

Trends in Air Pollutant Concentrations and the Impact of Meteorology in Shandong Province, Coastal China, during 2013-2019

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ABSTRACT

Although weather conditions significantly affect air pollutant concentrations, few quantitative studies have been conducted on the effects of long-term and seasonal changes in meteorology on air quality. Hence, in this study, the trends in Shandong Province, China, for six criteria pollutants (viz., sulfur dioxide [SO₂], carbon monoxide [CO], particulate matter [PM] with an aerodynamic diameter of < 10 μm [PM₁₀], PM with an aerodynamic diameter of < 2.5 μm [PM_{2.5}], nitrogen dioxide [NO₂], and ozone [O₃]) were analyzed for the period of 2013–2019, when overall emissions of air pollutants decreased, and the Weather Research and Forecasting model coupled with Chemistry (WRF/Chem) was applied to evaluate the role of inter-annual and seasonal meteorological changes. Five of the six criteria pollutants exhibited a sharp drop in concentration until 2017 and a gradual decline afterward, with the maximum and minimum annual values occurring during winter and summer, respectively. In contrast, the level of O₃ rose between 2013 and 2019 and displayed the opposite seasonal trend. Also, the diurnal concentrations of the first five criteria pollutants showed a typical bimodal distribution, whereas those of the O₃ showed a typical unimodal distribution. Furthermore, a trimodal distribution was observed for the ratios between the diurnal PM_{2.5} and PM₁₀ concentrations. Using 2013 as the baseline, the inter-annual meteorological changes accounted for only 3.4–18.6% of the decrease in the five criteria pollutants—with little effect on the O₃—between 2015 and 2019, indicating that emission control measures drove the long-term improvement in air quality during these years. However, seasonal meteorological factors, which favored diffusion during summer and winter but accumulation during spring and autumn, played a larger role in the short term for all six species, especially during winter, when they reduced concentrations (excluding those of SO₂ in 2019 and O₃ altogether) by 6.5–31.0%.

Keywords: Air pollution, Seasonal variation, Diurnal variation, WRF/Chem, Meteorological condition

1 INTRODUCTION

The air quality in China has been significantly improved since the implementation of the Action

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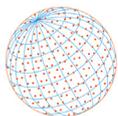
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Plan for Prevention and Control of Air Pollution in September 2013. As one of the most developed and populous provinces of China, the Shandong also issued the Prevention and Control Planning of Air Pollution (abbreviated as “Air Pollution Planning”) to mitigate air pollution in July 2013, which included controlling key industries (such as power plants and the steel industry), fugitive dust pollution (such as construction dust and straw burning), and vehicle emissions (such as strengthening emission standard and promoting the use of clean fuels).

The improvement of air quality benefits from the emission reduction of pollution sources over the past few years (Liu *et al.*, 2015; Li *et al.*, 2019; Wang *et al.*, 2020b). Zheng *et al.* (2018) reported that China’s anthropogenic emissions decreased by 59% for sulfur dioxide (SO₂), 21% for nitrogen oxides (NO_x), 23% for carbon monoxide (CO), 36% for PM₁₀ (particulate matter with aerodynamic diameters less than 10 μm), and 33% for PM_{2.5} (aerodynamic diameters less than 2.5 μm) during 2013–2017. The NO_x emissions over 48 Chinese cities increased by 52% from 2005 to 2011 and decreased by 21% from 2011 to 2015 (Liu *et al.*, 2017). The total emissions of six major atmospheric pollutants (PM_{2.5}, PM₁₀, SO₂, NO_x, volatile organic compounds (VOCs), and ammonia (NH₃)) in Beijing have also followed a downward trend, decreased by 35% in 2015 compared to 2006 (Xue *et al.*, 2019). Several studies have also focused on analyzing the trends in air pollutant concentrations (Shen *et al.*, 2018; Wang *et al.*, 2019b; Li *et al.*, 2020a) and chemical composition of PM_{2.5} (Lang *et al.*, 2017; Geng *et al.*, 2019; Kong *et al.*, 2020) in megacities and typical urban agglomerations in China. The annual average concentrations of particles and their chemical compositions (such as SO₄²⁻, organic matter, NH₄⁺) have exhibited a significant decreasing trend in 74 key cities and three major urban agglomerations (such as Beijing-Tianjin-Hebei, Pearl River Delta, and the Yangtze River Delta) in China during 2013–2017, while the elemental carbon concentration increased in the Pearl River Delta, Fenhe, and Weihe River Plain (Wang *et al.*, 2019b). Among the three major regions, the largest reduction in PM_{2.5} and its chemical constituents was observed in Beijing-Tianjin-Hebei during 2013–2017 (Geng *et al.*, 2019). Field studies on the trends of air pollutant concentrations in Shandong Province, the coastal region of China, have not yet been conducted.

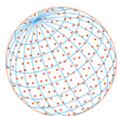
The meteorological conditions also play a critical role in air quality (Zhai *et al.*, 2019; Zhao *et al.*, 2020). For example, severe air pollution events occurred in northern China during January 2020, with the daily average concentration of PM_{2.5} exceeding 200 μg m⁻³, although the pollutant emissions decreased due to the Spring Festival holiday and coronavirus disease outbreak (Wang *et al.*, 2020d; Zhang *et al.*, 2020a). Several studies have explored the contribution of inter-annual meteorological changes to air pollutant concentration reductions (Hong *et al.*, 2019; Vu *et al.*, 2019; Xue *et al.*, 2020; Zhang *et al.*, 2020b). The decrease in the PM_{2.5} concentration due to emissions reduction increased annually from 2013–2017 in key regions of China, especially in 2016 and 2017 (Zhang *et al.*, 2019b). Changes in meteorology have led to an increase in the maximum daily 8-h average O₃ concentration over the central part of central-eastern China, while it decreased over the eastern part of the region in 2003–2015 (Sun *et al.*, 2019). However, the contribution of meteorological conditions may be influenced by the local climate due to the complexity of meteorological changes. Zhang *et al.* (2019b) found that the meteorological factors played more important role in the decrease of PM_{2.5} in winter in the Beijing-Tianjin-Hebei and the Yangtze River Delta. Field studies on the impact of seasonal meteorological conditions on air quality have seldom been reported. Therefore, evaluating the contributions of inter-annual and seasonal meteorological changes to air pollutant concentration reductions should be further studied.

