Characteristics of Ground Ozone Concentration over Beijing from 2004 to 2015: Trends, Transport, and Effects of Reductions

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Abstract: Based on the hourly averaged ozone monitoring data during 2004–2015 in urban area and at DL background station in Beijing, a comprehensive discussion of the characteristics of ozone concentration was conducted. Annual averaged concentration of daily maximum 1 h ozone (O_3 1h) was all increasing at urban sites (1.79 ppbv yr^{-1}) and DL background station (2.05 ppbv yr^{-1}) while daily maximum 8 h average ozone concentration (O_3 8h) was increasing in urban area (1.14 ppbv yr^{-1}) and slightly decreasing at DL background station (-0.47 ppbv yr^{-1}) from 2004 to 2015 due to different ozone sensitivity regimes and ratios of NO_2/NO. Diurnal variations of ozone peaks obtained at downwind DL station were about 1 hour later than that in urban area from May to October in different years and concentrations of ozone at a DL background station were much higher than those at urban sites. Moreover, the differences of ozone peaks between urban sites and DL background station were becoming significantly smaller in recent years, which may be related to the regional ozone transport and the expansion urbanization of Beijing. Based on the joint efforts of regional air pollution prevention and control, Beijing achieved the 2015 Grand Military Parade blue. Calculated average concentrations of CO, NO_2, and O_3 in S2 (August 20~31,2015) and S3 (September 01~03,2015) decreased by 31.48%, 43.97%, and 13.21% at urban sites, and by 20.93%, 57.10%, and 23.62% at DL station, respectively compared with those in S1 (August 01~19,2015) and S4 (September 04~30,2015). A reduction of local anthropogenic emissions such as VOCs and NO_x could decrease ozone efficiently especially in downwind areas of Beijing and made the ozone peaks appear 2~3hours earlier compared to the scenarios of no emission reductions. Compared to the increasing ozone during APEC period, average ozone concentration decreased significantly in the downwind Beijing due to larger ratios of VOCs/NO_x. In order to decrease the ozone concentrations in Beijing, VOCs

emissions should be reduced larger and be controlled stricter than those of NO_x in Beijing and the policy of regional air pollution joint prevention and control should still be promoted unswervingly and jointly.

Key words: $O_3$8h; trend; Beijing; regional transport; reductions

1. Introduction

Ground-level ozone, one of the most important secondary air pollutants in the atmosphere, is generated through photochemical reactions between nitrogen oxides (NO_x) and volatile organic compounds (VOCs) (Trainer et al., 2000; Sillman, 1999). High concentrations of ozone near the ground are harmful to human health, ecosystems, and global climate (Fiore et al., 2009).

In recent years, elevated regional ozone concentration and atmospheric oxidation capacity in China have attracted increasing attentions (Lin et al., 2008; Zhang et al., 2007). Numerous studies have analyzed the variations of ozone and its photochemical reactions with its precursors based on the measurements over a short period or satellite data (Chan et al., 2003; Wang et al., 2012; Vingarzan, 2004). Most studies in China were mainly concentrated in city cluster regions, such as Pearl River delta (Li et al., 2011;Wei et al., 2012; Zhang et al., 2013), Yangtze River delta (Li et al., 2014; Ding et al., 2013; Ran et al., 2009), and Beijing–Tianjin–Hebei regions(BTH) (Tang et al., 2009;Shao et al., 2009; Lu et al., 2010). These studies focused on the chemical characteristics of ozone, with few discussions on the variation trends,ozone transport and its influencing factors especially by regional reduction measures within a long period for the lack of observed data in Beijing (An et al., 2006; Chou et al., 2009; Yuan et al., 2009) and other limiting factors.

Different to the continuously decreasing ground ozone concentrations in urban sites in the US (Pollack et al., 2013), recent limited studies performed in China, particularly in BTH area, suggested that ozone concentrations in both regional background and urban areas are increasing (Meng et al., 2009; Wang et al., 2008) due to large NO_x emissions. Few long-term studies about the trends of ground-level ozone in Beijing were discussed (Lu et al., 2010),let alone analyze the trends of daily maximum 8 h averaged ozone concentration(O_3 8h) and daily maximum 1 h ozone concentration(O_3 1h) and the effects of urbanization and regional emission reduction measures on ozone concentrations. After the implementation of the new standard of "Ambient Air Quality Standard" (MEP, 2013) in 2013, the levels of O_3 1h and O_3 8h have a direct impact to the ranks of the air quality in Beijing. Furthermore, the increasing ozone pollution in Beijing have obtained much public concerns from Beijing Municipal Government and the whole society (Ding et al., 2013; Wang et al., 2013) especially in Summer. The executive meeting of the State Council examined and adopted ‘The Control Measures of Beijing Air Pollution during 2012–2020” (http://zhengwu.beijing.gov.cn/gzdt/gggs/t1225355.htm). According to the regulation, the non-attainment hours of ozone in Beijing will decrease by 30% than that in 2010 and should

be controlled at about 200 hours annually. Therefore, the results of previous studies were far from the current needs.

Air quality security programs were implemented from August 20 to September 3 in 2015 to guarantee the air quality for the parade on the 70th Victory Memorial Day for the Chinese People's War of Resistance against Japanese Aggression (the 2015 Grand Military Parade). Chinese government established numerous emission reduction measures, such as reducing coals, industrial adjustment, joint prevention measures, and limitation of vehicles (particularly heavy-duty buses and trucks from outside Beijing, and odd-even license plate policy on roads within urban Beijing). As regional emission reduction measures can not be copied and costs a lot of manpower and material resources, it offers a precious opportunity to study the changes in ozone and its precursors during the period of air quality assurance.

This paper aims to investigate the temporal trends of $O_3$1h and $O_3$8h in different sites in Beijing and verify the importance of ozone transport. Also, we evaluated the changes on ozone concentration after the reduction measures during the 2015 Grand Military Parade in 2015.

2 Materials and methods

2.1 Site distribution

Beijing is located at 115.7 °–117.4 °E, 39.4 °–41.6 °N. This area is at the northwest edge of the North China Plain and close to the edge of the semi-desert zone. Its terrain exhibits a dustpan shape, and it is surrounded by mountains in three directions. The average altitude of Beijing is 43.5 m, and the general altitude of mountains is in the range of 1 000–1 500 m, which is not conducive to air pollutants diffusion. The total area of Beijing is 16410.54 km^2, in which 62% are mountains. Its total forest coverage in the plain region is about 15%, which is lower than that in whole city (38%). Beijing exhibits a temperate continental monsoon climate, where it is hot and rainy in summer and cold and dry in winter. Over the past decade, the annual averaged rainfall is less than 450 mm, 80% of which is concentrated in June, July, and August (BJEPB,2014; Beijing Statistics Bureau, 2014).

As the capital of China, the air quality monitoring network in Beijing is more advanced than other regions of China (BJEPB, 2014). In 2001, an air quality monitoring network that obtains 35 stations was established by the Beijing Municipal Environmental Monitoring Center (BJMEMC, http://zx.bjmemc.com.cn/, **Fig. 1**). The 35 stations cover all districts that contain different environment types defined by regional background, such as suburbs, city, and residential. Twelve monitoring sites (DL, DS, GY, TT, WSXG, AT, NZG, WL, GC, SY,

CP, HR) in urban area and one background station DL were selected in Beijing and used in this study. DL station (116.22 °E, 40.29 °N, about 45 km northwest of Tiananmen square) is the background station of World Meteorological Organization Environmental Monitoring center in China and has conducted air pollutant monitoring work for decades.Meteorological sounding data in Beijing at Guanxiangtai station (GXT,54511, 116.46 ° E, 39.80 ° N) were downloaded from the Department of Atmospheric Science, College of Engineering, University of Wyoming (http://weather.uwyo.edu/upperair/sounding.html).

