

Shifting patterns of Ozone Season due to policy and human activity in Beijing between 2013 and 2023

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Abstract: Since 2013, Beijing has implemented a series of stringent measures to control air pollution, resulting in significant improvements in air quality. This study examines the decadal evolution of surface ozone in Beijing from 2013 to 2023 against the backdrop of continuous emission reduction policies. The Ozone Season in Beijing exhibited pronounced interannual variability, while linear trend analysis indicated a tendency toward prolonged duration. Low-percentile concentrations of the maximum 8-hour moving average ozone (MDA8) showed a statistically significant upward trend, whereas mean and high-percentile values exhibited no significant monotonic trends. The ozone exposure window shifted toward later hours, particularly during the cold season. Diurnal peak concentrations decreased, while trough concentrations increased, leading in a flattened diurnal variation. Furthermore, both the maximum ozone production and loss rates decreased in absolute magnitude, with the production peak tending to occur earlier in the day. These findings establish an observational baseline for understanding urban ozone evolution under intensified emission controls and provide implications for future policy formulation and public health protection strategies.

Keywords: Ozone Season, exposure window, production rate, ozone percentile concentration, diurnal variation.

Highlights:

1. The Ozone Season in Beijing prolonged.
2. Summertime ozone exposure extends into the nighttime.
3. Both diurnal production and destruction rates dropped

1.Introduction

Urban ozone poses a significant threat to public health, as long-term exposure to elevated ground-level ozone increasing the risks of respiratory, cardiovascular, and other diseases(Nuvolone et al., 2018; Crouse et al., 2015; Zhang et al., 2019; Wang et al., 2020b; Liu et al., 2018). Tropospheric ozone formation is governed by photochemical reactions involving its precursors, primarily, volatile organic compounds (VOCs) and nitrogen oxides (NOx), which are largely emitted from urban industrial activities and vehicular traffic. In addition to precursor emissions, meteorological and climate conditions exert substantial influence on the temporal

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37 variability and long-term trends of surface ozone (Sitch et al., 2007; Bais et al., 2018; Bernhard
38 et al., 2023).

39 Beijing, the capital of China, is situated in the northern North China Plain and has a
40 population of approximately 22 million. As a major megacity in northern China and international
41 metropolis, its environmental quality in Beijing has garnered substantial attention (Huang et al.,
42 2017; Qiang and Hu, 2022; Li et al., 2020; Streets et al., 2007). Since 2013, the city has
43 implemented comprehensive air pollution control measures (Li et al., 2020; Wen et al., 2021;
44 Wang et al., 2023), achieving remarkable reductions in primary pollutant concentrations that
45 have been recognized by the United Nations Environment Programme as the “Beijing Miracle”
46 (UNEP, 2019). However, improvements in ozone concentrations have lagged behind those of
47 other pollutants such as PM2.5, and the pronounced interannual variability of ozone levels
48 indicates that mitigating ozone pollution presents a more complex challenge.

49 Amid these pollution control efforts, substantial changes in the magnitude and composition
50 of atmospheric emissions have occurred in Beijing and its surrounding regions (Zhang et al.,
51 2016; Xue et al., 2020; Li et al., 2017; Wu et al., 2023). Consequently, a systematic examination
52 of ozone variations over the past decade is essential for evaluating the effectiveness of these
53 measures and informing future policy directions.

54 Previous research on ozone in Beijing has focused predominantly on the summer
55 months (Xu et al., 2011; Wang et al., 2020a; Zhang et al., 2021; Zhao et al., 2018), despite ozone
56 pollution occurring year-round, typically from March to October, with pronounced episodes in
57 spring and autumn (Chen et al., 2019; Wang et al., 2020c; Wang et al., 2015). Given the
58 substantial interannual variability in ozone levels, long-term trend analysis is essential for
59 advancing our understanding of overall evolution of surface ozone concentrations. This study
60 introduced the concept of the Ozone Season to delineate periods of concentrated ozone pollution
61 across different years and to facilitate examination of its long-term trends.

62 The Ozone Season was first established in North American studies (Cox and Shao-Hang,
63 1996), where it was identified as June to September for northern cities and May to October for
64 southern cities. Notably, Cardelino (Cardelino et al., 2001) indicated that the typical Ozone
65 Season in the United States spans early May through late September, while Qin (Qin et al., 2004)
66 defined it for Southern California as June to October. In China, studies have generally adopted
67 the warm season (April to September) as the analytical framework for ozone pollution (Lu et al.,
68 2020; Li et al., 2021b; Gao et al., 2021). However, owing to regional climatic and meteorological
69 differences, the duration of ozone pollution varies considerably. In northern Chinese cities,
70 ozone pollution typically occurs from April to September, whereas in southern cities such as
71 Guangzhou and Chengdu, it may extend through October or even into November and
72 December (Wang et al., 2017; Li et al., 2021a; Wang et al., 2022).

