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Spatial and temporal differentiation of air quality and its influence factors in 16 cities in Shandong Province from 2019 to 2020†

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Ambient air quality is a complex dynamical system that is affected by a number of subsystems, such as particulate matter emission, meteorological factors, and socioeconomic factors. However, the mechanism of action of meteorological factors and socioeconomic factors and correlation between cities are unclear. The spearman correlation coefficient was used to analyze and explore the spatial and temporal differentiation of air quality (SO_2, NO_2, PM10, PM2.5, CO-95per and O_{3-8h}), the proportion of days with heavy pollution (PDHP), the proportion of days without pollution (PDWP) and influencing factors (particulate matter emission, meteorological factors and socioeconomic factors) in 16 cities in Shandong Province in the period 2019–2020. The results indicated that the concentrations of SO_2, NO_2, PM10, PM2.5 and CO-95per showed a trend of "winter high, summer low", while the concentration of O_{3-8h} showed a trend of "winter low, summer high". The air quality of coastal cities was better than that of inland cities. Temperature and precipitation were negatively correlated with the concentrations of SO_2, NO_2, PM10, PM2.5, CO-95per ($P < 0.05$). There were significant positive correlations between SO_2 and the proportion of secondary industry, between PM10 and population density, and between PM2.5 and population density. The annual mean concentrations of PM2.5, PM10, NO_2 and SO_2 were positively correlated, and the positive correlation between PM10 and PM2.5 was extremely significant. The correlation between pollutants in the heating period and nonheating period was significantly different. PM2.5/O_{3-8h} and PM2.5/NO_2 had no correlation in the heating period but had a significant positive correlation in the nonheating period. The PDHP in the heating period was positively correlated with SO_2, NO_2, PM10, PM2.5 and CO-95per, indicating that the air pollutants released during the heating process in winter are important factors that affect the air quality. There was a significant positive correlation between different cities in Shandong Province, and the correlation was affected by the distance between cities and the type of pollutants.

Sustainability spotlight

It is noteworthy that air pollution is a serious problem in Shandong Province and special efforts are needed to overcome it. This study is environmentally significant as it reveals the spatial and temporal differentiation of air quality in 16 cities in Shandong Province and its influencing factors (particulate matter emissions, meteorological factors and socioeconomic factors). In this study, we found that there is a correlation between the concentration of pollutants in Shandong Province and environmental implication (pollution in the heating or nonheating period, types of pollutants, distance between cities). Our results provide important insights into improved air quality that could aid in the development of public policies associated with air pollution.

1 Introduction

Ambient air quality is a complex dynamical system that is affected by a number of subsystems, such as particulate matter emission, meteorological factors, and socioeconomic factors. With the advancement of industrialization and urbanization, air pollution problems are becoming increasingly prominent.[1–3] Serious air pollution has potential negative effects on the ecological environment and human health.[4,5] Facing the

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increasingly severe situation of air pollution control, the Chinese government has implemented a series of environmental treatment policies to continuously strengthen air pollution regulation.[6] In 2013, the State Council issued Air Pollution Prevention and Control Action Plan, which proposed that the concentration of fine particulate matter in cities at the prefecture level and above should be reduced by at least 10% from 2012 levels by 2017, the number of days with good air quality should be increased yearly, and the concentrations of fine particulate matter in the Beijing-Tianjin-Hebei region, the Yangtze River Delta and the Pearl River Delta should be reduced by approximately 25%, 20% and 15%, respectively.[7,8]

Shandong is one of the most prosperous and heavily polluted provinces in the country, with significant coal mining, oil refineries, and metallurgical and mechanical sectors.[9] In recent years, Shandong Province has carried out air pollution control work. The Shandong Provincial Government formulated the 2013–2020 Air Pollution Prevention and Control Plan of Shandong Province, specifying specific tasks in six aspects, including actively adjusting the energy structure, vigorously adjusting the industrial structure, deepening the pollution control of key industries, strengthening the comprehensive control of dust, and requiring that the environmental and air quality of the whole province be up to standard by 2020 (approximately 50% better than that in 2010).[10]

Improving the air quality in Shandong Province is critical to the success of the Three-year Action Plan to Win the Blue Sky Battle and the air quality improvement targets in the Beijing-Tianjin-Hebei region. According to the 2017–2018 Autumn and Winter Comprehensive Air Pollution Control Action Plan for the Beijing-Tianjin-Hebei Region and its Surrounding Areas issued by the Ministry of Ecology and Environment in 2017, Shandong cities such as Jinan, Zibo, Jining, Dezhou, Liaocheng, Binzhou and Heze were identified as transport routes for air pollution between Beijing, Tianjin and Hebei. Therefore, the pollutants emitted from Shandong can also be transported to Beijing-Tianjin-Hebei under the influences of southerly/southeasterly winds.[11] The control and prevention of air pollution in Shandong Province not only helps to improve the air quality in the cities and regions of Shandong Province, but also helps to reduce the transmission impact to surrounding areas such as Beijing, Tianjin and Hebei.[12] It is necessary and vital to know more about the spatial and temporal differentiation of air quality and its influence factors in Shandong Province to realize the coordinated development not only for Shandong Province, but also the Beijing-Tianjin-Hebei Region.

Domestic and foreign scholars have studied air pollutants and their influencing factors in Shandong Province. Wu *et al.*, 2022 proposed a geospatial extension relational analysis model to identify the drivers of ambient air quality.[13] The results indicated that the air quality in eastern coastal Shandong can be improved by increasing the urbanization ratios, per capita GDP, proportion of tertiary industry, internal expenditure on R&D, disposable income and coverage rate of urban green areas or reducing energy consumption per 10 000 yuan of GDP. Using PM2.5 data from 2014 to 2017 inversed from daily PM2.5 data in Shandong Province and employing spatial autocorrelation and geographical detector methods, Yu *et al.*, 2021 revealed the temporal and spatial evolution of PM2.5 concentrations in Shandong Province and their driving factors.[14] Geographic detection analysis indicated that the main driving factors for the change in PM2.5 concentration in Shandong Province were crop broadcast area and soot emissions. Zhang *et al.*, 2018 used samples collected simultaneously in one year at four sites in Shandong (Zibo, Zaozhuang, Qingdao and Jinan) to identify the source of PM2.5 and analyzed the related health risks.[15] Based on a positive matrix factorization (PMF) model, they concluded that secondary formation, coal combustion and industry emissions were the main sources of PM2.5 in Shandong.

However, the regional air quality is affected by natural environmental factors and human activities.[16] At present, most of the studies on the air quality in Shandong Province focus on the inter-annual or provincial scale, and there are few comprehensive research studies on the air quality in Shandong Province on multi-time scales (inter-monthly, seasonal and inter-annual scales), multi-spatial scales (regional and provincial scales) and multi-influencing factors (meteorological factors, industrial and domestic pollutant emissions and socio-economic factors). Therefore, the work of the air pollution prevention and control needs to enter a new situation of "season-year-round" and "city-region" coordination. Meanwhile, attention should be paid to the collaborative control between O_3 and PM2.5, so it is significantly important to study the spatial and temporal differentiation of air quality and its influence factors.

In this study, correlation analysis was conducted on the data of air pollutant concentration, meteorological factors and socioeconomic conditions in Shandong Province during 2019–2020 to clarify the spatiotemporal differentiation characteristics of air quality in Shandong Province, the correlation between pollutant indicators and particulate matter emissions, meteorological factors and socioeconomic factors, as well as the correlation between pollutants and pollutants and between different cities. This analysis provides a reference for national and Shandong Province air quality improvement and pollution control during the "14th Five-Year Plan☐.