Shandong Province (114.79–122.71°E, 34.38–38.40°N) is located in eastern China, and is surrounded by the Yellow and Bohai Seas on three sides. In this study, the seasonal and diurnal variations of six criteria pollutant (including SO₂, CO, PM₁₀, PM_{2.5}, NO₂, and O₃) concentrations from 2013 to 2019 in Shandong Province were analyzed. The contributions of the meteorological changes to the concentration reductions of six criteria pollutants were also evaluated, especially from the perspective of inter-annual and seasonal. The results are helpful in exploring the efficacy of control measures and policy making.

2 METHODS

2.1 Data Source

The hourly concentrations of six criteria pollutants at 74 monitoring stations covering 16 cities



were obtained from the Department of Ecological Environment of Shandong Province. Note that the concentrations of O₃ and CO were not released from January to September in 2013, excluding Jinan and Qingdao. Additionally, the meteorological conditions (ambient temperature [T], relative humidity [RH], precipitation [Pre], wind speed [WS], and wind direction [WD]) were obtained from the Meteorological Information Comprehensive Analysis and Process System (MICAPS) of the China Meteorological Administration. Fig. S1 illustrates the location of Shandong Province and the monitoring stations.

2.2 WRF/Chem Modeling System

In this study, the Weather Research and Forecasting model coupled with Chemistry (WRF/Chem) was applied to evaluate the contribution of meteorological changes to air pollutant concentration reductions in Shandong Province, which has been widely used for mesoscale simulation and has been proven to be reliable (Chen *et al.*, 2017, 2018; Lv *et al.*, 2020; Wang *et al.*, 2020e; Xing *et al.*, 2020). The initial and lateral meteorological boundary conditions for WRF/Chem were generated using the National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) data, which were available at a $1 \times 1^\circ$ resolution and temporal resolution of 6 h. A two-level nested-grid architecture was employed to implement the WRF/Chem modelling system (Fig. 1). Domain 1 covers most areas of north-eastern China with a grid resolution of 27×27 km, and Domain 2 covers Shandong Province and the surrounding regions with a 9×9 km grid resolution.

The target simulation periods were January, April, July, and October from 2013 to 2019, representing winter, spring, summer, and autumn, respectively. The scenarios were run separately using the meteorological conditions of 2013, 2015, 2017, and 2019, while other configurations (such as the emission sources, physical schemes, and chemical schemes) for all simulations were the same. The 2016 emission inventory used for simulation was obtained and processed from the Multi-resolution Emission Inventory for China (MEIC; <http://www.meicmodel.org/>). Thus, the differences between the scenarios can illustrate the impact of meteorological changes.

2.3 Model Evaluation

To evaluate the modeling performance, the simulated meteorological parameters (e.g., temperature at 2 m [T₂], relative humidity at 2 m [RH₂], wind speed at 10 m [WS₁₀], and wind direction at 10 m [WD₁₀]) and the concentrations of six criteria pollutants from the lowest layer were compared with the observations. The correlative coefficient (R), normalized mean bias (NMB), and normalized mean error (NME) were analyzed according to the United States Environmental Protection Agency (EPA) model evaluation protocol (U.S. EPA, 2007).

In general, the agreement of meteorological parameters between simulated and observed data was good with R, NMB, and NME of T₂ ranging from 0.5 to 0.9, -1.1% to -0.2%, and 10.8% to 41.3%, respectively in different seasons in 2013–2019 (Table 1). The simulated RH₂ were also compared with the observed data with R ranging from 0.5 to 0.8, and the NMB and NME ranging from -17.9% to 34.2% and 11.8–38.8%, respectively. The R, NMB, and NME of WS₁₀ ranged from

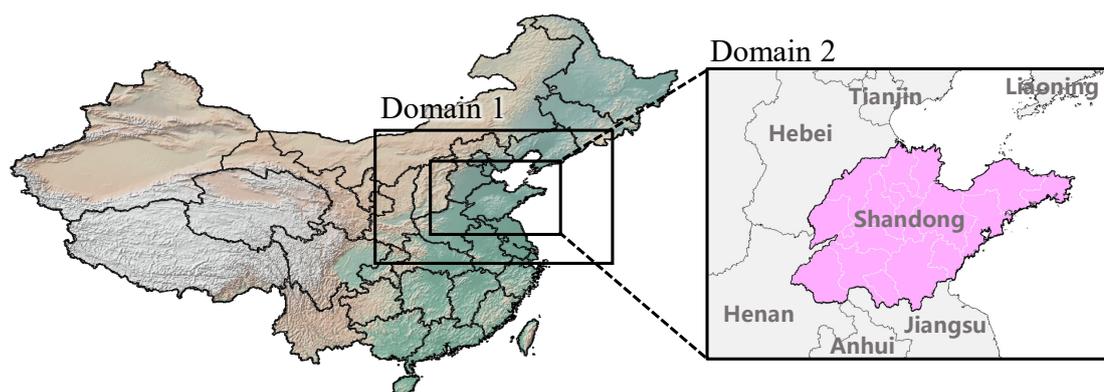


Fig. 1. Design of two-level modeling domains.

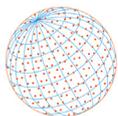


Table 1. Comparisons of meteorological parameters between simulated and observed data.

Year	Season	T ₂ (°C)			RH ₂ (%)			WS ₁₀ (m s ⁻¹)		
		R	NMB (%)	NME (%)	R	NMB (%)	NME (%)	R	NMB (%)	NME (%)
2013	Spring	0.9	-0.4	26.8	0.8	-5.4	38.8	0.7	-27.5	13.8
	Summer	0.8	-0.3	15.4	0.5	-13.7	22.9	0.6	-15.3	35.7
	Autumn	0.9	-0.6	19.6	0.5	8.6	29.3	0.5	-13	22.6
	Winter	0.8	-0.7	40.5	0.7	9.7	35.4	0.6	-10.5	18.9
2015	Spring	0.8	-0.6	24.4	0.8	-11.9	23.6	0.8	-20.8	20.4
	Summer	0.7	-0.5	11.5	0.5	34.2	33.3	0.5	-11.9	30.9
	Autumn	0.8	-0.4	38.5	0.5	24.7	38.1	0.5	-19.5	25.7
	Winter	0.8	-0.5	18.1	0.8	12.9	30.5	0.8	-14.2	16.4
2017	Spring	0.5	-0.6	26.3	0.6	-9.6	37.9	0.5	-22.1	24.1
	Summer	0.7	-0.4	12.8	0.7	8.4	11.8	0.7	-12.4	31.1
	Autumn	0.7	-0.2	22	0.7	-8.8	23.6	0.7	-15.5	14.2
	Winter	0.8	-0.3	41.3	0.7	-9.9	27.5	0.7	-13.3	16.9
2019	Spring	0.9	-0.7	16.5	0.8	-8.6	26.5	0.8	-17.6	23.3
	Summer	0.8	-0.6	10.8	0.7	22.4	17.7	0.7	-14.7	35.7
	Autumn	0.9	-0.8	17.6	0.8	-17.9	22.8	0.8	-16.2	22.6
	Winter	0.8	-1.1	36.6	0.7	-17.3	27	0.8	-9.8	20.4