In this study,we retrieved NO_2 and HCHO VCDs from the OMI products in urban Beijing (GY site, 116.33 ° E, 39.93 ° N) and combined the corresponding ratios to analyze the chemical sensitivity of PO_3 (ozone production rate) during both Parade and APEC periods.

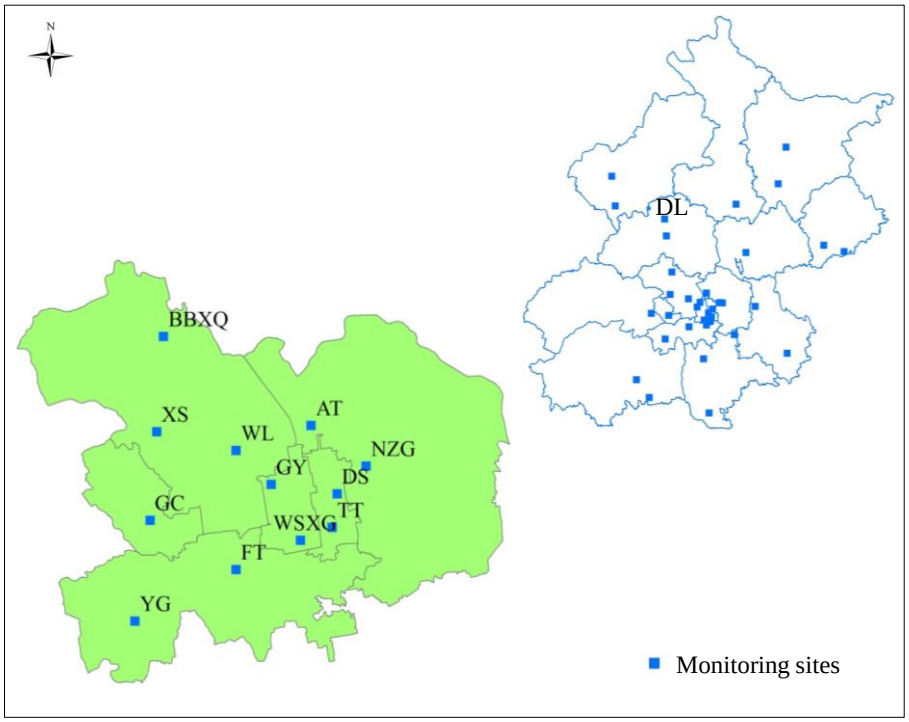


Fig. 1 Distribution and classification of observation sites in Beijing

2.2 Monitoring instruments

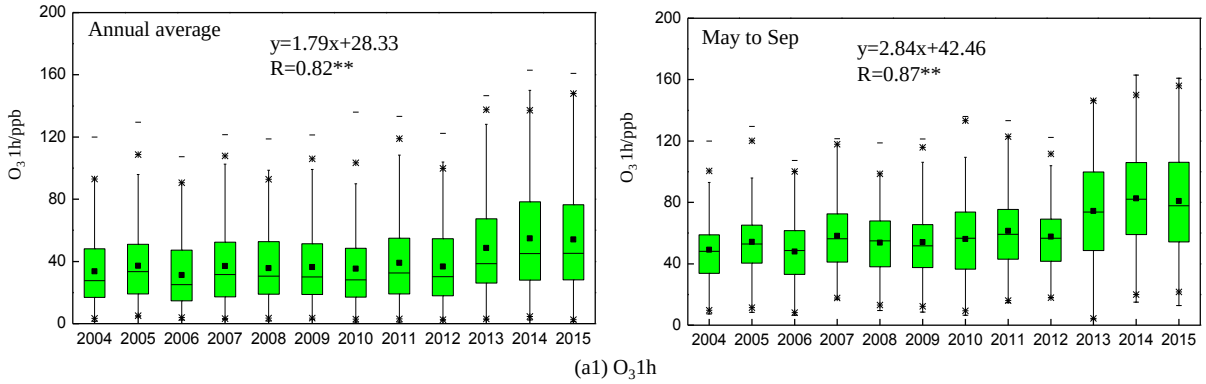
Ozone are mornitored by the 49C ozone analyzer instruments produced by Thermo Fisher Corporation (USA). The minimum limit of ozone analyzer instrument is 1×10^{-9}, and the zero and cross drifts are 0.4%/24 h and ±1% /24 h, respectively. An ozone calibrator (49IPS) traceable to the Standard Reference Photometer maintained by the WMO World Calibration Center was used to calibrate the ozone analyzers. Ozone monitoring instrument at each station had a zero cross calibration every three days, precision audit every three month, and an accuracy check every six months to ensure the accuracy of ozone monitoring in Beijing. Thermo Fisher 42C NO–NO_2–NO_x analyzer was used to monitor NO and NO_2

concentrations with a limit of 0.05×10^{-9}, zero drift of 0.025×10^{-9}/24 h, and span drift of ±1%/24h. Operation procedure strictly followed the "The Specification of Environmental Air Quality Automatic Monitoring Technology" (HJ/T193-2005, http://kjs.mep.gov.cn/hjbhbz/bzwb/dqhjbh/jcgfffbz/200601/t20060101_71675.htm), and the equipments were regularly calibrated and maintained by technicians.

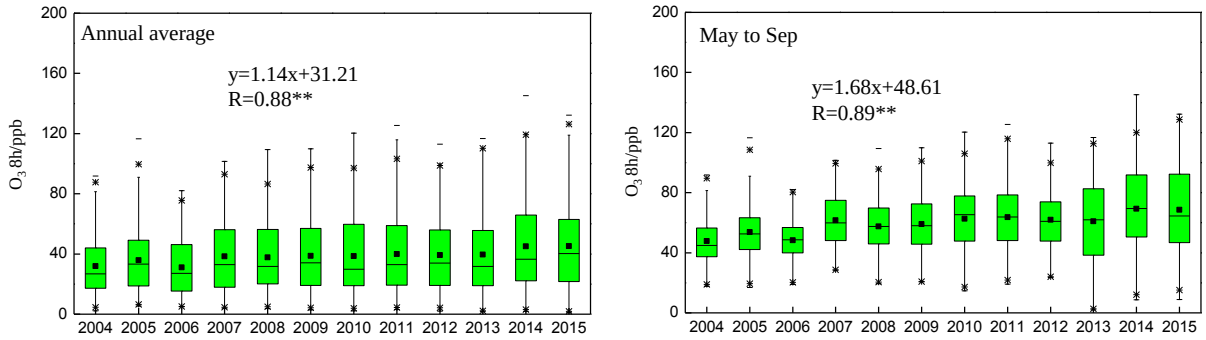
3 Results and Discussion

3.1 Variations trends

In Beijing, the nonattainment days of ozone is mainly concentrated in the months of May to September and the comparisons of trend analysis between nonattainment days and reaching standard days were very meaningful in some mega cities especially Beijing (http://www.bjepb.gov.cn/bjepb/323474/324034/324735/index.html). A simple linear regression and statistical tests, such as Pearson's correlation analysis were implemented to investigate the trends of $O_3$1h and $O_3$8h in urban area and at DL station in Beijing (**Fig. 2**), for the inter-annual variations of ozone concentrations in urban Beijing, $O_3$1h was in an evident upward trend with an annual averaged concentration growth rate (AAGR) of 1.79 ppbv yr^{-1} (correlation coefficient R=0.82,highly correlated) and an higher increase of 2.84 ppbv yr^{-1} during May to September (MSAGR,R=0.87,highly correlated) from 2004 to 2015.Variation of $O_3$8h was in an overall upward trend with AAGR of 1.14 ppbv yr^{-1} (R=0.88,highly correlated) and MSAGR of 1.68 ppbv yr^{-1} (R=0.85,highly correlated) during May to September, respectively from 2004 to 2015. For the variations of ozone concentration at DL background station, $O_3$1h was in an overall upward trend with AAGR of 2.05 ppbv yr^{-1} (R=0.81, highly correlated) and MSAGR of 0.14 ppbv yr^{-1} (R=0.10, micro relevant), whereas $O_3$8h was in a slightly downward trend (AAGR=−0.47 ppbv yr^{-1}, R=−0.42,weak correlation,real relevant; MSAGR=−0.70 ppbv yr^{-1}, R=−0.40, real relevant).