2 Materials and methods

2.1 Study domain and data source

Shandong Province (34°22.9′ – 38°24.01′ N, 114° 47.5′ – 122° 42.3′E) is located in the eastern part of the North China Plain, in the lower reaches of the Yellow River, near the Bohai Sea and the Yellow Sea, and includes inland and peninsula areas. It has a warm temperate monsoon climate, spring drought, little rain, and significantly varying daily temperatures. In summer, there is a prevailing southeast monsoon that is hot and rainy. Autumn is sunny and cool with less precipitation, and winter is cold and dry under the control of continental high pressure. The heating period is from November to March.

The distribution of 309 monitoring points in 16 cities in Shandong Province selected by this study is shown in Fig. 1. Air quality data (SO_2, NO_2, PM10 and PM2.5, O_{3-8h}, concentration of CO-95per proportion, proportion of days with heavy pollution

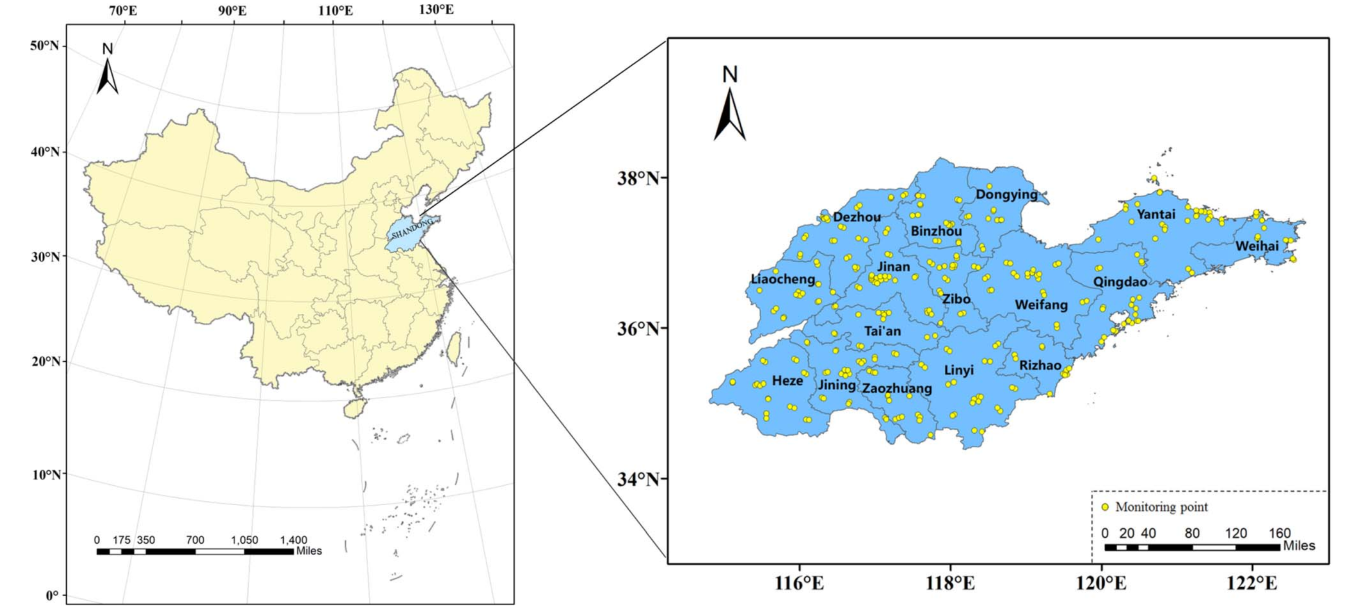

Fig. 1 Distribution of air quality monitoring stations in Shandong Province.

(PDHP), proportion of days without pollution (PDWP)) and the distribution of monitoring points were derived from the Shandong Province Ecological Environment Agency (**https://www.sthj.shandong.gov.cn/**). Particulate matter emissions, meteorological data and social economic data were obtained from the Shandong Province Bureau of Statistics (**https://www.tjj.shandong.gov.cn/**). Meteorological elements included air temperature and precipitation. Social and economic factors included the gross regional product (GRP), investment in environmental protection, proportion of secondary industry, population density, green coverage and population growth rate.

2.2 Calculation of AQI

The calculation of AQI was based on the Technical Regulation on Ambient Air Quality Index (HJ633-2012). Table S1† shows the individual air quality index (IAQI) and limited value of related pollutant. The IAQI and AQI were calculated as follows:

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

$$\mathrm{AQI} = \max\{\mathrm{IAQI_1}, \mathrm{IAQI_2}, \ldots, \mathrm{IAQI}_n\}$$

where $\mathrm{IAQI_P}$ was the IAQI of pollutant (P), C_P was the mass concentration of pollutant (P), $\mathrm{BP_{Hi}}$ was the high-value of the limited value of related pollutant for C_P in Table S1,† $\mathrm{BP_{Lo}}$ was the low-value of the limited value of related pollutant for C_P in Table S1,† $\mathrm{IAQI_{Hi}}$ was the IAQI for $\mathrm{BP_{Hi}}$ in Table S1,† $\mathrm{IAQI_{Lo}}$ was the IAQI for $\mathrm{BP_{Lo}}$ in Table S1,† and n was the number of the pollutant. The classification and level of AQI are shown in Table S2.†

2.3 Spearman correlation coefficient analysis

Using SPSS 27.0, the Spearman correlation coefficient was used to calculate the correlation between the pollutant concentration and particulate matter emissions, meteorological factors, socioeconomic factors, pollutant indicators and cities. The calculation process is as follows:

$$r_\mathrm{s} = 1 - \frac{6\sum_{i=1}^{n} {d_i}^2}{n(n^2 - 1)}$$

where r_s is the Spearman correlation coefficient, d is the rank difference of samples x_{ij} and y_{ij} from the smallest to the largest, n is the sample size, and $n = 16$. When calculating the correlation between the pollutant and particulate matter emissions for 2020, x_{ij} is the mean concentration of a pollutant in a city, $i = 1,2,3\ldots$, i means different cities, $j = 1,2,3\ldots$, and j means the mean concentration of SO_2, NO_2, PM10, PM2.5, CO-95per and O_{3-8h}. y_{ij} is the emission of certain particulate matter in a city in 2020, $i = 1,2,3\ldots$, i means different cities, $j = 1,2,3\ldots$, j represents total urban particulate matter emissions, domestic particulate matter emissions and industrial particulate matter emissions. The other correlation calculation process of other indicators is consistent with the correlation calculation process of pollutant-particulate matter emissions.

3 Results and discussion

3.1 Spatiotemporal variation of air quality in Shandong Province

The monthly changes in atmospheric pollutant (SO_2, NO_2, PM10, PM2.5, CO-95per and O_{3-8h}) concentrations, PDHP and PDWP in 16 cities in Shandong Province during 2019–2020 were statistically analyzed. The results are shown in Fig. 2, showing significant spatiotemporal differences in air quality in Shandong Province.

