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New indicators for air quality and distribution characteristics of pollutants in China

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ABSTRACT

China has been suffering from an increasingly worse issue of air pollution over these years, which has not only caused a severe impact on the indoor environment, but also presented a tough challenge to clean production and a serious threat to human health. At present, China continues to adopt Air Quality Index (AQI) for the assessment of outdoor air pollution, which takes into consideration only the most serious pollutant and thus neglects the effects of other pollutants during the same period. In consideration of the wide-ranging impact of various pollutants, a new Total Air Quality Index (TAQI) is proposed to describe the widespread effects of six criteria pollutants on air quality. Based on the current indicators applied in China, TAQI could reveal the contribution made by each kind of pollutant to the overall air quality as indicated by Contribution Rate (CR). In this study, the daily average values for the concentration of six pollutants across 103 individual cities nationwide as collected from 2015 to 2017 are taken as the data source. According to the new indicators, the CRs of six pollutants in China are first explored. Then, the pollution situation facing these 103 cities within three years is categorized into five different groups by cluster analysis, and the characteristics of each group are summarized. Finally, the five most representative cities are identified by correlation analysis to explore the seasonal variation characteristics of pollutants. As revealed by this study, particulate matter and NO_2 are the primary pollutants, which exhibit a range of obvious spatial and temporal variation characteristics. The findings of this study are expected to draw public attention to the gaseous contaminants (especially NO_2), guide the control measures of indoor and outdoor pollutants, and assist with the design of clean air conditioning system.

1. Introduction

Due to the rapid process of industrialization and urbanization in the most recent decades, China has encountered a series of severe air pollution problems due to the significant increase of energy consumption, which has resulted in a substantial amount of pollutant emissions [1–5]. The Environmental Performance Index 2018 demonstrates that the score of China is 14.39 for outdoor air quality (full score is 100), and China is ranked as the fourth worst country among 180 countries [6]. Outdoor air pollution has posed a severe threat to the sustainable development of society and economy, which thus causes a widespread concern over public health [7–9]. Not only will this lead to a deterioration in outdoor air quality, it will also affect indoor air environment by (natural and mechanical) ventilation or infiltration [10–15]. Plenty of studies have demonstrated that air pollutants including particulate matters and gaseous contaminants can put human health at risk either directly or indirectly [16–18]. PM_{10} can be inhaled and accumulate in the human respiratory system, while $PM_{2.5}$ can even enter the alveoli and bloodstream, which will further lead to respiratory symptoms, cardiovascular diseases and premature mortality [19,20]. Gaseous

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contaminants will cause irritation to the mucous membranes of the upper respiratory tract, affect lung function, and even cause toxin damage to the nervous system [21,22]. In addition, they also play a significant role in the yield of cleanroom products such as chips. Therefore, it is essential to measure air quality in a precise way as it could improve the understanding that people have as to the characteristics of pollutants, which is conducive to the containment of air pollution for the improvement to indoor environment.

The World Health Organization (WHO) has been particularly concerned about the impact of air quality on human health for more than half a century. Its air quality guidelines were first published in 1987, revised in 1997 and finally released in 2005 as the latest version [23], which covers five pollutants, including $PM_{2.5}$, PM_{10}, SO_2, NO_2 and O_3. The concentration limits of five pollutants as mentioned above are specified under both long-term exposure and short-term exposure conditions (O_3 is given the 8 h average concentration value), and it aims to provide a reference for other countries to formulate environmental air quality standards.

In order to measure air quality in a simple and intuitive way while making the ongoing status known to the general public, there are many countries around the world who have adopted integrated air quality index to make a quantitative assessment of air quality. The higher the index is, the worse air pollution becomes, and the more significant impact it will have on human health. Based on the original version of the PSI (Pollutant Standard Index), Air Quality Index (AQI) was proposed by the United States Environmental Protection Agency, and has been commonly used worldwide. The primary feature of AQI approach is to transform the concentration values of six criteria pollutants (including CO) into a comparable dimensionless index with the range from 0 to 500, respectively. In general, the concentration limit and the measured exposure time of AQI system vary from country to country and are aligned to the environmental air quality standards formulated by a specific country. On this basis, there is a necessity for each country to revise the concentration limit according to its practicalities of air pollution, as a result of which AQI systems are not exactly the same across different countries.

In some previous studies, AQI approach and the environmental air quality standards have been applied to analyze the current status of air pollution in many cities or regions. Generally speaking, those studies tend to focus on the spatial and temporal variations of air pollutants [24–28]. AQI value and standards were used to measure outdoor air quality, while chief pollutant (CP) was regarded as the leading factor for air pollution. Besides, attempt has been made in some studies to determine the correlation among the pollutants [29,30] and the association between pollutants and meteorological conditions [31–35].

Despite the common use of AQI approach, it is still questioned and criticized as it takes into consideration only the most serious pollutant and thus neglects the combined effects of multiple pollutants. In some studies, attempt was made to reveal the comprehensive effects of multiple pollutants by improving the AQI approach [36–40]. Swamee and Tyagi [41] proposed a cumulative function among pollutants based on AQI approach, which integrates the sub-indexes of six criteria pollutants into a single index range of 0–500, namely Aggregate Air Quality Index (AAQI). Kyrkilis et al. [42] adopted this index to Athens, based on which an evaluation standard of air quality was suggested in 2007. There is another comprehensive index, that is, Air Quality Health Index (AQHI), which factors the results of environmental epidemiology into the AQI approach. The relative risks for respiratory and cardiovascular diseases associated with air pollutants were obtained by performing the time-series studies, where the overall percentage of excess risks of the hospital admissions associated with pollutants was calculated and the total percentage was used to determine the pollution level of the day. At present, some Canadian cities are using AQHI proposed by Stieb et al. [43], with consideration given to three pollutants including $PM_{2.5}$, NO_2 and O_3. Meanwhile, Hong Kong developed AQHI on the basis of the Canadian approach as proposed by Wong et al. [44], with five pollutants of $PM_{2.5}$, PM_{10}, SO_2, NO_2 and O_3 taken into account.

