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Ground ozone variations at an urban and a rural station in Beijing from 2006 to 2017: Trend, meteorological influences and formation regimes

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ABSTRACT

Due to complicated ozone formation regimes controlled by both ozone precursors and meteorological conditions, the underlying drivers for soaring ozone concentrations in Beijing were limitedly examined. With ground observation and remotely sensed data, the variation of ground ozone concentrations at an urban AT (Aoti) station and a rural YF (Yufa) station in Beijing was examined and a notable upward trend of ozone concentrations at both stations was found from 2006 to 2017. Following this, through a statistical model, the relative contribution of meteorological conditions to long-term ozone variations at both stations was calculated as 2%–3%, indicating anthropogenic emissions were the major cause for long-term ozone variations in Beijing. Meanwhile, short-term ozone variations could be influenced significantly by seasonal and synoptic meteorological conditions. Furthermore, the long-term variations of ozone formation regimes across Beijing were analyzed using OMI (Ozone Monitoring Instrument) retrieved HCHO VCDs (Vertical Column Densities)/NO_2 VCDs, which were strongly correlated with ozone concentrations. The results suggested that the ozone formation regime for the AT urban station and YF rural station had changed from VOCs-limited and NO_x-limited to VOCs-NO_x-limited respectively. In this case, NO_x-oriented emission-reduction measures for reducing $PM_{2.5}$ concentrations might conversely enhance ozone concentrations in Beijing. Given the increasingly heterogeneous distribution of different ozone formation regimes across Beijing, emission-reduction strategies that balanced consider the reduction of VOCs and NO_x emissions should be better designed and implemented according to local ozone formation regimes.

1. Introduction

In recent years, ground ozone concentrations across China increased rapidly, especially for summer, 2017, when severe pollution episodes were observed in such mega cities as Beijing, Nanjing and Xi'an distributed in different regions of China. Due to its severe threat on human health (Chen et al., 2012; Li et al., 2015b; Brauer et al., 2015) and crop production (NRC, 1991; Feng et al., 2014; Wang et al., 2017), the monitoring and management of soaring ground ozone pollution have received growing emphasis.

Since 2005, with the growing availability of ground ozone observation data, a large number of studies concerning the spatio-temporal patterns and the influencing factors of ground ozone concentrations across China has been conducted. These studies mainly focused on those developed regions, including the Pearl River Delta region in the south, the Beijing-Tianjin-Hebei region (Zhang et al., 2015; Ma et al., 2016; Wang et al., 2018; Cheng et al., 2018) in the north, the Pearl River delta (Zhang et al., 2013) in the

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south and the Yangtze River delta (Li et al., 2012; Ding et al., 2013; Pu et al., 2017) in the east of China. Specifically, soaring summertime ozone pollution in Beijing receives a major research interest.

Beijing suffers from persistent and severe air pollution. Since January 2013, high-concentration $PM_{2.5}$ has become the dominant airborne pollutant during the past five years. To reduce $PM_{2.5}$ concentrations, the local government implemented a series of emission-reduction measures, which effectively reduced the annually mean $PM_{2.5}$ concentrations in Beijing by 35% from 2013 to 2017. However, with a dramatic $PM_{2.5}$ reduction, ground ozone concentrations in Beijing increased significantly. In June 2017, ground ozone became the dominant airborne pollutant whilst ozone pollution episodes started to occur in Beijing. As a result, a comprehensive understanding of the trend and attribution of ozone pollution in Beijing has become an urgent environmental issue. In terms of the long-term variations, many studies proved a notable upward trend of ground ozone concentrations in Beijing. Ding et al. (2013) quantified that the overall ground ozone concentrations in Beijing rose 2% per year from 1995 to 2005. Ma et al. (2016) suggested that ground ozone concentrations at a background station in Beijing, which received limited anthropogenic emissions, increased 1.13 ppbv/yr from 2003 to 2013. Similarly, Zhang et al. (2015) found the ground ozone concentrations in a rural and an urban station in Beijing rose 0.18 μg/m^3 and 0.35 μg/m^3 annually from 2005 to 2013. Cheng et al. (2018) found that both O_3_1 h and O_3_8 h at urban stations demonstrated a linear and stable upward trend whilst ozone concentrations at a background station in Beijing demonstrated no regular patterns from 2004 to 2015. In addition to the trend analysis, the potential drivers for soaring ozone concentrations in Beijing have been comprehensively examined. From the perspectives of ozone precursors, Zhang et al. (2014) suggested that local photochemical reactions might lead to a faster increase rate of ground ozone concentrations at an urban observation station in Beijing from 2005 to 2011. Li et al. (2015a) indicated that vehicle exhaust contributed majorly to VOCs and ozone formation in Beijing. By analyzing the variations of ground ozone concentrations and corresponding precursors during a pollution period in October 2014, Chi et al. (2018) found the ozone production in Beijing was mainly influenced by the VOC-limited regime and thus the VOC reduction from Beijing and its neighboring areas could mitigate ozone pollution. Liu et al. (2019) also pointed out that both regional transport and local emissions led to a heavy ozone pollution episode in Beijing in summer, 2015. However, as Wei et al. (2018) suggested, the analysis of VOCs and precursors alone could not clearly explain the ozone variation in Beijing from 2013 to 2017, which was a combined effect of anthropogenic emissions and meteorology. Therefore, increasing studies have been conducted on meteorological attribution of ground ozone pollution and suggested that temperature (Li et al., 2017; Pu et al., 2017), humidity (Li et al., 2017; Chen et al., 2019b), wind speed (Tu et al., 2007; Tong et al., 2017), wind direction (Tong et al., 2017) and air pressure (Ding et al., 2013) were closely related to ground ozone concentrations. Specifically, Chen et al. (2019b) indicated that temperature, humidity and solar radiation were major meteorological drivers for ozone concentrations in Beijing from 2006 to 2016.

Despite massive studies conducted, a better understanding of the meteorological and anthropogenic drivers for the soaring ozone pollution in Beijing is required for effectively predicting and managing ozone pollution in Beijing. Firstly, most studies mainly focused on individual meteorological factors whilst overall meteorological influences on ground ozone concentrations were limitedly examined. Furthermore, both meteorological factors and the mass concentrations of different ozone precursors affect ground ozone concentrations by influencing the reaction regimes and rate for ozone production and decomposition. Therefore, quantifying the relative contributions of meteorological conditions and anthropogenic emissions to long-term ozone variations in Beijing remains challenging. Secondly, the production and decomposition of ground ozone are controlled by photochemical reactions between NO_x and VOCs (Sillman, 1999 ; ; Trainer et al., 2000). The reactions between ozone precursors are complicated and unpredictable, controlled by the proportion of different precursors and meteorological conditions. Therefore, the $PM_{2.5}$-reduction oriented emission-reduction measures, which mainly focus on NO_x reduction, lead to unstable effects on ozone reduction. As a case, Cheng et al. (2018) revealed that contingent emission-reduction measures for mitigating $PM_{2.5}$ pollution during haze episodes and special events had a weak, even converse effect on ozone reduction. Few studies thoroughly analyzed the long-term variation of different ozone precursors that influenced the mechanisms of ozone formation across Beijing, leading to the difficulty in the design and implementation of ozone-reduction oriented emission-reduction measures in Beijing.

