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The relationships between $PM_{2.5}$ and aerosol optical depth (AOD) in mainland China: About and behind the spatio-temporal variations☆

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ABSTRACT

Satellite aerosol products have been widely used to retrieve ground $PM_{2.5}$ concentration because of their wide coverage and continuous spatial distribution. While more and more studies have focused on the retrieval algorithms, the foundation for the retrieval—relationship between $PM_{2.5}$ and satellite aerosol optical depth (AOD) has not been fully investigated. In this study, the relationships between $PM_{2.5}$ and AOD were investigated in 368 cities in mainland China from February 2013 to December 2017, at different temporal and regional scales. Pearson correlation coefficients and the $PM_{2.5}$/AOD ratio were used as indicators. Firstly, we established the relationship between $PM_{2.5}$ and AOD in terms of the spatio-temporal variations, and discuss the impact of some potential factors for a better understanding of the spatio-temporal variations. Spatially, we found that the correlation is higher in the Beijing-Tianjin-Hebei and Chengyu regions and weaker in coastal areas. The $PM_{2.5}$/AOD ratio shows an obvious north—south difference, with the ratio in North China higher than South China. Temporally, the correlation coefficient tends to be higher in May and September, with the $PM_{2.5}$/AOD ratio higher in winter and lower in summer. As for interannual variations, we detected a decreasing tendency for the $PM_{2.5}$-AOD correlation and $PM_{2.5}$/AOD ratio for recent years. Then, to determine the impact of the weakening of the $PM_{2.5}$-AOD relationship on $PM_{2.5}$ remote sensing retrieval performance, a geographically weighted regression (GWR) retrieval experiment was conducted. The results showed that the performance of retrievals is also decreasing while $PM_{2.5}$-AOD relationship getting weaker. Our study investigated the $PM_{2.5}$-AOD relationship over a large extent at the city scale, and investigated the temporal variations in terms of interannual variations. The results will be useful for the satellite retrieval of $PM_{2.5}$ concentration and will help us to further understand the $PM_{2.5}$ pollution situation in mainland China.

1. Introduction

Fine particulate matter or $PM_{2.5}$ (particles with aerodynamic diameters of less than 2.5 μm) has attracted the public's concern due to its adverse impact on human health (Lelieveld et al., 2015; Ho et al., 2018). The spatially continuous mapping of $PM_{2.5}$ is substantially required for determining the population exposure to $PM_{2.5}$, thus making the use of satellite aerosol optical depth (AOD) product for the retrieval of surface $PM_{2.5}$ popular (Al-Saadi et al., 2005; Zhang et al., 2009; Boys et al., 2014; van Donkelaar et al., 2015a; van Donkelaar et al., 2015b; Jung et al., 2017; Li et al., 2017a; Li et al., 2017b; Shen et al., 2018; Guo et al., 2017b de Hoogh et al., 2018; Xu et al., 2018; Li et al., 2018). An important premise and theoretical foundation for retrieving surface $PM_{2.5}$ concentration with satellite AOD is the strong correlation and connection between $PM_{2.5}$ and AOD (Li et al., 2015). However, the correlation between $PM_{2.5}$ and AOD is not always solid and there are many differences between $PM_{2.5}$ and AOD (Zheng et al., 2017; Zhang and Li, 2015). For example, $PM_{2.5}$ mainly represents the turbidity of the atmosphere near the ground. In contrast, AOD

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represents the whole atmospheric column, which extends from the ground surface to an altitude of several hundred kilometers. Furthermore, $PM_{2.5}$ mainly represents the dry mass concentration of fine particulate, which is hardly affected by water and coarse particles, but the value of AOD includes the influence of water vapor and coarse particles. Apart from this, in essence, the value of $PM_{2.5}$ represents mass concentration, whereas the AOD value represents the extinction ability. The connection between mass concentration and extinction ability can be either strong or weak as the composition of $PM_{2.5}$ and aerosol varies. Thus, the relationship between $PM_{2.5}$ and AOD can be affected by many factors like relative humidity, planetary boundary height, aerosol properties etc. (Zheng et al., 2017; Zhang and Li, 2015). In different conditions, these factors may affect the relationship to a different degree. Therefore, before retrieving $PM_{2.5}$ concentration through AOD in a large area for a long time, it is necessary to explore the relationship between $PM_{2.5}$ and AOD for the same large spatial and temporal extent, and to establish the spatial and temporal variations of the relationship at a fine scale. Only by doing this can we determine whether or not the foundation is always solid when retrieving $PM_{2.5}$ with AOD in a large area over a long period of time.

Mainland China has suffered serious $PM_{2.5}$ pollution in recent decades. The current $PM_{2.5}$-AOD relationship analysis studies for mainland China are either confined to a small area (Shao et al., 2017; Ma et al., 2016; Wang et al., 2018a) or have lacked detailed investigation of the spatial variations though with a large spatial extent (Guo et al., 2009; Guo et al., 2017a). Besides, the studies of the temporal variations of the $PM_{2.5}$-AOD relationship have mainly focused on the seasonal variation (Li et al., 2015; Ma et al., 2016), and interannual variations have been ignored, which is very worthy of study because the $PM_{2.5}$ pollution in mainland China has changed in recent years (Lin et al., 2018). To understand the $PM_{2.5}$-AOD relationship in mainland China more comprehensively and thoroughly, the study of a large area for a long time range, with comparisons for different cities and periods, is needed.

Our study was aimed at comprehensively investigating the relationships between $PM_{2.5}$ concentration and satellite AOD in mainland China, with an emphasis on the spatial distribution pattern and temporal variations, especially interannual variations. In this study, we explored the $PM_{2.5}$-AOD relationship in 368 cities and nine urban agglomerations based on a 59-month record of observations from February 2013 to December 2017, and the relationships were measured by Pearson correlation coefficient and the $PM_{2.5}$/AOD ratio. Mainly 3 aspects of work have been done: 1) spatio-temporal variations of $PM_{2.5}$-AOD relationship were explored in city, region, month and year scale; 2) the impact of some of the influencing factors for the $PM_{2.5}$-AOD relationship, including the aerosol type, relative humidity (RH), topography, and planetary boundary layer height (PBLH) were discussed; 3) the impact of the interannual variations of $PM_{2.5}$-AOD relationship on satellite retrieval accuracy for $PM_{2.5}$ concentration was validated. The conclusions of this study could provide useful information for $PM_{2.5}$ retrieval; for example, the spatial and temporal variation patterns of the $PM_{2.5}$-AOD relationship could provide a reference for the development of spatially and temporally self-adaptative retrieval algorithms. Furthermore, the investigation of the influencing factors for the $PM_{2.5}$-AOD relationship could help to improve our understanding of the formation mechanisms of air pollution.

