Trends in ambient air pollution levels and $PM_{2.5}$ chemical compositions in four Chinese cities from 1995 to 2017

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Abstract: An in-depth analysis of the specific evolution of air pollution in a given city can provide a better understanding of the chronic effects of air pollution on human health. In this study, we reported trends in ambient concentrations of particulate matter (PM) and gaseous pollutants [sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and ozone (O_3)] from 1995 to 2017 and $PM_{2.5}$ composition for the period of 2000–2017 in Guangzhou, Wuhan, Chongqing, and Lanzhou. We provided socio-economic indicators to help explain the pollution trends. SO_2 and PM (including PM_{10} and $PM_{2.5}$) concentrations showed a downward trend in recent years with the most notable reduction in SO_2 in Chongqing and $PM_{2.5}$ in Guangzhou. There was an overall flat trend for NO_2, while O_3 showed an upward trend in recent years except in Lanzhou. The majority of $PM_{2.5}$ mass was SO_4^{2-} (6.0–30 μg/m^3) and organic carbon (6.0–38 μg/m^3), followed by NO_3^- (2.0–12 μg/m^3), elemental carbon (2.1–12 μg/m^3), NH_4^+ (1.0–10 μg/m^3), K^+ (0.2–2.0 μg/m^3), and Cl^- (0.2–1.9 μg/m^3). Except for secondary inorganic aerosols in Wuhan, annual average concentrations of all $PM_{2.5}$ constituents showed a declining trend after 2013, corresponding to the trend of $PM_{2.5}$. The secondary sources in $PM_{2.5}$ were found to be most prominent in Wuhan, while the most abundant EC and Cl^- in Lanzhou was attributed to the use of coal. Despite temporal and spatial variabilities across the four cities, coal combustion, traffic emissions, and secondary pollution have been the major sources of $PM_{2.5}$ pollution. These trends in ambient air pollution levels and $PM_{2.5}$ composition may help understand changes in health outcomes measured at different times within the time period of 1995–2017 in the four cities.

Keywords: Air pollution; long-term variation; $PM_{2.5}$; chemical composition; source

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Introduction

The associations have been well established between ambient air pollutants, such as sulfur dioxide (SO_2), nitrogen oxides (NOx), aerosol particulate matter (PM), ozone (O_3), and human health (1-4). With rapid economic growth, China has suffered from substantial air pollution, resulting in many studies evaluating the impacts of air pollution on human health (5-8). For example, exposure to NO_2 or SO_2 has been associated with bronchitis, asthma, and emphysema, all of which were more pronounced in children and asthmatic patients (9). Numerous studies have reported that $PM_{2.5}$ and its specific chemical constituents were linked to the incidence of respiratory diseases and mortality as well as lung function (10-12).

In the period of 1993–1996, a cross-sectional study of children's respiratory health in relation to ambient air pollution was conducted based on a gradient in pollutant concentrations across the four Chinese cities of Lanzhou (LZ), Wuhan (WH), Chongqing (CQ), and Guangzhou (GZ) (13-17). The study found that higher air pollution levels were significantly associated with a greater risk for developing symptoms, respiratory disease, and reduced lung function in children. Parents had a greater risk of respiratory diseases. More than 20 years later, with significant changes in many aspects of the society and the population, it is important to understand the extent to which air pollution changes contributed to changes in respiratory health in children in these cities.

In general, heavy air pollution events were highly concentrated in four regions: North China Plain, Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin (18,19). Three of the four cities (WH, GZ, and CQ), located in YRD and PRD, have often been the sites for studying various characteristics of air pollution (20-24). Taking PM pollution as an example, improvement in the PRD region has been substantial in the past decade (making O_3 often become the primary pollutant in recent years) (25), while $PM_{2.5}$ remained the primary pollutant in the YRD region and the Sichuan Basin (26,27). Many studies have also been launched in LZ because of its unique meteorological and geographical conditions (near the desert with four distinct seasons) (28,29). The published reports provided valuable information to understand longitudinal changes in ambient air quality in the four cities.

In the present review, based on the background information of socio-economic development, we aim to systematically examine the changes in air pollution levels and $PM_{2.5}$ chemical compositions from the 1990s to 2017. Data for air pollutants ($PM_{2.5}$, PM_{10}, SO_2, NO_2, and O_3) in the four cities were collected from 1995 to 2017 to study the evolution of the air pollution in each city. In addition, spanning more than a decade we analyzed data on the chemical compositions of $PM_{2.5}$ to identify the temporal and spatial changes in the source apportionments of $PM_{2.5}$. We anticipate that the findings of this review can provide insights to help understand potential changes in health outcomes attributable to changes in air quality from the time of the original health study to the time of the current health study in the same cities. We also expect that the findings can provide historical perspectives on air quality evolutions to inform new control policies.

Data and methods

Study site description

Based on the studies conducted more than 20 years ago, our current analysis included four Chinese cities of LZ, WH, CQ, and GZ (see *Figure 1*). Located in the northeastern side of the Qinghai-Tibet Plateau, LZ is situated in a semi-enclosed Yellow River valley basin that narrows in the north and south and extends in the east and west direction. LZ is characterized by windy and arid springs, which is a high season of sand and dust weather, where the climate has clear vertical variation spectrum and transitional characteristics (30-32). WH is geographically situated at the confluence of the Yangtze and Han Rivers and lies in central China, where the north subtropical monsoon climate offers sufficient light and heat and abundant rainfall (33). Moreover, WH is the core city of the Yangtze River Economic Belt, which has developed as an important industrial base and comprehensive transportation hub in China. CQ is characterized as a mountainous city located in the southwestern part of China and the upper reaches of the Yangtze River. With the annual humidity upwards of 70%–80%, CQ is nicknamed the *City of Fog* (34). As the core city of the Pearl River Delta metropolitan area, GZ is located in the south-central part of Guangdong Province, which is the largest city in South China. GZ belongs to the subtropical monsoon climate zone with an average annual temperature of 20–22 ℃.