To fill these gaps, based on long-term ground observation and remotely sensed data, this research aims to (a) examine the long-term ozone variations at an urban and a rural station in Beijing (b) understand the relative contribution of meteorological conditions to long-term ozone variations in Beijing based on an advanced statistical model, Kolmogorov-Zurbenko (KZ) filtering (c) understand long-term variations of ozone formation regimes across Beijing based on Ozone Monitoring Instrument (OMI) data. The trend, meteorological influences and formation regimes of ground ozone concentrations in the urban and rural station provide important reference for effectively understanding and managing ozone pollution in Beijing and other mega cities across China.

2. Materials and methods

2.1. Data sources

To comprehensively understand the trend, meteorological drivers and formation regimes of ground ozone in Beijing, long-term ground ozone data, meteorological data and ozone precursor data are required for this research.

2.1.1. Ozone data

Ground ozone data for this study were obtained from two monitoring stations, Aoti (AT) station in the Northern urban area, and the other is Yufa (YF) station located at the southern border of Beijing. 49C ozone analyzers (Thermo Fisher Corporation, USA) were employed for monitoring and collecting ozone and its precursor data, including CO, NO_2 and NO. The 49C ozone analyzer works based on the principle that the 254 nm wavelength is majorly absorbed by ozone and closely related to the volume fraction of ozone. Ground ozone concentrations are calculated by analyzing the absorption levels of specific wavelength when samples pass through the ozone analyzer.

2.1.2. Meteorological data

This research attempted to examine the long-term ozone trend at an urban and a rural station without the influence of meteorological variations. Therefore, a set of meteorological factors, including temperature (Daily mean temperature), relative humidity (Daily mean relative humidity), Solar Radiation (Daily SSD, sunshine duration), wind speed (Daily mean wind speed), wind direction (Daily dominant wind direction), precipitation (Daily total precipitation) and air pressure (Daily mean pressure) from the Guanxiangtai meteorological station, Beijing, (GXT, 116.46° E,

39.80° N), was acquired from China Meteorological Bureau. The location of AT, YF and GXT station is demonstrated as Fig. 1.

2.1.3. OMI data

Compared with reliable ground observation data of ozone and meteorological factors, long-term monitoring of major ozone precursors, Oxynitride and VOCs, were collected based on remote sensing data. Oxynitride and VOCs include a diversity of components and a commonly employed approach is to extract NO_2 and HCHO concentrations for monitoring the magnitude of Oxynitride and VOCs (Millet et al., 2006; Wolfe et al., 2016; Kaiser et al., 2018). For this research, long-term NO_2 VCDs (Vertical Column Densities) and HCHO VCDs were retrieved from OMI DOMINO version 2.0 products. The OMI instrument monitors solar radiation backscattered in the UV–visible domain that ranges from 270 to 500 nm (Schoeberl et al., 2001; Levelt et al., 2006). Several studies (Boersma et al., 2011; Hains et al., 2010; Lamsal et al., 2010; Piters et al., 2012) have verified the reliability of this product, and suggested that DOMINO was highly consistent with ground observation data. The tropospheric NO_2 VCDs for Beijing were obtained from Level 3 OMI daily global products (http://giovanni.gsfc.nasa.gov/giovanni) with a spatial resolution of 0.25° × 0.25°, uncertainty of $\pm 0.5–1.5 \times 10^{15}$ molecules cm^{-2} and an overall relative error of 10%–40% (Boersma et al., 2009). This type of OMI product may lead to large bias for areas with such bright objects as snow, and cloud. For a reliable estimation, pixels with the parameter cloud emissivity larger than 0.5 were removed. In addition, OMI troposphere HCHO VCDs, which are binned into 0.25° × 0.25°grids with an overall error of 25–31% (Millet et al., 2008), from 2006 to 2017 was obtained using HDFView2.11 software from NASA website/Belgian space high atmosphere Institute (BIRA-IASB, https://www.nasa.gov/). Similar to the process of NO_2 VCDs data, pixels with cloud amount greater than 20% or relative bias larger than 24% were eliminated to reduce the influence of cloud cover.

2.2. Methods

In addition to long-term trend analysis of ground ozone concentrations based on ground observation data, HCHO VCDs/NO_2 VCDs from OMI data can be used directly to understand the variation of ozone formation regimes in Beijing. One major aim and challenge is to understand long-term ozone variations in urban and rural areas within Beijing controlled by anthropogenic emissions. Therefore, a robust statistical model, KZ (Kolmogorov-Zurbenko) filter, was used to remove the influence of meteorological variations on ground ozone concentrations. Meanwhile, a MK (Mann-Kendall) test was used to verify whether the extracted trend of ground ozone concentrations was significant.

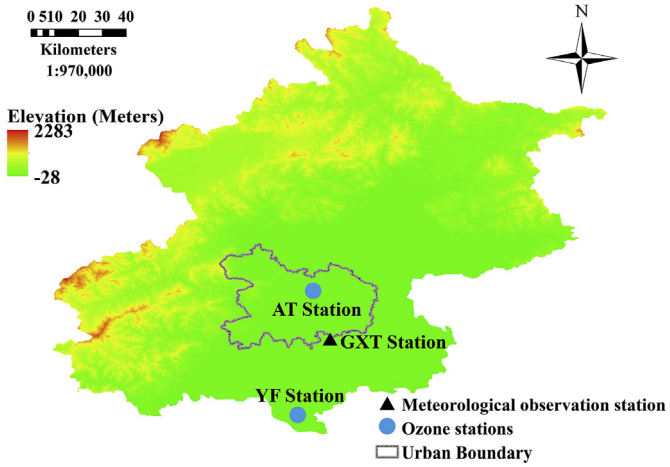

Fig. 1. The location of ozone and meteorological observation stations in Beijing.

