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ORIGINAL PAPER

Study of the meteorological influence on ozone in urban areas and their use in assessing ozone trends in all seasons from 2009 to 2015 in Tianjin, China

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Abstract
Ozone pollution in urban areas has increasingly become a topic of intense research in China. Assessing the impact of emission control strategies on O_3 levels is complicated by the disturbance of meteorological factors. This study employs a statistical methodology named generalized additive model (GAM) to characterize the relationship between meteorological factors and ozone levels as well as to meteorologically adjust the ozone trends in Tianjin from 2009 to 2015. The results indicate that the afternoon temperature and the morning solar radiation are the leading meteorological factors controlling O_3 in Tianjin. GAM proves to be an effective tool in predicting ozone levels, because it could capture 40–77% of the variance in the daily ozone maxima for different seasons. Only 3–5 parameters are incorporated into the final model for each season. During summer months, the most important explanatory variables are those influencing the photochemical production. Whereas in winter, ozone destruction by titration with NO is the dominant mechanism affecting the O_3 levels. After adjustment for meteorological effect, general upward trends are evident in ozone levels, which is − 0.34, 0.97, 0.70 and 0.12 ppb year^{-1} in spring, summer, autumn and winter, respectively, indicating that the emission reduction strategies in Tianjin from 2009 to 2015 appear to be more beneficial in reducing the effect of O_3 loss using titration rather than mitigating the photochemical pollution. The clear rising tendency of yearly medians of the meteorologically adjusted ozone (0.24 ppb year^{-1}) strongly suggests formulating a specialized mitigation strategy for regional O_3 pollution.

1 Introduction

Photochemical smog episode—characterized by elevated levels of surface ozone (O_3) has attracted considerable attention owing to its detrimental impact on forests, agricultural crops and human health (Bhatia et al. 2012; Feng et al. 2015; Goodman et al. 2015; Yang et al. 2018). Although worldwide research on urban ozone pollution began in the early 1980s, most early studies focused only on European and North American cities. Long-term (> 10 years) trend analysis of ozone pollution in mainland China has been rarely reported due to the limitation of data. Only a slight amount of systematic research and ozone measurement was conducted in China until the mid 2000s (Wang et al. 2017). Nevertheless, several extreme high ozone episodes have been reported in or around some Chinese cities (Wang et al. 2006; Zhang et al. 2007; Ran et al. 2012).

High ozone pollutions have always been associated with the degradation of human health (WHO 2000). Chen et al. (2017) pointed out that the health impact of ozone has always been a widespread problem in China in the recent years. The actual efficiency of emission control generally depends on several indicators concerning ozone formation regimes, like the ozone production efficiency (OPE), the ratio of VOC/NO_x, as well as ambient meteorological factors. Vestreng et al. (2004) investigated the time series of surface O_3 in Switzerland, and indicated that the emission reduction measures of O_3 precursors during the 1990s were insufficient to effectively reduce the daily maximum O_3 concentrations. It will be of great interest to examine the efficiency of current emission restrictions in China, which mainly aimed at primary pollutants, on the ambient O_3 levels in the future studies (Wang et al. 2017).

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Favorable meteorological conditions, for instance, high temperature, intense sunlight, light wind, always play a critical role in the formation of high O_3 episodes (Mao and Talbot 2004; Ding et al. 2004; Cheng et al. 2015; Jaffe and Zhang 2017). The interannual variation of O_3 concentration may result from the favorable/unfavorable weather conditions, from the emission changes of photochemical precursors, or from a combined effect of these. Hence, it is necessary to distinguish the impact of meteorological variability on O_3 levels in assessing O_3 trends (EPA 2005). Rao et al. (1997) indicated that changes of ozone concentration due to emission control policy may be relatively small hence difficult to detect unless they are isolated from the weather elements. Lou et al. (2015) employed the GEOS-Chem model to study the impact of variations in meteorological factors and anthropogenic activities on the surface ozone levels in China.

The meteorological adjustment of O_3 concentration provides an approach for the purpose of air quality forecasting and examining the underlying effects of control strategies on ozone time trends apart from the natural effects of meteorological factors (Thompson et al. 2001). Several statistical approaches have been adopted for the meteorological adjustment of ozone concentrations which could be broadly classified into regression-based, spatio-temporal modeling, and extreme value methods (Wikle et al. 1998; Ordonez et al. 2005). Such methods have been employed to evaluate the long-term trends in surface O_3 caused by emission control measures in Europe (Ordonez et al. 2005) and North America (Zheng et al. 2007; Camalier et al. 2007), whereas relevant work is quite scarce in China. Most of the relevant studies in China qualitatively described the relationships between ozone and meteorological factors, but the seasonal variation patterns have not been depicted explicitly (Wang et al. 2017). Moreover, the leading meteorological factors in different Chinese cities were still unclear (Tu et al. 2007; Shan et al. 2009). The generalized additive model (GAM) is a regression-based statistical model which could incorporate either linear or nonlinear relationships with continuous and discrete variables. As pointed out by several authors, GAM is an effective tool to deal with the complex linear or nonlinear relationship between air pollutants (e.g., CO, SO_2, $PM_{2.5}$ and O_3) and meteorological variables (Pearce et al. 2011; Cai et al. 2016; Gong et al. 2017). Nevertheless, such a study has rarely been involved with mainland China (Gong et al. 2018).

Tianjin (39°N, 117°E) is located in the eastern part of the Beijing, Tianjin and Hebei (BTH) region in the North China Plain (NCP). The region of BTH is one of the most important city clusters in China, accounting for 7.8% of the national population and 10.9% of the national GDP in 2010 (Zhao et al. 2013b). Tianjin, as the second largest city in the BTH region, covers an area of 11,300 km^2 with a population of 8 million. With the booming economic development, urbanization and population expansion, photochemical pollution has become increasingly prominent in Tianjin, due to the massive emission of ozone precursors. Nonetheless, ozone pollution in BTH outside of Beijing has not yet been investigated sufficiently and considerable gaps and uncertainties lie in the knowledge of properties of regional O_3 pollution and the corresponding mitigation strategies in this region (Tang et al. 2012). Bian et al. (2007) found that aerosol particles could have a strong impact on the ground-level O_3 in Tianjin. Yao et al. (2011) and Han et al. (2011) presented evidence that O_3 formation is strongly influenced by the prevailing levels of nitrogen oxidants (NO, NO_2 and NO_x). Due to the implementation of stringent emission restrictions under China's 12th Five-Year Plan (2011–2015), NO_x emission has substantially decreased in the major urban regions of China ever since (Zhao et al. 2013a). Whereas for VOCs, emission of which has been growing in mainland China since the 1980s is attributed to the lack of targeted regulations (Lu et al. 2013). How the continuing NO_x emission reduction and rising tendency of VOCs would influence the O_3 pollution in China, after removing the disturbance of meteorological factors, remain an active area of interest. To address this issue, Xing et al. (2011) analyzed multiple ideal emission reduction options based on the CMAQ model and indicated that solely reducing local NO_x or VOC emissions in Beijing (even by 90%) would still be insufficient to meet the O_3 Class 2 Ambient Air Quality Standard of China. What are the dominant meteorological factors controlling O_3 in different seasons, and how would local emission control strategies affect the O_3 time trends after removing the meteorological effects? Such studies were rarely involved with Tianjin, hence there is an urgent need for it in this area.