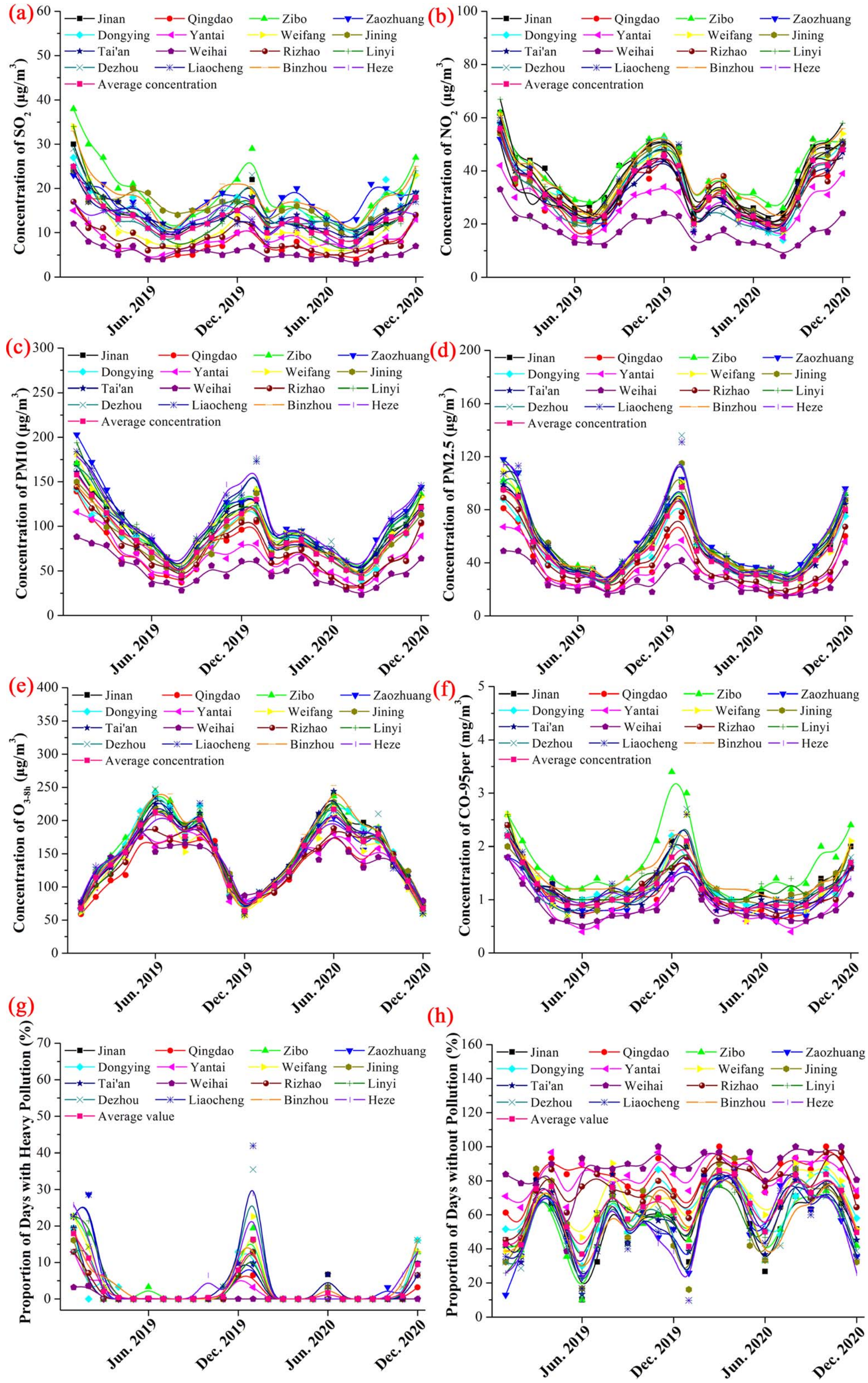

Fig. 2 Air quality in Shandong Province: concentration of SO_2 (a), NO_2 (b), PM10 (c), PM2.5 (d), O_{3-8h} (e) and CO-95per (f), PDHP (g) and PDWP (h).

In terms of time scale, the provincial index of pollutant concentration in 2020 was better than that in the same period in 2019, and the improvement in air quality from February to April of 2020 was the most prominent, which could be attributed to the COVID-19 outbreak in the winter of 2019. The government successively adopted mandatory quarantine and other control

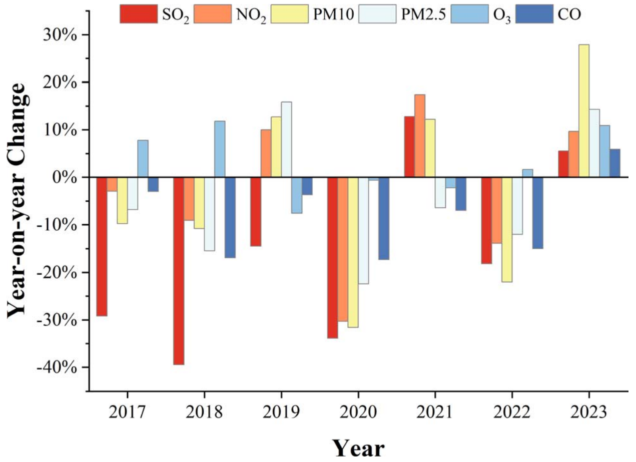

Fig. 3 The year-on-year change in the pollutant concentration from January to March (lockdown period in 2020) of 2017–2023 (year on year: comparison of the nth month of a year with the nth month of the previous year).

measures to restrict the outdoor activities of the population, and the province fell into an unprecedented state of closure, which lasted approximately 6 weeks.[17] This action reduced the use of fossil fuels and vehicles, thereby reducing emissions of pollutants from both domestic and industrial sources.[18,19] The year-on-year improvement in air quality during the 2020 lockdown period (January to March) was the best for the same period from 2017 to 2023 (Fig. 3). The pollutant concentration during January to March (lockdown period in 2020) of 2017–2023 is shown in the ESI (Table S3†). Among them, the year-on-year improvement of NO_2, PM10, PM2.5 and CO during the containment period in 2020 is the largest in recent years. At the same time, only the concentration of six basic pollutants decreased during the containment period in 2020.

In contrast to the other air pollutants, O_3 has remained at a similar level or even shown enhancements compared to pre-lockdown periods. Many studies from China and other countries also found that O_3 showed an increase during lockdowns.[20–24] However, overall, during the COVID-19 pandemic lockdowns, emissions from human activities were stalled. Improvements in the air quality in many cities around the world have been reported (Table 1).[25–32] O_3 is a secondary pollutant formed through photochemical reactions between volatile organic compounds (VOCs) and nitrogen oxides (NO_x) in the presence of high temperatures and sunlight.[33,34] Several factors may have contributed to the counterintuitive trend of increased O_3 levels. At the city scale, the O_3 production rate is strongly linked to the VOC/NO_x ratio.[35] Generally, urban areas are characterized by a low VOC/NO_x ratio due to high NO_x concentrations driven by emissions from vehicles.[36] However, due to the decrease in NO_x emissions from its main source (transport), it is plausible that the O_3 generation was enhanced because of a high VOC/NO_x ratio and its reduced loss through chemical titration with NO.[37] This decrease in the observed PM2.5 levels may have led to an increase in the incoming solar radiation, and thus enhanced photochemical production of O_3.[38,39] Additionally, the lower PM2.5 levels present a less efficient sink for hydroperoxyl radicals ($\cdot HO_2$), enhancing the O_3 generation *via* the peroxy radical pathway.[40]