Data collection

Data on socioeconomic indicators

It is known that air quality is generally associated with economic development stages. To help understand the long-term trend of ambient air pollution, we collected auxiliary data pertaining to the socioeconomic indicators from the *Statistical Bulletin on National Economic and Social Development* (35-38) in four cities. Annual values of four indicators between 1995 to 2017 included Gross Domestic Product (GDP), resident population, gross industrial output, and domestic car ownership. Measurement units for the indicator values were 100 billion yuan for GDP and gross industrial output, million people for the resident population, and ten thousand vehicles for domestic car

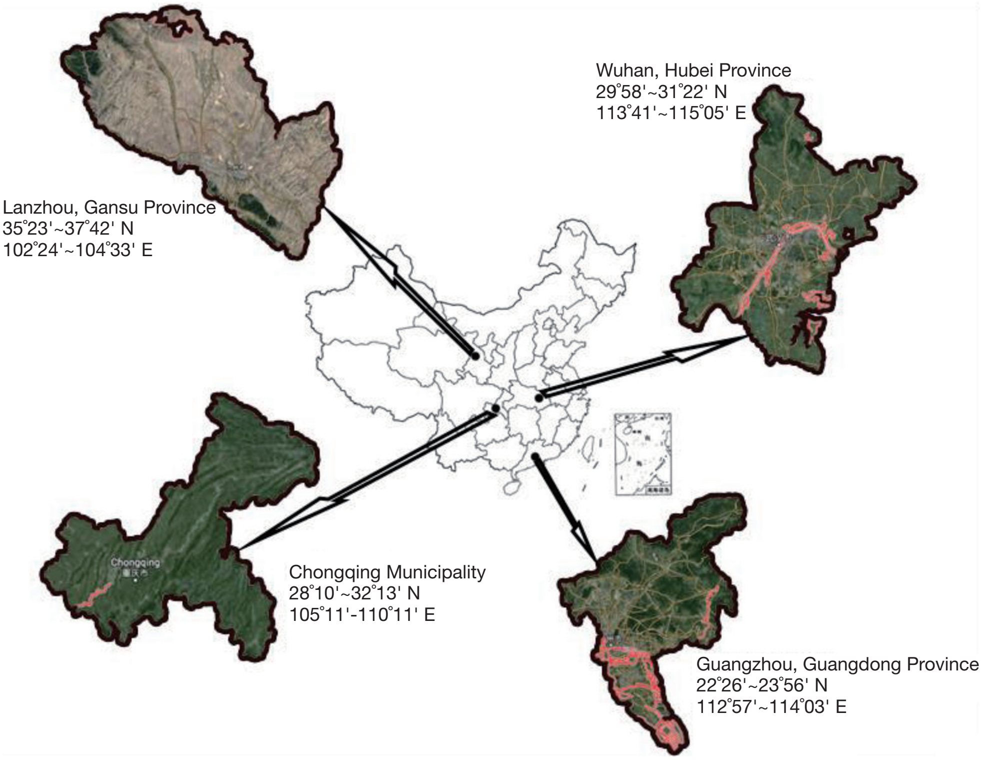

Figure 1 Map showing the four cities included in this study, including Lanzhou (LZ), Wuhan (WH), Chongqing (CQ), and Guangzhou (GZ).

ownership.

Data on air pollutants

The official data of air pollutants were annually averaged and extracted from the *Report on the State of Environment* (25,39-41) for the four cities during 1995 and 2017. Although we collected a significant amount of data, reports documenting annual concentrations of SO_2 and NO_2 in 1997 and 1998 were not available in WH and GZ (data was missing for CQ in 1997), while data from 1997 to 2001 were not available in LZ. Based on the *Chinese Ambient Air Quality Standards* formulated in 1996 (*GB3095-1996*) (42), the state requirement to monitor PM_{10} data began in 2000, replacing total suspended particulate (TSP). Thus, the PM_{10} data were first added into the report from 2000 in CQ, and other three cities began keeping records in 2001. Similarly, the $PM_{2.5}$ monitoring data were first added to the reports in 2012 based on the *Chinese Ambient Air Quality Standards* revised in 2012 (*GB3095-2012*) (43). We chose the daily averaged values in maximum 8-hour O_3 concentrations from 2014 on the website (44) where the data were collected from *China National Environmental Monitoring Centre*, due to the inconsistencies in the reports (such as the different year when data were first added and different type of monitoring O_3 including daily averaged in maximum 8-hour and one-hour averaged).