T₂: temperature at 2 m; RH₂: relative humidity at 2 m; WS₁₀: wind speed at 10 m; R: correlation coefficient; NMB: normalized mean bias; NME: normalized mean errors. The R, NMB, and NME were calculated based on the simulated and observed data. The simulated data were extracted from grids covering 16 cities in Shandong Province in 2013–2019. The observed data at 16 monitoring stations covering 16 cities were obtained from the Meteorological Information Comprehensive Analysis and Process system (MICAPS) of the China Meteorological Administration. The location of the meteorology monitoring stations can be found in Fig. S1.

0.5 to 0.8, -27.5% to -9.8%, and 13.8% to 35.7%, respectively. For WD₁₀, the mean bias (MB) and gross error (GE) were analyzed according to other studies (Hu *et al.*, 2016; Emery *et al.*, 2017; Wang *et al.*, 2019a) and compared with the benchmarks suggested by Emery *et al.* (2001) (Table S1). MB values of WD₁₀ were within the benchmark of $\leq \pm 10$ for 1, 3, 1, and 1 season in 2013, 2015, 2017, and 2019, respectively.

The comparison of observed and simulated criteria pollutant concentrations showed that the model reproduced the variations of concentrations (Table 2). The R, NMB, and NME of air pollutants ranged from 0.5 to 0.7, -18.4% to 15.9%, and 11.5% to 39.7%, respectively. This might be explained by the uncertainties inherent in emission inventories and the unavoidable deficiencies of meteorological and air quality models. By keeping these uncertainties in mind, the modeling performance of the WRF/Chem for simulating temporal and spatial distribution of meteorology and air pollutant concentrations in Shandong Province was reasonably good for the modeled period (Wang *et al.*, 2017a, b; Chen *et al.*, 2018).

3 RESULTS AND DISCUSSION

3.1 Trends in the Air Pollutant Concentrations

Concentrations of five criteria pollutants (exclude O₃) decreased in Shandong Province from 2013–2019 (Fig. 2). The annual average concentrations of SO₂, NO₂, O₃, CO, PM₁₀, and PM_{2.5} in 2013 were $69.6 \pm 26.9 \mu\text{g m}^{-3}$, $48.0 \pm 9.5 \mu\text{g m}^{-3}$, $43.2 \pm 15.4 \mu\text{g m}^{-3}$, $1.7 \pm 0.6 \text{mg m}^{-3}$, $161.4 \pm 43.0 \mu\text{g m}^{-3}$, and $96.5 \pm 27.8 \mu\text{g m}^{-3}$, respectively. The SO₂, NO₂, PM₁₀, and PM_{2.5} concentrations were 1.2–2.8 times higher than the annual secondary guideline value (GB3095-2012). However, no annual standards are proposed for CO and O₃ in China. The secondary guideline values for CO and O₃ in this study are the 24-h and 1-h average concentration standards, and the CO and O₃ concentrations met this standard in 2013. Six years after the implementation of Air Pollution Planning, this drove synergistic co-reductions in the SO₂, NO₂, CO, PM₁₀, and PM_{2.5} concentrations in 2019, with decreases of 80.1%, 26.8%, 54.5%, 39.2%, and 46.2% from the values in 2013, respectively, while the increasing trend of the O₃ concentration has not been sufficiently restricted.

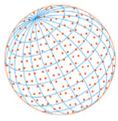


Table 2. Comparisons of air pollutants between simulated and observed data.

Pollutant	Parameter	Spring	Summer	Autumn	Winter
PM ₁₀	R	0.6	0.7	0.7	0.6
	NMB (%)	-6.8	5.9	-11.3	-18.4
	NME (%)	35.4	28.9	15.9	39.7
PM _{2.5}	R	0.7	0.7	0.6	0.6
	NMB (%)	-12.5	6.5	-10.2	-10.6
	NME (%)	20.5	15.8	30.5	38.4
SO ₂	R	0.6	0.7	0.6	0.6
	NMB (%)	-9.8	-14.6	6.8	5.6
	NME (%)	22.9	31.4	35.4	28.6
NO ₂	R	0.6	0.5	0.6	0.6
	NMB (%)	5.1	6.8	-8.4	-2.6
	NME (%)	26.4	20.6	21.4	32.4
O ₃	R	0.5	0.5	0.5	0.6
	NMB (%)	7.9	15.9	12.8	-8.9
	NME (%)	23.6	33.6	30.5	18.4
CO	R	0.7	0.6	0.6	0.6
	NMB (%)	-2.5	5.2	-3.8	6.7
	NME (%)	13.4	18.9	11.5	22.1

The simulated data were extracted from grids covering 16 cities in Shandong Province in 2017. The observed data at 74 monitoring stations covering 16 cities in 2017 were obtained from the Department of Ecological Environment of Shandong Province. The location of the air quality monitoring stations can be found in Fig. S1.

The notable improvements in five criteria pollutants' concentrations reflect the combined effects of various emission-reduction measures that have been implemented over the past few years, such as adjusting the energy structure, industrial restructuring, upgrading emission control technologies, and controlling pollution from key industries. For example, Shandong Province emits the most air pollutants from the Chinese power industry due to its high coal consumption rate and dense distribution of coal-fired plants (Xiong *et al.*, 2016). The CO, PM₁₀, PM_{2.5}, SO₂, and NO_x emissions from power plants in Shandong Province accounted for 10–11% of the total emissions in China (Tong *et al.*, 2018). China implemented an ultra-low-emission standard for coal-fired power plants in 2014 (MEE, 2018), and the installed capacity of ultra-low-emission coal units increased from zero in 2013 to 0.89 billion KW in 2019, accounting for 86% of the installed capacity of coal-fired units (MEE, 2020). This is considered as a significant achievement in air pollution control in China; therefore, the government is applying ultra-low-emission standards in other industries (Wang *et al.*, 2020a). Note that the high air pollutant concentrations in 2013 was also due to the extremely poor ventilation conditions, which might be linked to Arctic Ocean ice loss in the preceding autumn and extensive boreal snowfall in the earlier winter (Zou *et al.*, 2017).