2. Data and methods

2.1. Study area and period

The study domain covered 368 cities and nine urban agglomerations in mainland China, as shown in Fig. 1. The nine urban agglomerations were the Beijing-Tianjin-Hebei Urban Agglomeration (BTH), the Yangtze River Delta urban agglomeration (YRD), the Pearl River Delta urban agglomeration (PRD), the Central Plain urban agglomeration (CP), Chengyu urban agglomeration (CY), the Yangtze River Mid-Reaches urban agglomeration (YRM), the Shandong Peninsula urban agglomeration (SP), the Mid-Southern Liaoning urban agglomeration (MSL), and the Hachang urban agglomeration (HC), all of which are national urban agglomerations approved by the National Assembly of China. These nine national urban agglomerations are densely populated and economically developed areas in mainland China, and have an important position in the development of national strategy. Therefore, we chose them to be the objects of the regional-scale study. The detailed information for the nine urban agglomerations is listed in Supplementary Table 1. The time range of our study was from February 2013 to December 2017, i.e., nearly 59 months (5 years) in total.

2.2. Data collection and methodology

Hourly $PM_{2.5}$ concentration data for 2013 to 2017 were downloaded from the Data Center of the Ministry of Environmental Protection of China (http://datacenter.mep.gov.cn/index). The daily $PM_{2.5}$ concentration was averaged from the hourly values. The $PM_{2.5}$ concentration is measured by tapered element oscillating microbalance (TEOM) or beta attenuation monitor.

The satellite AOD product used in this study was the MODIS level 2 daily AOD data from the Terra MOD04_L2 Collection 6, which are reported at 10×10 km, with uncertainty levels of $\pm 0.05 \pm 0.20 \times$ AOD over land (Chu, 2002; Levy et al., 2007; Levy et al., 2013). In this product, the AOD data over land with low quality (quality flags = 1,2) has been removed to assure the data quality. The product was downloaded from the NASA Level-1 and Atmosphere Archive and Distribution System (LAADS) Distributed Active Archive Center (DAAC) (https://ladsweb.modaps.eosdis.nasa.gov/).

Meteorological data including Relative humidity (RH), air temperature (TMP), u-wind (UW), v-wind (VW), and pressure (PS) was also prepared in our study. They were downloaded from the NCEP/NCAR Reanalysis 1: Surface data (https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.surface.html) with a resolution of $2.5^{\circ} \times 2.5^{\circ}$.

The relationship between daily $PM_{2.5}$ concentration and AOD was measured by Pearson correlation coefficients (r) and the $PM_{2.5}$/AOD ratio (η). The ratio between $PM_{2.5}$ concentration and AOD was first promoted by van Donkelaar et al., in 2010 as a conversion factor (Van Donkelaar et al., 2010), and the parameter indicates the dry mass concentration of $PM_{2.5}$ per unit aerosol optical thickness. A previous study proved that the $PM_{2.5}$/AOD ratio is a good parameter to measure the relationship between $PM_{2.5}$ concentration and AOD (Zheng et al., 2017). Hence, we introduced this parameter in our work, in addition to the correlation coefficients, for a more comprehensive analysis of the $PM_{2.5}$ and AOD relationship.

3. Results and discussion

3.1. Spatial variations of the relationship

3.1.1. Relationship analysis at the city scale

Fig. 2(a) shows the correlation coefficients between $PM_{2.5}$ concentration and AOD in the 368 cities, where the strong correlation is mostly concentrated in eastern Sichuan, Chongqing, Yunnan, the BTH region, and some cities in southern Xinjiang. In contrast, the

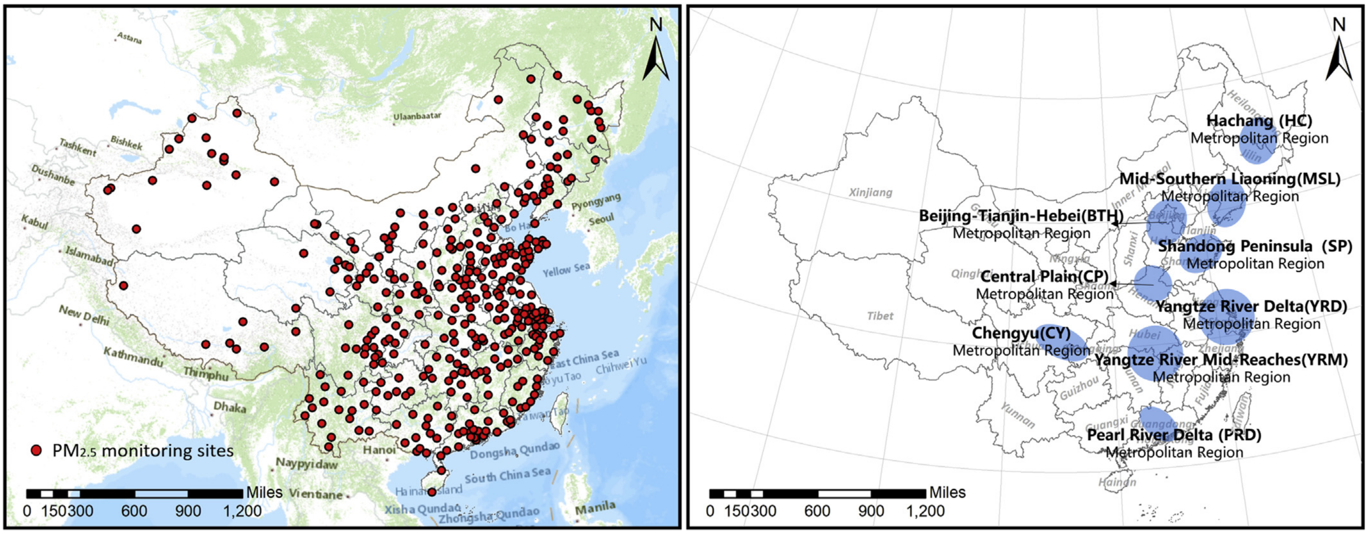

Fig. 1. Study area: (a) the 368 cities, and (b) the nine urban agglomerations in mainland China.

correlation in the PRD area, Hainan province, and some cities in the Qinghai-Gansu-Ningxia region (QGN) is quite low. This spatial difference may be the result of aerosol type and properties variations. In the PRD area, Hainan province, and QGN region, the aerosol types are mainly sea salt and dust (Mielonen et al., 2009) with a low fine mode faction (Supplementary Fig. 1), which means the AOD in these regions mainly comes from coarse particles like dust and sea salt. However, the main sources for $PM_{2.5}$ in these regions are secondary particles, coal burning, industrial emissions, and vehicle emissions (Tan et al., 2017; Wang et al., 2006). $PM_{2.5}$ and AOD have different kinds of sources, making the correlation between them low. In comparison, the aerosol type in Sichuan, Chongqing, and the BTH region is mainly the urban-industry type (Che et al., 2015), and the fraction of fine particles is high (Supplementary Fig. 1). Thus, the AOD and $PM_{2.5}$ in these regions tend to share more common sources, making the correlation higher.