73 Consequently, the warm season does not fully capture ozone pollution events across
74 different areas. Therefore, the Ozone Season is defined here as the period when photochemical
75 ozone production is most active, making it more suitable for characterizing the regional features
76 of ozone pollution. Ozone Season describes the interval during which photochemical ozone
77 production is maximized and surface ozone concentrations are frequently elevated. By design,
78 the Ozone Season is tied to the photochemical lifecycle of ozone rather than to a fixed calendar,
79 thereby naturally capturing the regional and seasonal heterogeneity of ozone pollution.

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According to China Ministry of Ecology and Environment, a day is designated as an ozone pollution day when the maximum 8-hour moving average (MDA8) exceeds $160\text{ }\mu\text{g m}^{-3}$ (Environment, 2012). Based on this standard, this study defines the Ozone Season as the continuous period from the first to the last MDA8 pollution event within a given year. Utilizing ground-level ozone monitoring data from Beijing between 2013 and 2023, this research examined changes in the Ozone Season and the long-term trends in ozone production and loss rates, with the aim of evaluating the efficacy of pollution control strategies in mitigating ozone pollution.

2. Data and methods

2.1 Data instruction

This study utilized daily and hourly ozone data from Beijing for the period 2013 to 2023, obtained from the China National Environmental Monitoring Centre (CNEMC; <https://www.cnemc.cn/en/>). Ozone concentrations were measured using the Thermo-Fisher 49C ozone analyzers, which employ ultraviolet photometry and report concentration in standard mass units ($\mu\text{g m}^{-3}$). Data was collected from 23 national air quality monitoring stations strategically distributed across urban and suburban areas to ensure representative spatial coverage. City-level hourly ozone levels were calculated as the arithmetic mean of measurements from all operational stations, with city MDA8 derived from MDA8 of stations. All measurements comply with the ambient air quality standards (GB3095-2012) established by the Ministry of Ecology and Environment of China.

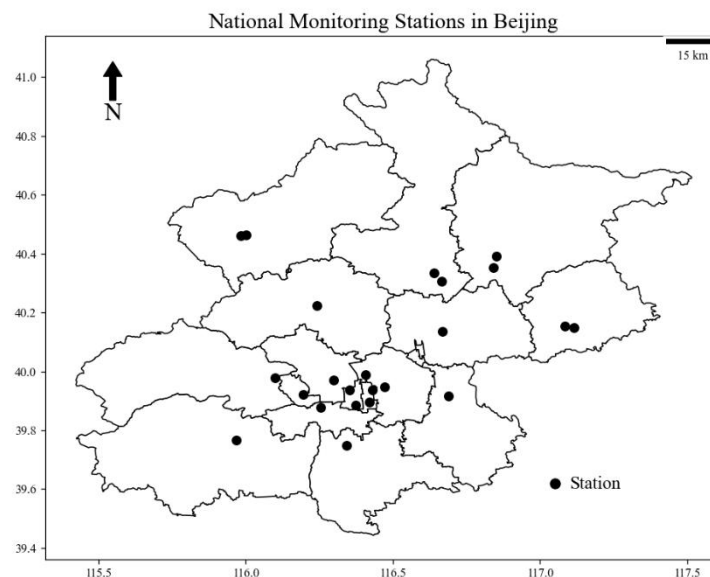


Figure 1 Spatial distribution of Beijing stations

2.2 Research methods

According to the standards set by the Ministry of Ecology and Environment of China, days on which the MDA8 exceeded $160\text{ }\mu\text{g m}^{-3}$ were classified as ozone pollution days. This research analyzed the temporal trends in the first and last pollution days of each year to determine the

duration of the Ozone Season, defined as the interval between these two dates. Additionally, the ozone concentration changes on representative pollution days over the past decade were examined to characterize shifts in ozone production and loss regimes in Beijing.

2.2.1 Ozone data quality control

Typical ozone days were selected to minimize the confounding effects of specific meteorological conditions, including precipitation, strong winds, and overcast skies, on daily ozone concentrations. Days with cumulative precipitation exceeding 0.1 mm or with sustained wind speeds above 5 m s⁻¹ for more than two hours were excluded from the analysis. the filtering is designed to identify photochemically typical days rather than to normalize meteorological conditions. Following this meteorological screening, the proportion of typical ozone days during the Ozone Season from 2013 to 2023 ranged from 54.3% to 76.7% across the study period (2013 – 2023). Notably, 2021 recorded the shortest Ozone Season and the lowest proportion of valid data after accounting for weather-based exclusions. Utilizing the retained hourly ozone data, interannual trends in typical diurnal ozone variation patterns were analyzed.