The various SO₂ emission reduction measures were most effective in 2013–2019, with a higher declining rate (~80%) than that of other pollutants (<55%) (Fig. 2). Data from the National Bureau of Statistics showed that the SO₂ emission in Shandong Province decreased by 55% during this period, while the emissions of NO₂ and total suspended particles only decreased by 30% and 21%, respectively (<http://www.stats.gov.cn/tjsj/ndsj/>). Owing to these measures, the number of clean days in Shandong Province exhibited an increasing trend, while the number of polluted days showed a decreasing trend (Fig. 3). Heavily polluted days have been almost eliminated, decreasing from 40 d in 2013 to 8 d in 2017 and 2019. The air pollution improvement objectives set in Air Pollution Planning were achieved in 2019 (DEE, 2018).

Despite the continuous improvement in the air quality of Shandong, the rate of the decrease in the concentrations of PM_{2.5} and PM₁₀ slowed in the late stage of Air Pollution Planning due to an increase in the difficulty of pollution control due to the cost of treatment and development of control technologies. Therefore, technological improvement is vital for pollutant removal and improving air quality. The PM_{2.5} and PM₁₀ concentrations decreased sharply in 2015 and 2017, with reductions ranging from 18.2% to 24.8% from the concentrations in 2013, while the reduction rates decreased to 8.8–9.0% during 2017–2019 (Fig. 2).

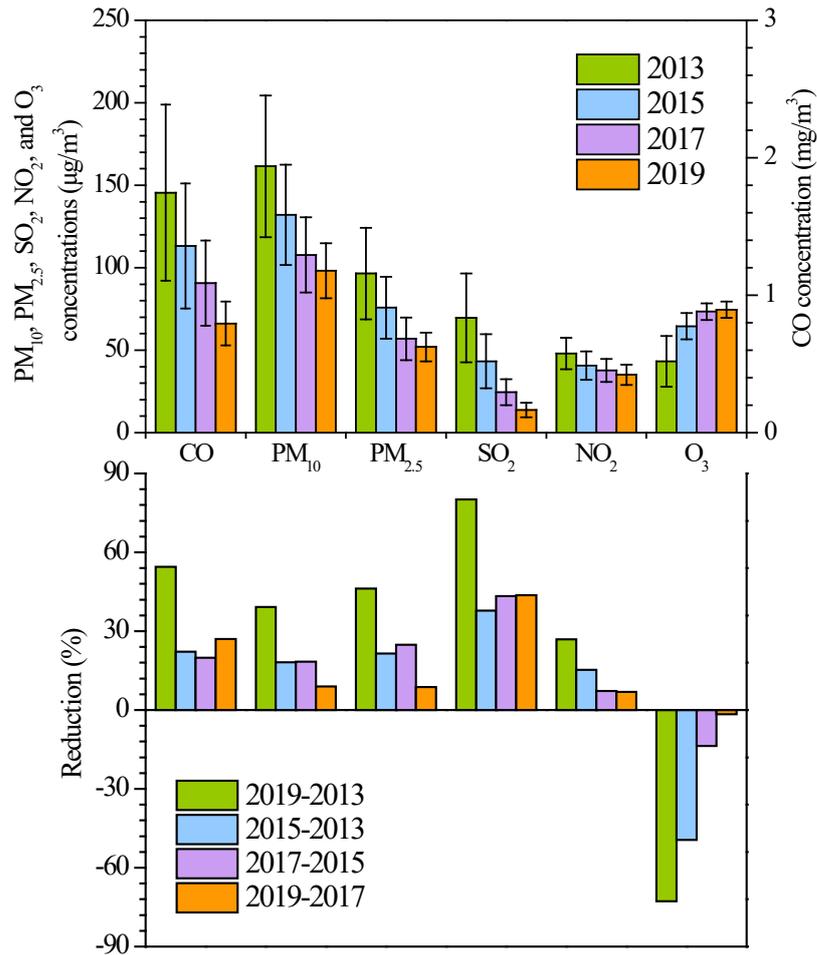
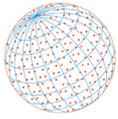


Fig. 2. Annual average concentrations and reductions of air pollutants in Shandong Province in 2013–2019. The concentrations are calculated based on the monitoring stations covering 16 cities of Shandong Province. 2019–2013 refers to lower ratios of air pollutant concentrations in 2019 than with that in 2013, and negative values indicate an increase in the concentrations.

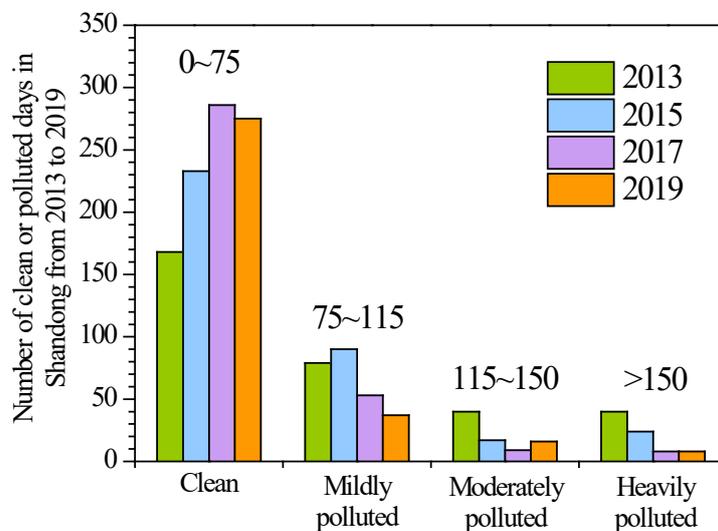
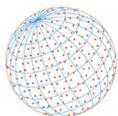


Fig. 3. Number of clean or polluted days in Shandong Province from 2013–2019. Days are divided into four categories based on the daily average PM_{2.5} concentrations covering 16 cities of Shandong Province according to the Technical Regulation on Ambient Air Quality Index (on trial; HJ633-2012). The values above columns are the guideline values for PM_{2.5} concentrations in each category.



