in urbanization (Fig. 5a). Further, based on the identified spatial association, the evidence from the urbanization- $PM_{2.5}$ evolutionary relationships showed that there was a positive correlation between urbanization and $PM_{2.5}$ in most Asian countries and some African countries with low urbanization rates, but there was a negative correlation between them in most European and American countries with high urbanization rates (Table 2), which also approved the EKC hypothesis.

There were several aspects of limitation in this study. The first was the issue of data uncertainty of $PM_{2.5}$. The raw data for $PM_{2.5}$ were the estimations based on AOD, this data showed different uncertainty in different regions (Van Donkelaar et al., 2016). The statistical average was used which neglected the internal spatial differences for each country. Second, the spatial weight can affect the result of spatial clusters and spatial outliers, the different spatial weight may lead some changes in the result. While, the countries were spread over multiple continents and islands, and had different areas, which affected the results of the spatial weight. Besides, not only the development of urbanization and other socio-economic factors could influence the $PM_{2.5}$ concentrations, but the natural factors could affect the interannual variation of $PM_{2.5}$ and affected the periodical variation trend, such as the increase in sandstorm would greatly increase the $PM_{2.5}$ concentrations. And, the time span of the $PM_{2.5}$ data was not long enough and could be used to reveal the long-term trend. Hence, the temporal variation of global mean $PM_{2.5}$ and the temporal variations of the mean values calculated according to urbanization stages do not seem to be powerful enough in supporting the urbanization- $PM_{2.5}$ EKC.

In conclusion, $PM_{2.5}$ concentrations presented a rising trend along with the rapid urbanization from 1998 to 2015, but the evolution relationship between $PM_{2.5}$ and urbanization rate was complex in different countries and regions. Most countries in East Asia, Southeast Asia, South Asia, and some African countries urbanized rapidly, together with high risk of $PM_{2.5}$ pollution, which are often countries with high-speed industrialization. The findings imply that some reasonable or healthy urbanization policies should be carried out to tackle $PM_{2.5}$ pollution. Furthermore, because of borderless and moving $PM_{2.5}$ pollution, international cooperation can contribute to combatting $PM_{2.5}$ pollution, especially in Asian countries.

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