2.2.1. Indicators of ozone formation regimes

NO_2 and HCHO concentrations are reliable proxies for in-situ observations of VOCs and NO_x during pollution episodes and HCHO/NO_2 is a reliable indicator for ozone formation regimes (Sillman, 1995; Duncan et al., 2010). Specifically, HCHO VCDs/NO_2 VCDs less than 1 suggests that ozone production reduces with the decline in VOCs (VOC-limited regime); HCHO VCDs/NO_2 VCDs greater than 2 suggests that ozone production reduces with diminishing in NO_x (NO_x-limited regime); HCHO VCDs/NO_2 VCDs between 1 and 2 suggests a transitional regime (VOCs-NO_x-limited regime) where ozone production can be affected by both VOCs and NO_x emissions (Liu et al., 2019). In this case, long-term satellite retrieved HCHO and NO_2 data provide solid support for understanding the variation of ozone production regime and ozone concentrations in Beijing.

2.2.2. Kolmogorov-Zurbenko (KZ) filtering

As introduced, meteorological conditions impose strong influences on ozone production. Therefore, seasonal components extracted from time series meteorological data should be filtered for understanding the adjusted trend of ground ozone concentrations mainly controlled by the variation of anthropogenic emissions (Eskridge et al., 1997). To extract time series data without disturbances from other influencing factors, Rao et al. (1994) proposed a Kolmogorov-Zurbenko (KZ) filter, which removes high-frequency variations through the iterative moving average. An intercomparison of four major trend detection models, including the KZ filter, PEST, anomalies, and wavelet transform, suggested that KZ filter achieved the highest confidence when processing long-term time series data (Eskridge et al., 1997). Given its reliable performances, growing studies (Zurbenko et al., 1996) employed KZ filter for extracting time series air pollutant data without seasonal signals of meteorological conditions. Specifically, Ma et al. (2016) employed the KZ filter to remove meteorological influences and extract long-term ozone trend at a background station, and suggested that meteorological conditions contributed limitedly to the rapid rise of ozone concentrations in Beijing. The reliable outputs from Ma et al. (2016)'s research proved that the KZ filter was a suitable approach for this research to reveal the long-term ozone trend without the influence of meteorological variations in urban and rural Beijing.

To avoid sharp breaks caused by the iterative moving average (m), an extended KZ filter with a dynamic m that varied according to the changing rate was proposed by Zurbenko et al. (1996). This extended KZ filter achieved reliable estimation of ozone concentrations in Beijing by removing the long-term influence of meteorological variations (Ma et al., 2016). This research employed this extended KZ filter, which is explained as follows:

The original time-series data is decomposed as:

$$X(t) = E(t) + S(t) + W(t) \quad (1)$$

$$Xb(t) = E(t) + S(t) \quad (2)$$

$$E(t) = KZ365,3(X) \quad (3)$$

$$S(t) = KZ15,5(X) - KZ365,3(X) \quad (4)$$

$$W(t) = X(t) - KZ15,5(X) \quad (5)$$

Where $X(t)$ presents the original time series. $E(t)$, $S(t)$ and $W(t)$ stand for the long-term, seasonal and short-term component respectively. The long-term component is mainly decided by long-term variations of anthropogenic emissions and climate changes. The seasonal component is mainly decided by seasonal meteorological variations, and the short-term component is mainly decided by short-term variations of anthropogenic emissions and small-scale weather systems. The $Xb(t)$ indicates the baseline component, presenting the general trend of the original time series. $KZi,j(X)$ features KZ filtering conducted on time series X using a moving window with the size of i and j iterations. Synoptic-scale episodes in Beijing usually last for 2 days to 3 weeks and thus can be filtered using a KZ filter with a 15-day window size and 5 iterations ($KZ15,5$). Meanwhile, $KZ365,3$ can filter the fluctuation of 632 days (Rao et al., 1994), which is suitable for filtering the long-term variation of meteorological conditions.

Through KZ filtering, $E(t)$ still retain some meteorological influences, which could be further filtered by multiple regression models established using meteorological factors highly correlated with ozone concentrations:

$$W(\mathrm{t}) = a_0 + \sum a_i \mathrm{w}_i(t) + \varepsilon_w(t) \tag{6}$$

$$X_b(\mathrm{t}) = \mathrm{b}_0 + \sum b_i x_i(t) + \varepsilon_b(t) \tag{7}$$

$$\varepsilon(t) = \varepsilon_w(t) + \varepsilon_b(t) \tag{8}$$

where $\mathrm{w}_i(t)$ and $x_i(t)$ stand for the short-term and baseline component of the ith meteorological factor. ε_w and ε_b are the regression residual of the short-term and baseline component. $\varepsilon(t)$ presents the total residual that includes the short-term variations of anthropogenic emissions and meteorological conditions neglected during the regression process.

For the calculated $\varepsilon(t)$, KZ filtering is conducted for acquiring the long-term component $\varepsilon_E(t)$. With filtered meteorological influences, the reconstructed time series data $X_{LT}(t)$ is computed as the sum of the mean value of $E(t)$ and $\varepsilon_E(t)$.

$$X_{LT}(t) = \overline{E(t)} + \varepsilon_E(t) \tag{9}$$

Hence, the relative contribution of comprehensive meteorological conditions, including multiple major meteorological factors to the long-term ozone variations is calculated as follows:

$$P_{contrib} = \frac{K_{\mathrm{org}} - K}{K_{org}} \times 100\% \tag{10}$$

where $P_{contrib}$ presents the relative contribution of meteorological conditions to long-term ozone variations, K_{org} stands for the changing rate of the original ozone time series; K indicates the changing rate of the restructured ozone time series with filtered meteorological influences.

2.2.3. MK (Mann-Kendall) test

M-K test (Mann, 1945; Kendall, 1975) has been widely used in trend analysis for precipitation, hydrology, and climate change (Gocic et al., 2013; Sang et al., 2014; Y. Chen et al., 2016; Yang et al., 2017). M-K test doesn't require the sample to follow specific patterns, and receives limited influence from outliers. Statistical value Z is computed as (11), with Z lower than zero represents the downward trend, and larger than zero represents the upward trend. |Z| larger than 1.28, 1.64 and 2.32 indicates that the sample passes the test at a 90%, 95% and 99% confidence level respectively.

$$Z = \begin{bmatrix} \dfrac{S-1}{\sqrt{Var(S)}} S > 0 \\ 0 \\ \dfrac{S+1}{\sqrt{Var(S)}} S < 0 \end{bmatrix} \tag{11}$$

where Z is the standard statistic; S is the statistic to be tested, Var is the variance. α represents the confidence level.