Given the effective role in exploring the complex nonlinearities of air pollutants, this work employs the GAM method to (1) characterize the relationships between different meteorological factors and O_3, (2) figure out the most important explanatory meteorological variables for each season and (3) meteorologically adjust the interannual variation trends of surface O_3 to investigate the effect of emission control strategies in Tianjin during the 2009–2015 period. Moreover, analysis in the present study would be done separately on a seasonal basis, considering that the mechanisms governing the relations between O_3 and meteorological factors differ substantially in different seasons, even change signs. Section 2 depicts the observation data and the statistical method applied in this study to characterize the association between ozone and meteorology and to meteorologically adjust the daily maximum O_3 concentrations. The results of the statistical analysis, involving the partial correlation between ozone and individual meteorological factor, the aggregate impact

of multiple meteorological factors on ozone, and the calculated trends of meteorologically adjusted O_3 are discussed in Sect. 3. Finally, Sect. 4 summarizes our findings.

2 Methodology

2.1 Site description

The meteorological and O_3 data employed in this study were collected at the urban meteorological monitoring site (39°04′N, 117°12′E) in Tianjin, China. This site is located in a mixed residential and traffic area, with no industrial source around. The terrain is flat with several low residential buildings and commercial buildings within a radius of approximately 1 km of the site. Two main roads (Heiniucheng road and Youyinan road) run E–W and S–N, respectively, about 100 m from the site.

2.2 Measurements and instrumentation

Surface O_3 concentration was continuously measured using the calibrated instrument from Thermo Environmental Instruments (US) through UV-absorption method. The concentration data was recorded every 5 min and used to calculate hourly mean value when the missing data were less than 25% of the hourly total, otherwise the hour would be treated as missing value. In this study, the daily ozone maxima was defined as the daily maximum 8-h average O_3 concentrations, and was calculated only if at least 75% of the data were available for the given day. Quality control checks were implemented every month involving calibration of the instruments as well as zero, precision and span checks. The standard samples of O_3 were supplied by National Standard Substance Institute of China. More detailed descriptions on data quality control of the urban meteorological monitoring site could be referred to Han et al. (2011).

Meteorological parameters, including air temperature, horizontal wind speed, wind direction, relative humidity and precipitation were measured with an automation weather station fixed at this meteorological site. In addition, the upward/downward longwave and shortwave radiation are measured by a net radiometer (CNR4, Kipp&Zonen) which were sampled every 1 min.

2.3 Statistical method

In this work, we explored the relationship between daily ozone maxima and the selected meteorological parameters for each season separately in Tianjin with the adoption of GAM in R software (R Development Core Team 2017). The GAM is a flexible and effective technique for incorporating linear or nonlinear relationships with continuous and discrete variables. The expression is as follows:

$$g(\mu_i) = X_i\theta + f_1(x_{1i}) + f_2(x_{2i}) + f_3(x_{3i}) + \cdots + \varepsilon_i \quad (1)$$

where i means the meteorological observation on the ith day. $g(\mu_i)$ is the link function specifying the relationship between the linear or nonlinear terms on the right-hand side of Eq. (1) and the response term μ_i. In this study, we employed a Gamma distribution and a log link function which has been successfully applied to ozone-related analysis in previous studies (Camalier et al. 2007; Pearce et al. 2011). $X_i\theta$ refers to the discrete variables which are not subject to linear/nonlinear relation and the corresponding coefficients. $f()$ represents the smoothing function and $f_j(x_{j,i})$ means the value of the smoothing function that corresponds to the observation of the variable j on the ith day. In addition, the penalized cubic regression splines (CRS) method was employed for the smooth functions $f_j(x)$ to incorporate the complex nonlinear response between different meteorological variables and daily ozone maxima. ε_i represents the residual.

The meteorological variables explaining most of the variability in the daily ozone maxima were selected from an extensive array of candidate meteorological variables. The important parameters could be identified using the F statistic in GAM. F statistic involves the comprehensive consideration of p value and degrees of freedom (e.d.f.) for each parameter. Those explanatory parameters with large p values (i.e., more likely to have a null coefficient hence lower impact on ozone levels) would be removed from the GAM model. A threshold value ($p < 10^{-3}$) was chosen to make sure that a not too large set of parameters could explain a considerable fraction of the fluctuations in daily ozone maxima. Only the parameters satisfying the threshold criterion ($p < 10^{-3}$) were finally incorporated into the GAM model for each season. Camalier et al. (2007) has employed F statistic to identify the dominant meteorological factors for 39 American cities. All the candidate meteorological variables considered in this study are listed in Table 1, as summarized in the associated researches focused on European (Ordonez et al. 2005) and North American cities (Camalier et al. 2007; Zheng et al. 2007). The main parameters considered in our analysis involve air temperature, shortwave radiation, wind speed/direction, air humidity and precipitation, which would influence ozone concentrations through photochemical formation, transport, diffusion, deposition and destruction processes. It is noteworthy that both the morning (afternoon) temperature and the square of it are taken into consideration in our analysis (as listed in Table 1). This is because the dependence of O_3 on temperature is generally quadratic rather than linear in Tianjin, as illustrated in Sect. 3.1. Besides, we also include the parameter 'day of the week (wd)' to test whether there is any clear difference in O_3 levels between weekdays and weekends (the so-called

Table 1 List of all the candidate meteorological variables for the adjustment model

Variable	Definition
mT[a]	Morning temperature (°C)
mT^2	Square of the morning temperature ($°C^2$)
aT	Afternoon temperature (°C)
aT^2	Square of the afternoon temperature ($°C^2$)
mRad	Morning downward shortwave radiation (W/m^2)
aRad	Afternoon downward shortwave radiation (W/m^2)
mSun	Morning sunshine duration (h)
aSun	Afternoon sunshine duration (h)
mWspeed	Morning wind speed (m/s)
aWspeed	Afternoon wind speed (m/s)
mWdir[b]	Morning wind direction
aWdir[b]	Afternoon wind direction
mSH	Morning specific humidity (g/kg)
aSH	Afternoon specific humidity (g/kg)
Precip[b]	Absence/occurrence of precipitation on the investigated day
pPrecip[b]	Absence/occurrence of precipitation on the previous day
wd[b]	Day of the week

[a]The morning (afternoon) temperature refers to the average value of air temperature from 06 to 12 h (12–18 h), and it was calculated only when at least 5 out of the 6 h values were valid on the respective day. Likewise, the morning (afternoon) averages of other meteorological variables were defined in the same manner

[b]Discrete variables

'weekend effect') as has been documented in some areas (Atkinson-Palombo et al. 2006). In this study, the correlation analysis and meteorological adjustment of the daily ozone maxima during the 2009–2015 period are all conducted for different seasons separately, namely, spring (MAM), summer (JJA), autumn (SON) as well as winter (DJF).

To further assess whether or not the emission control strategies are effective in reducing the ozone levels, variation in ozone that was caused by the meteorological influence needs to be removed. To address this issue, daily ozone maxima can be adjusted for meteorological effects as follows (Ordonez et al. 2005):

$$O_3(\text{adjusted}) = \text{mean } O_3 + \left[O_3(\text{measured}) - O_3(\text{predicted})\right] \quad (2)$$

where mean O_3 refers to the mean value of all the considered daily ozone maxima in the investigated spring, summer, autumn or winter of 2009–2015; O_3 (measured) refers to the measured daily maxima of O_3 concentrations in the investigated period; and the predicted (i.e., explained by meteorology) daily ozone maxima, O_3 (predicted), could be calculated based on Eq. (1). The motivation of meteorologically adjusted daily ozone maxima is for better understanding the ambient O_3 trends, especially in those megacities affected by the implementation of emission control programs.

The trends of the measured and meteorologically adjusted daily ozone maxima for different seasons could be given by the slopes of the linear regression of the respective yearly medians of the daily ozone maxima against the year Y:

$$O_3(\text{measured}) = d_1 + d_2 Y + \varepsilon \quad (3)$$

$$O_3(\text{adjusted}) = e_1 + e_2 Y + \varepsilon. \quad (4)$$

3 Results and discussion

3.1 Observed annual variation of surface ozone concentrations

Observed annual variation of surface ozone concentration during 2009–2015 in Tianjin exhibits evident seasonal pattern which peaks in summer (see Fig. 1). Daily ozone maxima close to 120 ppb was frequently observed during this period, with the highest hourly O_3 concentration reaching up to 162 ppb. Such high ozone episodes would always lead to an increase in the risk of acute health issues in a short time, for instance, asthma or heart attack.