In addition to the aerosol type and properties impact, it can be inferred that topography and climate also have a great impact on the $PM_{2.5}$-AOD relationship. For instance, in Sichuan province (marked by cyan boundary in Fig. 2(b)), the correlation is high in the Mideastern Sichuan Basin, but is low in the western Sichuan plateau. The dividing line of the high and low correlation displayed by the dotted cyan line in left of Fig. 2(b) is very close to the topography boundary of Sichuan province marked in the right of Fig. 2(b). In Xinjiang province, the correlation in the south is high and is weaker in the north. The dividing line between high and low correlation is very close to the temperature and climate zone boundary as shown in Fig. 2(c). However, this partitioning does not apply for the whole mainland China, because the $PM_{2.5}$-AOD relationship is simultaneously affected by many factors, in addition to topography and climate. The synthetic impact of all the influencing factors, such as topography, climate, emissions, aerosol optical and physical properties, and even environmental policies, results in the spatial variations found in our study.

Fig. 2(d) shows that the results of the ratio distribution, there is a conspicuous south−north difference in the figure, with higher $PM_{2.5}$/AOD ratios in the north and lower ratios in the south. We find that the distribution of $PM_{2.5}$/AOD ratio is quite similar to the distribution of RH as shown in Fig. 2(e), with high ratio in low RH regions and low ratio in high RH regions. We drew the scatterplot between the ratio and the RH (shown in the inset in Fig. 2(e)), and calculated the Pearson correlation coefficients. The correlation coefficient is −0.45, implying a strong negative correlation between $PM_{2.5}$/AOD ratio and RH. In the southern part of China, the RH is usually higher than in the north. When the environment relative humidity is high, the aerosol would be humidified and the particles tend to contain more water. The AOD represents the extinction ability of aerosol, the water contained in the particles will contribute a lot to AOD and makes the value of AOD larger. However, the concentration of $PM_{2.5}$ is dry mass concentration, where the water contained in the particles is evaporated and contributes little to $PM_{2.5}$ mass concentration in the measurements. That is to say, high humidity makes the particles in the air contain more water, thus making the AOD higher, but the impact on the concentration of $PM_{2.5}$ is relatively weak, resulting in a relatively lower $PM_{2.5}$/AOD ratio in the humid south. On the contrary, in the dry northern China, the relative humidity is low, the extinction ability of aerosol is more contributed by dry particles in the atmosphere, thus, the contribution of $PM_{2.5}$ to AOD is larger, making the ratio larger.

3.1.2. Relationship analysis at the regional scale: the study of nine typical urban agglomerations

The correlation for $PM_{2.5}$ concentration and AOD in the nine urban agglomerations are shown in Fig. 3 through the scatter plot. The Chengyu and Beijing-Tianjin-Hebei urban agglomerations have the highest correlations. Shandong Peninsula has a correlation coefficient of 0.42, following behind the CY and BTH regions. The YRM region has the lowest correlation among the nine urban agglomerations. As for the other urban agglomerations, the correlation coefficients are all between 0.37 and 0.39, which is very close to the average correlation coefficient of all the cities calculated in the last section. We note that the results at the regional scale have a slight difference with the results at the city scale. In the city scale analysis, cities in the PRD region have the lowest correlation, but the correlation is higher in the region-scale analysis, and the YRM urban agglomeration has the lowest correlation. This is because correlation at the regional scale is not a simple averaging of all the cities in the region, and the connections and relevance between these cities can affect the correlation at the regional scale. The fact that the correlation between the $PM_{2.5}$ and AOD of the PRD region is higher than the correlation of the individual cities indicates that cities in the PRD region have very similar pollution conditions. The