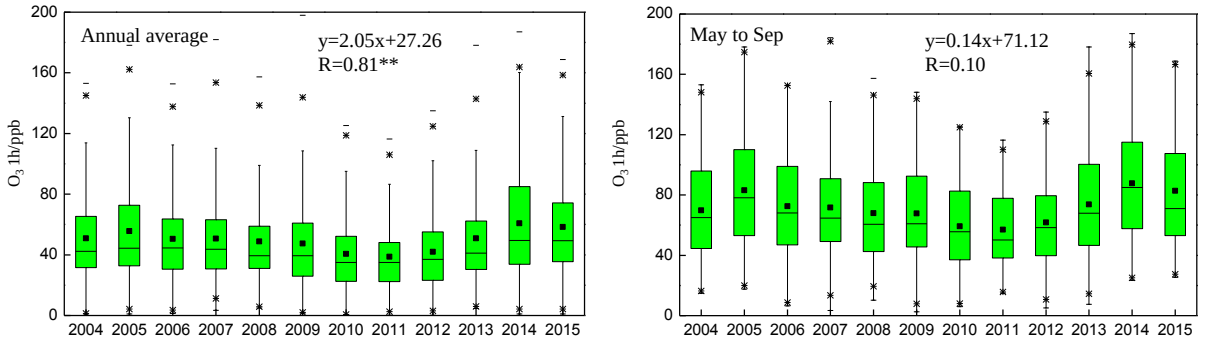


(a1) $O_3$1h

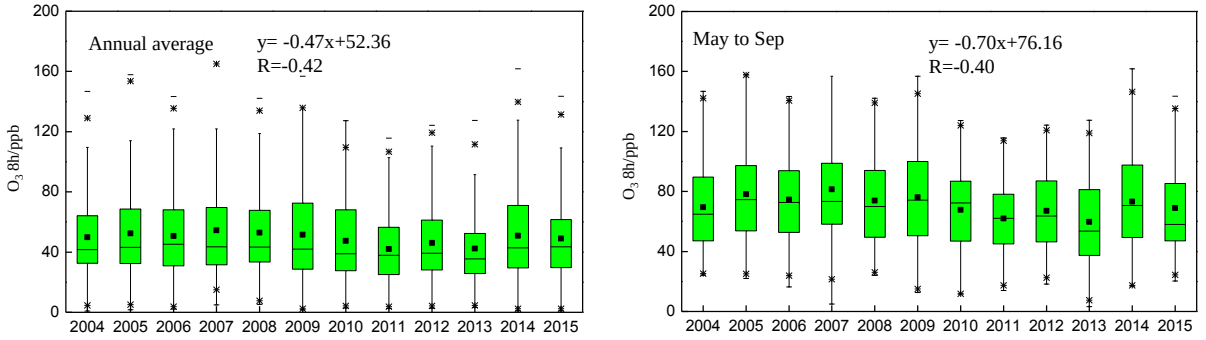


(a2) $O_3$8h

(a) Urban



(b1) $O_3$1h



(b2) $O_3$8h

(b) DL

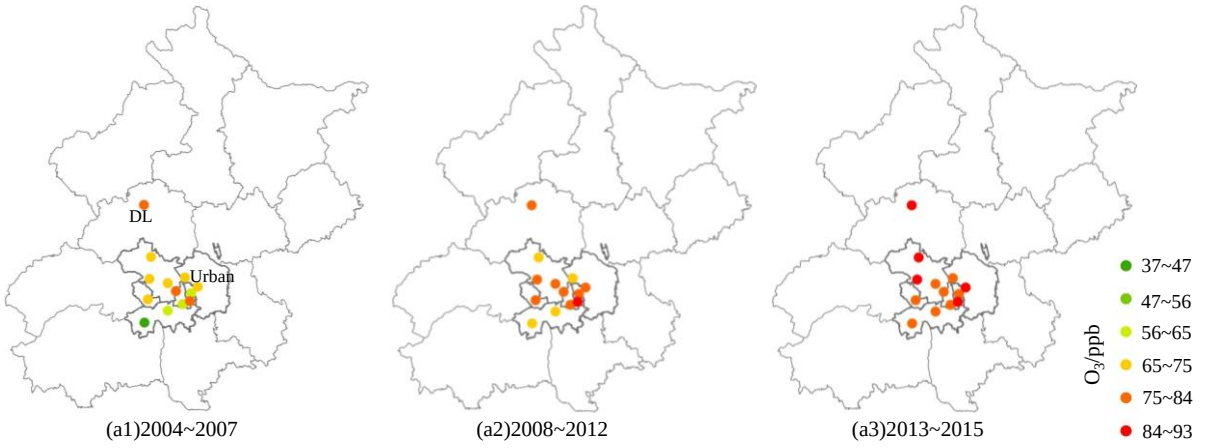
Fig. 2 Variation trends of $O_3$1h and $O_3$8h in urban Beijing (a1, a2) and at DL background station (b1, b2) from 2004 to 2015 (including annual trends and trends from May to September) and the linear fitting equations(95% confidence interval, two-tailed ,**highly relevant)

$$r = \frac{\sum (X - \overline{X})(Y - \overline{Y})}{\sqrt{\sum (X - \overline{X})^2 \sum (Y - \overline{Y})^2}} \quad (R1)$$

(r is the pearson correlation coefficient; X is the ozone concentration;Y presents the year;r between 0 and 0.3 representing the micro relevant, r between 0.3 and 0.5 representing the real relevant, r between 0.5 and 0.8 representing the significant relevant,and r between 0.8 and 1.0 representing highly relevant)

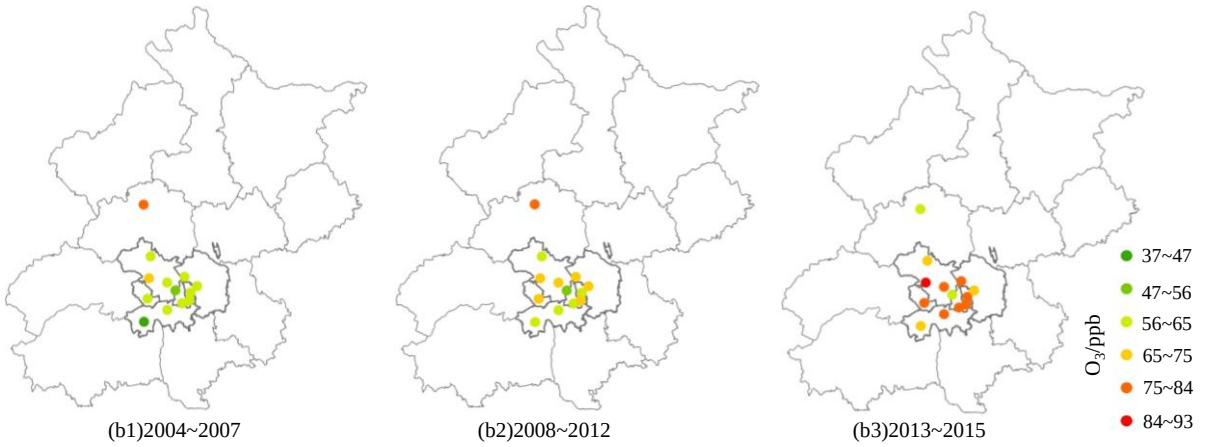
The Mann–Kendall trend detection test(Kendall, 1975) has been commonly applied to assess the significance of monotonic trends in ozone pollution data time series and we found the year 2013 was an intercept break point. In 2008, Beijing held the Olympic Games and the ozone in Beijing decreased significantly.Thegood air quality promote the awareness of environmental protection to take various common emission reduction mearures in the next few years. In