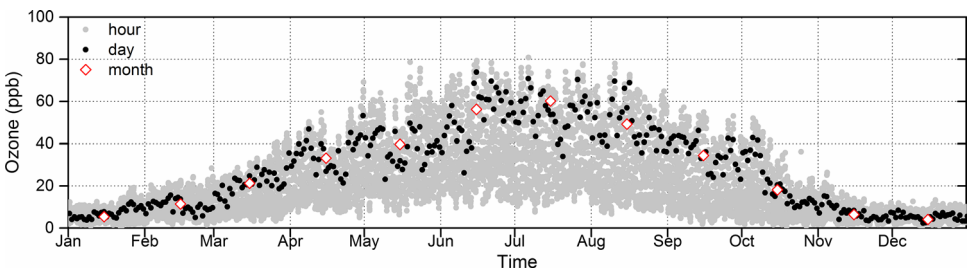

Fig. 1 Observed hourly ozone concentrations (gray dots) in Tianjin averaged during the 2009–2015 period. Also shown is the daily maximum 8-h average O_3 concentrations (i.e., daily ozone maxima; black dots) and the monthly medians of daily ozone maxima (red diamonds)

3.2 Impact of individual meteorological factor on ozone

Each meteorological factor plays its unique role in explaining the variations in ozone concentration through different physical and/or chemical mechanisms. Moreover, as pointed out by Thompson et al. (2001), the dependency of meteorology on ozone concentration may vary substantially in different regions and/or different seasons, as a result of various climatic conditions and background concentration of local pollution. In this section, the partial correlation analysis is employed to investigate the impact of individual meteorological factors on local O_3 levels in Tianjin for each season. Because the influence of meteorological conditions on ozone is an integrated effect, using the correlation coefficient may give misleading information if there is another confounding variable that is numerically related to both variables of interest. The partial correlation coefficient (PCC) measures the effect of a specific meteorological variable on ozone after removing the effects of a set of other variables.

Daily ozone maxima are always tightly associated with ambient air temperature (see Fig. 2). Moreover, according to our data, the dependency of surface O_3 levels on afternoon temperature is stronger than that on morning temperature, probably due to the fact that daily maximum temperature generally occurs in the afternoon. Figure 2 exhibits the scatter plots of daily ozone maxima against afternoon temperature on the respective day, along with the PCC and the fitted regression equation, for winter (Fig. 2b) and other seasons (Fig. 2a). All the PCC values shown in the figure are statistically significant at the 95% confident level (the same below). As can be seen from Fig. 2, daily ozone maxima are closely related to the afternoon temperature throughout the year (PCC = 0.41) except in winter (PCC = 0.15). In addition, the dependence of daily ozone maxima on temperature is more quadratic than linear. The strong correlation between daily ozone maxima and afternoon temperature is mainly due to the fast photochemistry combined with elevated biogenic volatile organic compound (BVOC) emissions under high temperatures (Curci et al. 2009).

Since solar radiation is the major driver of photochemistry, the level of O_3 and surface shortwave radiation fluxes would be inextricably linked. As expected, a quite strong positive correlation (PCC = 0.35–0.42) could be found between solar radiation and daily ozone maxima in all seasons (see Fig. 3). The dependency of daily ozone maxima on solar radiation is weaker than that on ambient temperature in spring and summer. Whereas in autumn and winter, solar radiation, rather than air temperature, plays a more dominant role in determining the O_3 level. In addition to the key role in photochemical reactions, solar radiation also affects the vertical mixing through surface heating and hence the dilution of air pollutants, resulting in less ozone destruction. While photochemical production is strongly restrained, ozone destruction prevails in winter, which could explain the much tighter association between daily ozone maxima and solar radiation in winter, rather than ambient temperature.

Another parameter concerning the influence of solar radiation is the sunshine duration. It is defined as the hourage during which the solar radiation flux exceeds 120 W/m^2. Similarly, both the morning and afternoon total sunshine duration are calculated. As presented in Fig. 4, daily ozone maxima generally increase with longer sunshine duration. Moderate positive PCC exists between daily ozone maxima and sunshine duration (PCC = 0.31–0.37).

The wind-related variables, including wind speed and direction, are also taken into consideration in the present study, as they influence the transport and dilution of air pollutants. Based on our data (figure is not shown), the dependency of O_3 on wind speed is quite poor (|PCC| < 0.2) and variable. In general, O_3 concentration exhibits a weak negative correlation with wind speed in spring, summer and autumn, but a weak positive correlation in winter. This is probably because stronger wind facilitates the dilution of O_3 and NO at the same time, while the reduction of NO would lead to lower O_3 titration counteracting the dilution effect to

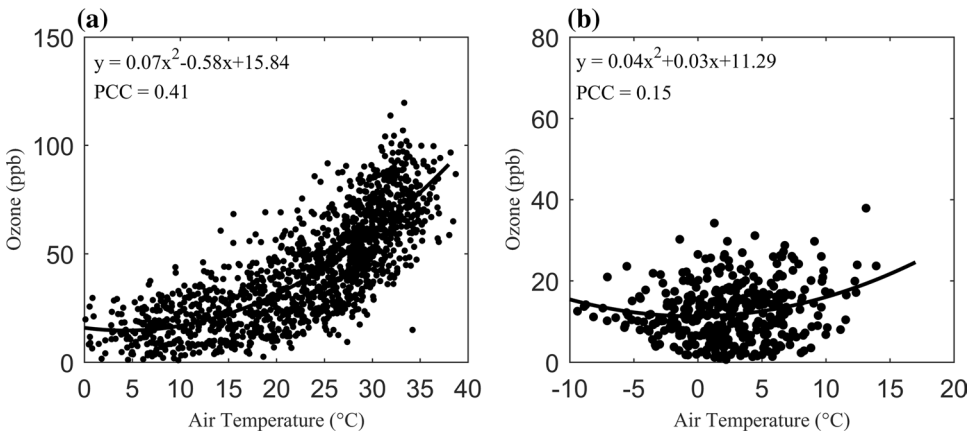

Fig. 2 Scatterplots of daily ozone maxima against afternoon temperature in spring, summer and autumn (**a**), and winter (**b**) during 2009–2015. The partial correlation coefficient (PCC) and fitted regression equation between daily ozone maxima and afternoon temperature are also shown in the figure