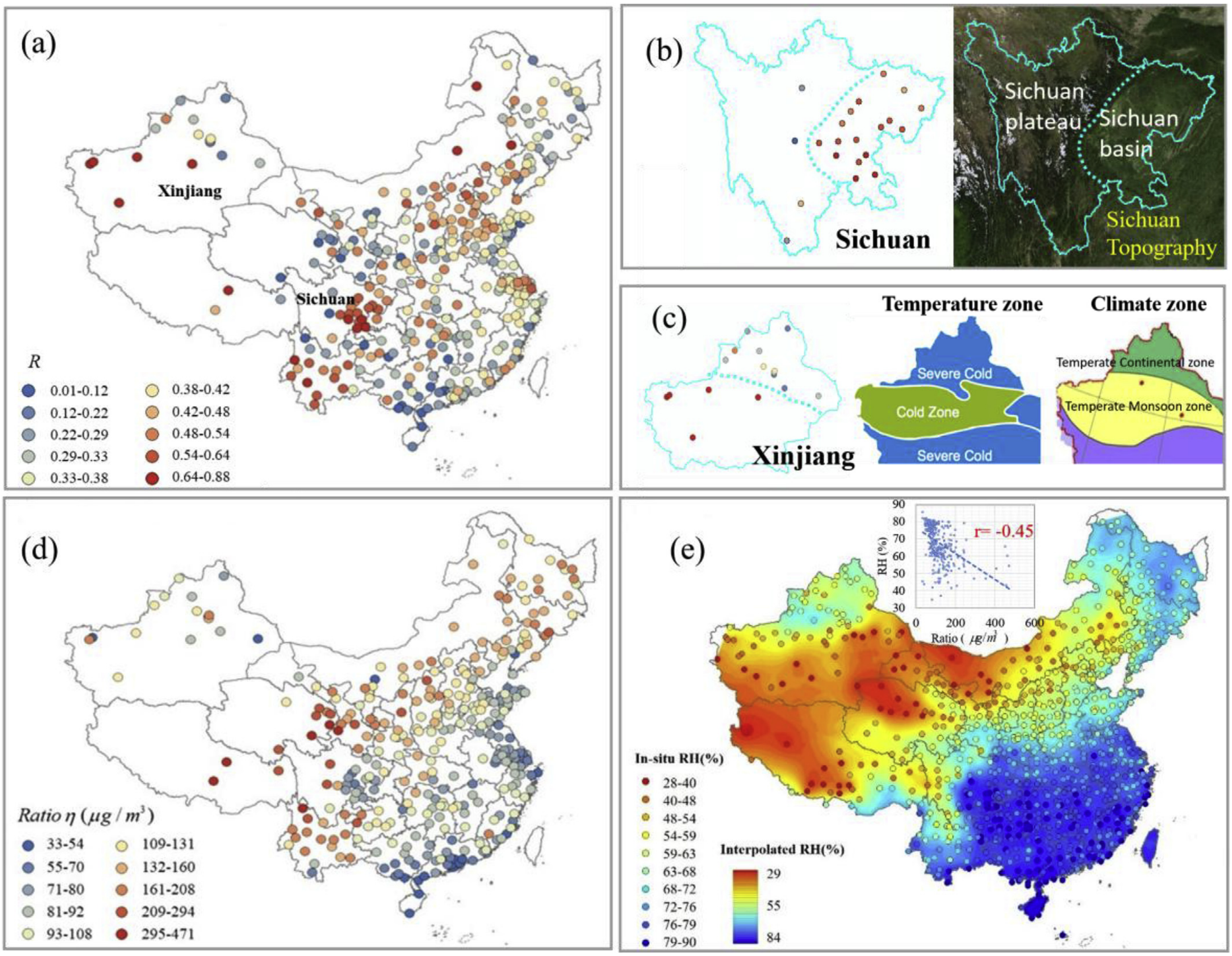

Fig. 2. The correlation coefficient (r) values and ratio (η) distribution in the 368 cities and relationships between r and topography and climate. (a) Correlation coefficients distribution. (b) Topography's impact on the distribution of r in Sichuan. (c) Climate's impact on the distribution of r in Xinjiang. (d) Ratio distribution in the 368 cities. (e) 2013–2017 five years average relative humidity distribution for mainland China. The colored dots represent the RH values measured by Chinese meteorological stations, the background color stands for the Empirical Bayesian Kriging (EBK) interpolated RH. Data source: China Meteorological Data Service Center (CMDC), http://data.cma.cn/. The inset is the scatterplot between RH and ratio. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

correlation between cities is high, making the regional correlation higher than the correlation at the city scale.

The results of the ratio calculation are shown in Fig. 3 as well, where the BTH region holds the highest ratio, followed by the HC and MSL areas, with ratios of 179 and 146, respectively. All three urban agglomerations with a high ratio are in North China, which is consistent with the results at the city scale. The ratios in the YRD, PRD, CY, and YRM areas are all below 100, which is a quite low level. As for the CP and SP areas, the ratio is at a moderate level, with $\eta = 139$ and $\eta = 111$.

To develop a comprehensive understanding of how do topography, aerosol type and meteorology influence the $PM_{2.5}$-AOD relationship, we made a simple classification of all the cities and the 9 urban agglomerations according to the values of the $PM_{2.5}$-AOD correlation and ratio, and summarized the features for each type of city and region, the results are shown in Fig. 4. The first type has a high correlation and high $PM_{2.5}$/AOD ratio, and the representative city and region are Beijing and BTH. The urban-industry type aerosol and dry climate make the correlation and ratio high. The second type includes cities such as Chengdu and Chongqing, which mainly located in or around CY region, and the correlation is high but the ratio low. Compared with BTH region, the climate in CY is more humid, which contributes to the lower ratio. Furthermore, the basin topography in eastern Sichuan also contributes to the high correlation. Features of the third type are low correlation and ratio. Representative cities include Zhuhai, Shenzhen, Haikou etc., which are mostly located in coastal areas in South China. The high proportion of sea salt in aerosol and humid climate result the low ratio and correlation. Representative region of the third type is the YRM, humid climate makes ratio low and the spatial heterogeneity make the correlation low. As for the forth type, cities of this type usually have a dry climate, and the proportion of coarse particles is high, the representative city is Lanzhou in Gansu province and Xining in Qinghai province.

3.2. Temporal variations of the relationship

3.2.1. Monthly variations

Fig. 5 shows the correlation coefficients for each month in the nine urban agglomerations from February 2013 to December 2017.