Table 1
The upper limit AQI values and corresponding pollutants, the health categories for AQI.

AQI	Pollutant concentrations						
	PM2.5, 24 h (μg/m3)	PM10, 24 h (μg/m3)	SO2, 24 h (μg/m3)	NO2, 24 h (μg/m3)	O3, 8 h (μg/m3)	CO, 24 h (mg/m3)	Health risk category
50	35	50	50	40	100	2	Excellent
100	75	150	150	80	160	4	Good
150	115	250	475	180	215	14	Lightly Polluted
200	150	350	800	280	265	24	Moderately Polluted
300	250	420	1600	565	800	36	Heavily Polluted
400	350	500	2100	750	1000	48	Severely Polluted
500	500	600	2620	940	1200	60	Severely Polluted

Back in 2012, the Chinese Ministry of Environmental Protection (MEP) started to adopt and develop the AQI system based on the US EPA AQI approach [45]. Obviously, this system is also subject to limitations on describing the comprehensive effects of pollutants as mentioned above. In this study, the daily average values for the concentration of six pollutants across 103 cities nationwide as obtained from 2015 to 2017 are taken as the data source. Based on the AQI system and Ambient Air Quality Standards adopted in China currently, Total Air Quality Index (TAQI) and Contribution Rate (CR) are proposed to describe the comprehensive effects of the aforementioned six criteria pollutants. After comparing two sets of indicators, the new indicators, TAQI and CR, are used to further explore the distribution of air pollutants. The pollution situation is classified, the characteristics of each category are summarized and the seasonal variations of pollutants are explored for each representative city. The new indicators proposed in this study play a significant role in the comprehensive evaluation of air quality and revealing the essential trends. The results are believed to increase public awareness of gaseous pollutants and provide guidance on how to control the indoor environment, especially for the design of clean air-conditioning system in China.

2. Methodology

2.1. Air quality indicators

2.1.1. Current indicators - AQI and chief pollutant

At present, China adopts the AQI approach to measure the levels of air pollution, which converts the concentration of each pollutant into the comparable dimensionless value known as Individual Air Quality Index (IAQI). The six criteria pollutants involved in this approach include $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO and O_3. IAQI of each pollutant is calculated using Eq. (1) and the corresponding thresholds of all pollutant are presented in Table 1.

$$IAQI_P = \frac{IAQI_{Hi} - IAQI_{Lo}}{BP_{Hi} - BP_{Lo}}(C_P - BP_{Lo}) + IAQI_{Lo} \tag{1}$$

where $IAQI_P$ indicates the individual air quality index of pollutant P, C_P denotes the mass concentration of pollutant P, BP_{Hi} represents the mass concentration break point that is $\geq C_P$ in Table 1, BP_{Lo} refers to the mass concentration break point that is $\leq C_P$ in Table 1, $IAQI_{Hi}$ stands for the $IAQI$ value corresponding to BP_{Hi} and $IAQI_{Lo}$ indicates the $IAQI$ value corresponding to BP_{Lo}.

After the concentration of all the six pollutant is converted into $IAQI$, the AQI value is obtained as the maximum value of all the six $IAQI_Ps$. The description of air quality corresponding to the AQI values is made in

Table 1. In the meantime, if AQI exceeds 50, the pollutant with the largest IAQI value is treated as a CP on that day. A pollutant that becomes CP for most times is determined as a CP of this year.

2.1.2. New proposed indicators - TAQI and cumulative chief pollutant (CCP)

From the learning of AQI approach, it can be known that AQI gives consideration exclusively to the most serious pollutant, as a result of which the effects of other pollutants during the same period tend to be neglected. In order to describe the comprehensive effects of the six criteria pollutants on air quality, TAQI is proposed on the basis of the AQI approach adopted in China. TAQI is defined as the sum of $IAQI_P$, while the concentrations of six criteria pollutants are the same between the concentrations of Grade I standards as stipulated by the Chinese Ambient Air Quality Standards (GB3095-2012) and the concentrations corresponding to $IAQI_P = 50$. It is considered that a pollutant does not affect human health when its $IAQI_P \leq 50$, so that TAQI value only aggregates the $IAQI_P$s which are in excess of 50. The TAQI value is calculated using Eq. (2):

$$\mathrm{TAQI} = IAQI_1 + IAQI_2 + IAQI_3 + \ldots + IAQI_n \text{ if } IAQI_P > 50 \tag{2}$$

The CR is further proposed to indicate the contribution made by a single pollutant to the overall air quality, and it is expressed as the ratio of $IAQI_P$ and TAQI, as shown in Eq. (3):

$$\text{Contribution rate} = \frac{IAQI_P}{TAQI} \times 100\% \tag{3}$$

The ratio of the total $IAQI_P$ value to the total TAQI value is referred to as the annual CR of a certain pollutant in this year. The pollutant with the annual largest CR is treated as the local Cumulative chief pollutant (CCP) this year.