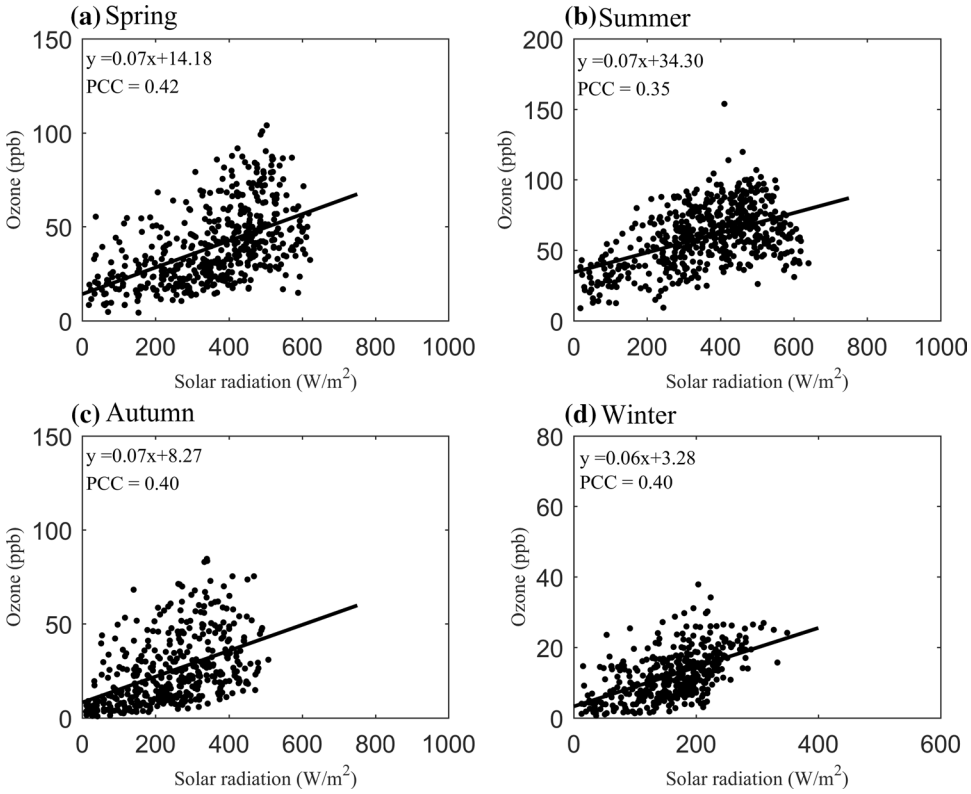

Fig. 3 Scatterplots of daily ozone maxima against morning solar radiation in spring (**a**), summer (**b**), autumn (**c**), and winter (**d**) during 2009–2015

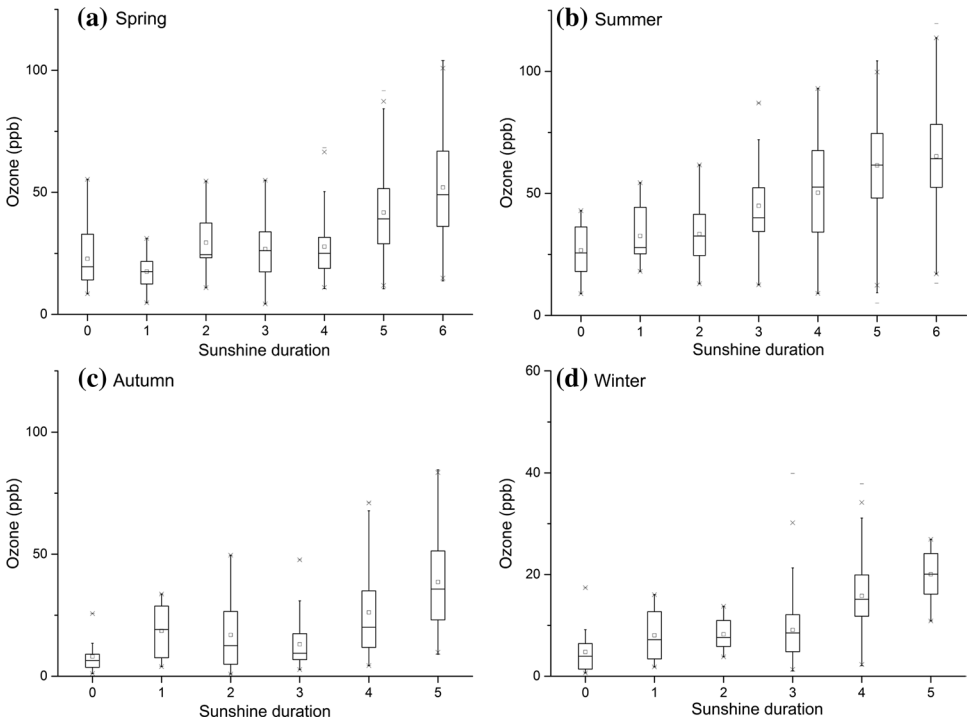

Fig. 4 Boxplots of the measured daily ozone maxima against morning sunshine duration for all seasons during 2009–2015. In each box, the line across the box stands for the median value ($q_{0.5}$), the edges of the box are the 25th ($q_{0.25}$) and 75th ($q_{0.75}$) percentiles. The squares are the mean values. The whiskers extend from the top and bottom of the box to show the extent of the main body of the data. Outliers are drawn as symbol '×' if they are larger than $q_{0.75}+1.5(q_{0.75}-q_{0.25})$ or smaller than $q_{0.25}-1.5(q_{0.75}-q_{0.25})$

some extent. Despite the weak dependency of O_3 on wind speed, the wind direction is likely to exert an impact on the occurrence of ozone episodes in Tianjin. To address this issue, Fig. 5 exhibits the daily ozone maxima in relation to wind direction in different seasons averaged from 2009 to 2015. In the present study, wind direction is treated as a discrete variable and is divided into eight sectors. Distributions of daily ozone maxima in winter are not presented, because

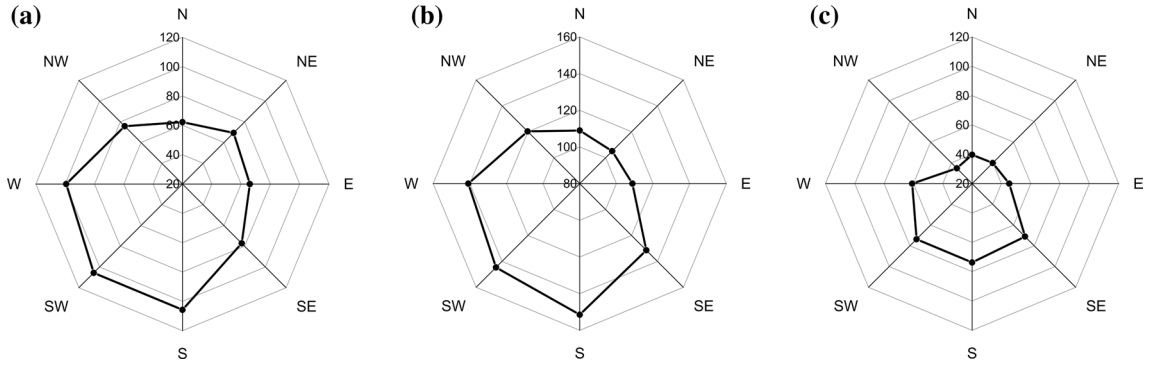

Fig. 5 Distributions of daily ozone maxima in eight wind directions in spring (**a**), summer (**b**) and autumn (**c**) averaged during 2009–2015. The radical coordinate refers to ozone concentration (μg/m^3)

there was barely any O_3 episode in winter and thus only a slight correlation with wind direction is seen. As illustrated in Fig. 5, when wind blows from W–SW–S sector, the O_3 levels are generally higher than the cases in which wind blows from other sectors in spring, summer and autumn. This indicates the important role of regional transport from upwind cities (e.g., Shijiazhuang, Baoding, Hengshui in Hebei Province) to Tianjin in these three seasons.