3. Results

3.1. Long-term variations of ground ozone concentrations in Beijing from 2006 to 2017

For both AT and YF station, monthly variations of O_3_8 h and O_3_1 h exhibited curves with a single peak, and high values appeared during May to September. Ozone concentrations increased significantly in April and May, reached the peak in June, and maintained a relatively high level in July and August. The decrease of ozone concentrations occurred in September and remained low from November to the next January. The long-term ozone trend at the AT and YF station from 2006 to 2017 was demonstrated as Fig. 2. For the AT station, the annually average O_3_8 h and O_3_1 h increased from 31.28 ppbv to 38.16 ppbv in 2006 to 49.3 ppbv and 58.11 ppbv in 2017. The annually mean increase rate for O_3_8 h and O_3_1 h was 1.64 ppbv yr^{-1} and 1.81 ppbv yr^{-1}. For the YF station, the annually average O_3_8 h and O_3_1 h increased from 31.63 ppbv to 39.14 ppbv in 2006 to 43.67 ppbv and 47.78 ppbv in 2017 respectively. The annually mean increase rate for O_3_8 h and O_3_1 h was 1.09 ppbv yr^{-1} and 0.79 ppbv yr^{-1}. According to the MK trend test, the long-term variation of O_3_8 h presented a significant upward trend at the AT ($n = 144$, $Z = 1.9886$, $\alpha = 0.05$) and YF ($n = 144$, $Z = 2.8115$, $\alpha = 0.01$) station. Meanwhile, O_3_1 h also demonstrated a significant upward trend at AT ($n = 144$, $Z = 1.8515$, $\alpha = 0.05$) and YF ($n = 144$, $Z = 2.4$, $\alpha = 0.01$) station.

Similar to previous studies (Ma et al., 2016; Cheng et al., 2018), this research revealed a significant upward trend of ground ozone concentrations at both AT and YF station from 2006 to 2017. The

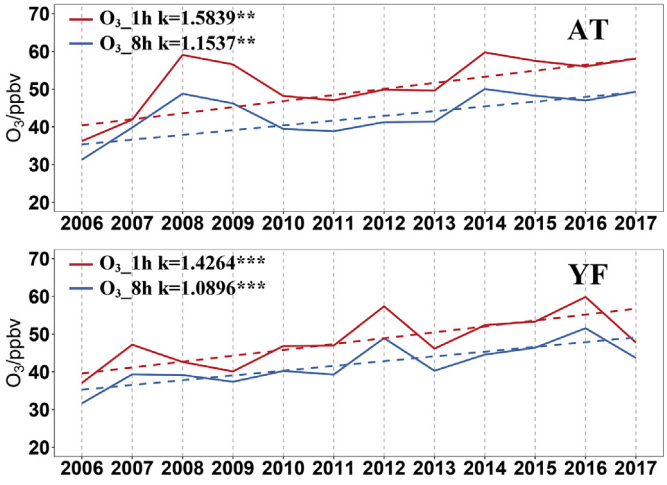

Fig. 2. The trend analysis of O_3_1 h and O_3_8 h from 2006 to 2017 at the AT and YF station based on MK test. ***, ** indicates the trend is significant at the a = 0.01, 0.05 confidence level.

initial annually mean ground ozone concentrations at the AT and YF station were very close in 2006. With a higher increase rate from 2006 to 2017, the annually mean O_3_1 h and O_3_8 h at the AT station were notably higher than O_3_1 h and O_3_8 h at the YF station in 2017. This could mainly be attributed to the rapid urbanization surrounding the AT station, where was not fully urbanized, for the preparation and conduct of 2008 Beijing Olympic Games, leading to successive soaring anthropogenic emissions and ozone concentrations at the AT station since 2006, especially in 2008 and 2009. For the YF rural station, the variations of ground ozone concentrations at the YF rural station from 2006 to 2017 was generally consistent, except for a sudden rise in 2012, which was consistent with Wan et al. (2014)'s research. Wan et al. (2014) also found a higher ground ozone concentration at rural stations than that at the urban stations in Beijing in 2012, which might result from the regional distribution and transport of ozone and precursors controlled by both anthropogenic and meteorological conditions.

Similar to previous studies (Ma et al., 2016; Cheng et al., 2018; Chen et al., 2019), this research revealed a significant upward trend of ground ozone concentrations at both AT and YF station from 2006 to 2017. The initial annually mean ground ozone concentrations at the AT and YF station were very close in 2006. With a higher increase rate from 2006 to 2017, the annually mean O_3_1 h and O_3_8 h at the AT station were notably higher than O_3_1 h and O_3_8 h at the YF station in 2017. This could mainly be attributed to the rapid urbanization surrounding the AT station, where was not fully urbanized, for the preparation and conduct of 2008 Beijing Olympic Games, leading to successive soaring anthropogenic emissions and ozone concentrations at the AT station since 2006, especially in 2008 and 2009. For the YF rural station, the variations of ground ozone concentrations at the YF rural station from 2006 to 2017 was generally consistent, except for a sudden rise in 2012, which was consistent with Wan et al.'s research (2014). Wan et al. (2014) also found a higher ground ozone concentration at rural stations than that at the urban stations in Beijing in 2012, which might result from the regional distribution and transport of ozone and precursors controlled by both anthropogenic and meteorological conditions.

In addition to the long-term trend analysis, the KZ filter was employed to decompose the original time series of monthly averaged O_3_8 h and O_3_1 h to reveal the major drivers for ozone variations in Beijing from 2006 to 2017 (Fig. 3). The larger the sum of the three components, the more independent the three components are. The long-term, seasonal and short-term components for decomposed O_3_8 h was 1.24%, 61.28%, 33.50% and 2.99%, 54.97%, 36.55% for AT and YF station respectively. Meanwhile, the long-term, seasonal and residual components of O_3_1 h were 0.92%, 61.23%, 33.80% and 2.86%, 48.54%, 42.41% respectively. Therefore, the sum of the three components for O_3_8 h and O_3_1 h at both stations was larger than 93.5%, indicating a satisfactory output of the KZ filtering. The relative significance of different components to both O_3_8 h and O_3_1 h was in the order of S(t) >W(t) > E(t), suggesting that ground ozone concentrations in Beijing were majorly affected by the variations of seasonal and short-term meteorological conditions.