However, particle control over Shandong Province still faces a great challenge as the annual average particle concentrations in 2019 still exceeded the annual secondary guideline value (GB3095-2012). The particles were dominated by PM_{2.5}, although the PM_{2.5}/PM₁₀ ratios decreased from 60% in 2013 to 53% in 2019. The decrease in the PM_{2.5}/PM₁₀ ratios (~7%) of Shandong Province was higher than that in other key regions (such as Beijing-Tianjin-Hebei and the Pearl River Delta), with a reduction of 2–3% (Wang *et al.*, 2019b), indicating that the stringent measures implemented for reducing particle emissions in Shandong Province are significant and effective. More measures should be implemented to reduce the emissions of particles and their gaseous precursors to mitigate particle pollution in Shandong Province, as particles can consist of various primary and secondary components (such as organic carbon, elemental carbon, sulfate, nitrate, and ammonium) (Wang *et al.*, 2017a; Zhang *et al.*, 2020b).

The O₃ concentration increased by 72.7% in 2019 compared with that of 2013, although the NO₂ decreased by 26.8% (Fig. 2). O₃ is a major atmospheric oxidant and is formed by the photochemical oxidation of hydrocarbons, CO, and NO_x under solar radiation (Sun *et al.*, 2019). It is more difficult to control O₃ than other pollutants, as it is a secondary pollutant with a non-linear relationship with its precursors (Tie *et al.*, 2013). Studies on the O₃ formation mechanism demonstrated that the sensitivity of O₃ to precursors differs between different regions or under different pollution conditions (Jeon *et al.*, 2014; Qiu, 2017; Fang *et al.*, 2020; Yu *et al.*, 2020). In this study, a negative exponential correlation was observed between the daily average concentrations of NO₂ and O₃ in 2013–2019 (Fig. 4). Control measures aimed at NO_x alone are insufficient to mitigate O₃ pollution, and synergistic control with a desirable NO_x/VOC reduction ratio is required for O₃ reduction, as an inappropriate ratio is most likely responsible to be responsible for the increase in O₃ (Yang *et al.*, 2019; Wang *et al.*, 2020c).

3.2 Seasonal Variation

The five criteria pollutants' concentrations exhibited clear seasonal variation characteristics, with the highest pollution occurring in winter and best air quality occurring in summer during 2013–2019, excluding O₃ (Figs. 5 and S1). The seasonal variations can be attributed to the climate and human activities. A stable atmospheric vertical structure weakens the turbulent atmospheric exchange and hinders the diffusion and dilution of air pollutants in the vertical direction, which is favorable for the accumulation of pollutants in a local area with low wind speed during winter (Zhang *et al.*, 2013; Shen *et al.*, 2018). Chow *et al.* (1993) also found that the elevated particles (e.g., carbonaceous aerosol) could result from increased vegetative burning (i.e., fireplace and woodstove) during winter. Motor-vehicle-related primary contributions to organic carbon and elemental carbon could also be larger owing to cold starts and restricted atmospheric mixing.

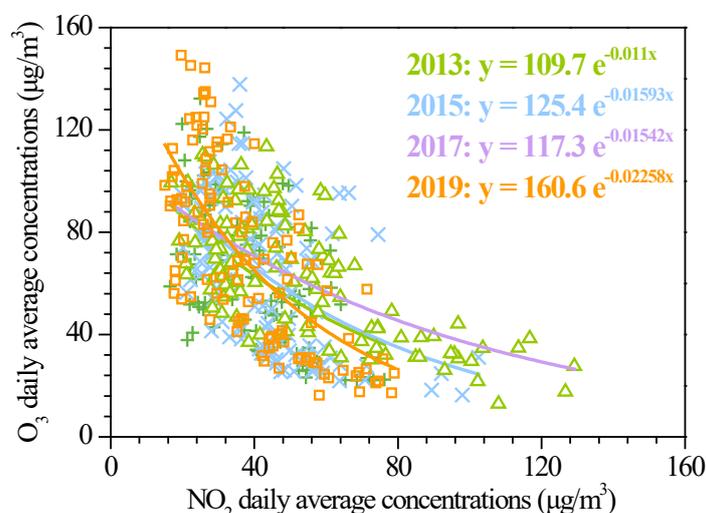


Fig. 4. Correlation between the daily average O₃ and NO₂ concentrations in Shandong Province in 2013–2019. The concentrations used in this figure are from January, April, July, and October in 2013–2019 covering 16 cities.

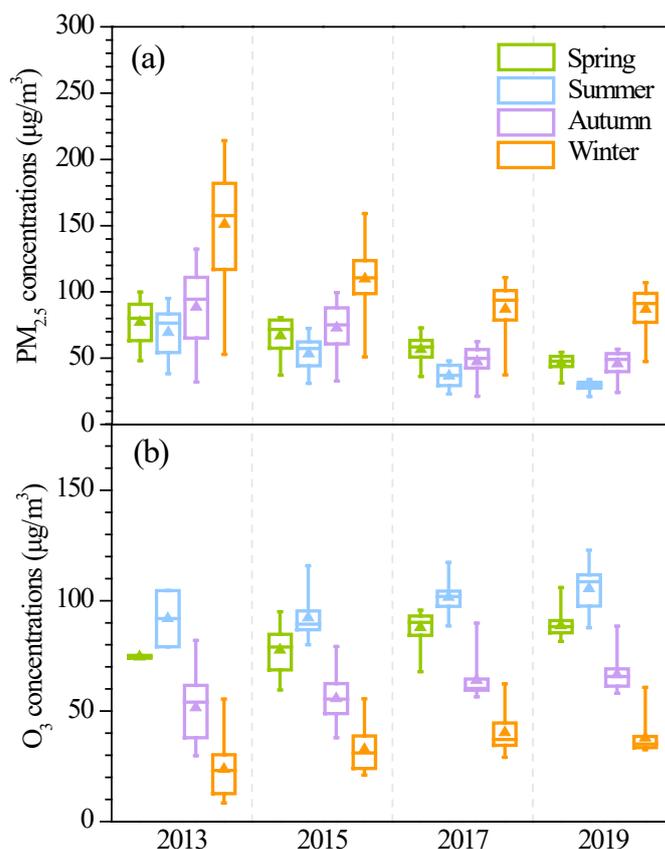
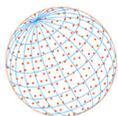
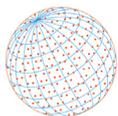


Fig. 5. Seasonal variations in the (a) $\text{PM}_{2.5}$ and (b) O_3 concentrations of Shandong Province during 2013–2019. The variations in the CO , PM_{10} , SO_2 , and NO_2 concentrations are presented in Fig. S2. The concentrations are calculated based on the monitoring stations covering 16 cities of Shandong Province. Spring: March–May; summer: June–August; autumn: September–November; winter: December–February. The box plots refer to the max, 75th, 50th, 25th, and min percentiles of the corresponding dataset, and the triangles indicate the mean values.