Table 1 Proportion of effective typical ozone days in Ozone Season from 2013 to 2023

Year	2013	2014	2015	2016	2017	2018	2019	2020	2021	2022	2023
Valid days	72	111	95	107	91	115	123	107	63	135	131
Total days	130	166	159	150	135	167	168	166	116	176	180
Effective rate (%)	55.4	66.9	59.7	71.3	67.4	68.9	73.2	64.5	54.3	76.7	72.8

2.2.2 Trends analysis method

The Mann-Kendall (MK) test is a non-parametric statistical method widely used for detecting monotonic trends in long-term environmental time series (Mann, 1945; Kendall, 1975; Sen, 1968). As a rank-based procedure, it does not require the data to conform to a specific probability distribution and is relatively robust to outliers, which makes it particularly suitable for atmospheric concentration records that are frequently skewed or contain extreme values (Gilbert, 1987).

For a time series containing *n* observations (*x*₁, *x*₂, ..., *x*_{*n*}), the MK test statistic *S* is computed as the sum of the signs of all pairwise differences:

$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^n \text{sign}(x_j - x_k) \tag{1}$$

where *sign*(*x*_{*j*} − *x*_{*k*}) is the signum function, taking values of +1 when (*x*_{*j*} − *x*_{*k*}) > 0, 0 when (*x*_{*j*} − *x*_{*k*}) = 0, and −1 when (*x*_{*j*} − *x*_{*k*}) < 0. Under the null hypothesis of no trend, the statistic *S* is approximately normally distributed for *n* > 10, with a mean of zero and a variance of:

$$\text{Var}(S) = \frac{n(n-1)(2n+5) - \sum_{p=1}^q t_p(t_p-1)(2t_p+5)}{18} \tag{2}$$

where *q* is the number of tied groups and *t*_{*p*} denotes the number of observations in the *p*-th tied group. The standardized test statistic *Z* is then calculated as:

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$$Z = \begin{cases} \frac{S-1}{\sqrt{\text{Var}(S)}} & \text{if } S > 0 \\ 0 & \text{if } S = 0 \\ \frac{S+11}{\sqrt{\text{Var}(S)}} & \text{if } S < 0 \end{cases} \quad (3)$$

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A positive (negative) Z value indicates an upward (downward) trend. The trend is deemed statistically significant at the 95 % confidence level when $|Z| > 1.96$. To quantify the magnitude of any detected trend, the Sen's slope estimator was additionally employed (Sen, 1968; Theil, 1950).

In this study, the MK test was applied to evaluate the long-term temporal trends in Ozone Season duration and ozone percentile concentrations, and to assess the robustness of interannual variations in the multi-year mean diurnal ozone cycles.

2.2.3 Ozone exposure window calculation

In recent years, urban ozone pollution has posed a persistent threat to human health, with the maximum daily average of ozone (MDA8) during the warm season widely adopted as a metric for assessing outdoor ozone exposure levels (Lu et al., 2018; Niu et al., 2022; Byun et al., 2022). MDA8 is defined as below.

$$MDA8 = \text{Max}_{t=1,2,3,\dots,17} \left\{ \frac{1}{8} \sum_{i=t}^{t+7} C_i \right\} \quad (4)$$

C_i denotes the hourly ozone concentration ($\mu\text{g m}^{-3}$) at hour i , where i is the starting hour of the sliding window, ranging from 01:00 to 17:00, yielding a total of 17 overlapping windows. The MDA8 is defined as the maximum of these 8-hour running averages. The corresponding 8-hour period beginning at the start time of this maximum window represents the daily peak ozone exposure window. We denote the sequential hours of this period as $t_1, t_2, t_3, \dots, t_8$, which serves as an effective indicator of health risk warnings for sensitive populations. This continuous exposure duration is designated as T_{MDA8} . For convenience in the subsequent interannual and inter-seasonal comparisons, T_{MDA8} is used to represent the temporal shift of peak exposure.

3. Results and discussion

3.1 Changes in the Ozone Season in Beijing

Over the past decade, the onset of the first annual ozone pollution event in Beijing has demonstrably shifted to an earlier time in the year. Figure 2 illustrates the variations in the Ozone Season from 2013 to 2023. Here, FPD denotes the first ozone pollution day of the year, marking the beginning of the Ozone Season, while LPD represents the last ozone pollution day, indicating the season's conclusion. The secondary axis displays the duration of the Ozone Season. Analysis shows that this event typically occurs between March and May. In 2023, the earliest date was recorded on March 30, whereas in 2021, the latest occurrence was on May 16. April exhibited the highest frequency of first pollution days, with seven occurrences, accounting for 66% of the total. May follows with three occurrences, and March with one. This trend reveals that the date of the first pollution event advanced from May 5, 2013, to March 30, 2023, indicating an average shift of approximately 3.5 days per year over the 11-year period.