2013,the state implemented a new environmental air quality standards, marking the work of environmental protection into a new stage. We divided the available the year of 2004-2015 of zone data into three parts: 2004-2007, 2008-2012, and 2013-2015. For the variations of $O_3$1h and $O_3$8h concentration in different periods at twelve sites in urban Beijing and DL background site (**Fig.3**), concentrations of $O_3$1h and $O_3$8h were all significantly increased during the period of 2013–2015 compared to those during the periods of 2004–2007 and 2008–2012. Average concentration of $O_3$1h during the period of 2013–2015 increased 3.71%~40.29% at urban sites in Beijing compared to that during the period of 2004~2012 while average concentration of $O_3$8h during the period of 2013–2015 increased 9.51%~62.58% at urban sites in Beijing compared to that during the period of 2004~2012. Average concentration of $O_3$1h during the period of 2013–2015 exceeded the standard about 77.53%~104.55% at urban sites in Beijing while it was 33.09%~92.32% for $O_3$8h. Therefore,the rising ozone concentration after the implementation of the new standards highlighted the terrible ozone pollution situation in recent years in Beijing.



(a1)2004~2007 (a2)2008~2012 (a3)2013~2015

(a)O_3 1h



(b1)2004~2007 (b2)2008~2012 (b3)2013~2015

(b)O_3 8h

Fig. 3 Average concentrations of $O_3$1h and $O_3$8h at urban sites and DL background site from May to September during the periods of 2004–2007, 2008–2012, and 2013–2015 in Beijing

From the frequency distributions (proportion rates in different ozone concentration intervals) of $O_3$1 h and O_3 8h in Beijing (**Fig. 4**), frequency of O_3 8h less than 10 ppbv increased significantly during the period of 2013 to 2015 compared with that during period of 2008–2012 and frequency of ozone concentration higher than 80 ppbv also became larger both at DL background station and urban sites. For O_3 1h,frequency of O_3 1h higher than 80 ppbv became significantly larger at both DL background station and urban sites during the period of 2013 to 2015 compared with that during period of 2008–2012 which indicated increasing frequencies of high ozone concentration caused the significant increase of $O_3$1h in Beijing in recent years; whereas the low values of $O_3$1h did not increased as $O_3$8h. This phenomenon was basically consistent with a few recent studies (Tang et al., 2009; Jonson et al., 2006; Xu et al., 2008) and could explain the increasing high ozone concentrations in Beijing due to the increasing frequency of higher ozone concentration at both background station and urban sites. Parrish et al. (2014) found that ozone volume fraction increases at an annual growth of 0.2–0.3 ppbv yr^{-1} in the Northern Hemisphere. Also, Meng et al. (2009) observed that ozone volume fraction increases at a rate of 1.0 ppbv yr^{-1} at the background station in Shanghai, China. In addition, Tang et al. (2009) found that ozone volume fraction increases at a rate of 1.1 $\pm$0.5 ppbv yr^{-1} during 2001–2006 in Beijing.

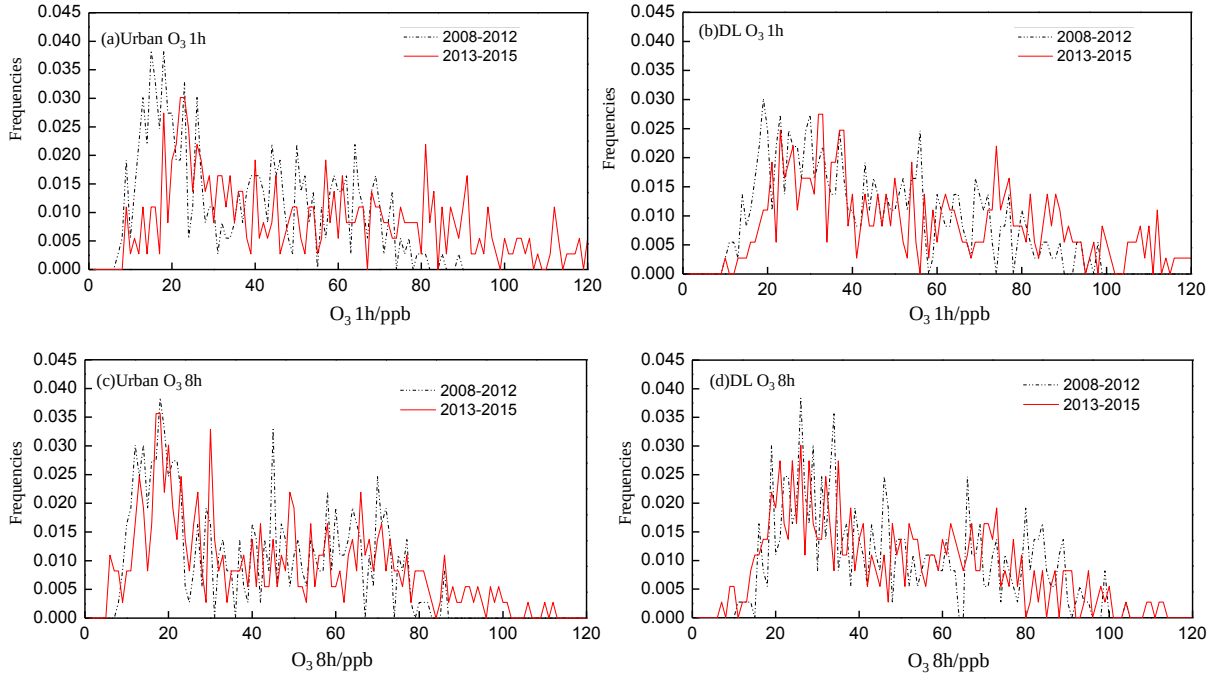


Fig. 4 Frequency distribution of the ozone concentrations($O_3$1h and $O_3$8h) at DL and urbanBeijing for the periods of 2008–2012 and 2013–2015

During 2008–2015, ratios between NO_2 and NO (lack of nitrogen oxide observed data during 2004–2007, **Table 1**) increased significantly with AAGR of 0.46 (R=0.85, highly relevant,95% confidence interval, two-tailed) and 1.87 (R=0.93,highly relevant,95% confidence interval, two-tailed) at urban sites and DL background station respectively, indicating ratios between NO_2 and NO were increasing larger at background station than urban sites in Beijing. Hourly averaged concentrations of NO_x were in the range of 29.21–39.76 ppbv,11.85–13.91 ppbv at urban sites and DL background station, respectively. NO_x concentration was in a downward trend with AAGR of −0.43 ppbv yr^{-1} (R=0.32, real relevant,95% confidence interval, two-tailed) and −0.21 ppbv yr^{-1} (R=0.24, micro relevant,95% confidence interval, two-tailed) at urban and DL background sites, respectively. This study could not further analyze the formation mechanism of ozone due to lack of observed data for VOCs during 2008–2015, but research indicated the VOCs concentration also declines in recent year in Beijing (Lu et al., 2010; Wang et al., 2015a). Wang et al.(2015a) found mixing ratios of NMHCs measured at PKU university site decreased by 37% during August increased by 28% from 2004 to 2012 and the measured NMHC/NO_x ratios declined by 14% during August from 2005 to 2012.Since,Non-methane hydrocarbons (NMHCs) accounts for the vast majority of VOCs and plays a critical role in the photochemical production of ozone, variations of VOCs should be further investigated for a thorough understanding of ozone trends in the future.