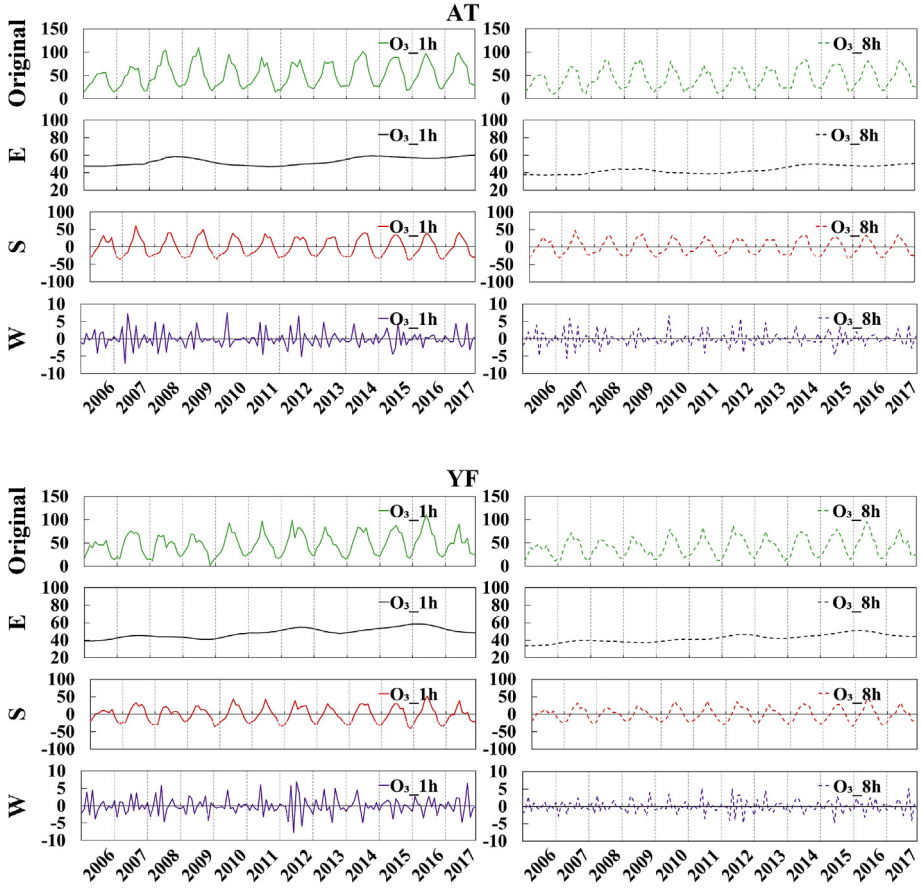

Fig. 3. The decomposition of monthly averaged O_3_8 h and O_3_1 h (ppbv) in Beijing from 2006 to 2017 using KZ-filtering. E: long-term component, S: seasonal component, W: short-term component.

3.2. The relative contribution of meteorological conditions to the long-term variations of ground ozone concentrations from 2006 to 2017

According to correlation analysis and MK test, wind speed, temperature, relative humidity, air pressure and solar radiation were major meteorological factors strongly correlated with ozone concentrations in Beijing (as shown in Table 1).

To remove the influence of other meteorological factors, multiple linear relationships between the long-term component of ground ozone concentrations and those correlated factors were established as follows (Take temperature as an instance).

$$E_X(t) = aE_T(t+17) + b + \varepsilon(t) \quad (12)$$

Where $E_X(t)$ presents the O_3_8 h (O_3_1 h) time series filtered by$KZ15,5$; $E_T(t)$ is the temperature that are filtered and lagged by 17 days; a and b present fitted parameters; $\varepsilon(t)$ represents other emission and meteorological factors not considered. Similarly, the influence of other factors on long-term ozone variations was computed. The comprehensive meteorological influence on ground ozone concentrations was thus calculated. Following this, the trend of original ozone time series and adjusted ozone time series with filtered meteorological variations at the AT and YF station were demonstrated in Table 2. Meanwhile, the relative contribution of meteorological conditions to long-term ozone variations at the AT and YF station was calculated. According to Table 2, the relative contribution of meteorological conditions at both stations was limited to around 2–3%, indicating that the anthropogenic emitted precursors was crucial for long-term ozone variations in Beijing.

3.3. Long-term variations of ozone formation regimes across Beijing

To comprehensive understand the anthropogenic driver for ozone variations in Beijing, both long-term variations of ozone formation regimes and ozone formation regimes during specific periods with emission-reduction measures were examined for this research.

Table 1
The trend of meteorological factors strongly correlated with ozone concentrations in Beijing.

Meteorological Factors	N	K (per month)	Z	Trend	Rs	p
Temperature	144	0.0032 °C **	2.20	up	0.567	0.000
Air Pressure	144	0.0180 hPa***	9.26	up	0.469	0.000
RH	144	0.0260%***	5.55	up	0.456	0.000
Speed	144	−0.0015 m/s***	−11.17	down	−0.478	0.000
SSD	144	−13.42 h**	−1.85	down	0.410	0.000

N: number of months. K: the variation trend. Z: standard statistic. Rs: the correlation coefficient between ozone concentrations and the individual meteorological factor. P: the significance of the correlation.
***, ** indicates the extracted trend is significant at the 0.01, 0.05 level.

Table 2
The relative contribution of meteorological conditions to long-term ozone variations in Beijing from 2006 to 2017.

Stations		Original trend pptv/month	Adjusted trend without meteorology influences pptv/month	The relative contribution of meteorological influences %
YF	O_3_8 h	0.095	0.093	2.76
	O_3_1 h	0.104	0.106	2.11
AT	O_3_8 h	0.057	0.055	3.52
	O_3_1 h	0.062	0.060	3.22

3.3.1. Variation of ozone formation regimes across Beijing from 2006 to 2017

Reactions between NO_x and VOCs to produce ozone are highly complicated and different proportion of NO_x and VOCs can lead to either the production or decomposition of ozone under different meteorological conditions. To effectively monitor the variations of ozone formation regimes across Beijing, major ozone precursors HCHO VCDs and NO_2 VCDs were retrieved from OMI data. The inter-annual variations of HCHO VCDs and NO_2 VCDs are presented as Fig. 4. Generally, the NO_2 VCDs increased since 2006, peaked in 2011 and decreased from 2013 to 2017, consistent with the long-term trend of HCHO VCDs (Wang et al., 2017). Furthermore, the correlations between ground ozone concentrations and different ozone precursors at the AT and YF station were examined.