Data sources of chemical components and source apportionments of $PM_{2.5}$

We reviewed articles regarding chemical composition and source apportionment of $PM_{2.5}$ to summarize spatial and temporal evolution of source emissions in the four cities. We integrated the absolute values and relative proportions of chemical compositions from $PM_{2.5}$ across different years (from 2000), including major water-soluble ions [sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), potassium (K^+), and chloride (Cl^-)], carbonaceous components [elemental carbon (EC), organic carbon (OC)], main toxic and source-characteristic metals [zinc (Zn), manganese (Mn), lead (Pb), copper (Cu), and chromium (Cr)] and others (relative proportion not shown). All relevant studies discussing individual cities are summarized in *Table 1*. Strict screening throughout the entire reviewed processing for considering the variable methods of data presentation in each document

Table 1 Published studies reporting chemical compositions of $PM_{2.5}$ in the four cities during 2000 and 2017

City	References
LZ	Tao, 2009 (45); Li *et al.*, 2016 (46); Shen *et al.*, 2016 (47); Wang *et al.*, 2015 (28); Wang, 2017 (29); Wei *et al.*, 2017 (48); Yang, 2013 (49); Li *et al.*, 2015 (50)
WH	Cheng *et al.*, 2012 (51); Zhang *et al.*, 2012 (52); Qiu, 2014 (53); Zhang, 2014 (54); Cao, 2017 (55); Li *et al.*, 2017 (56); Zhang, 2017 (57); Hao *et al.*, 2018 (20); Zhang *et al.*, 2015 (21); Huang *et al.*, 2016 (12); Cao *et al.*, 2012 (22)
CQ	Cao *et al.*, 2012 (22); Lv *et al.*, 2006 (24); Yang *et al.*, 2011 (24); Zhang, 2007 (58); Zhang *et al.*, 2011 (59); Li *et al.*, 2012 (60); Yu *et al.*, 2014 (61); Jiao *et al.*, 2013 (62); Chen *et al.*, 2017 (63); Li *et al.*, 2014 (64); Huang *et al.*, 2018 (65); Cao, 2017 (55); Lan, 2018 (66)
GZ	Cao *et al.*, 2012 (22); Lai *et al.*, 2007 (67); Cao *et al.*, 2004 (68); Wang *et al.*, 2006 (69); Feng *et al.*, 2011 (70); Tao, 2009 (45); Ma, 2017 (71); Lai *et al.*, 2016 (72); Xiao *et al.*, 2014 (73); Liu *et al.*, 2018 (74); Tao *et al.*, 2017 (1); Zhao, 2018 (75); Li *et al.*, 2016 (46); Tao *et al.*, 2014 (76)

PM, particulate matter; LZ, Lanzhou; WH, Wuhan; CQ, Chongqing; GZ, Guangzhou.

Table 2 Comparison of social and economic development for four cities between 1995 and 2017

Indicators	LZ		WH		CQ		GZ	
	GR*	Range	GR	Range	GR	Range	GR	Range
GDP (100 billion yuan)	11	0.21–2.5	21	0.61–13	16	1.1–20	16	1.3–22
Population (million people)	0.20	2.7–3.3	0.53	7.1–10	0.13	30–34	0.39	6.5–9.0
Gross industrial output (100 billion yuan)	6.3	0.31–2.2	18	0.77–14	27	0.77–21	12	1.7–23
Domestic car ownership (ten thousand vehicles)	19	5.0–102	17	14–261	21	25–567	1.9	87–249

*, each indicator includes the reported value and growth rate (GR) between 1995 and 2017. LZ, Lanzhou; WH, Wuhan; CQ, Chongqing; GZ, Guangzhou.

was reflected in the selection of long-term research data for one subject group in a given city. We found that the data for EC and OC were not available before 2013 in LZ based on the above screening principles. Additionally, normalization methods were used to eliminate gaps between the data with a wide range of sources. The model equation can be written as follows:

$$y\%_{ij} = \sum_{k=1}^{n} (x/\mathrm{PM}_{2.5})_{ijk}/n \qquad [1]$$

where $y\%_{ij}$ is the normalized proportion of one component in $PM_{2.5}$ of city j in year i; x is the absolute concentrations of component, while n represents the article counts of city j in year i.

To better describe the local pollution characteristics during the examined period, we also reviewed the annual environmental protection regulations and specific implementation methods from the *State and Urban Report on the State of Environment* in the *Environmental Protection Bureau*'s official website (25,39-41).

An overview of social and economic development in four cities

Since entering into the 21st century, China has experienced rapid urbanization and an exponential economic growth (77). Based on the available accounting, the four cities presented an obvious gap in the urban development for diverse socioeconomic indicators summarized in *Table 2*.

By 2017, GZ was recognized as one of the first-tier cities with the largest regional GDP (2,150 billion yuan) and gross industrial output (2,269 billion yuan) compared to other cities. Even in 1996, the number of domestic vehicles in GZ reached 871 thousand vehicles, resulting in the lower growth rate compared to other cities. The resident population has increased by 2.51 million people, and the growth rate since 1995 was only second to WH (where the growth rate was up by 53.4%, from 7.10–10.89 million people). The available data suggest that WH experienced very significant development owing to the highest growth rates of GDP by 21 times and resident population growth