The time change of the pollutant concentration is significant, and the difference is large: SO_2, NO_2, PM10, PM2.5 and CO-95per change monthly and present a "unimodal" trend: the summer concentration is low and the winter concentration is high, as coal heating in the winter causes the SO_2, NO_2, PM10, PM2.5 and CO-95per emissions to increase.[41,42] At the same time, the winter in Shandong Province is cold and dry, and the relatively stable atmospheric stratification allows the inversion layer to easily appear near the surface, which promotes the accumulation of air pollutants near the surface.[43,44] Moreover, industrial and fossil fuel sources could play significant roles in particulate matter and gaseous pollutant accumulation.[45,46] However, the concentration change of O_{3-8h} is opposite to that

Table 1 List of different studies around the world that claimed improved air quality during COVID-19 lockdowns

Study	Location	Period	Findings/remarks
Aman *et al.*, 2020	Ahmedabad, India	2015–2020	Decrease in suspended particulate matter
Mihăilă *et al.*, 2023	NE Romania	March 16 to May 14, 2020	NO_2 and SO_2 decreased for the entire region by 8.6 and 14.3%
Blackman *et al.*, 2023	Bogotá, Columbia	March 2020 to June 2021	Cut PM2.5 pollution by 15% and NO_2 pollution by 21%
Kumari *et al.*, 2022	Dublin, Ireland	March 27 to June 5, 2020	27.7% improvement in AQI and 28% reduction in average NO_2 level
Agami *et al.*, 2021	Israel	March 8 to May 2, 2020	The biggest reduction was observed in NO_x, which was 41%
Polednik *et al.*, 2021	Lublin, Poland	April 16–17, 2020 compared to April 4–5, 2017	Decrease of the average PM2.5 and PM10 concentrations by ~30% and ~14%, respectively
Rahman *et al.*, 2020	Dhaka city, Bangladesh	March 8 to May 15, 2020	26, 20.4, 17.5, 9.7 and 8.8% declined in PM 2.5, NO_2, SO_2, O_3, and CO concentrations, respectively
Chen *et al.*, 2020	China (367 cities)	January 5 to January 20, 2020	NO_2: decreased 12.9 μg m^{-3}, PM2.5: decreased 18.9 μg m^{-3}

of SO_2 and other pollutants, with high concentrations in summer and low concentrations in winter. The summer temperatures are high and the sunlight is strong, which are conducive to the formation of O_3 in the atmosphere.[47,48] Shandong Province at higher latitudes has short days in winter, which leads to a short illumination time and weak light intensity, thus greatly reducing the formation of O_3.[49]

In terms of the spatial scale, the air quality gradually improved from inland areas to coastal areas (as shown in Table S4† and Fig. 4). Taking 2019 as an example, the city with the highest concentration of SO_2 was Zibo (20 μg m^{-3}), followed by Binzhou (19 μg m^{-3}), Zaozhuang and Jining (17 μg m^{-3}), all exceeding the primary concentration limit, while Weihai had the lowest concentration (6 μg m^{-3}). The city with the highest concentration of NO_2 was Zibo (42 μg m^{-3}), followed by Jinan (41 μg m^{-3}) and Binzhou (39 μg m^{-3}), all exceeding the secondary concentration limit. The city with the lowest concentration was Weihai (20 μg m^{-3}). The city with the highest concentration of PM10 was Zaozhuang (113 μg m^{-3}), followed by Heze (112 μg m^{-3}) and Liaocheng (111 μg m^{-3}), both exceeding the secondary concentration limit. The city with the lowest concentration was Weihai (56 μg m^{-3}), but it still exceeded the primary concentration limit. The city with the highest PM2.5 concentration was Zaozhuang (59 μg m^{-3}), followed by Liaocheng (58 μg m^{-3}), Linyi and Heze (57 μg m^{-3}), all exceeding the secondary concentration limit. The city with the lowest PM2.5 concentration was Weihai (29 μg m^{-3}), but it still exceeded the primary concentration limit. The cities with the highest concentrations of O_{3-8h} are Zibo, Liaocheng and Binzhou (204 μg m^{-3}), all exceeding the secondary concentration limit. The city with the lowest concentration is Qingdao (147 μg m^{-3}), but it still exceeds the primary concentration limit. The highest concentration of CO-95per was in Zibo (1.9 mg m^{-3}), followed by Weifang, Liaocheng and Binzhou (1.7 mg m^{-3}), and the lowest concentration was in Weihai (1.1 mg m^{-3}). All of these cities were within the primary concentration limit. Overall, the air quality in the coastal areas of the peninsula was good, with 83.3% of the days in Weihai, 80.3% of the days in Yantai and 78.9% of the days in Qingdao having good air quality in 2019 and only 0.5%, 2.5% and 1.9% of the days having heavy pollution, respectively. The air quality in inland areas was poor: Zibo, Liaocheng and Zaozhuang had only 51.0% days with good air quality in 2019, 49.9% and 51.0% days with good air quality, respectively, while the number of heavily polluted days was 4.7%, 5.5% and 4.7%. According to Yao's statistics on the distribution of particulate matter and gaseous pollutants in 2015, the annual mean concentrations of PM10 and PM2.5 showed a downward trend from west to the east.[44] The concentrations of SO_2 and NO_2 in central China were higher than those in other regions, while the annual mean concentration of O_3 in coastal areas was slightly lower than that in inland areas. In summary, the main polluted areas were in the middle and western parts of Shandong. Coastal areas have better air quality than inland areas. The peninsula area is close to the Yellow Sea and Bohai Sea, and is subject to the influence of sea wind, sea waves, and extratropical cyclones year-round. Its high relative humidity is conducive to the diffusion and settlement of fine particles.[50] There is frequent and persistent air convection disturbance between land and sea. There are few heavy polluting industries, such as metal smelting and fuel processing, in coastal areas, while the central and northwestern areas are important industrial centers in Shandong Province. The landform of Shandong Province is undulating and hilly, which is not conducive to the diffusion and transfer of pollutants.

3.2 Effect of particulate matter emission on air quality

Fig. S1† shows the industrial sources, domestic sources and total amount of particulate matter emissions from 16 cities in Shandong Province in 2019–2020. Table S5† shows the correlation analysis results of the pollutant index (annual mean concentration of SO_2, NO_2, PM10, PM2.5, O_{3-8h} and CO-95per, PDHP and PDWP) and particulate matter emission index (total particulate matter emission, particulate matter industrial source emission, particulate matter domestic source emission) in 2020. The correlation between the pollutant index and particulate matter emission index is not strong. The main anthropogenic sources of coarse particulate matter are road dust emissions and soil tilling-induced dust emissions in

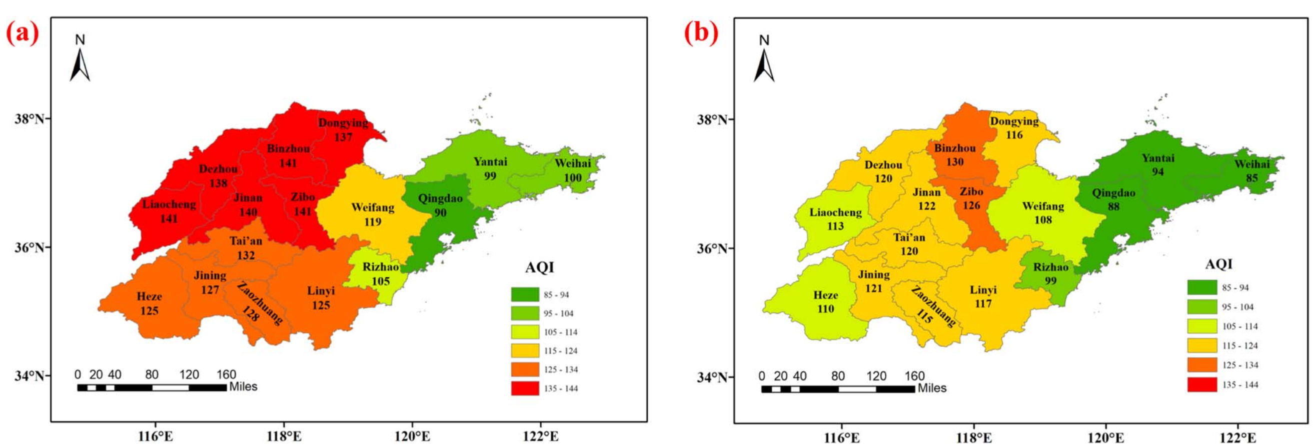

Fig. 4 AQI of 16 cities in Shandong Province in 2019 (a) and 2020 (b).