For the AT station, the Spearman correlation coefficient (Rs) between O_3_8 h and HCHO VCDs, NO_2 VCDs and HCHO VCDs/NO_2 VCDs was 0.64 (P = 0.00), −0.68 (P = 0.00) and 0.83 (P = 0.00) respectively. The Spearman correlation coefficient (Rs) between O_3_1 h and HCHO VCDs, NO_2 VCDs and HCHO VCDs/NO_2 VCDs was 0.68 (P = 0.00), −0.66 (P = 0.00) and 0.86 (P = 0.00) respectively. For the YF station, the Spearman correlation coefficient (Rs) between O_3_8 h and HCHO VCDs, NO_2 VCDs and HCHO VCDs/NO_2 VCDs was 0.56 (P = 0.00), −0.75 (P = 0.00) and 0.75 (P = 0.00) respectively. The Spearman correlation coefficient (Rs) between O_3_1 h and HCHO VCDs, NO_2 and HCHO VCDs/NO_2 VCDs was 0.58 (P = 0.00), −0.75 (P = 0.00) and 0.76 (P = 0.00) respectively. Despite different ozone formation regimes, the long-term ozone trend demonstrated strong and significant correlations with major ozone precursors NO_x and VOCs, OMI traceable NO_2 VCDs and HCHO VCDs at both the AT urban station and YF rural station. Specifically, HCHO VCDs/NO_2 VCDs demonstrated a stronger correlation with ground ozone concentrations than HCHO VCDs or NO_2 VCDs alone, especially for the AT urban station, indicating that HCHO VCDs/NO_2 VCDs was an effective indicator for monitoring long-term ground ozone concentrations at a large scale.

For the AT urban station, the notable decrease of NO_x emissions and slight decrease of VOCs emissions contributed to the higher ratio of HCHO/NO_2 in urban Beijing, causing an upward trend of HCHO/NO_2 and the ozone formation regime had changed from VOCs-limited to VOCs-NO_x-limited. For the YF rural station, a smaller decrease rate of NO_2 led to a downward trend of HCHO/NO_2, indicating the ozone formation regime in rural Beijing had changed from NO_x-limited to VOCs-NO_x-limited.

Given the indicative value of HCHO VCDs/NO_2 VCDs, the long-term variations of the distribution of HCHO VCDs/NO_2 VCDs across Beijing were further examined (Fig. 5). Areas with VOCs-limited regime were mainly concentrated in central Beijing whilst areas with NOx-limited regime mainly appeared in the northern part of Beijing. The concentrations of oxynitride were relatively high in urban areas due to intensive industrial and traffic emissions. On one hand, high-concentration NO reacts with ozone and leads to a reduced ozone concentration. On the other hand, high-concentration NO_2 reacts with ·OH to form HNO_3 and terminates the chain reaction of ·OH that produced ozone. Therefore, ground ozone concentrations in central Beijing might be lower than those in suburban areas, which was consistent with previous studies (Cheng et al., 2018). According to Fig. 5, areas with VOCs-NO_x-limited regime increased significantly and the distribution of different ozone regime areas became more and more fragmented. Therefore, considering the complicated ozone formation mechanisms subject to the proportion of NO_x and VOCs, specific emission-reduction measures that fully consider the local ozone formation regime should be accordingly designed to mitigate ozone pollution in Beijing and its surrounding areas.

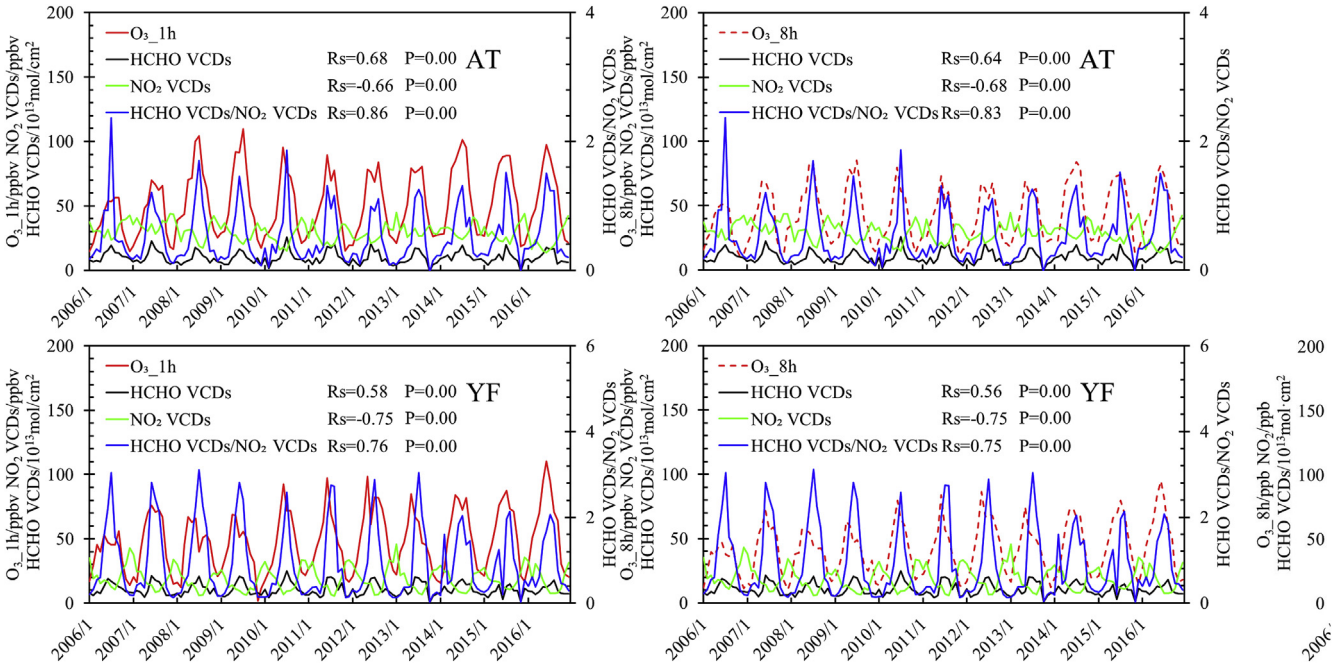

Fig. 4. Correlations between ground ozone concentration and HCHO VCDs and NO_2 VCDs, HCHO VCDs/NO_2 VCDs in Beijing from 2006 to 2017. Rs: The Spearman correlation coefficient. P: the significance of the correlation.