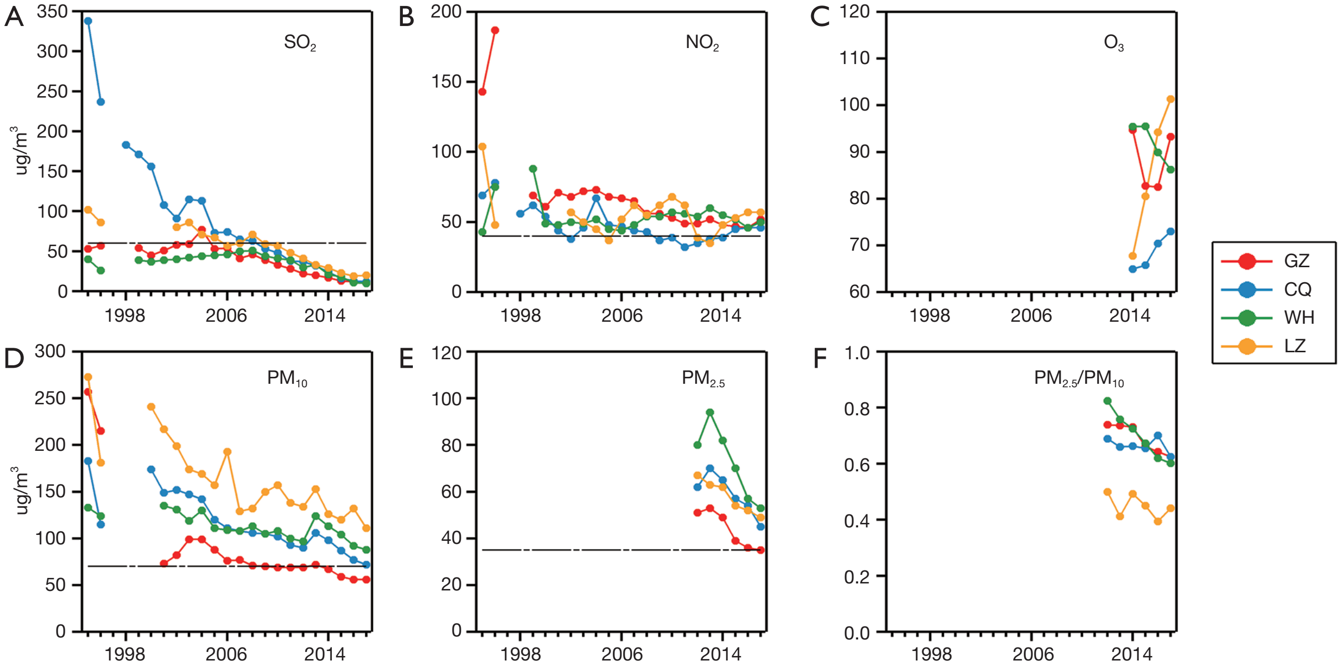

Figure 2 Temporal variations in five major air pollutants in four cities: (A) SO$_2$, (B) NO$_2$, (D) PM$_{10}$ for 1995–2017, (C) O$_3$, (E) PM$_{2.5}$, (F) PM$_{2.5}$/PM$_{10}$ for 2012–2017. The black dotted line indicates the second-level concentration standards of pollutants according to the government *GB3095-2012* (43) standard. SO$_2$, sulfur; O$_3$, ozone; PM, particulate matter.

by 53.4%. Although GZ and CQ were growing at the same rate in GDP (a multiple of 16) from 1995 to 2017, the development strength of CQ ranked number 5 among all large Chinese cities, reflected in approximately a 9.5% increase in GDP growth rate in 2017 compared to 2016 (78), reaching almost 2 trillion yuan, significantly more than GDP growth rate observed for GZ. In contrast, the slowest development was observed for LZ which had the lowest values for all indicators in 2017 compared to the Yangtze River Delta and PRD cities (40).

Long-term variations in major air pollutants

Using available research data on the effects of air pollution on human respiratory health across 20 years (79,80), we analyzed the long-term trends of air pollutants including SO$_2$, NO$_2$, PM$_{10}$ [1995–2017], PM$_{2.5}$ and PM$_{2.5}$/PM$_{10}$ [2012–2017] and O$_3$ [2014-2017]. The results are summarized in *Figure 2*.

SO$_2$

Data presented in *Figure 2A* indicate a significant downward trend for SO$_2$ annual concentrations among the four cities from 1995 to 2017. In 2017, SO$_2$ levels in all the cities dipped below 20 μg/m^3, which were approximately one fourth of the values reported in the late 1990s. The SO$_2$ levels in CQ showed the greatest improvement over the years (81), with a 96.4% decrease from 338 μg/m^3 in 1995 to 12 μg/m^3 in 2017. A similar change was observed in LZ, where the declining rate of SO$_2$ was 80.4% (from 102 μg/m^3 in 1995 to 20 μg/m^3 in 2017). The trends in SO$_2$ levels were drastically different for GZ and WH which showed gradual rise prior to 2004 and 2008, respectively. Then we saw the values in all four cities descending below the second-level concentrations (60 μg/m^3) after 2008. It was related to the rigorous investigation of reducing pollutant emissions in the Eleventh Five-Year Plan (25,39-41). Despite air quality standards revision in 2012, SO$_2$ was still considered as one of the six major pollutants, resulting in sustained efforts put into monitoring of SO$_2$ performed across the country (43). The type of pollution has shifted from soot-pollution (PM$_{10}$ associated with SO$_2$) to single type of PM (PM$_{2.5}$ and PM$_{10}$ pollution) in some cities (e.g., the primary pollutant in LZ was PM$_{10}$ and PM$_{2.5}$, while in CQ and WH was

$PM_{2.5}$), which resulted from a series of measures to reduce air pollution, such as the implementation of coal-to-gas projects and clean coal-fired technologies (82).