Ozone precursors (VOCs and NO_x) were decreasing, whereas average concentrations of $O_3$8h were still increasing in urban Beijing, which may be caused by the particular sensitivity regimes and other related factors. Although previous studies indicated the distribution of the sensitivity regimes of $O_3$8h concentration was similar to that of $O_3$1h concentration (Zhang et al., 2008), this distribution was a little different in urban Beijing (Shao et al., 2006). The important reason was that the distribution of ozone sensitivity regimes is closely related to meteorological condition and emission distribution (Zhang et al., 2008; Sillman, 1999). Generally,ozone reactions are mainly VOCs-sensitive and NO_x-sensitive in urban Beijing and suburban areas or more remote areas of Beijing, respectively (Tang et al., 2009). In urban Beijing, a reduction of anthropogenic NO_x could increase local ozone efficiently while a reduction of anthropogenic NO_x in urban and suburban areas could reduce ozone efficiently in downwind suburban areas. Except for different regimes,the other factors caused by the increasing ozone concentration may be related to a significant increase in regional tropospheric NO_x concentrations, particularly in BTH area (Richter et al., 2005; Van der A et al., 2006), or high concentrations of the regional zone and its precursors transport (Parrish et

al., 2014). Also,the rapid growth of population and industrialization have driven substantial increases in ozone background concentrations in BTH area (Willem et al., 2015). Restricted by P(Ox=O_3+NO_2) calculation and observation,we fail to present comprehensive explanations to compare the characteristics of O_3 production between local Beijing and DL site. But according to the analysis (Zhang et al.,2014), VOCs and NOx both decreased between 2006 and 2011 and the decrease in VOCs reactivity ($-5\%yr^{-1}$) was slightly larger than the decrease in NOx ($-4\%yr^{-1}$), leading to a slight decrease in P(Ox). The sunshine hours and visibility are also the important factors influencing O_3 production. Hence, variations of P(Ox) need to be further investigated for a better understanding of ozone trends.

Table 1 Statistics of NO_2/NO in urban areas and at DL site during 2008–2015 in Beijing

Parameter		2008	2009	2010	2011	2012	2013	2014	2015
NO_x /ppbv	Urban sites	32.93±23.45	36.52±23.52	39.76±38.32	37.42±23.17	35.01±21.42	37.71±23.85	36.26±21.89	29.21±18.83
	DL site	13.91±11.95	16.30±14.17	15.12±9.33	17.95±14.59	16.35±14.45	17.66±16.72	14.42±11.95	11.85±10.74
NO_2/NO	Urban sites	3.61±2.46	2.87±1.87	3.01±2.13	3.33±2.62	3.74±3.32	4.05±3.83	5.07±4.96	5.13±4.30
	DL site	4.89±3.01	3.68±2.24	4.27±2.15	4.75±2.17	8.12±6.67	8.41±5.48	10.83±6.88	12.18±10.97

3.2 Diurnal variations and regional transport

Fig. 5 was the diurnal variations of ozone in urban Beijing and at DL station from May to September during 2004–2015 and **Table 2** presented the statistical ozone peaks at DL station and urban sites from May to September during 2004–2015. In general, ozone concentration at DL station was higher than that of urban sites, and ozone peaks at DL station from May to September in different years was 1.01–1.56 times that of urban sites. For the spatial distribution of ozone,it was lower in central urban area and relatively higher in the northern and western area with good vegetation. Ozone has a lifespan of several days; consequently, high ozone concentrations can be found in regions distant from precursor emission sources (Seinfeld 2004; Kalabokas *et al.*, 2000), and several chemical ozone destruction reactions existing in urban center, such as R2–R3, are absent in background areas (Saitanis 2003; Pablo *et al.*, 2013). In Beijing, NO_x concentration in the urban center of the city was typically higher because of the large amount of vehicle population, which consumed and titrated a certain amount of ozone. The ozone peaks at DL station from May to September in different years was obviously 1 hour behind than that of urban sites, which was closely related to the regional ozone transport (R4). Most of the ozone was generated during the transport of its precursors

from emission districts to surroundings or background sites. In summer, high temperature, strong solar radiation, low humidity, and small southwest wind in Beijing strengthen photochemical pollution; moreover, ozone and its precursors, such as NO_x, CO, and VOCs, are transported to the downwind area, hence the reduced ozone peak concentration in downwind area (Carnero et al., 2010; Shan et al., 2010).

In addition,the differences of ozone peaks between DL station and urban sites were significantly decreased from 18.20 ppbv to 2.72 ppbv during 2004–2010 and 2011–2015. This change may be related to the expansion of urbanization of Beijing. With city expansion and economic development, the district near DL station was urbanized and easily influenced by anthropogenic emissions. Santini et al.(2010) found Beijing urban extent estimated from Landsat data was from 1105km^2 to 4139km^2 between 2000 and 2009. Jacobson et al. (2015) pointed that urbanization decreases the concentrations of many surface chemicals due to their vertical dilution but increases near-surface ozone.

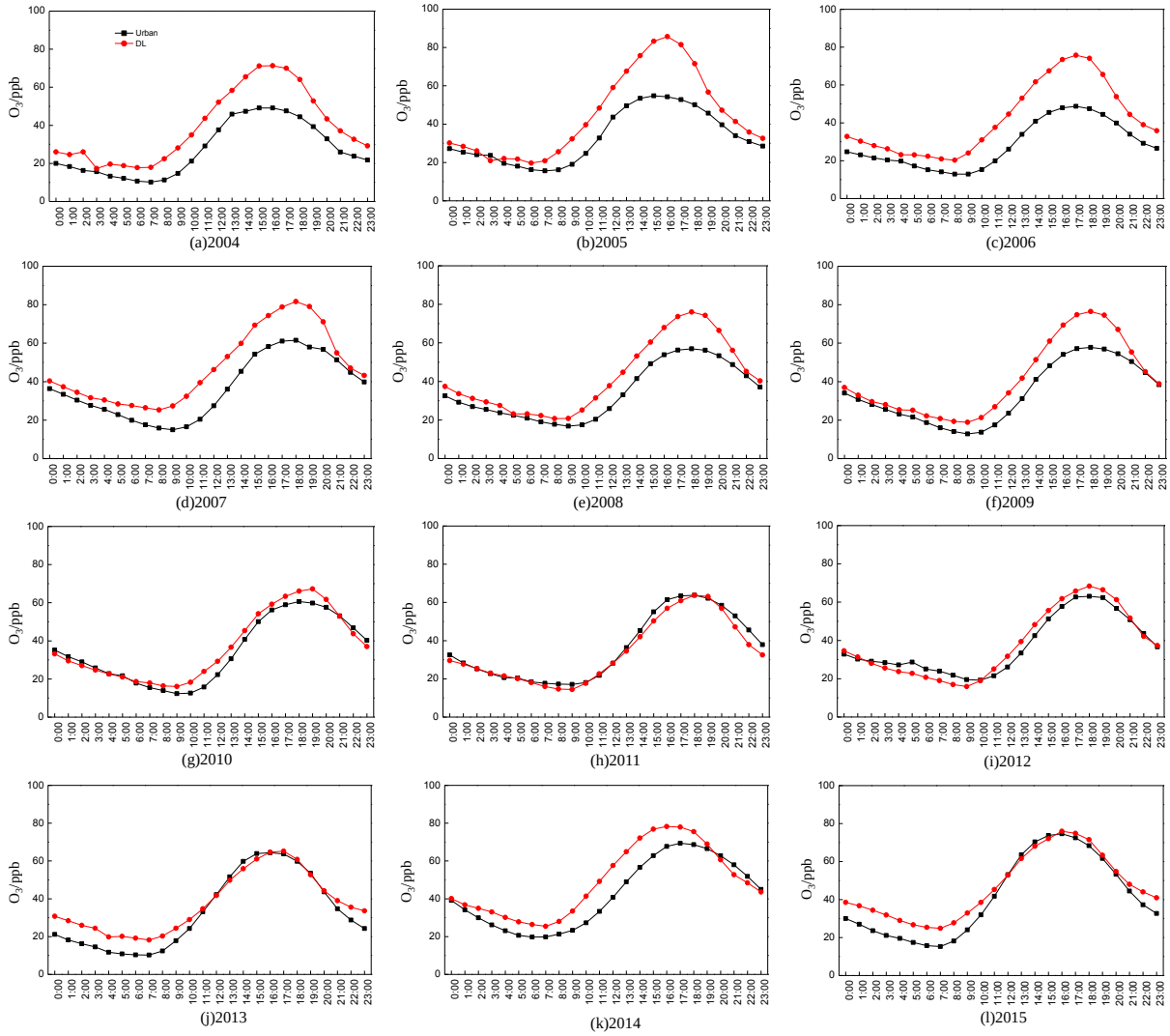


Fig. 5 Diurnal variations of ozone in urban areas and at DL station from May to September during 2004–2015 in Beijing