China.[51] Studies have proven that air is fluid, and air quality is not only affected by local pollution sources, but also related to air transport in surrounding areas and even at medium and long distances.[52] At the same time, the chemical composition and hygroscopic properties of atmospheric fine particles determine their environmental effects and surface heterogeneous reactions; that is, if atmospheric particles stay in the air for an extended period of time, they easily react with other substances to form secondary pollutants.[53] W. S. Chow *et al.* investigated the 10 year trend of PM2.5 components and source tracers in an urban station in Hong Kong, China, from 2008 to 2017, and found that the decreasing trend of gaseous pollutants (SO_2 and NO_x) and particulate pollutants (sulfate and nitrate) generated by secondary transformation did not show a direct proportional correlation. This situation reflects the complex nonlinear chemical relationship between the precursor and product.[54] However, in the heating period, there was a significant positive association between NO_2 and the industrial sources/total amount of particulate matter, and there was a significant negative association between PDWP and the industrial sources/total amount of particulate matter, as shown in Table S6.† The results indicated that the heating period and NO_2 pollutant played key roles in the improvement of the air quality.

3.3 Effect of the meteorological factor on air quality

In terms of the meteorological conditions, the temperature and precipitation are selected as research topics in this paper. The change trends of temperature and precipitation in 16 cities in Shandong Province during 2019–2020 are shown in Fig. S2.† Shandong Province has a warm temperate monsoon climate. The winter is controlled by continental high pressure, and is cold and dry. Meanwhile, summer is affected by the southeast monsoon, which is hot and rainy. The temperature difference in the 16 cities is small. December or January has the lowest temperature, reaching −1.5–1.5 °C, and July or August has the highest temperature, reaching 25–29 °C. The precipitation of 16 cities presents the characteristics of "more summer and less winter", with significant seasonal changes and significant precipitation differences among cities. In 2020, the annual precipitation of Rizhao reached 1232.9 mm, while that of Binzhou was only 625.8 mm.

Correlation analysis of the monthly pollutant index changes and monthly temperature changes in Shandong Province from 2019 to 2020 was conducted. As shown in Table 2, there was a significant negative correlation between the air temperature and the concentrations of SO_2, NO_2, PM10, PM2.5 and CO-95per and the proportion of heavy pollution days ($P < 0.01$). Furthermore, there was a significant positive correlation between the air temperature and O_{3-8h} ($P < 0.01$). These results indicated that the concentrations of SO_2, NO_2, PM10, PM2.5 and CO-95per and the proportion of heavily polluted days were higher, and the concentration of O_{3-8h} was lower when the temperature was low. The air quality in summer is better than the air quality in winter. In winter, due to the influence of thermal conditions, most atmospheric junctions exist in a stable structure, resulting in temperature inversions. The stronger the inversion is, the weaker the vertical convective movement of the atmosphere and the worse the diffusion conditions of air pollution.[55,56] Summer can increase the degree of atmospheric instability, which is conducive to the diffusion of gaseous pollutants. In cold weather in the winter, urban residents burn coal for heating, which further aggravates air pollution.[42] O_3 has a strong correlation with air temperature, which is an important factor affecting the photochemical reaction rate. With the increase in solar ultraviolet radiation, the air temperature gradually rises, promoting the photolysis reaction of NO_2 and accelerating the generation rate of O_3, leading to an increase in the mean concentration of O_{3-8h}. In summary, the elevated temperatures enhanced the formation of O_3, as previously reported.[47,48]

Correlation analysis of the monthly pollutant index changes and monthly precipitation changes in Shandong Province from 2019 to 2020 was conducted. Table 2 shows that the monthly precipitation is negatively correlated with the concentrations of SO_2, NO_2, PM10, PM2.5 and CO-95per and the proportion of heavily polluted days, and the correlation is significant in 2019 ($P < 0.05$). This result indicates that the higher the monthly precipitation is, the lower the concentrations of SO_2, NO_2, PM10, PM2.5 and CO-95per and the better the air quality condition. Atmospheric pollutants are removed from the atmosphere mainly through dry deposition and wet deposition.

Table 2 Correlation analysis between the pollutant index and meteorological factor index in 2019 and 2020[a]

	Temperature in 2019		Temperature in 2020		Precipitation in 2019		Precipitation in 2020	
Pollution index	Correlation	*P* value	Correlation	*P* value	Correlation	*P* value	Correlation	*P* value
Annual mean concentration of SO_2	−0.928**	≤0.001	−0.799**	0.002	−0.882**	≤0.001	−0.686*	0.014
Annual mean concentration of NO_2	−0.874**	≤0.001	−0.662*	0.019	−0.636*	0.026	−0.673*	0.017
Annual mean concentration of PM10	−0.953**	≤0.001	−0.853**	≤0.001	−0.809**	0.001	−0.559	0.059
Annual mean *l* concentration of PM2.5	−0.961**	≤0.001	−0.930**	≤0.001	−0.807**	0.002	−0.476	0.118
Annual mean concentration of O_{3-8h}	0.937**	≤0.001	0.909**	≤0.001	0.545	0.067	0.538	0.071
Annual mean concentration of CO-95per	−0.920**	≤0.001	−0.811**	0.001	−0.625*	0.03	−0.453	0.139
PDHP	0.014	0.966	0.168	0.602	0.35	0.265	−0.385	0.217
PDWP	−0.920**	≤0.001	−0.499	0.099	−0.638*	0.026	−0.078	0.81

[a] $P < 0.05$; $**P < 0.01$.

The flushing effect of precipitation on pollutants and the sedimentation and diffusion effect of strong convective movement accompanied by precipitation on pollutants make annual precipitation an important factor in restraining air pollution.[10] PM2.5 and PM10 are the main atmospheric pollutants in Shandong Province, exceeding the second-level concentration limit, but the improvement is obvious in summer, mainly because of the precipitation effect on the removal of atmospheric particles.[48,57] However, the correlation coefficient shows that PM10 ($P = 0.118$, 2020) has a more significant correlation with precipitation than PM2.5 ($P = 0.059$, 2020), indicating that raindrops have a stronger scouring effect on large particulate matter particles and a weaker scouring effect on small particulate matter.[58]

3.4 Effect of socioeconomic factor on air quality

The level of regional economic development may have an impact on its air quality, and the results are shown in Tables S7, S8, 3, S3 and S4.†

Fig. S4† shows the proportion of secondary industry, population density and green coverage rate of 16 cities in Shandong Province during 2019–2020. In terms of industrial structure, the proportion of primary, secondary and tertiary industries in 16 cities in Shandong Province is relatively average and reasonable, among which the proportion of secondary industry in most cities is concentrated at 35–45%. However, in 2020, the proportions of secondary industry in Dongying and Zibo were 56.3% and 48.4%, respectively, far exceeding the provincial mean. In terms of the population density, there is a significant difference among the 16 cities in Shandong Province. In 2020, the population densities of Jinan, Qingdao and Dongying will reach 902.1 people per square kilometer, 894.9 people per square kilometer and 266.1 people per square kilometer, respectively. The green coverage rate of these 16 cities in Shandong Province in 2019–2020 was concentrated at 40–46%, with no significant difference among the cities.