NO_2

Based on the data in *Figure 2B*, the NO_2 levels have not really changed between 1995 and 2017, showing a relatively flat trend compared with SO_2. Given the NO_2 concentration values from 1995, the reductions of 91, 23, and 47 μg/m^3 were seen in GZ, CQ and LZ, respectively, while there was a rise of 7 μg/m^3 in WH. However, in recent years, the level of NO_2 pollution has far exceeded the second-level concentration (standard) after 2014 in all four cities. The main anthropogenic source of nitrogen oxides in cities is the burning of fossil fuels, two-thirds of which were emitted from mobile sources such as motor vehicles, and one-third from fixed sources such as factories and power plants (83). The available data indicate that the popularity of clean fuels has increased over the years, and exhaust emissions of motor vehicles have been continuously reduced (84). Nevertheless, the rapid urbanization has led to a sustained increase in the number of motor vehicles, dramatically increasing the emission of NO_2 to some extent (49,64). The evolution of NO_2 pollution and factors associated with NO_2 emissions remain complicated.

O_3

Ozone, with annual mean concentrations between 64 to 102 μg/m^3 now, has become the main risk restricting the optimization of urban air quality after $PM_{2.5}$ level declined in many cities in recent years. Based on the result in *Figure 2C*, the daily average in maximum 8-hour O_3 concentrations showed upward trend since 2015 in GZ, CQ, and LZ. Especially, LZ has experienced a remarkable increase leading to the highest values (101.3 μg/m^3) in 2017. It is tangible that volatile organic compounds (VOCs) and NOx provide important precursors for ozone formation (33) largely deriving from process of petroleum refining and vehicle exhaust emissions (85). The rising trend of ozone was also highly consistent with the rise of NO_2 (*Figure 2B*) in the same period, which have been reported in previous studies (86,87). In general, longer daylight hours and stronger solar radiation contribute to ozone levels (22). The PRD region has subtropical climate that favors ozone formation when precursor pollutants are present. This made O_3 pollution receiving particular attention in GZ and Shenzhen (88). Although O_3 concentration in WH appeared to be on a decreasing trend, it was in a high concentration range (between 80 and 100 μg/m^3).

PM_{10}

A clear reduction in PM_{10} levels has been found between 1995 and 2017 in the four cities (see *Figure 2D*). Compared to 1995, the levels of PM_{10} in 2017 were 78.2%, 60.7%, 59.3%, and 33.8% lower in GZ, CQ, LZ, and WH, respectively. However, the pollution level in 2017 was still higher than the second-level concentration (70 μg/m^3) in CQ, LZ, and WH, while the value in GZ dropped below the standard for the first time in 2014. These reductions are likely a reflection of the active rectification of PM_{10} carried out at the nationwide level which has achieved good results (44). The exception occurred in 2013, when the concentration of PM_{10} was significantly increased in all four cities corresponding to the worst smog in China that occurred the same year, which spread to 25 provinces and affected more than 100 different cities (44). Interestingly, LZ was heavily polluted with PM_{10}, which was mainly from the natural and meteorological conditions (89-91). Additionally, LZ suffered from a very dusty weather all year with frequent dust storms in the city resulting in high PM_{10} concentrations (92). In general, the technological transformation, tightening of environmental management policies and increased funds (25,39-41) for controlling dust and coal emission were efficiently utilized in all four cities.

$PM_{2.5}$

Monitoring of $PM_{2.5}$ levels began in 2012 and showed very comparable trends to PM_{10} pollution. We also calculated the ratio of $PM_{2.5}/PM_{10}$ that represents the composition of particulate pollution. The larger the ratio, the higher the mass concentration of fine particles (respirable particles) in total inhalable particles (all particles with an aerodynamic diameter ≤10 μm). For the same inhaled mass of PM_{10}, a higher $PM_{2.5}/PM_{10}$ ratio means that more fine particles can reach and deposit in the deep lung and cause more health damages (93). Specifically, there was a clear downward trend in $PM_{2.5}$ levels from 2013 in GZ, CQ and WH, with decreases of 18 μg/m^3, 25 μg/m^3, and 42 μg/m^3 respectively (concentrations dropped 18 μg/m^3 since 2012 in LZ). Unlike with PM_{10}, the most serious pollution with $PM_{2.5}$ was observed in WH. Data suggest that the transport of local air masses from the northeast of WH may have contributed the