Table 2 Peak ozone concentration from May to September during 2004−2015 at DL station compared with the urban city of Beijing

Parameter	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2004~2015
delay hour/h	1	1	0	0	0	0	1	1	0	1	0	1	0
concentration difference /ppbv	22.18	30.95	27.00	20.11	19.09	18.67	6.64	0.33	5.28	0.78	8.83	0.24	12.38

O_3+OH→HO_2+ O_2(R2)

O_3+NO→NO_2 (R3)

NO_x+VOCs→O_3 (R4)

3.3 Emission reductions on ozone concentrations

Prevention and control of air pollution in BTH region and its surrounding areas, such as Shandong,Shanxi,and Inner Mongolia,effectively reduced the air pollutants' emission intensity caused by "human activities" during the parade on the 70th Victory Memorial Day for the Chinese People's War of Resistance against Japanese Aggression. Local enhanced reduction measures in Beijing were implemented firstly in S2 (August 20~31,2015) stage, and regional enhanced reduction measures in Beijing and its surrounding areas were implemented subsequently in S3 (September 01~03,2015) stage. Calculated average concentrations of CO, NO_2, and O_3 in S2 and S3 stages decreased by 31.48%, 43.97%, and 13.21% at urban sites, and by 20.93%, 57.10%, and 23.62% at DL site, respectively (**Table 3; Fig. 6**) compared with those in S1 (August 01~19,2015) and S4 (September 04~30,2015) stages. After the implementation of enhanced reduction measures in the surrounding area in S3 stage, average concentrations of NO_2 and O_3 at the urban sites decreased by 3.95% and 8.05%, respectively compared with those in S2 stage, and average concentrations of CO in S3 were close to those in S2. Overall, the enhanced reduction measures decreased most air pollutants in Beijing significantly and effectively.

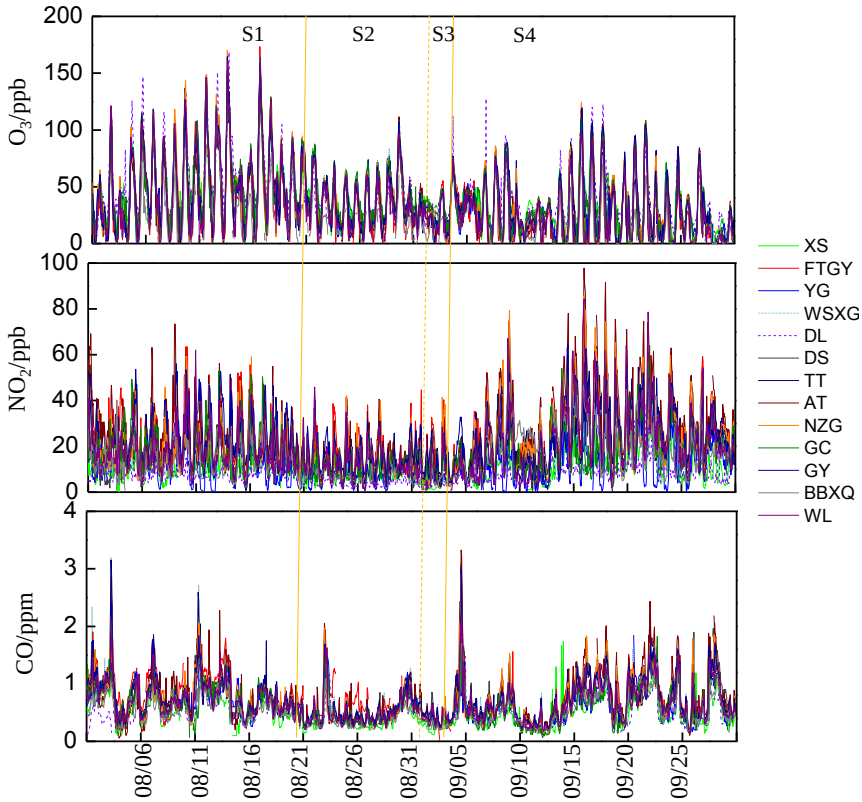


Fig. 6 Concentrations of main air pollutants in urban Beijing from August 01to September 30 in 2015.

Table 3 Average concentrations of main air pollutants at urban sites at four stages in Beijing in 2015

Pollutants	Stage	XS	FTGY	YG	WSXG	DS	TT	AT	NZG	GC	GY	BBXQ	WL	DL
O_3/ppb	S1	52.96	50.47	48.86	51.41	46.89	48.37	50.17	53.21	52.23	50.88	34.26	51.33	56.37
	S2	38.02	34.67	38.89	39.73	35.14	36.14	37.78	39.22	42.64	36.92	17.66	32.97	33.55
	S3	36.59	34.61	33.72	34.38	31.36	33.23	35.54	34.43	37.46	33.97	13.40	30.47	36.93
	S2~S3	37.31	34.64	36.31	37.05	33.25	34.68	36.66	36.83	40.05	35.44	15.53	31.72	35.24
	S4	33.66	31.82	28.95	28.09	27.49	25.23	32.30	29.77	28.69	31.97	16.78	26.69	35.90
NO_2/ppb	S1	9.89	25.72	12.17	19.60	17.95	19.72	24.95	19.42	19.04	24.64	15.00	18.32	8.63
	S2	6.02	16.38	6.59	13.20	12.89	14.87	14.98	13.16	10.53	15.17	7.55	12.90	3.71
	S3	5.78	16.45	7.67	12.98	12.69	13.85	13.60	12.33	11.50	14.42	7.16	9.70	3.97
	S2~S3	5.90	16.42	7.13	13.09	12.79	14.36	14.29	12.74	11.01	14.79	7.35	11.30	3.84
	S4	11.68	27.32	14.35	22.42	25.06	25.01	31.52	26.26	20.27	23.90	21.34	24.27	9.26
CO/ppm	S1	0.58	0.98	0.73	0.85	0.78	0.80	0.79	0.78	0.74	0.82	0.57	0.75	0.54
	S2	0.35	0.73	0.47	0.50	0.45	0.46	0.49	0.45	0.45	0.49	0.34	0.42	0.38
	S3	0.36	0.63	0.52	0.58	0.55	0.56	0.57	0.51	0.49	0.56	0.38	0.50	0.44
	S2~S3	0.36	0.68	0.50	0.54	0.50	0.51	0.53	0.48	0.47	0.52	0.36	0.46	0.41
	S4	0.50	0.74	0.61	0.78	0.76	0.74	0.82	0.74	0.58	0.74	0.57	0.68	0.50

In order to eliminate the influence of meteorological factors, we counted the observed variations of meteorological elements from August 20 to September 05 at GXT station in Beijing between 2010 and 2015 (We only collected meteorological data for the past five years). From **Table 4,**the temperature and wind speed at ground which can affect ozone concentration directly changed slightly during the study periods. Average temperature was

fluctuating between 23.6~24.3℃ and it was was lowest in 2011 and highest in 2013. Average wind speeds was fluctuating between 1.4~1.9 m s^{-1},suggesting the atmosphere was generally stable in August in Beijing. Average relative humidity and suface pressure was also possessing the same characteristics. Frequency of the north wind at 850hPa directly affects vertical diffusion of ozone. Average frequency of the north wind at 850hPa was between 12.1% and 41.2% and it changed to 24.3% from August 20 to September 05,2015 indicating the atmosphere was also relatively stable in the vertical direction. It is reported that both sunshine hours and visibility in BTH region have been decreasing in the past decades (Yang et al., 2009; Zhao et al.,2011). But we could not detect the significant decrease of sun-shine hours due to lack of observed meteorological data. Briefly, we suppose that the meteorological elments might play only minor role in the ozone concentration changes in Beijing, and then focus our discussion on the effects of regional emission reduction measures.