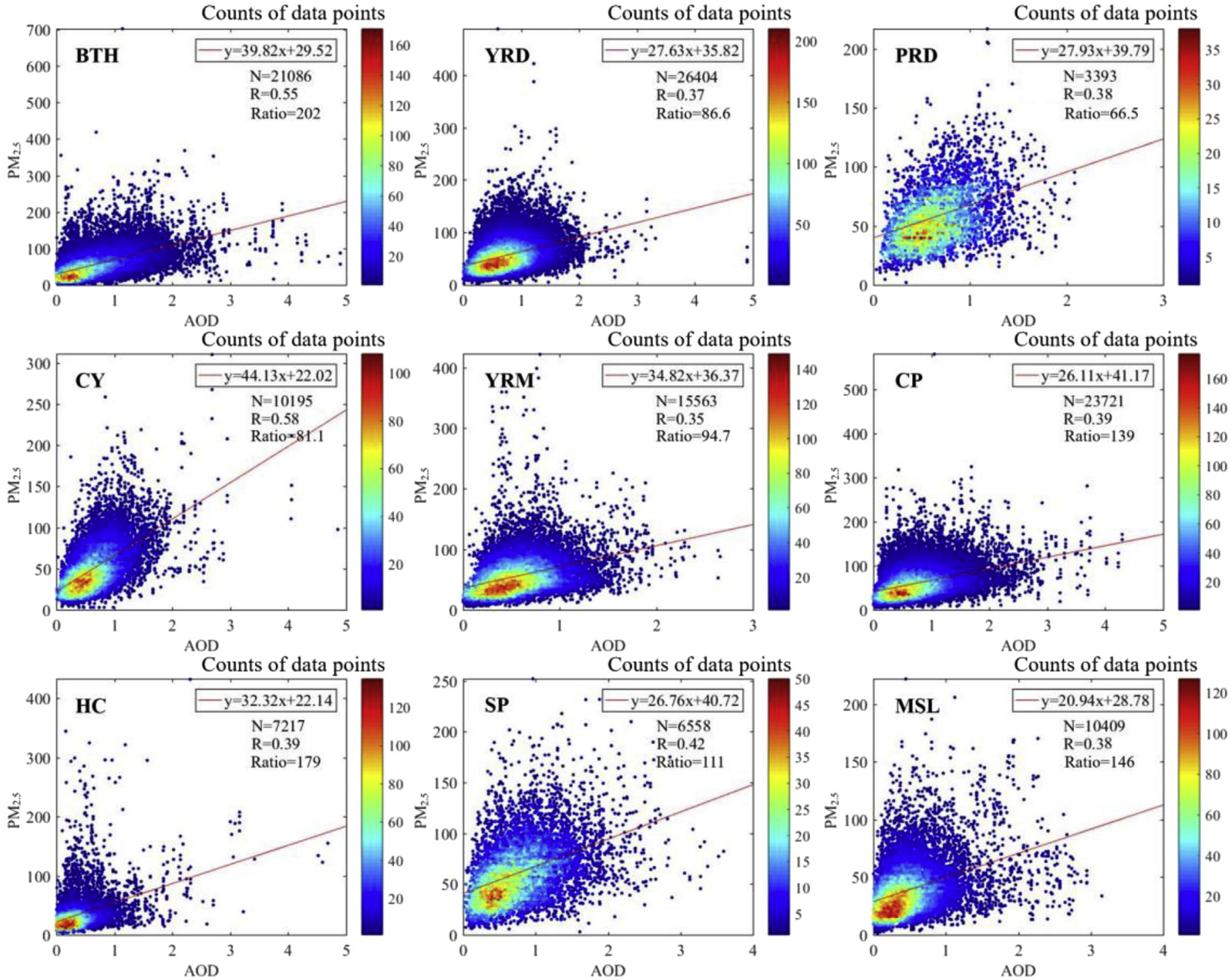

Fig. 3. Scatter plots for $PM_{2.5}$ concentration and AOD in the nine urban agglomerations (the unit of $PM_{2.5}$ concentration is μg/m^3). The number of samples, correlation coefficients and ratios of $PM_{2.5}$ and AOD are displayed in the upper right corner. The color for dots represents the density of dots. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Because the AOD is commonly missing in winter due to cloud/snow cover or high surface reflectance (Xiao et al., 2017), the correlation coefficients in the cold season are mostly missing (the small number of samples makes the significance test hard to pass). The periodic variations of *r* are not obvious. However, we can still find that different regions tend to share similar monthly patterns every year. For example, in 2014 and 2016, the monthly curves are bimodal for most regions, one peak is in May and the other in September. As for 2013, the monthly curve is unimodal for most regions, the high correlation appears in August and September. In 2017, the high correlation tends to appear in September and then reaches a trough in November. The variations of the first half of the year are weak and the curve is nearly flat and smooth. As we can see, the monthly pattern is different for each year, although in the same year, the varying pattern can be different in certain regions. On the whole, the monthly variations of *r* are quite complex and we cannot come to an easy conclusion, such as the correlation is high in spring and low in winter. However, in the terms of statistics, there is a higher probability that high correlation will appear in May and September.

We then calculated the ratios in the nine urban agglomerations. The results are shown in Fig. 6. There are obvious periodic variations, in that the ratio is usually lower in warm season and becomes higher in cold season. This difference may result from the seasonal variations of pollution and meteorology. On the one hand, some publications had shown that $PM_{2.5}$ concentration is high in winter and lower in summer (Li et al., 2017b), and for AOD the reverse (Sogacheva et al., 2018; de Leeuw et al., 2018), therefore, making the ratio high in winter and low in summer. On the other hand, the low PBLH in winter (Supplementary Fig. 2) makes the fine particles mostly concentrate in the lower atmosphere, and thus the surface $PM_{2.5}$ concentration, which is measured by ground sites and used in our paper, can account for a higher ratio in the $PM_{2.5}$ concentration for the whole atmospheric column. Hence, the $PM_{2.5}$/AOD ratio can be higher in winter. Similar results were also found in a previous work (Zheng et al., 2017). Furthermore, the RH in winter is usually lower than summer (Supplementary Fig. 3), that may also contribute to the low ratio in summer and high ratio in winter.

3.2.2. *Interannual variations*

The interannual variations of $PM_{2.5}$-AOD correlation are displayed in Fig. 7(a) and Fig. 7(b) (the results for the nine urban

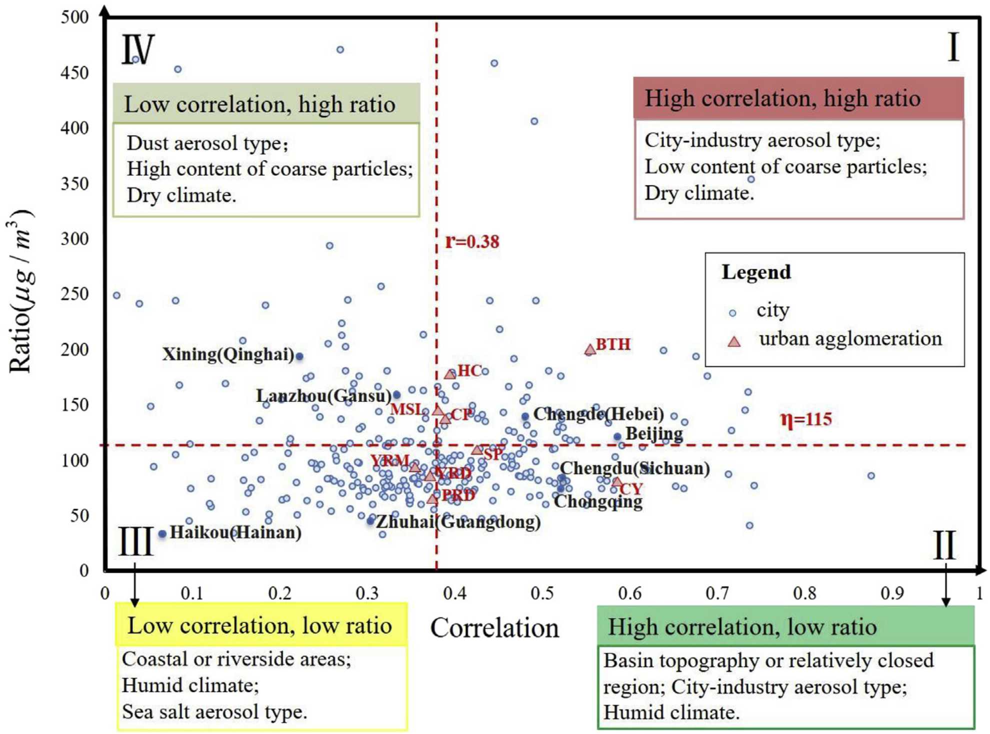

Fig. 4. The four types of correlation/ratio, and the features for each type. (Each light blue circle represents a city, and dark blue point stands for the representative city, the position of each point is determined by the r and η values of the city. The red triangles represent urban agglomerations, where the positions are determined in the same way. The rectangle is divided into four parts using the average *r* and η value of all the cities as thresholds, which are displayed as the red dotted lines in the figure, where the four parts represent the four types of correlation/ratio. The features of each type are summarized in the corresponding part.). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

agglomerations are shown in two pictures, for a better display of the data). Because the correlation coefficients are often missing in the cold season, and the periodic variations are not obvious, we selected the highest correlation in the warm season (May to October each year) in each year for the nine urban agglomerations to study the interannual variation of the correlation. In most regions, the correlation gets stronger from 2013 to 2014, but starts to decrease after 2014, indicating a weakening of the $PM_{2.5}$-AOD correlation.

Fig. 7(c) and (d) show the interannual variations of the ratio. Overall, there is a decreasing tendency from 2013 to 2017, although with some fluctuations, which indicates that $PM_{2.5}$ is accounting for a smaller part in AOD. To quantitatively evaluate the rate of decrease, we conducted simple linear fitting for the ratios from 2013 to 2017, and represent the rate of decrease with the slope of the fitting line. The linear slope for the BTH area is −16.6, and the absolute value is the largest among the nine urban agglomerations. The linear slope for the three urban agglomerations in the north, which have higher ratios than the other regions, are all less than −7. However, the slope values for the other regions are all larger than −5, representing a slower rate of decrease. Among all the nine urban agglomerations, the Chengyu urban agglomeration is the only region where the ratio is increasing slowly overall, and the slope of the fitting line is 0.73. Combining the results about correlation and ratio, we can infer that the relationship between $PM_{2.5}$ and AOD is getting weaker.