The time required for the cold start of motor vehicles also increased due to the low T in winter ($0.5\text{--}2.6^\circ\text{C}$), facilitating the conversion of semi-volatile substances to particles and increasing exhaust emissions (Chow *et al.*, 1993). During monsoon seasons, abundant Pre ranging 1312–4696 mm in Shandong Province during 2013–2019 along with high winds from the sea can alleviate air pollution compared with other seasons ranging 94–856 mm (Wang *et al.*, 2015). Regarding human activities, fire points were observed based on satellites in Shandong Province in winter (<http://satsee.radi.ac.cn:8080/index.html>), indicating the increased combustion (coal and biomass) for heating resulted in the highest air pollutant concentrations in winter (Li *et al.*, 2020b; Zhou *et al.*, 2020). All of these factors resulted in great fluctuations in the five criteria pollutants' concentrations during the four seasons.

The decreases in SO_2 and CO concentrations were more prominent in autumn and winter during 2013–2019, while in summer for $\text{PM}_{2.5}$, PM_{10} , and NO_2 (Table S2). As one of the main sources of SO_2 and CO in autumn and winter, coal consumption has been reduced by over 30 million tons, and seven cities in Shandong Province replaced coal power with gas and electricity over the last few years. Furthermore, fireworks and firecrackers are important contributors to air pollution during the Spring Festival, and the impact is more severe when stable atmospheric conditions occur during winter. Fireworks and firecrackers have been forbidden in some cities to reduce the air pollutant emissions in Shandong Province, leading to a significant decrease in the pollutant concentrations.

The O_3 concentrations in different seasons followed the order of winter < autumn < spring < summer in 2013–2019 (Fig. 5). T and solar radiation are important factors affecting O_3 formation by photochemical reactions (Tang *et al.*, 2009). Taking Jinan as an example, a strong positive



correlation between the T and O₃ concentrations was observed during the summers of 2013–2019 with the correlative coefficient of 0.62 (Fig. 6(a)). The high T during these periods led to strong solar radiation and accelerated the photochemical reaction rate. Furthermore, as a major precursor, the evaporation of VOCs emissions from vegetation and painting increases under higher T (Squires *et al.*, 2020). Sun *et al.* (2019) reported that increased levels of anthropogenic non-methane VOCs are the main cause of the increases in O₃ over the eastern part of central-eastern China. The O₃ concentration exhibited a negative correlation with the RH in the summer with the correlative coefficient of –0.68 (Fig. 6(b)). High RH (such as summer Pre) has a scavenging effect on the precursors of O₃, affects solar radiation, and indirectly inhibits the formation of O₃. Therefore, high T, low RH, and low wind are favorable for the formation of O₃ near the ground.

3.3 Diurnal Variation

The diurnal variations in five criteria pollutants' concentrations exhibited a typical bimodal distribution, except for O₃, with the maximum concentrations occurring from 08:00 to 10:00 and 20:00 to 00:00, and minimum concentrations occurring from 0:00 to 07:00 and 14:00 to 18:00 in 2013–2019 (Figs. 7 and S2), which was generally consistent with those in other megacities and urban agglomerations (Zhou *et al.*, 2015; Shen *et al.*, 2018; Kuerban *et al.*, 2020). This variation was affected by the diurnal emissions and meteorological conditions. There are two traffic rush hours in the morning and evening, and the morning peak of the air pollutant concentrations was closely related to the increased vehicle emissions and road dust during the first traffic rush hour. Air pollutants were accumulated after the second rush hour in the evening under a decreased boundary layer height, which suppressed the diffusion of pollutants (Zhang *et al.*, 2014; Xie *et al.*, 2019). The lower values in the afternoon were attributed to the increased boundary layer height, which was conducive to the diffusion of air pollutants.

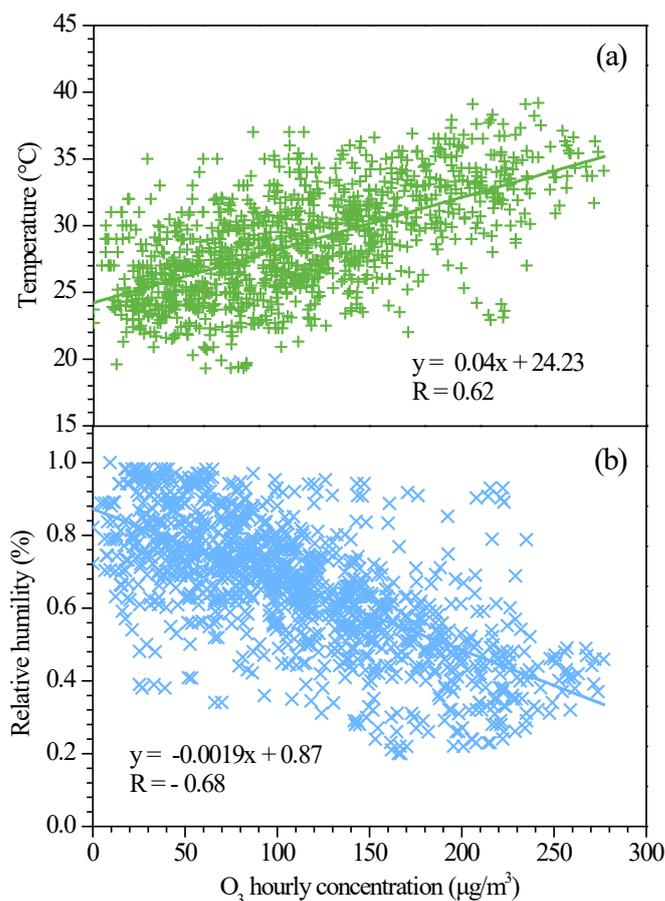


Fig. 6. Correlations between the O₃ concentration and (a) temperature or (b) relative humidity in Jinan during summer 2013–2019.