2.2. Pollutant concentration data

The concentrations of the aforementioned six criteria pollutants, including $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO and O_3, are measured on the national air quality monitoring sites distributed in different cities, suburbs and rural areas. Due to the uneven distribution of monitoring sites across the country, compared with suburbs and rural areas, the number and density of monitoring sites in cities are relatively large. As the number varies significantly from one area to another, only the average value obtained from multiple monitoring sites is taken as the daily concentration for each area.

In order to research the overall air quality and the characteristics of pollutant distribution across China, a total of 103 cities nationwide are selected considering the impact of urbanization on air quality and data integrity and reliability (from first-, second-, and third-tier cities [46]). The 24-h average concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO and 8-h moving average (maximum) of O_3 from 1st January 2015 to 31st December 2017 of 103 cities are included in this study, and the data is sourced from the Beijing Meteorological Bureau.

2.3. Statistical analysis

2.3.1. Cluster analysis

Cluster analysis has been used to explore the spatial distribution characteristics of air quality frequently [47–49]. It provides a method to classify objects based on their characteristics. The samples or variables can be classified into different clusters depending on the nature of their relationship, while the objects in the same cluster are more similar (in some sense) to each other than to those in other clusters.

In this study, the pollution situation in 103 cities over the past three years is classified with the assistance of the SPSS 19.0 software (SPSS, Inc., Chicago, IL, USA). The total number of samples is 309, and the variables include the annual average TAQI and the annual CRs of the six pollutants, respectively.

2.3.1.1. Data standardization. Data standardization is the first step in cluster analysis, which could determine the accuracy of clustering results. The dimensions or orders of magnitude of variables differ significantly. In order to compile the data for comparison purpose, transformations are required on a frequent basis. In this study, the annual average TAQI and the annual CRs of six pollutants are transformed into a robust z-score, as shown in Eq. (4):

$$z_{ij} = \frac{x_{ij} - \mu_j}{\sigma_j} \tag{4}$$

where z_{ij} indicates the z-score of the variate j in city i, x_{ij} represents the value of the variate j in city i, μ_j denotes the arithmetic mean value of the variate j in all city and σ_j refers to the standard deviation of variate j in all cities.

2.3.1.2. Hierarchical cluster method. There are various types of cluster analysis. As the classification number is not determined in advance, the hierarchical cluster method is applied in this study. Hierarchical cluster method can be categorized into Q-cluster for sample clustering and R-cluster for variable clustering. Q-cluster is selected in this study to process data as the ultimate aim is to classify the pollution situation.

2.3.1.3. Distance calculation. If the annual average TAQI and the annual CRs of the six pollutants are treated as a 7-dimensional space, the difference among the 309 samples in this 7-dimensional space would be measured by distance. A variety of distances have been developed and widely used to measure the difference among the individuals. The Squared Euclidean distance indicated as the sum of the squares of the differences between two variables is employed to measure the difference among the 309 samples, and the mathematical definition is shown as Eq. (5):

$$d_{xy} = \sum_{i=1}^{n} (x_i - y_i)^2 \tag{5}$$

where d_{xy} indicates the Squared Euclidean distance among pollution situation x and pollution situation y on 7-dimensional space, x_i refers to the i-th variate value of pollution situation x, y_i denotes the i-th variate value of the pollution situation denoted as y.

2.3.1.4. Ward's method. In addition, the distance between different clusters needs to be calculated as well considering the distance between the samples. There are various methods to calculate the distance between different clusters, of which is Ward's method [50]. The advantage of Ward's method is that it can produce relatively compact clusters, which is easy to classify. In this study, Ward's clustering method is applied, where two clusters with the most insignificant increase in the error sum of squares of deviations will be selected for combination until all the samples are clustered together.

2.3.2. Correlation analysis

There are totally 103 cities involved in this study, and it is complicated to analyze the seasonal characteristics of the aforementioned six pollutants in each city in detail. In order for reduction in the number of research objects, a number of representative cities are selected to represent the average severity of pollution for the corresponding category based on the classification results of the pollution situation.

Therefore, the average value of seven variables in each category is treated as the reference value for the representative city falling into this category to be chosen by correlation analysis. Distance analysis, a kind of correlation analysis, is carried out to measure the similarity between the variables of all the samples and the reference values within the same category. The approach to measuring distance is still reliant on the

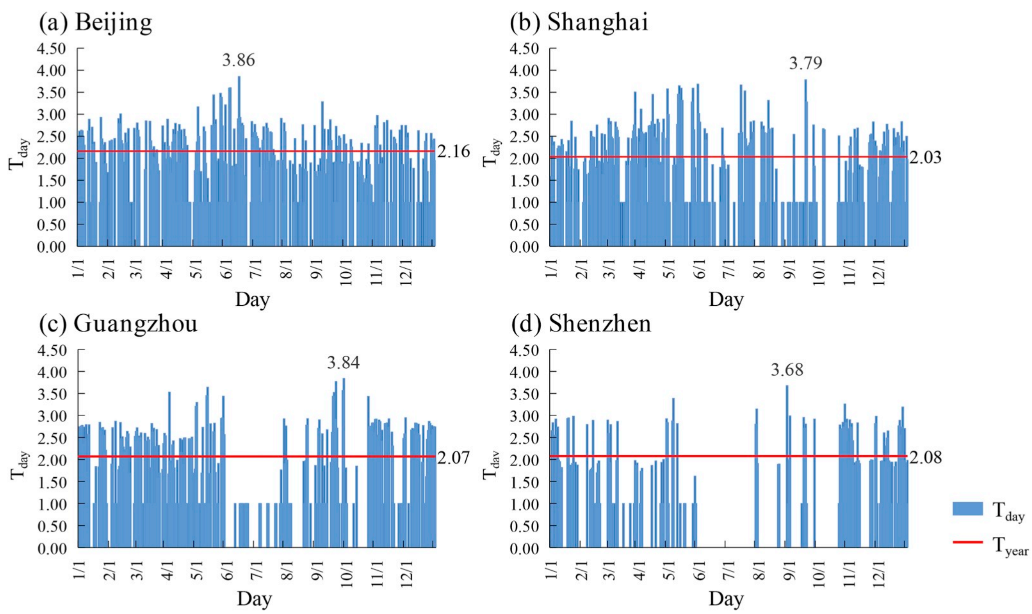

Fig. 1. T_{day} and T_{year} of four cities in 2017.