Figure 6 indicates that there is also a moderate effect of water vapor on O_3 concentrations in spring (PCC = 0.24) and autumn (PCC = 0.31), whereas in summer and winter, the correlation is very weak (|PCC| < 0.02). The modeling study by Dawson et al. (2007) also found complicated impact of water vapor on ozone, and attributed it to the compensating interactions between water vapor and O_3. High air humidity could enhance the production of OH radicals yielding higher O_3 concentrations in high-NO_x regions (e.g., urban areas). Whereas in another way, increasing water vapor also increases O_3 loss through the consumption of excited oxygen atom $O(^1D)$ (Jacob and Winner 2009):

$$O_3 + h\nu \rightarrow O_2 + O(^1D) \quad (5)$$

$$O(^1D) + H_2O \rightarrow 2OH \quad (6)$$

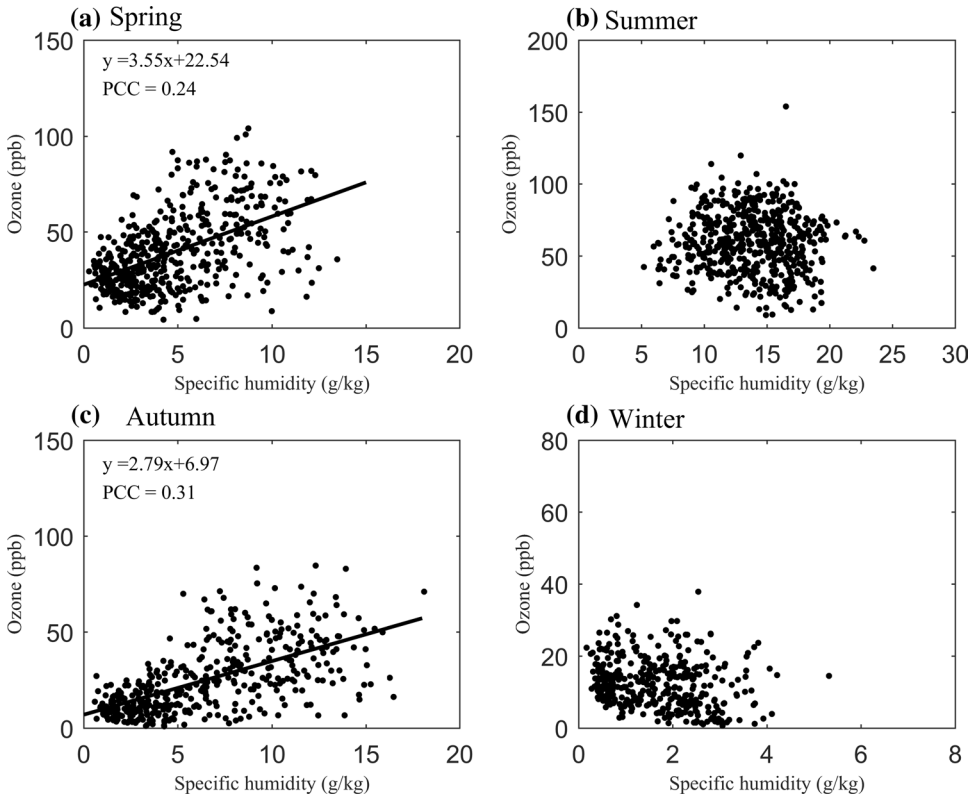

Fig. 6 Scatterplots of daily ozone maxima against morning specific humidity in spring (**a**), summer (**b**), autumn (**c**), and winter (**d**) during 2009–2015

Besides, some researches have also pointed out an additional effect under very dry conditions, i.e., the drought stress on vegetation, which would suppress the stomatal uptake of O_3 and hence its dry deposition; this effect is suggested to be a key factor leading to the high O_3 episodes over European cities during summer months (Solberg et al. 2008).

In addition, the absence/occurrence of precipitation is considered as a discrete variable both on the investigated day and the previous day to give account for the impact of wet removal of air pollutants. Nevertheless, data analysis in our study shows no clear difference of O_3 levels whether or not precipitation occurred on the investigated/previous day (figure is not shown). This indicates that wet deposition could be negligible for ozone mainly due to the hydrophobic nature of ozone as well as its major precursors (Jacob and Winner 2009).

Apart from the above-mentioned meteorological factors, the day of the week (i.e., weekday: Monday–Friday, weekend: Saturday and Sunday) is also taken into consideration to account for the weekly cycle of the anthropogenic emission of O_3 precursors, as reported in some areas of the world (Qin et al. 2004; Sakamoto et al. 2005). Based on our observations in Tianjin, however, there are no obvious distinctions of O_3 level between weekends and weekdays for all seasons (figure is not shown).

3.3 Aggregate impact of multiple meteorological factors on ozone

In the present study, the GAM method was employed to depict the aggregate impact of multiple meteorological factors on daily ozone maxima for different seasons during the 2009–2015 period in Tianjin. All the variables listed in Table 1 were initially introduced in the GAM model for each season separately.

All the meteorological factors selected by the final model, i.e., the most important explanatory factors, for each season are listed in Table 2. On the basis of Eq. (1), the predicted (i.e., explained by meteorological factors) daily ozone maxima of Tianjin could be calculated for each season with the selected variables (see Table 2) and the corresponding coefficients.

Table 2 Most important explanatory variables involved in the GAM for each season

Spring	Summer	Autumn	Winter
aT^2	aT^2	aT^2	aRad
mRad	aRad	aRad	aSun
aSH	mSH	aSH	aWspeed
mWdir	mWspeed mWdir	mWdir	

Table 3 Number of predictors selected by the GAM model for each season and the seasonal averages of the explained variance and correlation coefficient

	Predictors	Expl. var. (%)	Corr. coef.
Spring	4	75.7	0.87
Summer	5	59.6	0.77
Autumn	4	77.4	0.88
Winter	3	40.7	0.64

The overall model performance is evaluated by statistical analysis (see Table 3). The statistical metrics applied in this study included the correlation coefficient (R) to determine the degree of correlation between the predicted and measured ozone levels, and the explained variance (EV) to quantify the ratio to which the GAM model accounts for the variation of observed values. As can be seen from the statistical results, GAM could explain the O_3 level pretty well, especially in spring, summer and autumn, with EV ranging from 59.6% to 77.4%, and R higher than 0.77. That is to say, for spring, summer and autumn in Tianjin, approximately 60–77% of the variance in the daily ozone maxima could be explained by the meteorological parameter-based GAM model. Whereas in winter, the model performance is relatively poor (EV = 40.7%, $R = 0.64$). This is probably because the effect of meteorology on O_3 levels is greatly limited for low-temperature conditions (e.g., ~ 20 °C), as pointed out by Camalier et al. (2007). Given that there is scarcely any photochemical episode in the winter months, the relatively poorer model performance in winter seems to be acceptable. Besides, it can still depict the variation trend of daily O_3 maxima pretty well ($R = 0.64$) in winter.

To further assess the prediction capability of the GAM model, the coefficients calculated with Eq. (1) for each season in Tianjin during the period 2009–2015 are reserved for the calculation of daily ozone maxima in 2016. Subsequently, the overall model performance is evaluated by both visual comparisons (see Fig. 7) and factor 2 analysis (see Fig. 8). Comparisons show that GAM could reproduce the variation trends of daily ozone maxima well ($R \in [0.58, 0.87]$) for all seasons of 2016 in Tianjin. This also confirms the key role of meteorological factors in affecting surface ozone concentrations.