3.3. *Discussion*

3.3.1. *Impacting factors for the $PM_{2.5}$-AOD relationship*

In the last section, we compared the spatial and temporal variations of the $PM_{2.5}$-AOD relationship with the spatio-temporal variations of some psychical geographical variables such as aerosol type, topography, RH, and PBLH etc., and through the combined analysis, we developed some understandings on how are these factors influencing the $PM_{2.5}$-AOD relationship, so we simply summarize the impact of these factors here.

1) Aerosol type and properties impact

The high correlation and high FMF in BTH, CY region and the low correlation and low FMF in PRD, QGN shows that the aerosol types and properties can influence the $PM_{2.5}$-AOD correlation. The urban-industry aerosol type, in which sulfates and nitrates are the main species, tends to cause high correlation between $PM_{2.5}$ and AOD. This is because this type of aerosol tends to have low FMF and can share more common sources with $PM_{2.5}$. In contrast, soil dust and sea salt aerosols, in which coarse particles make up a larger proportion and the FMF is small, usually have a different source from $PM_{2.5}$.

2) Topography impact

Topography is a very important influencing factor for $PM_{2.5}$ and the $PM_{2.5}$-AOD relationship, but the study of it is lacking since the quantitative description of complex topography structure is often difficult. In our study, we found that the basin terrain can often result in a high correlation between $PM_{2.5}$ and AOD in local regions. For example, Sichuan Basin and Tarim Basin both have high *r* values. Therefore, we infer that basin areas tend to form a regional pollution environment, and $PM_{2.5}$ and AOD can share a larger amount of influencing factors, and thus have a higher correlation.

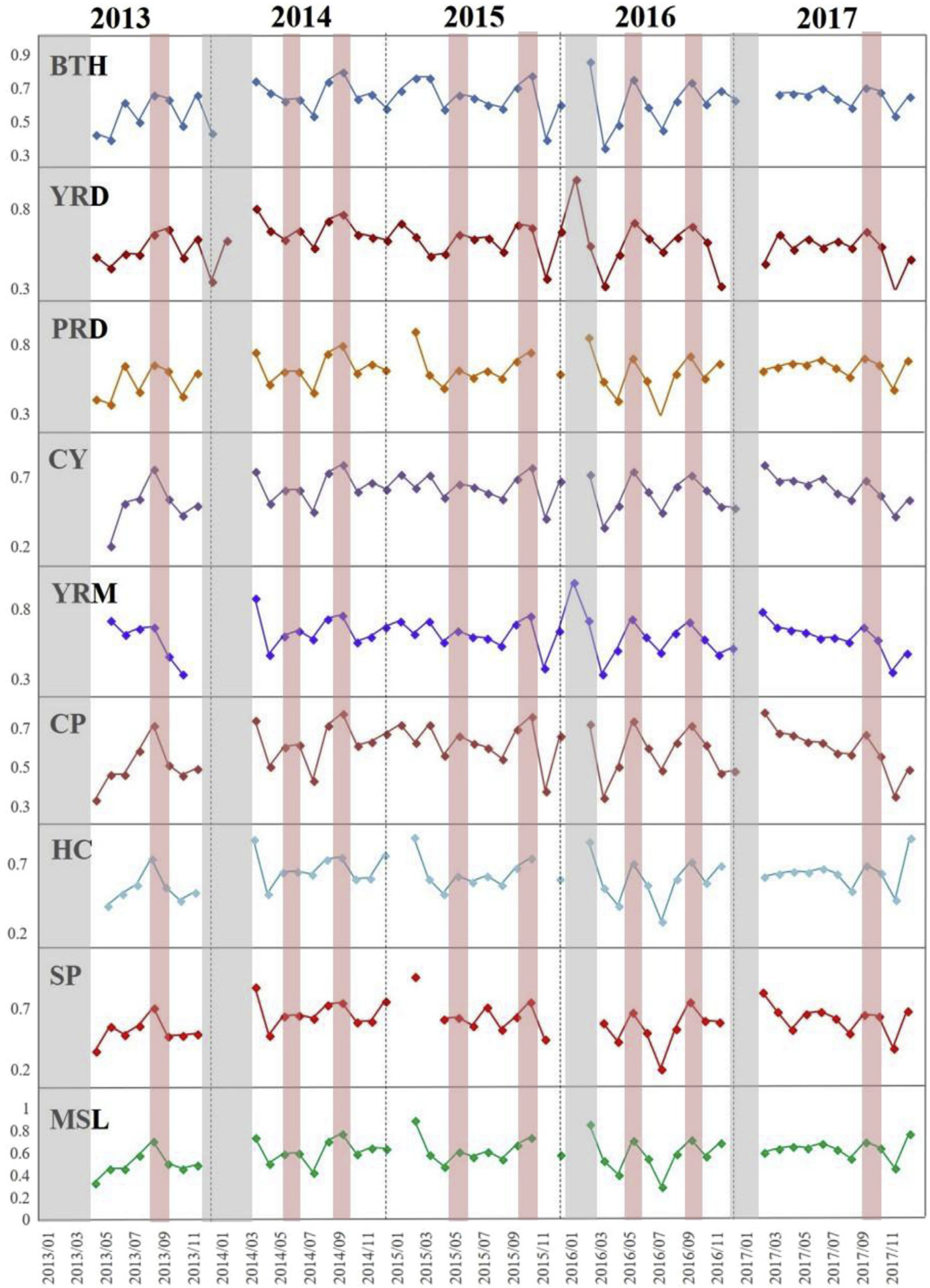

Fig. 5. The monthly variations of the significant (p-value<0.05) correlation coefficient *r* in the nine urban agglomerations (the gray bars represent the data missing months and the red bars highlight the high correlation months). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

3) Relative humidity (RH) impact

The high consistency between the ratio distribution and RH distribution shows that RH can influence the $PM_{2.5}$-AOD relationship a lot. High humidity represents a high content of water vapor in the environmental atmosphere and in suspended particles. Water vapor in particles can contribute a lot to aerosol extinction ability and make AOD increase. However, the concentration of $PM_{2.5}$ is mainly dry mass concentration, which means that the water vapor in particles will be evaporated, and thus contributes little to $PM_{2.5}$ mass concentration. Hence, high humidity tends to make the ratio between $PM_{2.5}$ concentration and AOD lower.