Square Euclidean distance, as shown in Eq. (5).

3. Results

3.1. Comparison between the existing and proposed indicators

3.1.1. AQI and TAQI

TAQI represents a new index and it is necessary to compare it with AQI. However, AQI and TAQI are defined in different ways, which makes it difficult to compare two indexes directly. Times are defined as $T = TAQI/AQI$, $T_{day} = TAQI_{day}/AQI_{day}$ can be obtained by daily data, and T_{year} is the annual average of T_{day} of a city. T_{year} is treated as the standard to measure the variation of T_{day} and figure out the difference between AQI and TAQI, while the fluctuation can be calculated using the following Eq. (6)

$$\text{Error} = \frac{|T_{day} - T_{year}|}{T_{year}} \times 100\% \tag{6}$$

The data on 4 cities as collected in 2017, including Beijing, Shanghai, Guangzhou, and Shenzhen, are selected as examples to facilitate the

Table 2
The details of cities with different CP and CCP.

Year	City	Annual CP	CCP	Year	City	Annual CP	CCP
2015	Deyang	$PM_{2.5}$	PM_{10}	2017	Changchun	$PM_{2.5}$	PM_{10}
	Guiyang				Chongqing		
	Haikou				Fuoshan		
	Lianyungang				Guiyang		
	Nanjing				Huanggang		
	Qujing				Hangzhou		
	Sanya				Jieyang		
	Wenzhou				Jinzhou		
	Yantai				Langfang		
	Yancheng				Lianyungang		
	Yueyang				Nanning		
	Zhuhai				Nantong		
	Shenzhen	NO_2	PM_{10}		Ningbo		
2016	Anshan	$PM_{2.5}$	PM_{10}		Ningde		
	Changchun				Putian		
	Changde				Shangrao		
	Deyang				Shenzhen		
	Nanjing				Tangshan		
	Nanning				Wenzhou		
	Ningbo				Wuxi		
	Shangrao				Wuhu		
	Shenyang				Xi'an		
	Xiangtan				Xuchang		
	Yancheng				Xinxiang		
	Zhenjiang				Xuzhou		
	Zhumadian				Nanyang	PM_{10}	$PM_{2.5}$
	Zunyi				Shanghai	NO_2	$PM_{2.5}$
	Zhuzhou				Suzhou		
	Shenzhen	NO_2	PM_{10}		Taizhou		
	Zhuhai				Zhuhai		
					Qinhuangdao	NO_2	PM_{10}

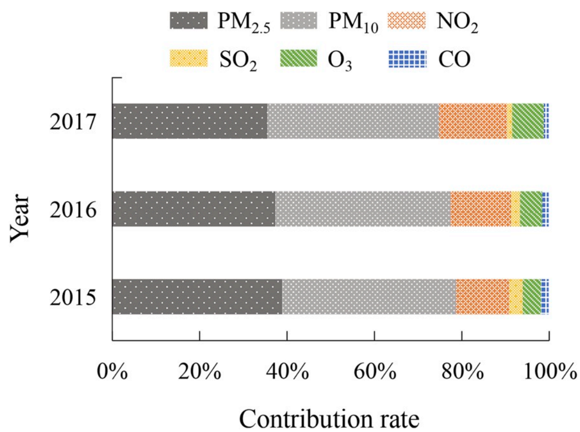

Fig. 2. Average CRs of six pollutants of 103 cities, 2015–2017.

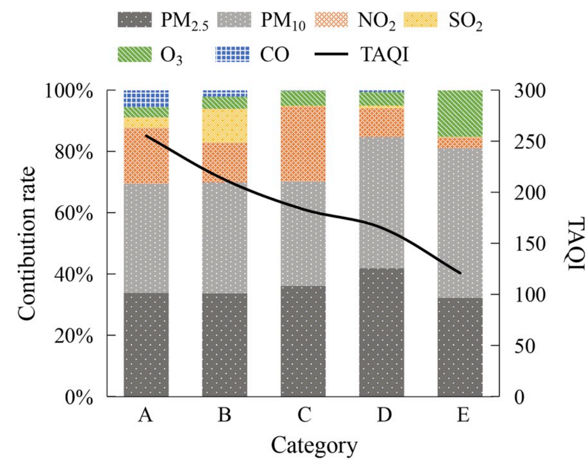

Fig. 3. The average TAQI and the average CRs of six pollutants in five pollution situation categories.

analysis. In this analysis, when AQI≤50, then TAQI = 0, which means there is no pollutant causing pollution on that day (all of $TAQI_P \leq 50$). T_{year} values and the fluctuation conditions of T_{day} in the four cities are shown in Fig. 1.