Table4. Average meteorological elements from August 20 to September 05 at GXT station in Beijing between 2010 and 2015.

Year	RH/%	Suface Pressure/hPa	T/℃	Speed/(m s^{-1})	Frequency of the north wind at 850hPa /%	T at 850hPa /℃
2010	70.7	1008.0	24.1	1.6	20.6	16.0
2011	73.2	1007.4	23.6	1.7	12.1	16.1
2012	68.6	1006.8	23.8	1.9	41.2	15.9
2013	66.2	1005.8	24.3	1.9	25	16.1
2014	67.6	1006.5	24.2	1.7	14.7	16.7
2015	69.1	1009.9	24.1	1.4	24.3	13.9

We furher analyzed the year-on-year comparisons of diurnal variations of ozone from 2004 to 2015 during the same periods of air quality assurance stages (S2 and S3) (**Fig.7ab**). Comparing the ozone peaks in urban Beijing during S2 and S3 stages between 2004-2014 and 2015 (the year of taking emission reduction measures to ensure the regional air quality), the ozone peak in 2015 was 3 hours earlier and 0.91 ppbv lower compared to the average ozone peaks during the period of 2004-2014. Whereas comparing the diurnal variations of ozone at DL site during S2 and S3 stages between 2004-2014 and 2015, the ozone peak in 2015 was 10.98 ppbv lower and 2 hours earlier compared to that during the period of 2004-2014. The ozone peaks between urban sites and DL site were much closer (only about 0.48ppbv) after the a reduction of anthropogenic emissions in 2015. The earlier ozone peaks indicated the approximate photochemical equilibrium (R5-R7) of O_3,NO and NO_2 was moved up in Beijing during the day due to regional emission reductions. Therefore,a reduction of anthropogenic emissions such as VOCs and NO_x in urban areas made the ozone peaks decrease significantly and appear 2~3hours earlier compared to the scenarios of no emission reductions

which was a very interesting phenomenon and first found in Beijing and it could also reduce ozone concentration efficiently especially at background sites or downwind areas by the weakened regional transport which was coincident with the study of Seinfeld (Seinfeld ,2006) . Higher VOCs emission reduction caused the slight decrease in urban area but significant decrease at downwind DL background station during the 2015 Grand Military Parade periods.

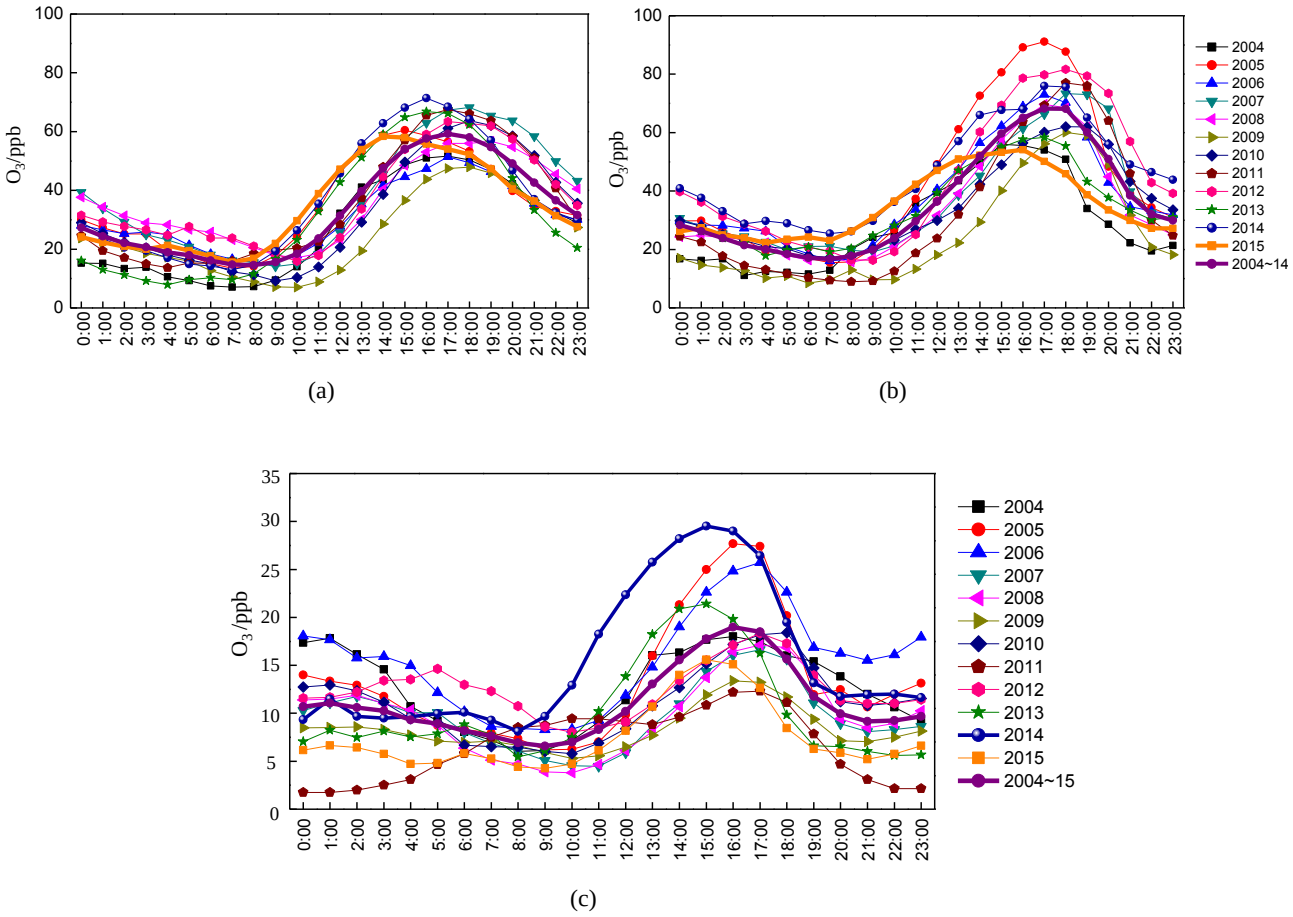


Fig. 7 Diurnal variations of ozone in urban Beijing (a) and at DL station (b) during S2 and S3 stages from 2004 to 2015 compared to that of APEC meeting air quality assurance period (Nov 1~Nov 12,2014) in urban Beijing (c) from 2004 to 2015

$$NO_2 + h\nu \rightarrow NO + O \quad \text{(R5)}$$

$$O_2 + O \rightarrow O_3 \quad \text{(R6)}$$

$$O_3 + NO \rightarrow NO_2 + O_2 \quad \text{(R7)}$$

Compared to the variations of ozone during Asia-Pacific Economic Cooperation (APEC) meeting air quality assurance period (Nov 1~Nov 12,2014;**Fig7c**) in urban Beijing,the ozone peak in 2014 was 1hour earlier and about 9.6 ppbv higher compared to the average ozone