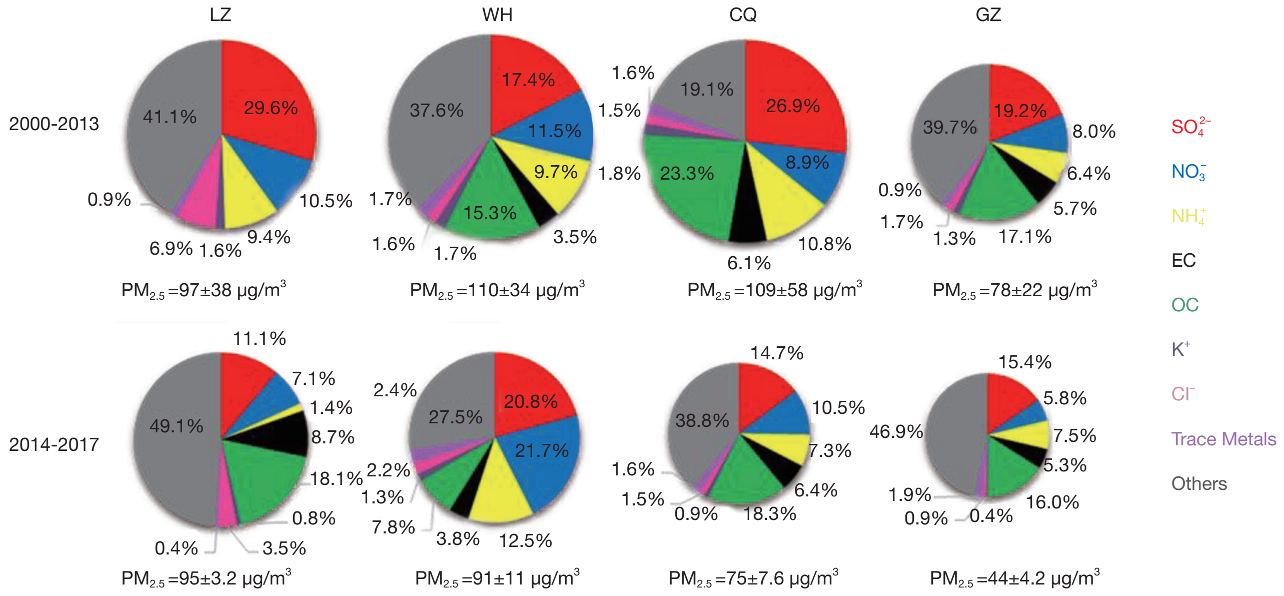

Figure 3 Spatiotemporal variation of mass fractions in major chemical composition of $PM_{2.5}$ in the four cities prior to and after 2013. Digital label for every sector indicates the corresponding component proportion, and the size of each fan corresponds to the absolute concentration of $PM_{2.5}$. PM, particulate matter.

most to $PM_{2.5}$ pollution, which originated from the cluster of steel plants located in the northeast region of WH (94). As shown in *Figure 2F*, we observed continuously decreasing trends of $PM_{2.5}/PM_{10}$ ratio from 2012 in both GZ and WH, while LZ had the lowest value because of serious PM_{10} pollution (shown in *Figure 2D*). Although the pollution level was overall reduced, the other three cities did not meet the second-level standard (35 μg/m^3) formulated in GB3095-2012 (43), while GZ has reached the value of 35 μg/m^3 in 2017. The strict prevention and control during the multi-sport Asian Games played an important role, leading to reductions in PM levels in the southern China compared to other regions of the country (95).

Spatiotemporal variation of chemical composition and sources of $PM_{2.5}$ in four cities during 2000 and 2017

Based on the available literature, the chemical characteristics of $PM_{2.5}$ in China are considered to be a mixture of organic and inorganic matter including water-soluble ions, elemental carbon, crustal material, and hydrocarbons (63,96-98), mainly originating from meteorological evolutions and potential human activities, such as transportation, household activities, vehicular movement and industrial sector (99). Due to the regional economic development, changes in industrial and energy structures, and an increasing number of vehicles, the composition ratios of $PM_{2.5}$ vary with location and time (100). Based on the policy—*Air Pollution Prevention and Control Action Plan*, introduced by the Chinese government in 2013 (101), we compiled the available data representing the average $PM_{2.5}$ concentrations and the relative composition of $PM_{2.5}$ during two periods (2000–2013 and 2014–2017) in the four cities (*Figures 3* and *4*).

Review of $PM_{2.5}$ concentrations

The averaged $PM_{2.5}$ concentrations after 2013 were 34.5, 34.4, 19.1 and 2.3 μg/m^3 lower than previous values at first periods [2000–2013] in GZ, CQ, WH and LZ, respectively (*Figure 3*). The data were in agreement with the results presented in *Figure 2E*. A significant decrease in the average values of $PM_{2.5}$ occurred geographically from north to south after the overall rectification in 2013, ranging from 39 to 102 μg/m^3.

SO_4^{2-}, NO_3^-, and NH_4^+

The proportions of three ions, crucial elements of the secondary inorganic component of air pollution, including SO_4^{2-} (8.4–57%), NO_3^- (4.2–21%) and NH_4^+ (2.3–19%),