Correlation analysis was conducted between pollutant indicators of 16 cities in Shandong Province and the proportion of secondary industry/population density/green coverage rate during 2019–2020. As shown in Table S7,† there was no significant correlation between the pollutant indicators of Shandong Province and socioeconomic factors, such as the proportion of secondary industry/population density/green coverage rate. However, there was a significant positive correlation between SO_2 and the proportion of the secondary industry, between PM10 and the population density, and between PM2.5 and the population density, respectively. The results indicated that the proportion of secondary industry played a key role in the gaseous pollutant (SO_2), but the population density played a key role in the particulate matter pollutant (PM10 and PM2.5). Using cross-sectional data of 282 cities and the BMA (Bayesian Model Average) method population density, Boqiang Lin found that the proportion of the secondary industry, the possession of civil motor vehicles and the annual mean temperature have a PIP (posterior inclusion probability) greater than 0.5 for each pollutant, which indicated that the population density, possession of civil motor vehicles, the proportion of secondary industry, and the annual mean temperature are the main influencing factors of air pollution.[59] Bai *et al.*, 2019 used the GWR method to discuss the spatial differences of different influencing factors on AQI in the Yangtze River Economic Belt, and concluded that there was a significant negative correlation between green coverage and AQI, indicating that the improvement of the urban green coverage had a significant effect on the improvement of AQI.[60] There is a significant positive correlation between the population density and AQI; that is, cities with high population density have a greater intensity of human activities, consume more resources, and release a larger number of pollutants.[61]

3.5 Correlation analysis between different pollution indices

Correlation analysis was conducted on the pollutant concentration, PDHP and PDWP in three stages (nonheating period, heating period and annual mean) in Shandong Province in 2019 and 2020. The results are shown in Fig. 5.

Fig. 5a and b shows the correlation analysis of the annual mean concentration of pollutants in Shandong Province from 2019 to 2020. It was found that gaseous pollutants are more susceptible to the interaction among pollutants than fine particles. O_{3-8h} is significantly positively correlated with NO_2,

Table 3 Correlation analysis between the pollutant index and social situation from 2019 to 2020[a]

Pollution index	Proportion of secondary industry		Population density		Green coverage rate	
	Correlation	P value	Correlation	P value	Correlation	P value
Annual mean concentration of SO_2	0.437	0.014**	0.157	0.399	0.173	0.351
Annual mean concentration of NO_2	0.173	0.353	0.159	0.394	0.105	0.573
Annual mean concentration of PM10	0.125	0.504	0.307	0.092*	−0.134	0.474
Annual mean concentration of PM2.5	0.088	0.638	0.376	0.037**	−0.098	0.600
Annual mean concentration of O_{3-8h}	0.290	0.113	0.078	0.677	0.171	0.357
Annual mean concentration of CO-95per	0.042	0.821	0.079	0.674	0.132	0.480
PDHP	−0.136	0.465	−0.217	0.240	0.028	0.882
PDWP	−0.258	0.161	−0.212	0.252	0.112	0.550

[a] *$P < 0.05$; **$P < 0.01$.

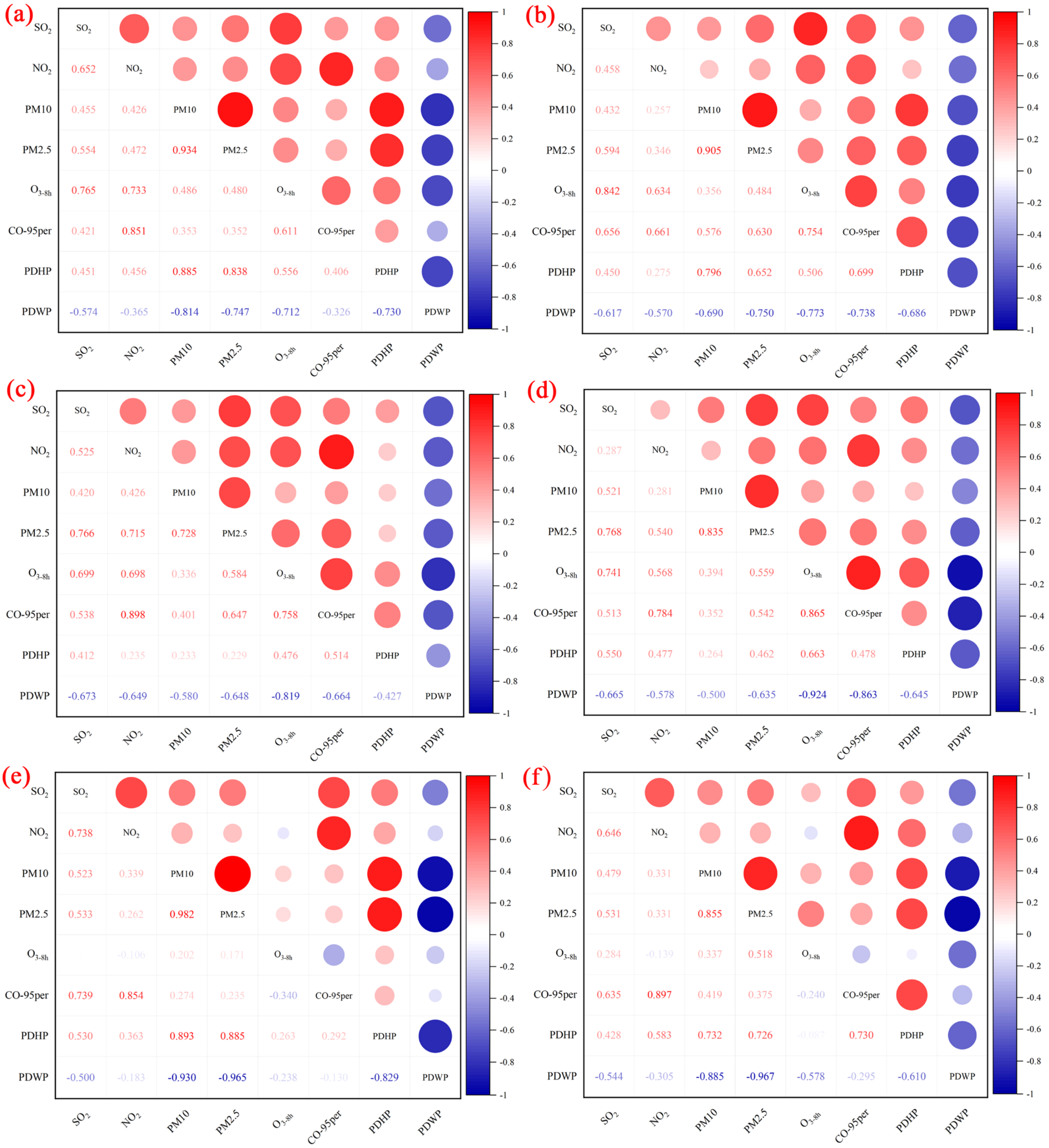

Fig. 5 The correlation analysis between different pollution indices: annual mean value of 2019 (a) and 2020 (b); annual mean value in the nonheating period of 2019 (c) and 2020 (d); annual mean value in the heating period of 2019 (e) and 2020 (f).