From Fig. 1, it can be seen that the times between T_{day} and T_{year} across the four cities are roughly 2.10, and the fluctuation of T_{day} could be recognized instantly for all the cities. The maximum value of T_{day} is 3.86 (Beijing), 3.79 (Shanghai), 3.84 (Guangzhou) and 3.68(Shenzhen) with the Error of 78.7%, 86.7%, 85.6%, and 76.9%, respectively. Conversely, the minimum value of T_{day} is 1.00, which suggests that there is only one pollutant causing pollution on that day with TAQI = AQI = $IAQI_P$. The error in these four cities is 53.7%, 50.7%, 51.7%, and 51.9%, respectively, which leads to the conclusion that the AQI value shows a massive difference with TAQI, and that AQI is incapable to evaluate air quality accurately as it ignores many other pollutants.

3.1.2. CP and CCP

In general, the CP is used to guide the control measures against air pollution in a season or a whole year in many studies, and this study proposes CCP for comparison with the annual CP. Based on the definition, the annual CP cares much more about the times of a certain pollutant, while CCP cares much more about the cumulative amount of a pollutant. As the definition differs, the annual CP and CCP could be inconsistent even when the same data is applied. In this case, the annual CPs and CCPs are calculated and compared within a total of 309 (103 cities with three years) years' data. It can be found out that the inconsistency between the annual CP and CCP appear in some cities during the period from 2015 to 2017, as shown in Table 2.

It can be seen from Table 2 that 61 years of data show some inconsistency. As revealed by the results, the difference between CP and CCP is mainly reflected in $PM_{2.5}$, PM_{10} and NO_2. It is the case across 47 cities for a total of 61 years, which accounts for approximately 45.63% of the total of 103 cities and for 19.74% of 309 years. The trend could also be concluded as such an inconsistency occurs on an increasingly frequent basis in recent years, involving 13 cases in 2015, 17 cases in 2016, and 31 cases in 2017, respectively. NO_2 is taken into consideration, and CCP becomes a new reasonable index for the identification of CP in the future.

3.2. Contribution rates of pollutants

CR is capable to express the extent to which each pollutant has an impact on the overall air quality. The average CRs of the aforementioned six pollutants among 103 cities over the past three years are counted to describe the average impact of each pollutant on a national scale for China, as shown in Fig. 2.

It can be seen from Fig. 2 that the composition of the six pollutants as mentioned above has yet to show a significant change over the past three years. Although the CR of particulate matter ($PM_{2.5}$ and PM_{10}) consistently stays above 70%, the CRs of gaseous contaminants are on the increase year on year. It is worth noting that the CR of NO_2 increases year on year and exceeds 15%, which is significantly higher compared to the other three gaseous pollutants (with total CR of 10%). Aside from the pollutant NO_2, it could generate secondary particles by chemical or physical reactions with primary particles (direct emission) [51], and nitrates have been one of the main secondary particles [52–54]. Reducing the NO_2 concentrations plays a significant role in reducing particulate matter pollution [55], for which it is essential to raise public awareness of NO_2. Apart from reducing the emission of NO_2, making full use of ecological regulation of water body can be effective in reducing NO_2 [56].

3.3. Classification of pollution situation

3.3.1. Category characteristic

Due to the impact of geographical environment and social policies, the pollution situation may be clearly different across many cities, and it is necessary to understand the characteristics of pollution for accurate control of air pollutants. The pollution situation in each city is represented by TAQI and CRs of six criteria pollutants, and the data of 103 cities in three years are classified into several different categories according to the similarity of pollution situation. The tree diagram of the hierarchical cluster is illustrated in Fig. S1, which is obtained by SPSS 19.0 software. According to the tree diagram, 309 groups of data are classified into five categories of the pollution situation. The information of years and cities included in each category is shown in Table S1.

The average TAQI and the average CRs of six pollutants are obtained in five categories respectively and the results are presented in Fig. 3 to recognize the characteristics of five categories clearly.

As shown in Fig. 3, the level of air quality improves from A to E, as indicated by the black solid line. Meanwhile, the types of pollutants are reducd. All of the six pollutants fall into categories A and B, while the types of pollutants in C, D and E are usually particulate matter, NO_2 and O_3. The CR of gaseous contaminants is roughly 20% in the categories with good air quality, while it is about 30% in other categories.

Combined with 309 samples, the characteristics of five categories are described in detail with the CRs of the six pollutants. All of six pollutants exist in category A, among which four are gaseous contaminants. The CR of NO_2 rises to 18.3%, which exceeds the average value in China, and the CR of CO reaches as high as 5.5% in category A. The CR of particulate matter in category B shows similarity to category A, with the CR of NO_2 and SO_2 being 13.0% and 11.0% respectively, while the CR of SO_2 reaches the maximum level in category B. In category C, there is barely any pollution caused by SO_2 and CO. However, the CR of NO_2 reaches

Table 3
Characteristics of five pollution situation categories.

Category	Air quality (relatively)	Category characteristic
A	**Seriously**	All of six pollutants cause pollution; NO_2 exceeds the average value (18.3%)
B	**to**	All of six pollutants cause pollution SO_2 outstanding (11.0%)
C		Few pollution caused by SO_2 and CO NO_2 outstanding (24.1%)
D		Few pollution caused by SO_2 and CO Particulate matter outstanding (84.7%)
E	**Good**	Few pollution caused by SO_2 and CO Particulate matter outstanding (81.0%) O_3 outstanding (15.2%)

24.1%, and 41.3% for Guangzhou in 2017. In categories D and E, the CR of particulate matter is significantly higher compared to gaseous contaminants, and the CRs of NO_2 and O_3 count for the majority in gaseous contaminants. The characteristics shown by the five categories of the pollution situation are summarized in Table 3.

3.3.2. Spatial and temporal variations

In order to explore the spatial characteristics shown by the categories of the pollution situation, the classification results of 103 cities over three years are shown in Fig. 4. Map shows the results of 2015, cities which transferred to different categories are selected and marked with transferred results over three years.