Factor 2 analysis is one of the most commonly applied methods to validate whether the predicted concentrations of air pollutants are acceptable (Li et al. 2011). Factor 2 (FAC2) indicates the percentage of the ratios of the predicted value to the observed value that range between 0.5 and 2. The expression is as follows:

$$\mathrm{FAC2} = \frac{N_{[1/2,2]}}{N_t} \tag{7}$$

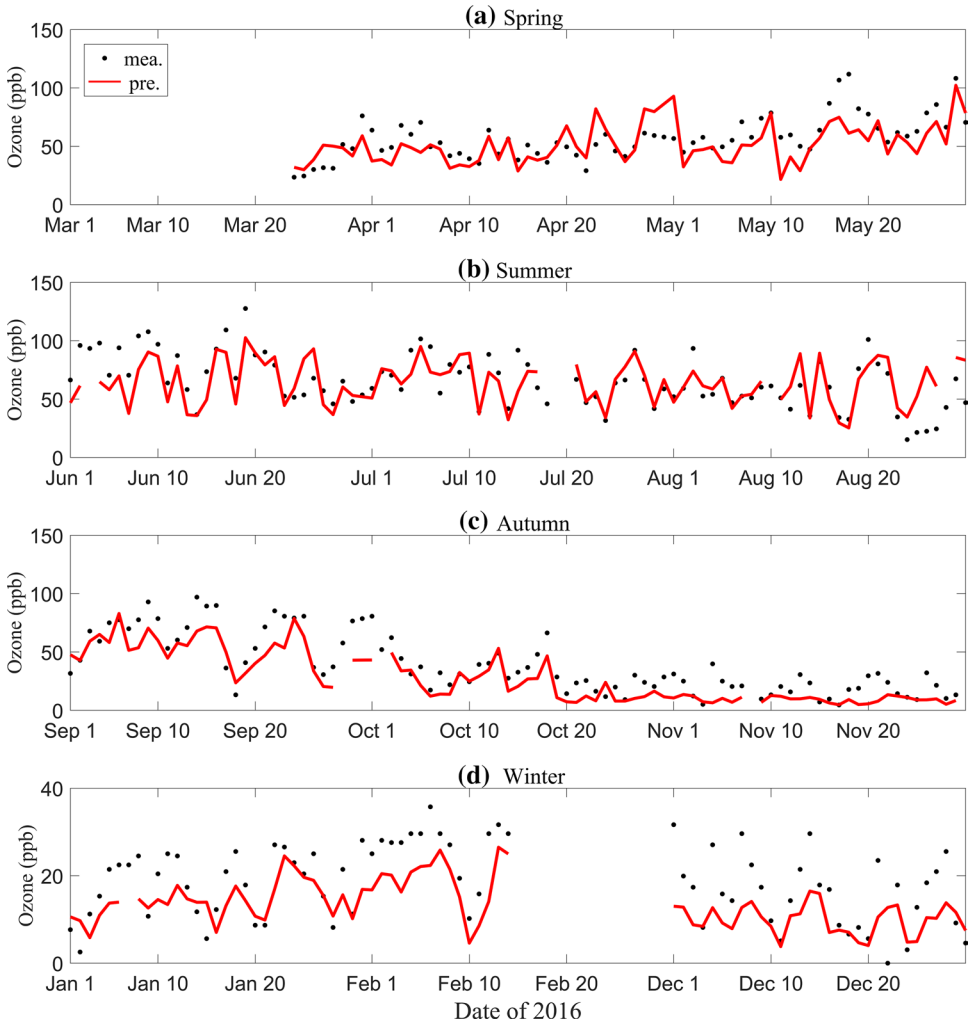

Fig. 7 Comparison of predicted daily ozone maxima (red line) against observations (black dots) for all seasons in 2016

where $N_{[1/2,2]}$ is the number of the ratios ranging between 0.5 and 2; and N_t is the total number of sample points. FAC2 = 100% means that the model could satisfactorily predict the observed values. The larger the FAC2 value is, the better the model performs. Figure 8 exhibits the results of factor 2 analysis for daily ozone maxima in Tianjin for all seasons of 2016. Results show that the FAC2 values of daily ozone maxima are within the range of [0.77, 0.99] for all seasons throughout the year, indicating that the multiple linear model performs quite well in the prediction of daily ozone maxima in Tianjin.

On the basis of model validation, Tables 2 and 3 show that only about 3–5 parameters are incorporated into the final model for different seasons. Overall, the afternoon temperature is the leading meteorological factor for controlling O_3 levels throughout the year except in winter. High temperatures are usually related to enhanced chemical O_3 production. In addition, the dependence of daily ozone maxima on temperature is more quadratic (aT^2) than linear (aT). Also as expected, parameters that associated with the properties of solar radiation (i.e., mRad, aRad, aSun) are also reserved in the final GAM for most seasons, as solar radiation is one of the main drivers of photochemical reactions and it can also affect the vertical mixing, namely the dilution of air pollutants. Wind field parameter (wind speed and/or direction), due to its impact on the transport and dilution of air pollutants, are also kept in the final model for all seasons. It is interesting that a negative correlation was found between wind speed and daily ozone maxima in summer, mainly due to the more dominant role of local photochemical production than other seasons so that the wind flow primarily acts to lower O_3 concentrations by dilution; whereas the opposite effect could be found in winter as the stagnation of air masses (i.e., low wind speed) favors the O_3 loss by NO titration. The role of water vapor is significant only in spring, summer and autumn. This is because higher air humidity enhances the formation of OH radicals, which would subsequently yield higher O_3 concentrations in high-NO_x regime (Vogel et al. 1999). Other parameters

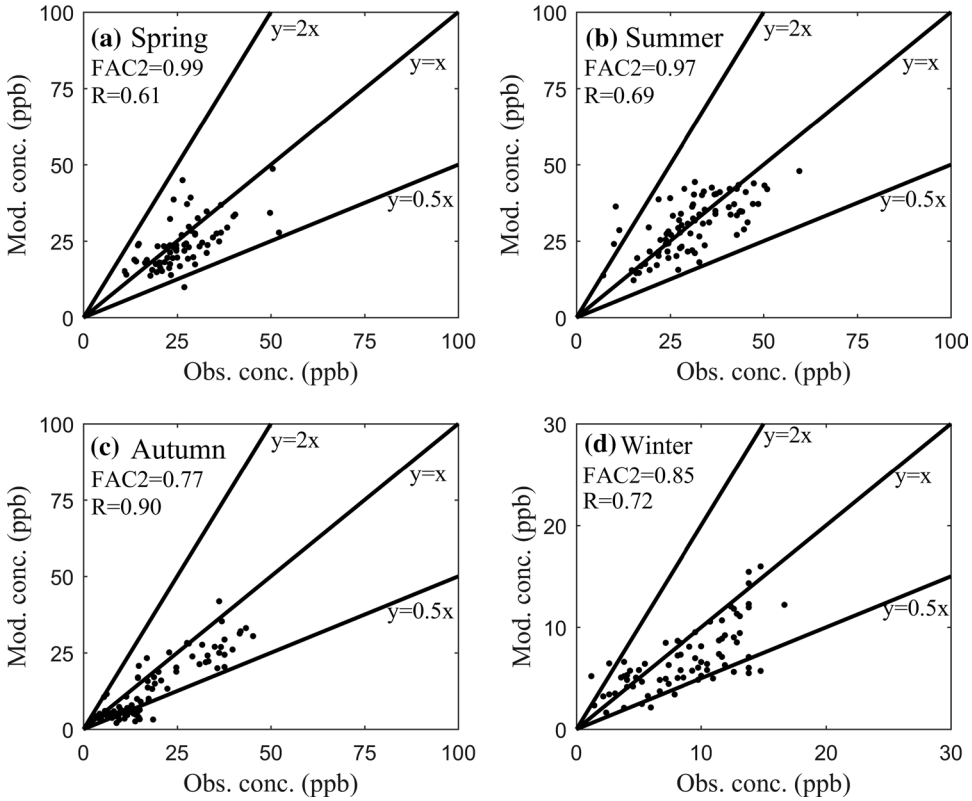

Fig. 8 Factor 2 analysis of predicted daily ozone maxima in Tianjin for all seasons in 2016

that are not involved in the final regression model for each season, because there is either a poor dependence of the daily ozone maxima on these parameters or their effects have already been explained by other correlated meteorological variables.

In general, the number of meteorological variables considered is higher in the summer months, due to the importance of local photochemical effects as well as more variable meteorology in summer. During the summer months, the most important explanatory variables are those influencing the photochemical production. High temperatures, as well as stronger solar radiation, are generally associated with increasing biogenic and anthropogenic VOC emissions, enhanced thermal decomposition of PAN, and faster photochemical reaction rate. In contrast, only three parameters are significant in determining the daily ozone maxima in winter, reflecting the weak dependency of ozone levels on meteorology, as already noted above. As photochemical reactions are greatly restrained in winter, the most important explanatory parameters are those that affect the vertical mixing process of air pollutants. The stability of atmospheric stratification is beneficial for ozone loss through titration with NO and dry deposition. Less ozone destruction, hence higher ozone levels are expected for stronger solar radiation as well as larger wind speed, because they favor vertical mixing.