391 peaks during the period of 2004-2015 although Beijing and its surrounding areas adopted
392 different levels of emission reduction measures.
393 As we all know,NO_x and VOCs emission control can considerably affect the O_3
394 concentration, but ozone generation is not a simple linear relationship with its precursors
395 (Sillman, 1999). Ozone pollution is mainly concentrated in summer, and biogenic emissions
396 accounted for a majority of the total VOCs. Therefore, the emission reduction of VOCs via
397 anthropogenic measures connot make it higher than that of NO_x.
398 The previous studies had proved that HCHO and NO_2 from the OMI serve as appropriate
399 indicators for in *situ* observations of total reactive nitrogen and VOCs (Liu et al.,2016;Duncan
400 et al.,2010). OMI tropospheric HCHO/NO_2 Ratio (Ratio)<1 represents PO_3 reduces with
401 diminishing in VOCs (VOC-limited conditions), and Ratio>2 represents NOx-limited
402 conditions. When the ratio is between 1 and 2 indicates a transition regime (mixed VOCs-
403 NOx-limited regime) where the instantaneous PO_3 could be affected by both VOCs and NOx
404 emissions. Three episodes are separately defined in this study: 1st episodes is defined as the
405 period of Parade (from August 20th to September 3rd 2015); 2nd and 3rd episodes are defined
406 as the "pre-Parade" from August 1st to 19th and the "post-Parade" from September 4th to
407 30th. Similarly, the three episodes of APEC are respectively defined as pre-APEC period
408 (from October 15th to 30th), APEC period (from November 1st to 12th) and post-APEC
409 period (from November 13th to 30th).
410 With a series of strict emission control measures during Parade, the ratio retrieved from
411 OMI had changed from 1.70 to 3.72. It means the PO_3 conditions had also changed from
412 mixed VOCs-NOx-limited to a predominantly NOx-limited condition due to the sharp drop of
413 NO_2 during Parade periods. After the strict emission control measures, the NO_2 returned to
414 relatively high values as pre-Parade and the ratio was also diminished (Ratio = 0.90, < 1),
415 which indicated the PO_3 was turned into a VOCs-limited condition during post-Parade. To
416 ensure the air quality during the military parade in 2015, NO_x and VOCs emission control in
417 Beijing and its surrounding areas lasted for almost a month, and VOCs emission control
418 measures was much stricter than NO_x; thereby, ensuring the reduction of VOCs emission
419 (45%) is higher than that of NO_x (30%)(MEP, 2015). Higher ratios by emission control
420 measures during Parade were not only work effectively for NO_2 pollution control patterns but
421 also effective for O_3 controlling.
422 Compared with pre-APEC, the ratio was changed from around 1.21 (VOCs-limited and
423 mixed VOCs-NOx-limited) to around 1.60 (mixed VOCs-NOx-limited and NOx-limited) in
424 Beijing during APEC. NO_2 and HCHO had a certain reduction during APEC which should

lead the O_3 diminishing. Conversely, the ozone concentration was increasing compared to pre-APEC. Regional VOCs emission (about 30%) was equal to that of NO_x (about 30%) (MEP, 2015) during APEC periods and it was easily affected by the relatively unfavorable diffusion conditions in Autumn in Beijing which lead to the concentrations of NO_x and CO two times larger than those of the 2015 Grand Military Parade. So different emission reduction ratios between NO_x and VOCs and different weather conditions led to different VOCs (ppbv)/NO_x (ppbv)ratios during the 2015 Grand Military Parade periods and APEC meeting. These results indicated that emission controls in this case maybe not strict enough or worked well to lessen the levels of ozone. This phenomenon of concentrations of most of the air pollutants decreased, whereas concentrations of ozone increased during APEC meeting period which was consistent with the study of Wang (Wang et al,2015b; Liu et al.,2016).

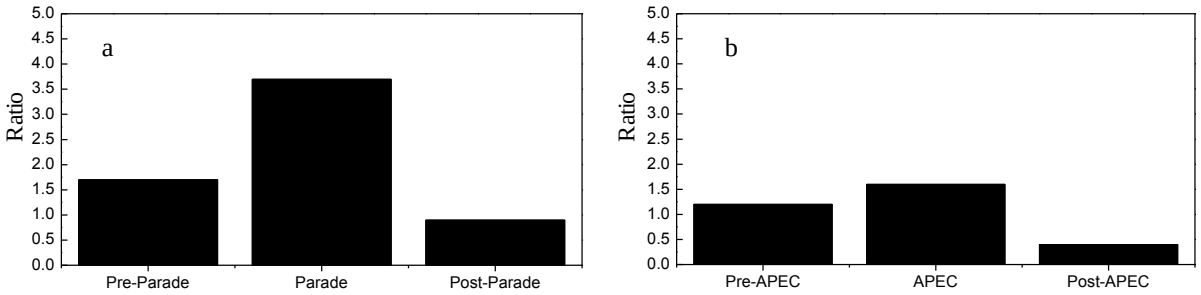


Fig. 8 The variations of averaged Ratio of three periods during Parade (A) and during APEC (B) at Beijing urban sites

Above all, success of air quality protection during the 2015 Grand Military Parade proved that the current governance policy is correct and far-sighted. Moreover, ozone pollution is typically a regional rather than a local issue. Thus, in the future, clean air action plan in Beijing should be implemented on the basis of the lessons from regional air pollution prevention and control mechanism to promote the continuous improvement of regional air quality unswervingly and jointly. Combined with multiple observation stations of ozone in a long period, the numerical models should also be combined to further analyze the ozone formation, so as to develop effective ozone pollution control measures.

4 Conclusions

Although Beijing local government has spent considerable efforts in Beijing to improve air quality and concentrations of the main air pollutants declined significantly. Resluts showed that nnual averaged concentration of daily maximum 1 h ozone ($O_3$1h) was all

increasing at urban sites (1.79 ppbv yr^{-1}) and DL background station (2.05 ppbv yr^{-1}) while daily maximum 8 h averaged ozone concentration ($O_3$8h) was increasing in urban area (1.14 ppbv yr^{-1}) and slightly decreasing at DL background station (-0.47 ppbv yr^{-1}) from 2004 to 2015 due to different ozone sensitivity regimes and ratios of NO_2/NO.

Diurnal variations of ozone peaks obtained at the downwind DL station were about 1 hour later than those of the urban sites from May to October in different years and concentrations of ozone at downwind background station were much higher than those at urban sites. Moreover, differences of ozone peaks between urban sites and DL background station were becoming significantly smaller in recent years, which may be related to regional ozone transport and the expansion urbanization of Beijing.

During several major activities held in Beijing such as the Asia-Pacific Economic Cooperation (APEC) Summit in 2014 and the Parade on the 70th Victory Memorial Day for the Chinese People's War of Resistance against Japanese Aggression in 2015,Beijing and its neighboring cities implemented numerous control strategies, including the suspension of factory operations and odd-and-even license plate rules. A reduction of anthropogenic emissions such as VOCs and NO_x could reduce ozone efficiently especially in downwind areas of Beijing and made the ozone peaks decrease significantly and appear 2~3hours earlier compared to the scenarios of no emission reductions.

On the basis of the discussion and analyses, several recommendations have been made for understanding the heavy air pollution in Beijing：

(1)Compared to the increasing ozone during APEC period, average ozone concentration decreased significantly in the downwind areas of Beijing due to larger ratios of VOCs/NO_x. In order to decrease the ozone concentration in Beijing, emissions of VOCs should be reduced larger than those of NO_x in Beijing. The collaborative control of various pollutants is becoming very important in Beijing.

(2)As air pollution is a regional problem, therefore, the simultaneous implementation of a regional prevention and control mechanism is necessary to promote continuous air quality improvement in Beijing.

(3)Many of the world's thriving cities are struggling with serious air pollution, Beijing's experience in controlling ozone against a backdrop of rapid expansion during air quality assurance periods is a story that should be shared with other emerging economies and burgeoning cities.

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The English in this document has been checked by at least two professional editors, both native speakers of English.

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