The spatial characteristics of the five categories can be seen clearly in Fig. 4. The cities in category A are concentrated in the Beijing-Tianjin-Hebei region. The cities in category B are scattered across the country. The cities in category C are mostly distributed along the Yangtze River Delta and Pearl River Delta regions. A majority of the cities falling into category D are located in the south of China. The cities in category E are concentrated in the southeast coastal and plateau areas. In general, there are clear differences in the spatial distribution for the pollution situation, which are mainly reflected in the cities between the north and the south. Moreover, industrialization is also a significant influencing factor for the pollution situation.

The phenomenon of cities transferring in categories occurs during the past three years, 13 cases from 2015 to 2016 and 15 cases during 2016–2017. The phenomena are frequent to occur in A to D (4 cases), D to E (6 cases), and D to C (8 cases). The number of cities in category A remains unchanged basically over past three years, the number of cities in categories B and D shows a decreasing trend, and the number of cities in categories C and E continues an increasing trend. At present, cities usually transfer through categories with an obvious change in gaseous contaminants, which makes it necessary to raise concerns about gaseous contaminants in the future.

3.4. Seasonal variation of pollutants in representative cities

The average value of 7 variables (TAQI and six CRs) in each category is taken as the reference value to choose a representative city by distance analysis. The representative city is required to stay in the same category for three years, and then its characteristic value is required to be closest to the feature value of the category. Finally, five cities are chosen from five categories, with three years' distance indicated in Table 4.

As revealed by plenty of studies, pollutants exhibit obvious seasonal characteristics by analyzing the concentration of pollutants, which is primarily related to the source of pollutants [57–59]. For instance, the concentration of particulate matters will increase in the heating period or due to sand storm or industrial and traffic-related emissions [60–62], as a result of which it will reach a high level during spring and winter months. However, the use of concentration along to describe the seasonal characteristics of pollutants is incapable to demonstrate the

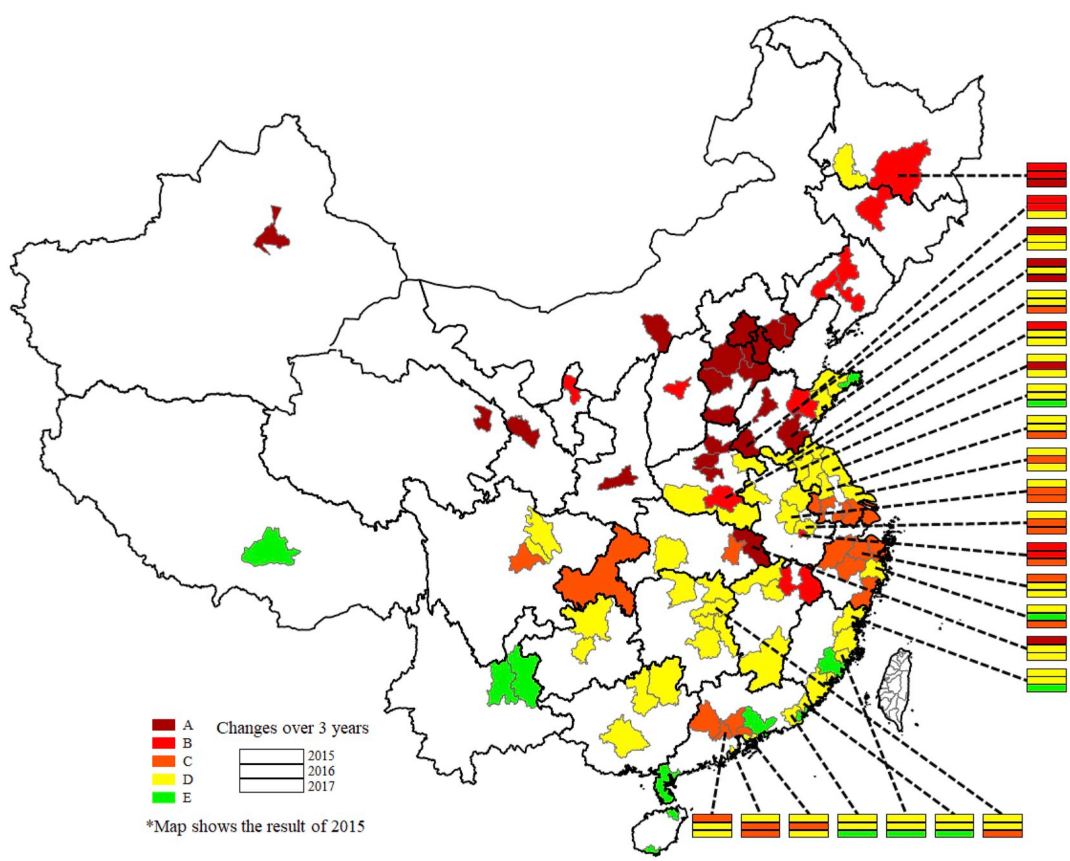

Fig. 4. The classification results of 103 cities from 2015 to 2017.

Table 4
Representative cities and the three years' distance between the. characteristic value and the reference value.

Categories	Representative cities	Year	Distance
A	Langfang	2015	3.040
		2016	1.332
		2017	6.064
B	Shenyang	2015	5.532
		2016	1.333
		2017	3.118
C	Hangzhou	2015	2.707
		2016	1.629
		2017	1.776
D	Xinyang	2015	3.366
		2016	0.805
		2017	0.773
E	Shantou	2015	1.229
		2016	12.032
		2017	0.465

comprehensive effect of various pollutants, for which the seasonal characteristics of pollutants are realized by combining concentrations and CRs of pollutants in this study.