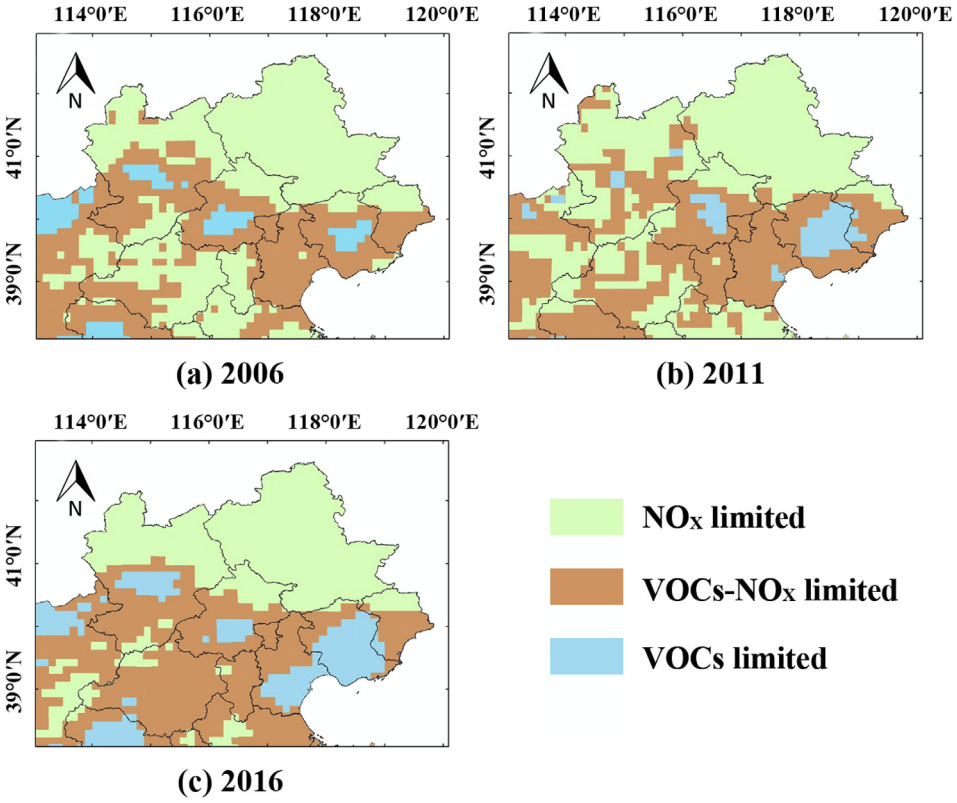

Fig. 5. Variations of ozone formation regimes across Beijing from 2006 to 2016.

3.3.2. Ozone formation regimes in Beijing during APEC period with emission-reduction measures

From November 1st to November 12th, 2014, Beijing implemented strict emission-reduction measures to improve local air quality during the Asia-Pacific Economic Cooperation (APEC) annual meeting. Since major meteorological factors in Beijing, including temperature, relative humidity, wind speed and surface pressure, ranged 6.9–11.6 °C, 43.2–77.7% 1.4–2.2 m s^{-1} and

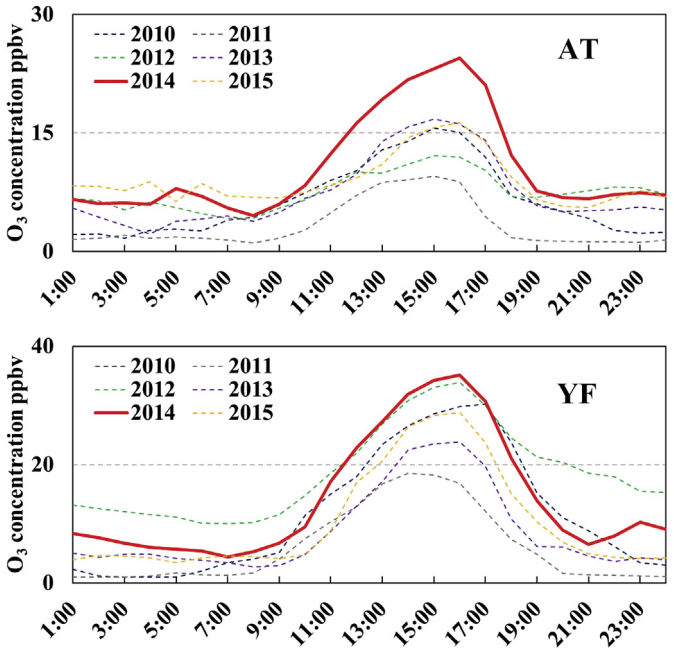

Fig. 6. Mean diurnal ozone concentrations in Beijing From November 1st to November 12th (2010–2015).

1015.1–1023.0 hPa, respectively, were generally consistent from November 1st to 12th during 2010–2015, the comparison between the ozone variation during the APEC period and that during the same period in other years (Fig. 6) provides useful reference for evaluating the performance of emission control measures (ECMs). According to Fig. 6, ECMs implemented during the APEC period, which targeted for reducing $PM_{2.5}$ concentrations, conversely significantly enhanced the diurnal peak ozone concentration, especially for the AT urban station with stricter ECMs. $HCHO/NO_2$ in Beijing rose from around 1.21 (VOCs-limited and VOCs-NO_x-limited) during the pre-APEC period to around 1.60 (VOCs-NO_x-limited and NO_x-limited) during the APEC period. With a series of ECMs, which exerted a much stronger influence on the reduction of NO_x than the reduction of VOCs, NO_2 VCDs decreased significantly, leading to a notable increase in $HCHO/NO_2$ and the ozone production rate during the APEC period. Previous studies (Wang et al., 2010; Witte et al., 2011) also revealed increased ozone concentrations in Beijing during periods with strict ECMs. During the 2008 Olympic ECMs period, Witte et al. (2011) found a transition from VOCs-limited to predominant NO_x-limited ozone formation regimes in Beijing. Wang et al. (2010) suggested that ground ozone concentrations in Beijing increased by 16% during the 2008 Olympic Games, due to the decreased NO-titration of ground ozone and regional transport of photochemically aged air. Given complicated ozone formation regimes, ozone-oriented ECMs that comprehensively consider the reduction of NO_x and VOCs are required for better managing ozone pollution in Beijing.

4. Discussion

Ma et al. (2016) revealed the relative contribution of meteorological conditions to notably increased ozone concentrations in Beijing was very limited through the KZ filter. Similar to Ma et al. (2016)'s findings, this research quantified that the relative contribution of meteorological factors to long-term ozone variations in Beijing was around 2%–3%. On the other hand, meteorological influences on long-term $PM_{2.5}$ variations are much stronger (Chen et al., 2016, 2017, 2018). Based on the KZ model, Chen et al. (2019a) quantified the relative contribution of meteorological conditions to long-term $PM_{2.5}$ variations in Beijing from 2013 to 2017 was around 20%. Meanwhile, the relative contribution of discomposed components to $PM_{2.5}$ variations was in the order of W(t) (66.8–83.8%) > S(t) (9–23.8%) >E(t)(1.2–3.5%), indicating that sudden change of meteorological conditions was the major meteorological driver for $PM_{2.5}$ pollution in Beijing. By comparison, the relative contribution of discomposed components to ozone variations was in the order of S(t) (48.54–61.28%) > W(t) (33.50–42.41%) >E(t) (0.92–2.99%), indicating that both seasonal and synoptic-scale variations of meteorological conditions exerted strong influences on O_3_1 h and O_3_8 h. Chen et al. (2019b) suggested that the comprehensive meteorological influences on ozone concentrations in Beijing were generally consistent from 2006 to 2016, and temperature was the dominant driver for ground ozone concentrations in Beijing. Lu et al. (2019) pointed out that the variation of anthropogenic emissions in 2017 solely would cause the decrease of ozone concentrations in Beijing in 2017, and the conversely soaring ozone pollution could be attributed to summertime high-temperature and low-humidity conditions induced increase of biogenic VOC emissions (Wang, H., 2018) and enhanced ozone production rate (Lu et al., 2019). In summary, despite the relatively small contribution of meteorological conditions to long-term ozone variations in Beijing, dramatic seasonal and sudden change of major meteorological conditions, especially temperature, may cause rapid variations of ground ozone concentrations in Beijing. Therefore, in addition to anthropogenic emission-reduction measures, summertime ozone pollution in Beijing may be mitigated through meteorological means. For instance, the forthcoming Beijing wind-corridor project, which aims for bringing in more strong northerly winds and reducing wintertime $PM_{2.5}$ concentrations, has the potential to reduce long-term summertime ozone pollution in Beijing by mitigating severe urban heat island effects. Meanwhile, since precipitation and high humidity are unfavorable conditions for ozone production (Chen et al., 2019b), artificial precipitation can be a contingent approach to reduce short-term ozone concentrations during pollution episodes.