3.4 Trends of meteorologically adjusted daily ozone maxima

On the basis of Eqs. (3) and (4), Fig. 9 shows the trends of the measured and meteorologically adjusted daily ozone maxima for all seasons during 2009–2015 in Tianjin. As shown in the figure, the year-to-year variability in the measured daily ozone maxima (the left panel in Fig. 9) can be effectively reduced after the meteorological adjustment (the right panel in Fig. 9). Here we employed the mean squared error (MSE) to quantify the extent to which the distribution of residuals spreads loosely (large MSE) or clusters tightly (small MSE) around the regression line. The MSE can be calculated as:

$$\mathrm{MSE} = \frac{\mathrm{SSE}}{n-2} \tag{8}$$

where SSE refers to the sum of squared errors in the simple linear regression models (Eqs. 6 and 7) and n is the sample size. The MSEs of the yearly medians of measured ($\mathrm{MSE}_{\mathrm{meas.}}$) and adjusted ($\mathrm{MSE}_{\mathrm{adj.}}$) daily ozone maxima for all seasons are listed in Table 4. In general, the variability in the yearly medians of measured daily ozone maxima is relatively higher for all seasons as indicated by the larger values of $\mathrm{MSE}_{\mathrm{meas.}}$ and thus small $\mathrm{MSE}_{\mathrm{adj.}}/\mathrm{MSE}_{\mathrm{meas.}}$ ratios. After meteorological adjustment, MSE could be reduced by

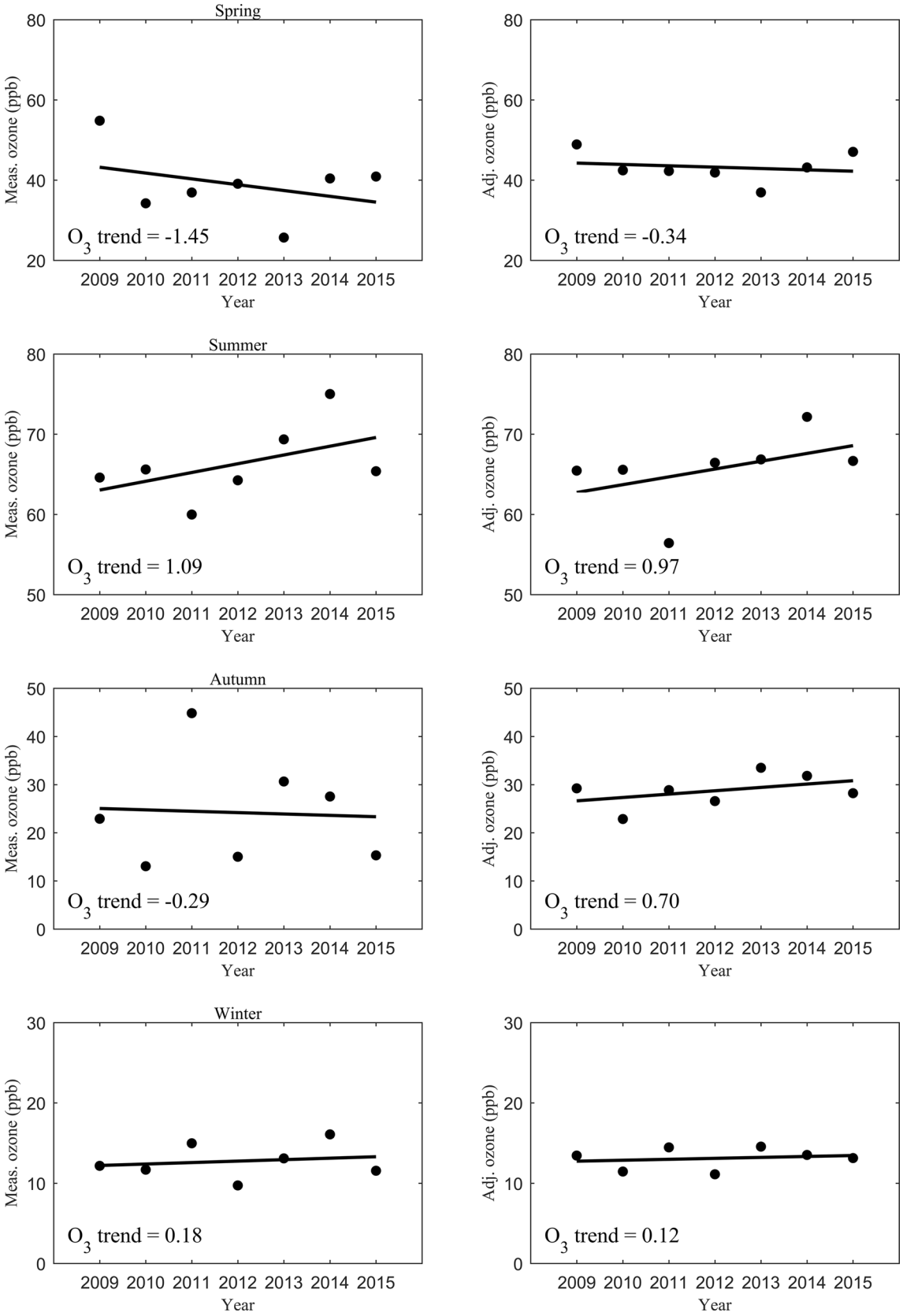

Fig. 9 Yearly seasonal medians of the measured (the left panel) and meteorologically adjusted (the right panel) daily ozone maxima for each season in Tianjin during 2009–2015. Also shown is the annual variation tendency of the seasonal medians of daily ozone maxima (unit: ppb year^{-1})

Table 4 Mean squared error (MSE) of the regressions of the yearly median of measured ($MSE_{meas.}$) and meteorologically adjusted ($MSE_{adj.}$) daily ozone maxima, and the MSE ratios—$MSE_{meas.}/MSE_{adj.}$—for each season in Tianjin during 2009–2015

	$MSE_{meas.}$	$MSE_{adj.}$	MSE ratio
Spring	43.01	9.78	0.23
Summer	14.02	5.29	0.38
Autumn	64.73	11.73	0.18
Winter	5.46	2.11	0.39

up to 82% in autumn, 77% in spring, and about 60% in summer and winter.

As shown in the left panel of Fig. 9, the seasonal medians of measured daily ozone maxima in Tianjin exhibit clear rising tendency only in summer (1.09 ppb $year^{-1}$), whereas downward trends are seen in spring (− 1.45 ppb $year^{-1}$) and autumn (− 0.29 ppb $year^{-1}$). Ozone trend in winter is not so evident (0.18 ppb $year^{-1}$). The variation trend of measured daily ozone maxima could be induced by both the changeable meteorological conditions and the artificial abatement efforts. Here we investigate the influence of emission control strategies on daily ozone maxima by checking the variation trend of meteorologically adjusted ozone levels in different seasons (the right panel in Fig. 9). After the meteorological adjustment, namely, removing the meteorological influence on O_3, the seasonal medians of daily ozone maxima exhibit a general upward tendency for most of the year. On average, the variation trend of adjusted daily ozone maxima is − 0.34, 0.97, 0.70 and 0.12 ppb $year^{-1}$ in spring, summer, autumn and winter, respectively. It is noteworthy that the rising tendency of O_3 concentration is the most evident in summer, when the occurrence of photochemical smog episodes is more frequent and induces greater risk on human health. This meteorologically adjusted ozone tendency suggests that the emission reduction strategies for primary pollutants in Tianjin during the 2009–2015 period were inappropriate to mitigate the photochemical pollution. Contrarily, based on our analysis, the emission reduction strategy appears to be more beneficial in limiting the effect of O_3 loss through titration with NO as a consequence of the decreased emission of primary pollutants, which result in the overall upward trend of adjusted O_3 for most seasons throughout the year. In addition, it has been recognized that the ozone formation in the urban area of BTH region is under VOC-limited condition, due to the abundance of NO_x emissions from traffic and industrial sources (Chou et al. 2009; Wang et al. 2010). That is to say, under this situation, decrease in NO_x concentration would lead to increase in O_3 levels. This could support our conclusions from a different view.