SO_2 and CO-95per in 2019 and 2020 ($P < 0.05$). However, PM10 only has a significant positive correlation with PM2.5 in 2019, and a significant positive correlation with PM2.5 and CO in 2020. Except for NO_2, most pollutants show a significant negative correlation with PDWP ($P < 0.05$), indicating that the main pollutants inhibiting good weather are SO_2, NO_2, PM10, PM2.5 and O_{3-8h}. In 2019 and 2020, the proportion of heavy pollution weather and the concentrations of PM10, PM2.5 and O_3 all passed the significance test ($P < 0.05$), showing a positive correlation. Therefore, heavy pollution weather is mainly affected by PM2.5, PM10 and O_3. The influence of fine particulate matter PM2.5 and inhalable particulate matter PM10 is more significant ($P < 0.01$), and the concentration of CO-95per shows a strong positive correlation with heavy pollution weather in 2020 ($P < 0.01$).

The results in Fig. 5c–f show that there are significant differences in the correlation between various pollutants in the heating period and nonheating period. The correlation of most pollutants is stronger in the nonheating period. PM2.5/O_3 and PM2.5/NO_2 have no correlation in the heating period, but have a significant positive correlation in the nonheating period. The main pollutants of heavy pollution weather are different in heating and nonheating seasons. The main sources of heavy pollution weather in the heating season are PM10, PM2.5 and CO-95per ($P < 0.01$, 2020), while the concentration of pollutants in the nonheating season has little correlation with the proportion of heavy pollution weather. Only CO-95per is significant ($P < 0.05$). There is a significant correlation between the PDWP and the six pollutants. As a secondary pollutant, O_{3-8h} is closely related to the emissions of other pollutants. In the nonheating season with severe O_{3-8h} pollution, O_{3-8h} has a significant positive correlation with CO-95per and NO_2 ($P < 0.05$), which may be related to the formation of O_3 by atmospheric photochemical reactions. O_3 formation is caused by the photochemical reaction of CO, NO_x, and volatile organic compounds (VOCs) under ultraviolet irradiation.[62]

SO_2 is positively correlated with CO-95per and NO_2 in both the heating and nonheating seasons. Primary gaseous pollutants (SO_2, NO_2, and CO) are widely used as tracers of emissions from anthropogenic sources.[63] However, the correlation between SO_2 and CO-95per and NO_2 increases after the nonheating period enters the heating period, and the correlation coefficient between SO_2 and CO-95per increases from 0.513 ($P < 0.05$) in the nonheating period to 0.635 ($P < 0.01$) in the heating period in 2020. The correlation coefficient between SO_2 and NO_2 increases from 0.287 in the nonheating season to 0.646 in the heating season ($P < 0.01$), indicating that the emissions of the two are synergistic, which may be due to the gradual increase in pollution contribution from coal burning and industrial production in winter. PM2.5, PM10, NO_2 and SO_2 show positive correlations to different degrees in heating and nonheating periods, indicating that these four pollutants have strong homology and synergy. The positive correlations observed between PM (PM2.5 and PM10) and NO_2 and SO_2 could be because the main components (nitrate and sulfate) of PM are primarily formed from NO_x and SO_2 (the gaseous precursors). The positive correlation between PM10 and PM2.5 in the heating period and nonheating period is extremely significant ($P < 0.01$), and the positive correlation between PM10 and PM2.5 in the heating period in 2019 is the highest, with the correlation coefficient reaching 0.982. Many recent studies have found that PM10 and PM2.5 are mainly derived from local high emissions from coal-fired power plants, iron and steel enterprises, cement industries, vehicles, and diesel and gasoline engines. PM2.5 is a component of PM10, so they have similar source and pollution characteristics.[64–66] This result is consistent with the findings of Sulaymon *et al.*, 2021.[66] In the nonheating period, O_{3-8h} has a high correlation with SO_2, and the correlation coefficients in 2019 and 2020 reach 0.699 and 0.741, respectively ($P < 0.01$), mainly because SO_3 is generated after SO_2 meets O_3, so the concentration of SO_2 has a high positive correlation with the concentration of O_{3-8h}.[67]

There is a significant correlation between the SO_2 and PM2.5 concentrations, NO_2 and CO-95per concentrations, PM10 and PM2.5 concentrations in the annual mean, heating period and nonheating period, while there is a significant correlation between O_{3-8h} and NO_2 in the nonheating period and annual mean. Therefore, it is meaningful to clarify the regression equation for different pollutants in the annual mean, heating period and nonheating period in order to understand the relationship between the pollutants and predict the development trend of the pollutant concentration. The fitting equations are as follows:

Annual mean: $\rho(SO_2) = 0.343\rho(PM2.5) - 3.448$

Heating period: $\rho(SO_2) = 0.278\rho(PM2.5) - 3.441$

Nonheating period: $\rho(SO_2) = 0.501\rho(PM2.5) - 5.566$

Annual mean: $\rho(NO_2) = 22.846\rho(\text{CO-95per}) - 1.385$

Heating period: $\rho(NO_2) = 23.263\rho(\text{CO-95per}) + 2.523$

Nonheating period: $\rho(NO_2) = 20.508\rho(\text{CO-95per}) + 7.717$

Annual mean: $\rho(PM10) = 1.733\rho(PM2.5) + 4.348$

Heating period: $\rho(PM10) = 1.061\rho(PM2.5) + 60.356$

Nonheating period: $\rho(PM10) = 1.929\rho(PM2.5) + 5.877$

Annual mean: $\rho(O_{3-8h}) = 2.410\rho(NO_2) + 99.717$

Nonheating period: $\rho(O_{3-8h}) = 2.037\rho(NO_2) + 124.991$

Fig. S5† shows the heating area from 16 cities in Shandong Province in 2019 and 2020. The heating area in different years and cities varies greatly. On the one hand, Qingdao and Jinan

Table 4 Correlation analysis between the pollutant index and heating area from 2019 to 2020[a]

	Heating area in 2019		Heating area in 2020		Heating area in 2019–2020	
Pollution index	Correlation	*P* value	Correlation	*P* value	Correlation	*P* value
Annual mean concentration of SO_2	−0.057	0.840	−0.204	0.467	−0.19	0.307
Annual mean concentration of NO_2	−0.122	0.666	−0.05	0.859	−0.13	0.487
Annual mean concentration of PM10	−0.443	0.098*	−0.543	0.037**	−0.448	0.011**
Annual mean concentration of PM2.5	−0.432	0.108	−0.629	0.012**	−0.512	0.003***
Annual mean concentration of O_{3-8h}	−0.436	0.104	−0.4	0.139	−0.412	0.021**
Annual mean concentration of CO-95per	0.12	0.669	0.063	0.825	−0.021	0.909
PDHP	−0.475	0.074*	−0.468	0.078*	−0.477	0.007***
PDWP	0.543	0.037**	0.632	0.011**	0.556	0.001***

[a] *$P < 0.05$; **$P < 0.01$; ***$P < 0.001$.