Ma et al. (2016) indicated that the long-term VOCs variation was a main cause for increased ozone concentrations in Beijing. Cheng et al. (2018) found that NO_2/NO was a reliable indicator for ozone concentrations in Beijing. However, due to the rapid transmission between different oxynitrides, the concentration of NO_2 and NO can hardly be precisely measured. Similarly, although $NO_2/HCHO$ were proved to be closely related to ozone concentrations through small-scale experiments (Sillman, 1995; Duncan et al., 2010), long-term consistence between the trend of $NO_2/HCHO$ and ground ozone concentrations at a large (city) scale has rarely been tested. To fill this gap, the correlation between ground ozone and ozone precursors in Beijing from 2006 to 2017 was examined using OMI retrieved NO_2 VCDs and HCHO VCDs, and HCHO VCDs/NO_2 VCDs proved a better indicator than the sole use of NO_2 VCDs and HCHO VCDs for ground ozone concentrations. However, due to the large variation and uncertainty of ozone formation mechanisms, quantitative regression models between ground ozone concentrations and major ozone precursors NO_2 VCDs and HCHO VCDs suitable for large regions were limitedly investigated. To fill this gap, statistical analysis based on massive data sources, field experiments, smog-chamber -based experiments and model simulation should be comprehensively employed to establish robust regression models for better estimating and managing ground ozone concentrations using OMI traceable ozone precursors.

With the gradual decline of $PM_{2.5}$ concentrations and rapid increase of ozone pollution episodes in Beijing, the comprehensive management of $PM_{2.5}$ and ground ozone concentrations has

become an urgent environmental issue. Different from specific and effective emission-reduction measures for reducing $PM_{2.5}$ concentrations, no specific strategy has been implemented to intendedly mitigate high ozone concentrations. Cheng et al. (2018) further suggested NO_x-oriented emission-reduction measures might conversely increase ozone concentrations in Beijing. On the other hand, this research revealed that the distribution of ozone formation regimes in Beijing had became more and more heterogeneous, leading to additional difficulties in employing unified emission-reduction policies for effectively mitigating ozone pollution across Beijing. To better manage ozone pollution in Beijing, field surveys and laboratory experiments should be conducted to propose and implement context-related ozone-reduction strategies according to the distribution of ozone formation regimes in Beijing. For now, without a thorough understanding of mechanisms how different emission-reduction measures and ozone formation regimes may quantitatively influence ozone concentrations, an alternative strategy can be employed for ozone reduction in Beijing. According to long-term experience of ozone management in US, USEPA (2004, 2011) suggested that NO_x-oriented emission-reduction was more effective for ozone management at the regional scale whilst VOCs-oriented emission-reduction was more effective for reducing ozone concentrations at the local scale. This conclusion was highly consistent with other studies that indicated NO_x-oriented emission-reduction policies led to increased ozone concentrations in Beijing (Cheng et al., 2018; Lu et al., 2019). For effectively reducing ozone pollution in Beijing, more emphasis should be placed on the balanced reduction of both NO_x and VOCs emissions.

Although the long-term meteorological influences on and formation regimes of ground ozone concentrations in an urban and a suburban site were examined, the mechanisms how overall meteorological conditions and anthropogenic emissions work together to quantitatively influence ground ozone concentrations remain unclear. Given the complicated ozone formation mechanisms under different meteorological conditions and ozone precursor components, multiple models, such as chemical transport models as WRF-CAMx (http://www.camx.com/), statistical causality models as Convergent Cross Mapping (Sugihara et al., 2012) and Granger test (Granger, 1969) and field experiments should be properly integrated to better quantify the combined effects of meteorological conditions and anthropological emissions on ground ozone concentrations at a large scale.

5. Conclusions

Based on ground observation and remotely sensed data, long-term ozone variations at an AT urban and a YF rural station in Beijing from 2006 to 2017 were examined and the major findings and suggestions are concluded as follows:

1) The annually average O_3_8 h and O_3_1 h at the AT urban station increased from 31.28 to 38.16 ppbv in 2006 to 49.3 and 58.11 ppbv in 2017 whilst the annually average O_3_8 h and O_3_1 h at the YF rural station increased from 31.63 to 39.14 ppbv in 2006 to 43.67 and 47.78 ppbv in 2017, indicating a significant upward trend of ozone concentrations across Beijing from 2006 to 2017.
2) From the meteorological perspective, the relative contribution of meteorological conditions to long-term ozone variations at both stations were around 2%–3%, suggesting anthropogenic emissions were the main driver for the long-term ozone variations in Beijing. Meanwhile, short-term ozone concentrations were influenced significantly by seasonal and synoptic variations of meteorological conditions. Therefore, long-term meteorological means such as wind-corridor projects that reduce summertime high-temperature and contingent meteorological means such as artificial precipitation that increases ambient humidity have the potential to mitigate soaring ozone pollution in Beijing.
3) From the perspective of ozone precursors, HCHO VCDs/NO_2 VCDs proved a better indicator than the sole use of NO_2 VCDs and HCHO VCDs for ground ozone concentrations. The ozone formation regimes for the AT and YF station changed from VOCs-limited and NO_x-limited to VOCs-NO_x-limited respectively. Given the increasingly heterogeneous distribution of different ozone formation regimes across Beijing, emission-reduction strategies that balanced consider the reduction of VOCs and NO_x emissions should be better designed and implemented according to local ozone formation regimes.

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