4) Planetary boundary layer height (PBLH) impact

The seasonal variations of $PM_{2.5}$/AOD ratio takes an opposite trend against PBLH, that means PBLH can have a great impact on $PM_{2.5}$-AOD relationship as well. A high PBLH makes particles able to suspend in a higher vertical space, but the $PM_{2.5}$ concentration measuring instruments are usually located on the ground surface, so only the surface $PM_{2.5}$ is measured. When the PBLH becomes higher, the $PM_{2.5}$ concentration we acquire from in-situ equipment

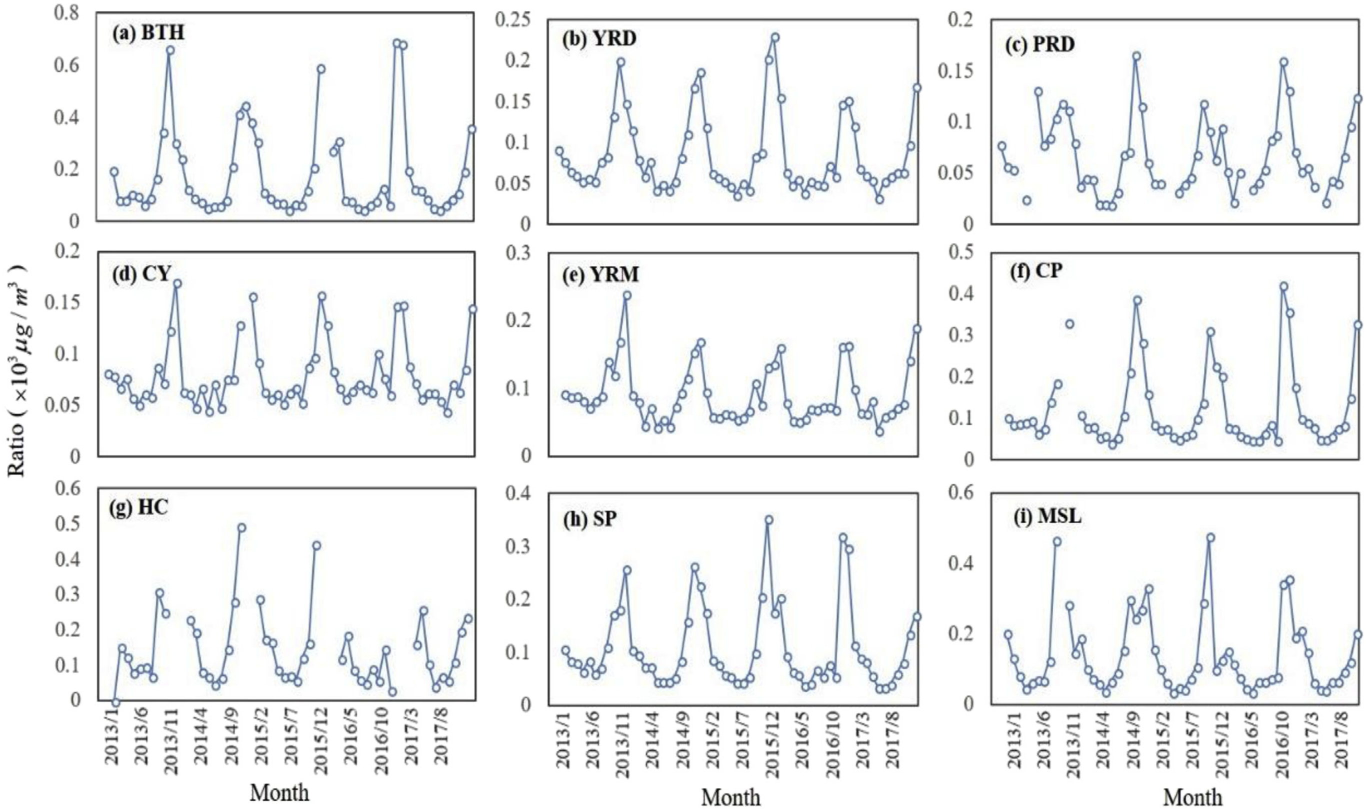

Fig. 6. The monthly variations of the $PM_{2.5}$/AOD ratio in the nine urban agglomerations.

will account for a reduced proportion of the total $PM_{2.5}$ in the atmospheric column, thus making the ratio between $PM_{2.5}$ and AOD lower. To verify the impact of PBLH, we conduct a vertical correction (Zhang and Li, 2015) for AOD to see whether the corrected AOD can be more correlated with $PM_{2.5}$, the result is shown in Supplementary Fig. 4, and for most of the regions, correlations are improved after AOD being vertical corrected by PBLH.

3.3.2. Implications for $PM_{2.5}$ retrieval through satellite AOD

MODIS AOD has been widely used for $PM_{2.5}$ concentration retrieval. However, as our results show, $PM_{2.5}$ is accounting for a smaller part of AOD, and their correlation has been decreasing since 2014. Therefore, AOD's prediction ability for $PM_{2.5}$ concentration needs to be further validated. Thus, we conducted a $PM_{2.5}$ retrieval experiment to see how the retrieval performance is varying as the $PM_{2.5}$-AOD relationship gets weaker. The GWR model is a popular regression model proposed by Brunsdon et al., in 1996 (Brunsdon et al., 1996). Recently, it was widely used for retrieving $PM_{2.5}$ concentration from satellite AOD and shows a good performance (Hu et al., 2013; He and Huang, 2018). Therefore, it was used in our experiment (for a more detailed theory of GWR, please refer the Supplementary material). The *GWR4* software developed by Tomoki Nakaya et al. (2005) was used to conduct the GWR calculation. AOD and five metrological factors (RH, TMP, UW, VW, PS) were used as independent variables in the model while $PM_{2.5}$ concentration is the dependent variable. The kernel type is adaptive bi-square and the optimal bandwidth is determined by minimizing the corrected Akaike information criterion (AICc). For each year from 2013 to 2017, a GWR model is established. The retrieval R^2 and adjusted R^2 are listed in Table 1 and the boxplots for the local R^2 of each environmental station can be seen in Supplementary Fig. 5. The R^2 and adjusted R^2 both decrease from 2013 to 2017, indicating that the performance of the GWR model is getting worse.

With time passing by, the $PM_{2.5}$ pollution in mainland China has been reduced (Supplementary Fig. 6). Meanwhile, our results show that the relationship between $PM_{2.5}$ and AOD are getting weaker, and the performance of AOD retrieval is also deteriorating. This implies that with the change of the pollution situation, the relationship between $PM_{2.5}$ and AOD are changing, and the predicting ability of AOD for $PM_{2.5}$ are also getting different. In the long term, we should keep a discreet attitude to the use of AOD product for $PM_{2.5}$ retrieval. In our study, we detected the start of the change. In the future, more studies should be made to capture the full picture and essence of this change.