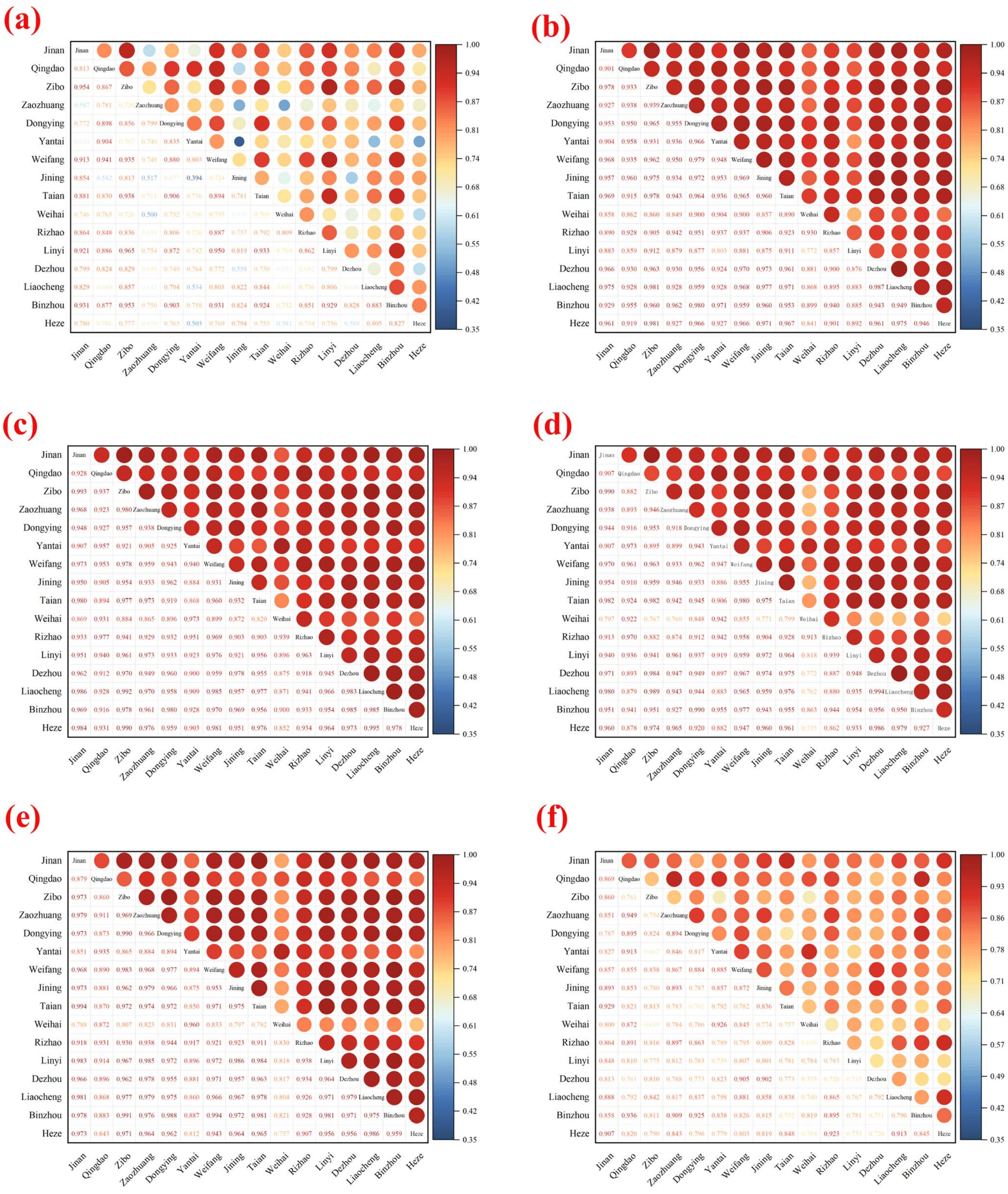

Fig. 6 The correlation analysis of SO_2 (a), NO_2 (b), PM10 (c), PM2.5 (d), O_{3-8h} (e) and CO-95per (f) concentrations in 16 cities in Shandong Province.

had the largest heating areas (> two hundred million km^2), and Heze had the smallest largest heating area (ten million km^2). On the other hand, the heating areas of fifteen cities were increased; only that of Jining was reduced from one hundred million km^2 to fifty million km^2. As shown in Table 4, there were significant positive correlations between PDWP and the heating area (2019, 2020, mean), significant negative correlations between PM10/PDHP and the heating area (2019, 2020, mean), significant negative correlations between PM2.5 and the heating area (2020, mean), and significant negative correlations between O_{3-8h} and the heating area (mean).

3.6 Correlation analysis between different cities

Correlation analysis was conducted on the mean concentrations of SO_2, NO_2, PM10, PM2.5, O_{3-8h} and CO-95per in 16 cities in Shandong Province during 2019–2020. The results are shown in Fig. 6.

With the continuous development of urbanization, the agglomeration effect between the urban agglomerations is gradually strengthened, and the air quality between the urban agglomerations influences each other.[68] Based on the above analysis results, most pollutants have significant positive correlation ($P < 0.05$) and extremely significant positive correlation ($P < 0.01$) among cities, indicating that the six pollutants have strong correlation and synergy among the cities. The correlation coefficient of each pollutant concentration is closely related to the spatial distance between cities. The smaller the spatial distance between cities, the more beneficial it is to the transmission of most pollutants. In addition to the distance between cities, the correlations are closely related to the types of pollutants. The correlation coefficient of the PM10 and PM2.5 concentrations between cities is generally greater than that of the SO_2 concentration between cities, indicating that the PM10 and PM2.5 concentrations are more susceptible to intercity transmission than the SO_2 concentration.

4 Conclusion

Spatial and temporal differentiation characteristics of SO_2, NO_2, PM10, PM2.5, O_{3-8h} and CO-95per, PDHP and PDWP and its influencing factors were analyzed in 16 cities in Shandong Province in 2019–2020. The intermonthly variation trends of SO_2, NO_2, PM10, PM2.5 and CO-95per were consistent, with low concentrations in summer and high concentrations in winter. The concentration of O_{3-8h} was high in summer and low in winter. Air quality gradually improved from inland areas to coastal areas. Mean monthly temperature/precipitation was negatively correlated with the concentrations of SO_2, NO_2, PM10, PM2.5, CO-95per and the proportion of heavy pollution days, and positively correlated with O_{3-8h}. The annual mean concentrations of PM2.5, PM10, NO_2 and SO_2 were positively correlated, and the positive correlation between PM10 and PM2.5 was extremely significant. There was a significant difference in the correlation between pollutants in the heating period and nonheating period. PM2.5/O_{3-8h} and PM2.5/NO_2 had no correlation in the heating period and a significant positive correlation in the nonheating period. The proportion of heavy pollution days in the heating period was significantly positively correlated with SO_2, NO_2, PM10, PM2.5 and CO-95per. There was a significant positive correlation between the pollutants in 16 cities in Shandong Province, and the correlation was affected by the distance between cities and the types of pollutants. The smaller the spatial distance between cities was, the more beneficial it was for the transmission of most pollutants. PM10 and PM2.5 were more susceptible to intercity transmission than SO_2.

In future research, the first challenge in the improvement of air quality will be the regional effect. The air quality is influenced by the distances between different cities and regions. The second challenge in the improvement of the air quality is the time effect. For the heating period and nonheating period, summer and winter, the key pollutants are different. The key pollutants in winter are SO_2, NO_2, PM10, PM2.5 and CO-95per, but the key pollutant in summer is O_3. The last challenge in the improvement of the air quality is the synergistic effect. It is not effective to reduce one pollutant; the cooperative governance for SO_2, NO_2, PM10, PM2.5, O_3 and CO is the striving direction.

Conflicts of interest

The authors declare no competing financial interest.

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