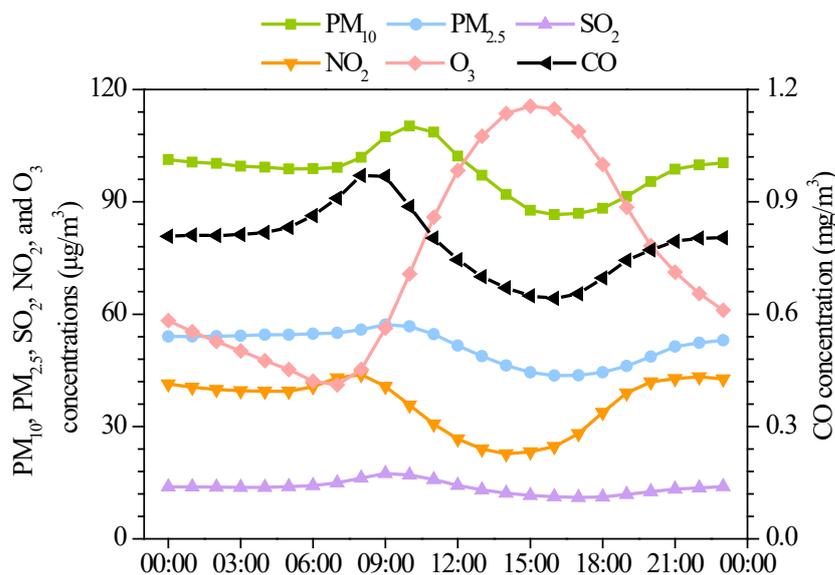
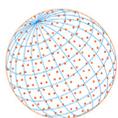


Fig. 7. Diurnal variations in the air pollutant concentrations of Shandong Province during 2019. The variations of the pollutant concentrations in 2013–2017 can be found in Fig. S3. The concentrations are calculated based on the monitoring stations covering 16 cities of Shandong Province.

The diurnal variations in the $PM_{2.5}/PM_{10}$ concentration ratio exhibited a trimodal distribution, with peaks at 00:00, from 06:00 to 07:00, and from 13:00 to 14:00, and minimum values from 01:00 to 02:00, 12:00 to 13:00, and 17:00 to 19:00 during 2013–2019 (Fig. S4). The higher ratios at the first two peaks were consistent with the diurnal variations in the particle concentrations, while the third peak (13:00–14:00) was contrary to the increasing concentration variation. Although the meteorological conditions were conducive to pollutant diffusion during the third peak, atmospheric oxidation was enhanced by the increases in solar radiation and the O_3 concentration (Fig. 7). As the dominant contributor to $PM_{2.5}$, the formation rate of secondary particles is accelerated by the photochemical and catalytic oxidation reactions of gaseous precursors (such as SO_2 , NO_x , NH_3 , and VOCs; Wang *et al.*, 2017a, b), leading to a high $PM_{2.5}/PM_{10}$ ratio.

The diurnal variation of the O_3 concentration exhibited a typical unimodal distribution, with the peak value at 15:00 and minimum value at 07:00. The higher value of the O_3 concentration was mainly due to the photochemical oxidation reactions in the presence of intense solar radiation, which was consistent with the decreases in the contents of precursors, such as NO_2 and CO (Ordonez *et al.*, 2005; Yin *et al.*, 2019).

3.4 Contribution of Meteorological Changes to Air Pollutant Concentration Reductions

Taking 2013 as the baseline, the inter-annual meteorological changes contributed 3.4–18.6% to the concentration reductions of six criteria pollutants in Shandong Province during 2015–2019 (Fig. 8). The concentration reductions due to meteorological improvement were much lower than the observed reductions in 2013–2019 ranging from 26.8–80.1%, indicating that emissions reduction was the dominant factor in the improvement in air quality during this period. The meteorological conditions had little impact on the changes in the O_3 concentrations, indicating that synergistic control of NO_x and VOCs was critical to mitigating O_3 pollution (Fig. 8). Lu *et al.* (2019) also reported that more stringent emission control measures were necessary to reduce surface O_3 pollution over China, particularly in years with unfavorable meteorological conditions. Overall, the contributions of inter-annual meteorological changes to concentration reductions of five criteria pollutants were similar in 2015 and 2017, with values of 7.9–18.6% and 7.6–15.9%, respectively, and higher than that in 2019 (3.4–15.7%). This indicates that the synoptic circumstances in 2015–2017 were similar, and more effort should be devoted to reducing emissions from anthropogenic sources to improve air quality, as the meteorological conditions in 2019 were less conducive to pollutant diffusion than those in 2015–2017. The results also indicated that the

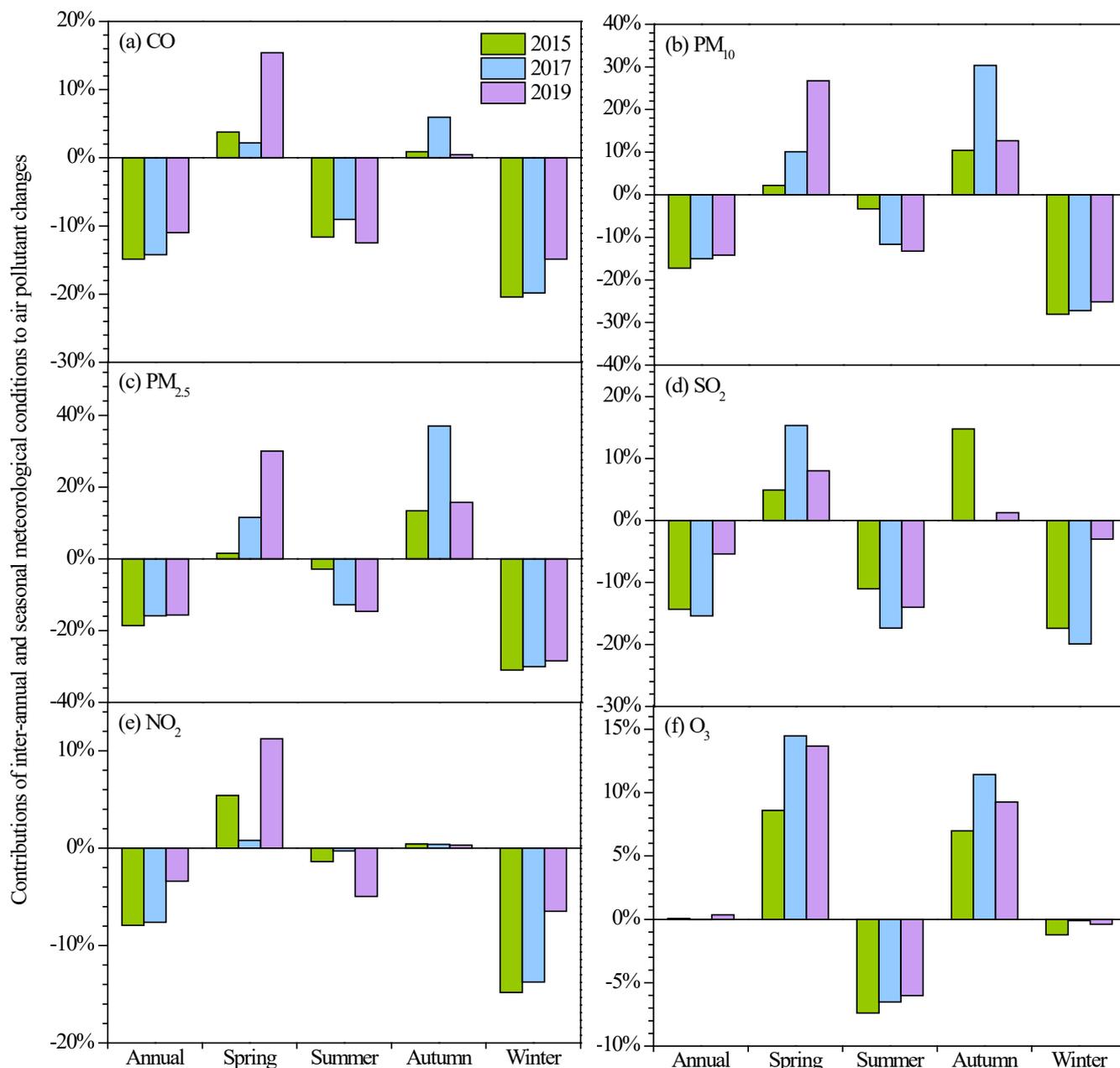
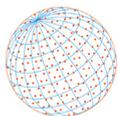