As China is located in the temperate zone of the northern hemisphere, spring lasts from March to May, summer lasts from June to August, autumn lasts from September to November, and winter lasts from December to February next year. The concentrations and CRs from March 2015 to November 2016 and December 2016 to February 2017 are selected to conduct analysis, with the results shown in Figs. 5 and 6. There is less pollution caused by CO as revealed by the above-mentioned analysis, and the magnitude of CO concentrations is shown to be different from other pollutants. For this reason, the results of CO concentrations are discounted from Figs. 5 and 6.

The concentration of particulate matters shows a significant seasonal variation during spring and winter months, which is less significant in summer and autumn months. Based on the results of Langfang (Figs. 5a and 6a) and Shenyang city (Figs. 5b and 6b), which are polluted by SO_2, the SO_2 concentration reaches its maximum in winter and shows a similar seasonal variation to particulate matters, which is primarily attributed to the same pollutant sources [63,64]. NO_2 concentration shows only insignificant change in four seasons, while O_3 concentration shows an opposite seasonal variation with higher in spring or summer and the lowest in winter, as the formation of O_3 is related to solar radiation [65,66]. The performance of O_3 concentration in Shantou (Figs. 5e and 6e) is contradictory to the above-mentioned characteristics, which is speculated to be associated with the meteorological conditions and topographic conditions in Shantou.

From the results of the CRs, it can be seen that particulate matters and NO_2 cause pollution in five cities. The types of pollutants that cause pollution are the most in winter and the least in summer. The CRs of particulate matters in winter are lower than in other seasons. Compared with the obvious characteristics of gaseous contaminants (category A, B and C), the CRs of SO_2 show a significant seasonal variation in Shenyang (class B), as it is almost 0 in summer and autumn but is as high as 20% in winter. NO_2 causes pollution in all of the four seasons for Hangzhou (category C), the CRs of which fail to show a significant seasonal variation. Shantou (category E) exhibits the highest CRs of O_3. Affected by

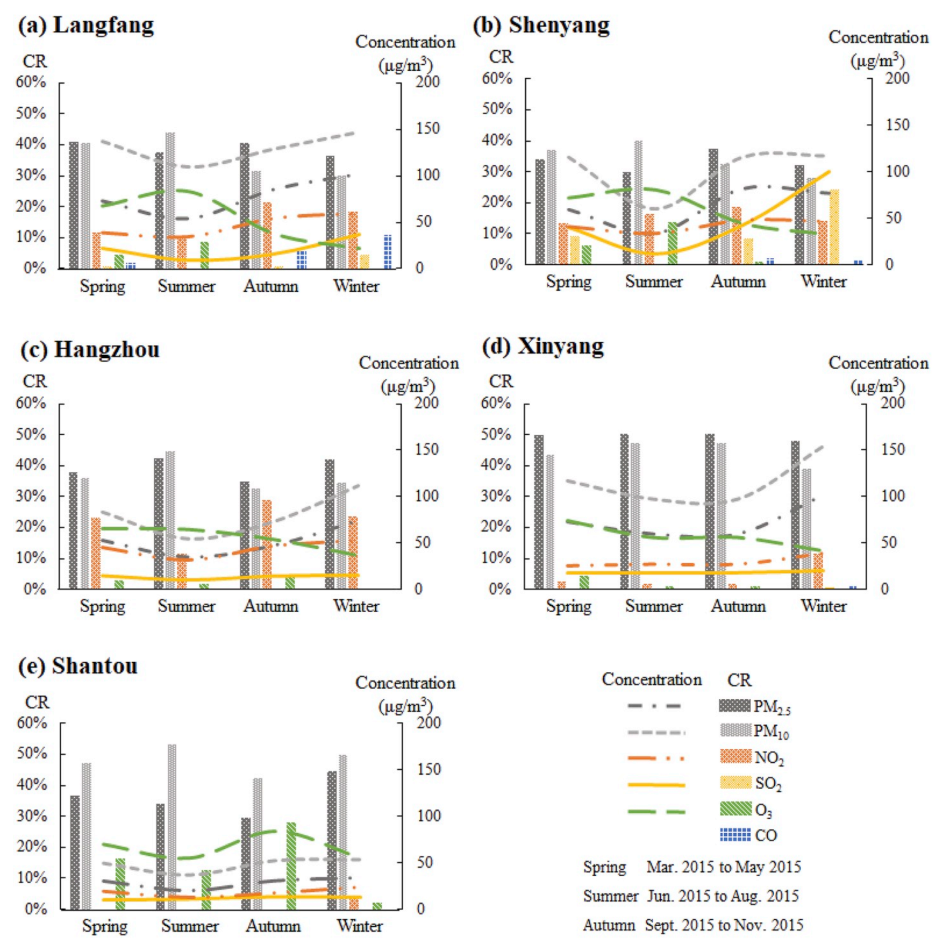

Fig. 5. Concentrations and CRs of six pollutants in five cities in four seasons, 2015–2016.