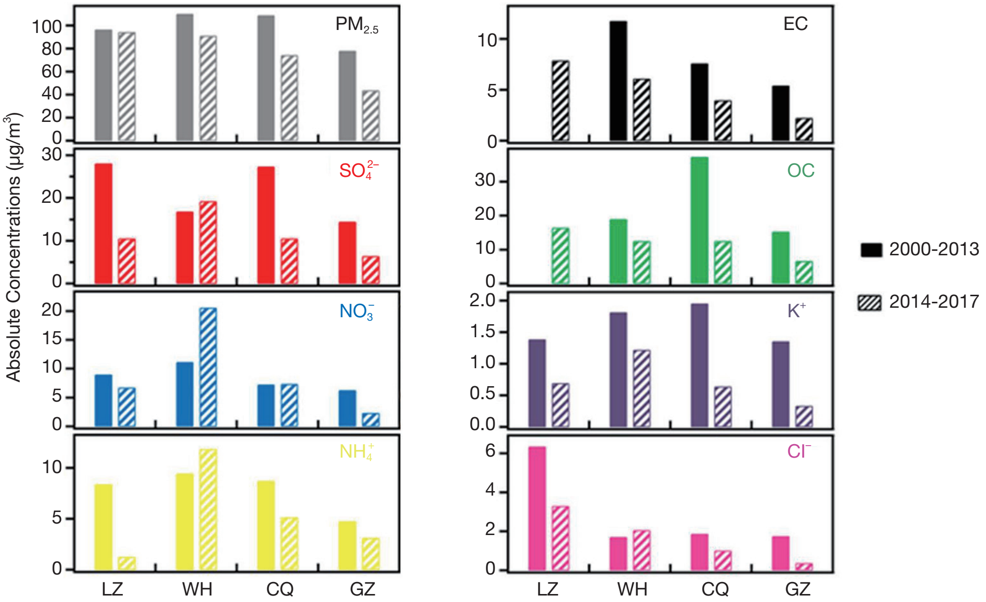

Figure 4 Variations in the absolute concentrations (μg/m^3) of examined $PM_{2.5}$ and seven components before and after 2013 in LZ, WH, CQ, and GZ. Solid column represents the first period [2010–2013] while the dotted line column indicates the second period [2014–2017] in each component across four cities. PM, particulate matter; LZ, Lanzhou; WH, Wuhan; CQ, Chongqing; GZ, Guangzhou.

showed different changes across different dimensions. Considered as the first major anthropogenic source, SO_4^{2-} contributed the most. The data indicate that LZ had the largest absolute levels of sulfates (28±18 μg/m^3) prior to 2013, followed by CQ, where the absolute concentration was upwards of 27±15 μg/m^3. Given high levels of SO_4^{2-}, there was a possibility that the sulfides from the coal burning had undergone a secondary conversion, indicating that CQ and LZ experienced more serious coal combustion emission prior to mandatory rectification in 2013. The long-term use of coal-fired heating in LZ and clusters of factories in CQ contributed significantly to high SO_4^{2-} levels (32,102). Importantly, recent concentrations of SO_4^{2-} in GZ, CQ and LZ decreased sharply, with average values of 6.5±2.2, 10±3.2 and 11±3.7 μg/m^3, respectively.

Based on the literature evidence suggesting that approximately 50% of nitrate mass can be attributed to coal combustion (103), NOx, as the precursor of nitrate, is mainly derived from urban anthropogenic activities such as traffic and factory emissions. The continuously upward trend occurred in WH during the two periods (average values 11–20 μg/m^3). A slightly upward trend in the levels of NO_3^- also occurred in CQ mainly due to the surge in the number of motor vehicles (*Table 2*), while LZ and GZ showed considerably lower levels of NOx emissions.

The trends of NH_4^+ in four cities were somewhat comparable to trends seen for SO_4^{2-} and NO_3^-. Specifically, we observed that the secondary inorganic pollution sources dominated the composition of $PM_{2.5}$ in WH. Cheng *et al.* (104) found stronger oxidation process of SO_2 and NO_2 in the atmosphere from research between 2016–2017. Due to the massive burning of fossil fuels, coal and biomass, the secondary inorganic aerosols accounted for a large proportion in the industrial area of WH, while the soil source dominated the large traffic volume and frequent urban construction throughout the year (29). Notably, in LZ, only 1.3±0.8 μg/m^3 of concentration value was recently normalized and similar results were found for ammonium levels which were lower than 3% in Northwest China during 2006–2013 based on a previous study (105). Overall, the data suggest that the secondary source of $PM_{2.5}$ in the northwest region (like LZ) was relatively small due to the yearly dry climate, which is not conducive to the occurrence of secondary reactions (48).

EC and OC

The levels of OC ranked first or second among the constituents shown in *Figure 3* in four cities, accounting for approximately 7–30% in terms of fraction. The proportions

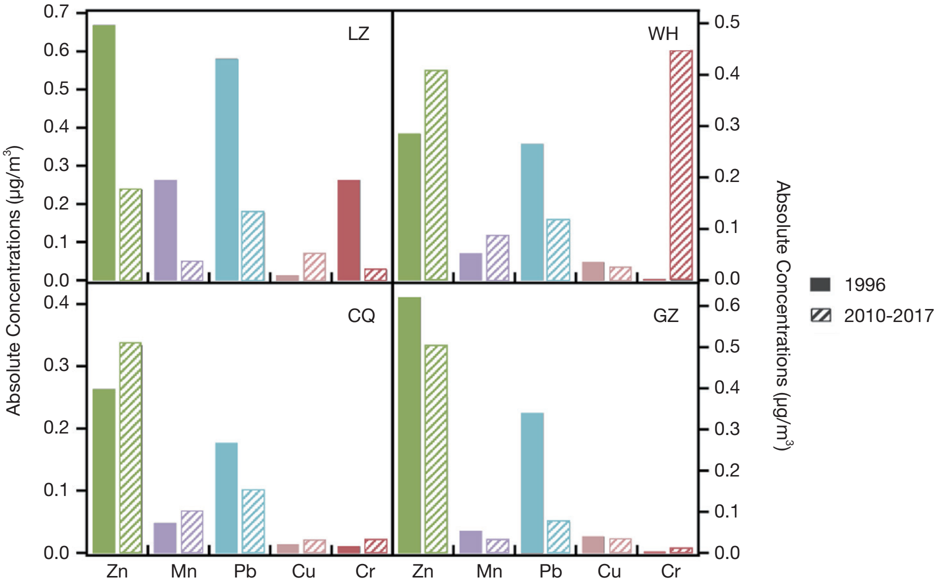

Figure 5 Comparisons of concentrations of five major metal elements during two periods [1996 and 2010–2017]. Solid column represents the first period [1996] while the dotted line column indicates the second period [2010–2017] for each component across four cities.