3.3.3. Limitations and future work

We studied the relationship between $PM_{2.5}$ and AOD using the 10 km MODIS AOD product in a large spatial extent and long time series, but there exist several uncertainties. One is the product quality of MODIS AOD. Though only the highest quality of AOD retrievals were used in our analyses, the quality of AOD over complex land surfaces and near water bodies can still be a problem (Wang et al., 2018b). Besides, there are many kinds of AOD product apart from MODIS AOD, such as the 750 m VIIRS AOD product (Jackson et al., 2013) and AOD product from Himawari-8, which have a temporal resolution of 10min (Zang et al., 2018). The differences among these products may also bring uncertainties to the relationship between $PM_{2.5}$ and AOD. In the future, we will pay more attention to the use of AOD product at a higher spatial and temporal resolution. With higher resolution, the $PM_{2.5}$ data and AOD product can match better at temporal and spatial scales. Actually, there are also some possible methods that can improve the $PM_{2.5}$-AOD correlation, such as the introduction of fine mode aerosol or filtering the aloft plumes, but the effect of these methods

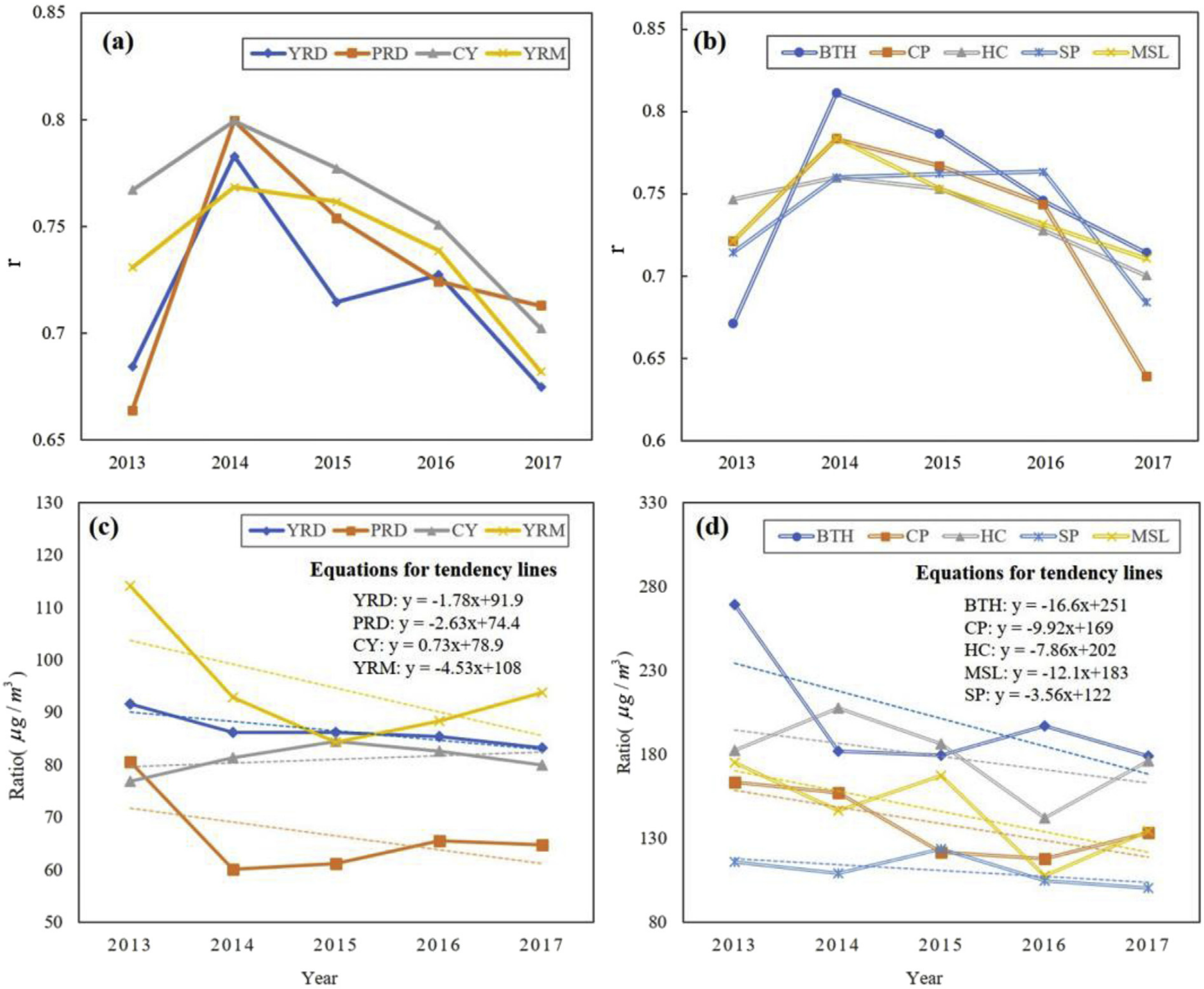

Fig. 7. The interannual variations of $PM_{2.5}$-AOD correlation (a, b) and their ratio (c, d) in the nine urban agglomerations. (The dotted lines in light color in (c) and (d) represent the fitted tendency lines). The scales of the subgraphs are different for a better display of the data, and r in (a, b) is the highest correlation coefficient in the warm season (May to October each year). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 1
The R^2 and adjusted R^2 of the GWR model for $PM_{2.5}$ retrieval from 2013 to 2017.

	2013	2014	2015	2016	2017
R^2	0.89	0.86	0.82	0.78	0.73
Adjusted R^2	0.87	0.83	0.81	0.75	0.69

in the vast mainland China needs further validation. In the future, we would also like to make more investigations on this problem and explore the $PM_{2.5}$-AOD relationships considering more potential factors like the temporal and spatial scales and the vertical structures of aerosols. At last, though we have explained the spatio-temporal variations of the $PM_{2.5}$-AOD relationship considering multiple kinds of factors, there are still several phenomena that we cannot fully explain, we would also like to make more investigations using more complex methods like model simulation, to explore the inner physical connections between these factors and $PM_{2.5}$-AOD relationship in the future.

4. Conclusions

The $PM_{2.5}$-AOD relationship is the cornerstone for $PM_{2.5}$ satellite retrieval. However, the foundation is not always solid. In this study, we found the $PM_{2.5}$/AOD ratio tends to be higher in dry regions like the drier north China, and the correlation between $PM_{2.5}$ and AOD tends to be larger in places that are serious polluted and where aerosols are fine mode dominated, like the BTH and CY regions. In recent years, the relationships between $PM_{2.5}$ and AOD are getting weaker, and the performance of $PM_{2.5}$ retrieval is getting worse as well. The spatio-temporal variations of the $PM_{2.5}$-AOD relationship are results of the synthetic impact of multiple factors including meteorology, topography and aerosol properties etc. The findings could provide useful instructions and important implications for satellite retrieval of $PM_{2.5}$ concentration, and will help with improving our understanding of the $PM_{2.5}$ pollution situation in China.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.02.071.

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