Fig. 8. The contributions of inter-annual and seasonal meteorological changes to air pollutant concentration reductions. This figure presents the relative changes (%) in the annual and seasonal average air pollutant concentrations in 2015–2019, taking 2013 as the baseline, using the WRF/Chem model. Negative values indicate that the meteorological conditions are conducive to the reduction of air pollutant concentrations in 2015–2019.

meteorological conditions in 2015–2019 were more favorable for reducing PM_{2.5}, with a reduction of 15.7–18.6%, followed by PM₁₀ (14.2–17.2%), CO (11.0–14.9%), SO₂ (5.4–15.4%), and NO₂ (3.4–7.9%).

Although the contribution of meteorological conditions to the changes of the six criteria pollutants in Shandong Province has not been reported before, several studies have evaluated this contribution in China and key regions, and their results can be compared to those obtained here. Table 3 summarizes the studies that have evaluated the contributions of inter-annual meteorological changes to PM_{2.5} reduction. Similar to this study, meteorological variations also reduced the PM_{2.5} concentrations in China (~9–12%), Beijing-Tianjin-Hebei (~5–16%), the Yangtze River Delta (~13%), and Beijing (~20%) during different periods, and also played almost equally important roles in reducing air pollution in Beijing during the Asia-Pacific Economic Cooperation

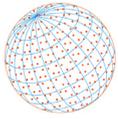


Table 3. Contribution of meteorological changes to PM_{2.5} reductions in other studies.

Study area	Study period	Contribution	Literature
China	2013–2017	9%	Zhang <i>et al.</i> (2019a)
China	2013–2018	12%	Zhai <i>et al.</i> (2019)
BTH	2014–2017	9%	Dong <i>et al.</i> (2020)
BTH	2013–2017	16%	Zhang <i>et al.</i> (2019a)
BTH	2013–2017	5%	Zhang <i>et al.</i> (2019b)
YRD	2013–2017	13%	
Beijing	2013–2017	20%	Chen <i>et al.</i> (2019)
Beijing	APEC 2014	30%	Wang <i>et al.</i> (2017b)
Beijing	Parade 2015	33%	

BTH: Beijing-Tianjin-Hebei; YRD: Yangtze River Delta; APEC: Asia-Pacific Economic Cooperation; Parade: Grand Military Parade.

in 2014 (~30%) and Grand Military Parade in 2015 (~33%). This further demonstrates that the contribution of meteorological changes to air pollutant concentration reductions may be influenced by the geographical location, topography, and local climate, indicating the importance of this study in Shandong Province for developing new policies.

The contributions of meteorological changes to the concentration reductions of the six criteria pollutants exhibited clear seasonal variation characteristics. The meteorological conditions were favorable for air pollutant diffusion in summer and winter, and accumulation in spring and autumn during 2015–2019. Overall, during winter, the contribution of the meteorological changes to the pollutant concentration reductions was larger, with a reduction of 6.5–31.0%, excluding SO₂ in 2019 and O₃. Zhang *et al.* (2019b) also found that the meteorological factors played a more important role in the decrease of PM_{2.5} during winter 2017 in the Beijing-Tianjin-Hebei and Yangtze River Delta regions, as the meteorological conditions improved the reductions in PM_{2.5} by 20% and 30% in the two regions, respectively. Note that the meteorological conditions increased the PM₁₀ and PM_{2.5} concentrations in autumn 2017 by 30.4% and 37.0%, respectively. Therefore, more attention should be paid to avoid heavily polluted days in autumn and winter due to the large volume of anthropogenic emissions.

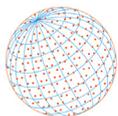
4 CONCLUSIONS

This study analyzed the trends in concentration for six criteria pollutants from 2013 till 2019 in Shandong Province and quantified the effects of inter-annual and seasonal meteorological changes. Whereas the levels of five of the species decreased by 26.8–80.1% over this period, with the maximum and minimum annual values occurring during winter and summer, respectively, that of O₃ increased, with the inverse seasonal pattern. Additionally, we observed typical bimodal distributions for the diurnal concentrations of the first five pollutants but a typical unimodal distribution for the O₃ and a trimodal distribution for the diurnal PM_{2.5}/PM₁₀ concentration ratios.

Further investigation revealed that inter-annual differences in the weather conditions from 2015 till 2019 decreased the levels of the five criteria pollutants by merely 3.4–18.6% (compared to the data from 2013) and produced even less of an effect on the O₃, indicating that emission control measures were primarily responsible for the enhancement in air quality between 2013 and 2019. However, seasonal meteorological factors, which favored diffusion during summer and winter but accumulation during spring and autumn, played a larger role in the short term for all six species. Therefore, to consistently improve the air quality, more effort must be devoted to reducing emissions from anthropogenic sources, especially during spring and autumn.

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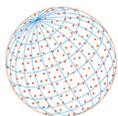


SUPPLEMENTARY MATERIAL

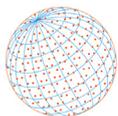
Supplementary data associated with this article can be found in the online version at <https://doi.org/10.4209/aaqr.200545>

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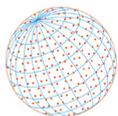
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