of OC were relatively reduced after 2013 in all regions, especially in CQ (from 37±21 μg/m^3 in first period to 13±2.2 μg/m^3 in second period). Other studies suggested that OC can be used to estimate the organic matters (OM), which is typically obtained by multiplying OC by a specific coefficient (63). Additional indirect data suggest that VOCs, the precursor of organic matter, were recently increasing. Taken together, CQ had the highest organic pollution compared to other cities prior to 2013 and the most effective control over VOC emissions. As a primary burning indicator, EC accounted for approximately 5–9% of emissions in the four cities. There were clear downward trends in WH, CQ and GZ, presenting the absolute concentration changes of 12±7.1 to 6.1 μg/m^3, 7.6±3.3 to 4.0±1.3 μg/m^3 and 5.4±2.9 to 2.3±0.4 μg/m^3, respectively. Geographically, values of EC decreased from north to south. The EC had a similar source like particulate organic carbon (POC), and was not a major chemical fraction in aerosol particles found in China (105).

K^+ and Cl^-

The highest average concentrations of Cl^- were seen in LZ (6.4±1.9 μg/m^3 and 3.3±0.6 μg/m^3 before and after 2013, respectively) while the trends for other cities were relatively flat. Combined with the highest proportion of Cl^- (3–7%) in LZ, the data further suggested that coal burning was the main source of aerosol pollution in this local area. Primary component K^+, as a biomass indicator, showed significant decreases in average concentrations across all cities (ranging from 0.5 to 2.1 μg/m^3), suggesting that the overall control of biomass burning has achieved very good results.

Trace metal elements

Data summarized in *Figure 5* show the comparison of trace metal concentrations in 1996 and in recent years (from 2010 to 2017) in four cities, including Zn, Mn, Pb, Cu, Cr, that are either specific source indicators or trace elements with serious impact on human health (106-109).

The concentrations of metal elements were generally low, ranging from 0.003 to 2.8 μg/m^3 more than 20 years ago, compared to recent values in the range of 0.012–1.7 μg/m^3. Data suggested that Mn, Zn, Pb were mainly derived from metal smelting (110) or combustion processes (111). Additionally, we observed the highest concentration values of Zn in all cities due to background content in soil and supernumerary content of vehicle exhaust emissions and tire wear (112). A small increase in the concentration of Zn and Mn was found in CQ and WH, while LZ and GZ showed an obvious decrease. The levels of Cu in CQ and LZ showed significant rise, while they were relatively flat in WH and GZ. Two toxic metals, Pb and Cr (113), showed variable trend concentrations across the four cities. At present, coal burning and industrial production (such as smelting and sintering process) have become the main sources of Pb due to mandatory use of unleaded gasoline since late 1990s (114). During the 12th Five-Year Plan period for controlling heavy metal pollution, Pb was included as one of the key target pollutants (115). Emission of Pb from factories seem to be effectively controlled in recent years (114). However, emissions of Cr from coal burning and industrial production appear to have increased significantly in WH, possibly due to the specificity of the study site (e.g., near the factory) in the reviewed literatures.

Conclusions and perspectives

Inter-annual variations in five major pollutants across two decades and chemical components of $PM_{2.5}$ in the recent years in the four cities were comprehensively summarized. It helped to provide a better understanding of the evolution of pollution sources for studying changes in health outcomes related to changes in air quality in populations with similar ages living in the same cities. The findings can be summarized as follows:

(I) All four cities experienced rapid growth over the last two decades; however, there were obvious geographical differences. For example, GZ, as one of the first-tier cities, was ranked first in GDP among all cities, while LZ showed slow development. Of note, the growth rate of CQ was the highest as of 2017 from the prior year.

(II) The evolution of five conventional pollutants in GZ, LZ, WH and CQ varied with space and over time. Clear downward trends occurred in SO_2 and PM (including PM_{10} and $PM_{2.5}$) levels. SO_2 concentrations in all four cities were below the second-level concentration (standard) of 60 $\mu g/m^3$ after 2008, while $PM_{2.5}$ and PM_{10} were still not up to the standard, except for PM_{10} in Guangzhou. In particular, the greatest improvement of SO_2 pollution occurred in CQ (a decline of 96.4%), and GZ showed the best results for reduction in particulate pollution. The levels of NO_2 showed relatively flat trends compared to other pollutants. Importantly, O_3 concentrations have been on rise, which should be considered in examining the effects of ambient air pollution on human respiratory health.

(III) Among the chemical components of $PM_{2.5}$, organic carbon and SO_4^{2-} dominated $PM_{2.5}$ mass concentration in all cities. The overall levels of pollutants showed decreases after 2013 in LZ, CQ and GZ, but the trend was the opposite in WH showing an upward trend in SO_4^{2-}, NO_3^-, NH_4^+, and Cl^- between 2014 and 2017. Moreover, WH had the highest mass fraction of SO_4^{2-}, NO_3^- and NH_4^+, indicating that the control of coal combustion and vehicle emissions should be stricter in this city. Finally, LZ, among the four cities, had the lowest proportion of secondary components and highest levels of EC and Cl^- mainly emitted from perennial coal-fired emissions.

(IV) The concentrations of different metals showed different long-term trends, ranging from 0.003 to 2.8 $\mu g/m^3$ more than 20 years ago, compared to recent values in the range of 0.012–1.7 $\mu g/m^3$.

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Footnote

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