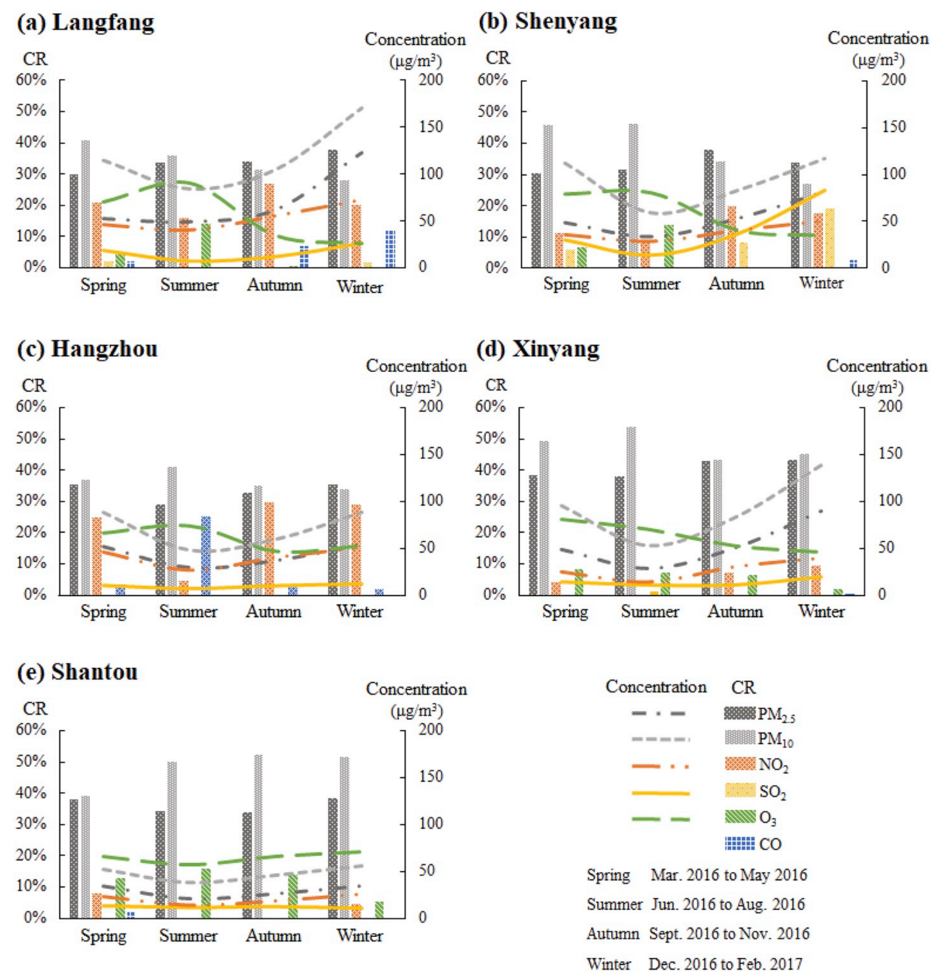

Fig. 6. Concentrations and CRs of six pollutants in five cities in four seasons, 2016–2017.

the origin of O_3, the CR of O_3 in winter is significantly reduced and shows an obvious seasonal variation.

It can be noticed by combining the results of pollutants concentrations and CRs that the seasonal variations of concentrations and CRs are similar for SO_2, NO_2 and O_3. However, particulate matters have the highest concentrations in winter but the lowest CRs, which is due to the comprehensive effects of the variety of pollutants in winter.

4. Discussion

Different from the AQI approach, TAQI is capable to contain more information, because of considering the comprehensive influence of six criteria pollutants. Furthermore, the CRs of pollutants can be obtained by TAQI, which can provide more valuable information to discover some phenomena as neglected by the AQI approach. For example, as indicated by the results of this research, NO_2 should be taken into account in China, but it was neglected in some research of air pollution based on the AQI approach. Besides, TAQI and CRs can implement the classification for the pollution situation, which can provide guidance on how to control air pollution. There are some limitations faced by TAQI at present. To be specific, TAQI is only applicable to compare air quality among different cities, as a result of which it cannot describe the air quality of a city independently. TAQI will be categorized into several levels in the future, which can assist in classifying air pollution and its impact on human health. Moreover, it could be consummated through comparison with other indexes in the future.

It is worth mentioning again that the representative cities just represent the average severity of pollution corresponding to each category. Actually, due to meteorological conditions and topographic features, it is possible for the seasonal variation of pollutants in some cities to be significantly different even when they fall into the same category of the pollution situation. Therefore, the representative cities are used to find out about the common characteristics of pollutants in these cities and study the characteristics of certain pollutants (such as SO_2 and O_3). The source of pollutants and the movement of pollutants due to meteorological or topographic factors will be taken as another research direction with new indicators in the future.

5. Conclusion

With the limitations imposed on the current AQI and CP indicators for the assessment of outdoor air pollution, a new air quality index TAQI is proposed based on AQI approach used in China, which takes the comprehensive effects of multiple pollutants on air quality into consideration, and CR is further proposed as the contribution of each pollutant. As revealed by the comparison results, TAQI and AQI have different descriptions of air quality, and the number of the inconsistence time between CP and CCP have been found to be on the increase year on year, which indicates that the new indicators (TAQI and CR) are more effective as indicator of outdoor air quality.

The distribution characteristics of pollutants in China are described by using new indicators. The results have demonstrated that $PM_{2.5}$, PM_{10} and NO_2 are the primary pollutants, and NO_2 becomes shining in recent years which should raise public awareness. The pollution situation can be classified into five categories with different characteristics in China. Spatiotemporal variations reveal that the pollution situation is clearly different between north and south China, which makes it necessary to raise public awareness of gaseous contaminants in the future. The types of pollutants that cause pollution show difference in the four seasons, and the comprehensive effects of pollutants are clearly manifested in winter. Those results will provide guidance on the control of outdoor pollutants, the creation of indoor air environment, and the design of clean air-conditioning system in China.

Declaration of competing interest

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

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Appendix A. Supplementary data

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