Figure 10 gives the annual variation of the measured and meteorologically adjusted daily ozone maxim with their respective regression lines. During the 2009–2015

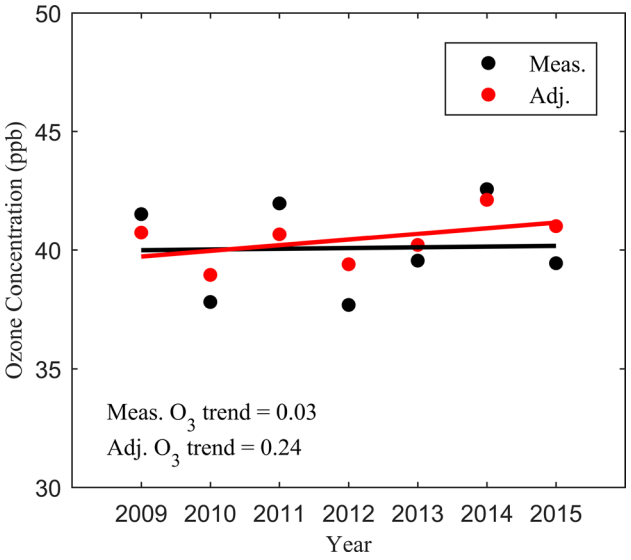

Fig. 10 Interannual variation of the yearly medians of measured (the black dots) and meteorologically adjusted (the red dots) daily ozone maxima in Tianjin during 2009–2015. Also shown is the interannual variation tendency of the yearly medians of daily ozone maxima (unit: ppb $year^{-1}$)

period, the measured O_3 levels show no clear tendency of deterioration (0.03 ppb $year^{-1}$), which makes it seem like the photochemical pollution is under control in Tianjin. Nonetheless, the trend analysis of the yearly medians of adjusted daily ozone maxima exhibits clear rising tendency (0.24 ppb $year^{-1}$) after removing the impact of meteorological variability. This positive annual trend indicates that recent emission reduction measures are more likely to lower the O_3 loss through titration by NO, rather than restrain the photochemical formation process. Hence, our analysis strongly suggests formulating a specialized mitigation strategy for regional O_3 pollution within the Tianjin area. For instance, Ou et al. (2016) recommended that synchronous reductions in anthropogenic VOC and NO_x with a ratio of 1:2 are needed to effectively reduce the peak ozone levels in PRD urban areas.

4 Conclusions

A statistical model named GAM has been employed to assess the impact of meteorology on daily ozone maxima and to meteorologically adjust the interannual variation of ozone levels in Tianjin during the 2009–2015 period. The analysis is done separately on a seasonal basis due to the fact that the mechanisms governing the relationship between ozone and meteorological factors may vary substantially in different seasons and even change signs. The observed surface O_3 concentration in Tianjin shows a clear seasonal

pattern, with the highest seasonal mean O_3 level occurring in the summer (JJA). During the 2009–2015 period, the maximum hourly O_3 concentration reached up to 162 ppb and the daily maximum 8-h average O_3 concentration close to 120 ppb could be frequently observed in the summer months during this period. Such high ozone episodes often cause a detrimental effect on public health.

Each meteorological parameter plays a unique role in affecting surface ozone levels through its own way. In the present study, partial correlation analysis is employed to investigate the impacts of individual meteorological factors on ozone and the significance level is tested. The statistical results indicate that daily ozone maxima is closely related to the afternoon temperature throughout the year except in winter and this dependence is more quadratic, rather than linear. A strong positive correlation could be found between solar radiation and daily ozone maxima in all seasons. Especially in autumn and winter, solar radiation, rather than air temperature, plays a more dominant role in determining the O_3 level. This is mainly because solar radiation is not only the main driver of photochemistry but also affects the ozone destruction which prevails in winter months. Wind direction also exerts an impact on the occurrence of ozone episodes in Tianjin. When wind blows from W–SW–S sector, the O_3 levels are generally higher than the cases in which wind blows from other sectors, suggesting the important role of regional transport from upwind cities to Tianjin. In addition, water vapor exhibits a moderate impact on ozone levels in spring and autumn.

GAM is an effective tool to explore the complex linear or nonlinear relationships between ozone and meteorological factors, and to figure out the important impact factors controlling ozone concentrations. Statistical results suggest that the model could explain the O_3 level pretty well with the explained variance ranging from 40.7% in winter to 77.4% in autumn and the correlation coefficient higher than 0.64. That means as much as 40–77% of the variance in the daily ozone maxima in Tianjin during 2009–2015 could be explained by the meteorological parameter-based GAM model for different seasons. Moreover, time series of the observed daily ozone maxima in 2016 are applied to validate the prediction capability of GAM. Both visual comparisons and statistical analyses demonstrate that the model performance is reliable in forecasting the daily ozone maxima in Tianjin. The selection of the most important explanatory parameters indicates that only about 3–5 parameters are incorporated into the final model for different seasons. During summer months, the most important explanatory variables are those influencing the photochemical production. Whereas in winter, ozone destruction by titration with NO and dry deposition is the dominant mechanism affecting O_3 levels.

Meteorological adjustment of O_3 concentration provides an approach to remove the meteorological influence on ozone variability, so as to assess whether or not the emission control strategies are effective in reducing the ozone levels during the investigated period. The year-to-year variability of daily ozone maxima could be reduced by up to 82% in autumn, 77% in spring, and about 60% in summer and winter after the meteorological adjustment. On average, the variation trend of meteorologically adjusted daily ozone maxima is − 0.34, 0.97, 0.70 and 0.12 ppb year^{-1} in spring, summer, autumn and winter, respectively. This general rising tendency in the seasonal medians of daily ozone maxima suggests that the emission reduction strategies for primary pollutants in Tianjin during the 2009–2015 period were insufficient to mitigate the photochemical pollution. On the contrary, the decreased emission of primary pollutants appears to be more beneficial in reducing the effect of O_3 loss through titration with NO, so as to result in the overall upward trend of adjusted O_3. On a yearly basis, despite the measured O_3 levels showing no clear worsening tendency (0.03 ppb year^{-1}) during the 2009–2015 period, the overall upward trend (0.24 ppb year^{-1}) of the meteorological adjusted daily ozone maxima strongly suggests formulating a specialized mitigation strategy for regional O_3 pollution for Tianjin areas.

As mentioned earlier, ozone measurements before mid 2000s were quite scarce in mainland China. Because of the lack of systematic ozone monitoring data in earlier years, this study only considered the variation trend of daily ozone maxima in Tianjin from 2009 to 2015, so that some years with very high/low O_3 levels may exert a strong impact on the calculation of the linear trend, especially when such years are located at the start/end of the investigated period. As the time span of measured O_3 data continues to grow, more comprehensive studies and longer time series of the ozone trend analysis need to be done in the future. Although limitations exist, our study still has an important scientific value, as it could provide a statistical methodology for characterizing the relationships between meteorology and ozone, improving the capacity of ozone forecasting, as well as meteorologically adjusting ozone trends to examine the underlying effects of emission control strategies on ozone levels apart from the natural effects.

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