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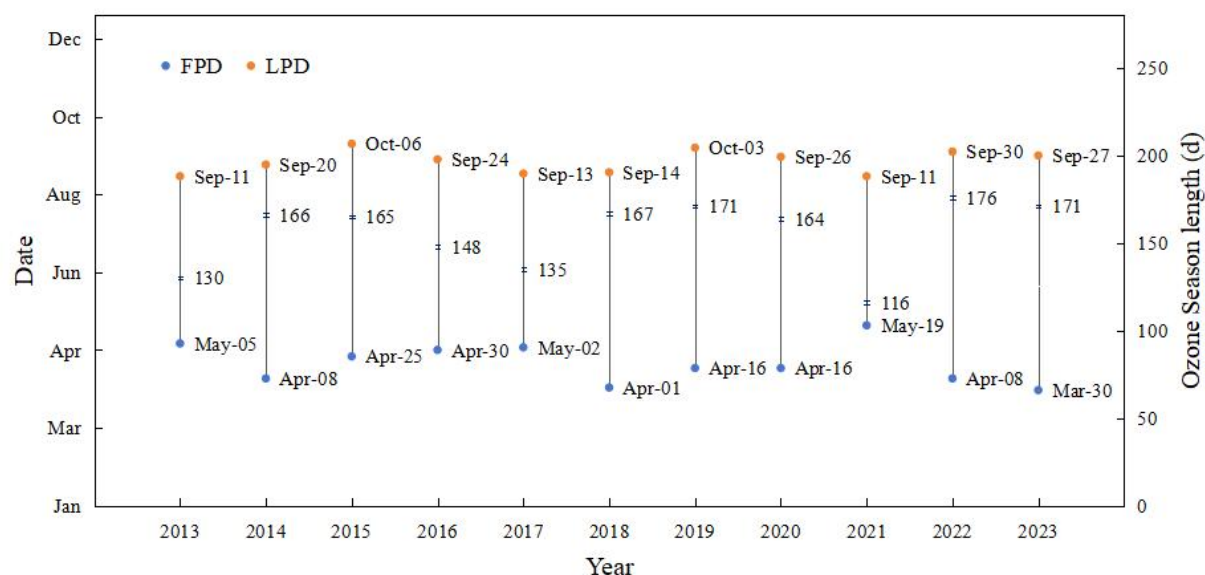


Figure 2 Changes in Beijing Ozone Season from 2013 to 2023

In contrast, the date of the last pollution event (LPD) in Beijing has been delayed over the same period. The final pollution occurrences are recorded between September and October, with the earliest date on September 11, 2013, and the latest on October 6, 2015. Unlike the first pollution days, the timing of the last pollution days exhibited substantially less variability, with 88% of occurrences in September. The last pollution date was postponed from September 11, 2013, to September 27, 2023, representing a delay of 16 days and an average trend of 1.5 days per year, which is slower than the trend observed for the first pollution event.

Furthermore, the duration of the Ozone Season in Beijing has increased over the past decade. Defined as the interval between the first and last pollution days, the season ranged from 116 to 176 days, with an average length of 155 days. The length of the Ozone Season varied considerably from year to year, with 2021 being the shortest and 2022 the longest. The earlier occurrence of the first pollution event and the later occurrence of the last pollution event have collectively extended the Ozone Season by an average of 5.0 days per year, representing approximately 3.2% of the average season length.

To assess the robustness of Ozone Season variations, we applied the Mann-Kendall (MK) test to the interannual trends of the first pollution day (FPD), last pollution day (LPD), and Ozone Season length. None of these metrics exhibited a statistically significant monotonic trend at the $\alpha = 0.05$ level. This is not entirely unexpected, as ozone photochemical production is highly sensitive to meteorological conditions, including temperature, solar radiation, and synoptic-scale circulation patterns (Li, K. et al., 2019; Lu et al., 2020; Wang et al., 2025). Such climate-driven variability can introduce significant noise that may mask underlying long-term trends, especially in a relatively short (11 year) time series (Weatherhead et al., 1998). We identified 2017 and 2021 as anomalous years with markedly shorter Ozone Seasons, likely attributable to such meteorological anomalies. To test the underlying trend with reduced noise, we excluded these two outlier years. After this adjustment, the MK test Z values for FPD and Ozone Season length increased to 1.80 and 1.89, respectively, approaching the critical value of

1.96 for significance at the 95 % confidence level. This suggests a marginally significant tendency toward an earlier FPD and a prolonged season length once meteorological noise is reduced, whereas the LPD still exhibited a relatively weak Z_c value. Thus, after filtering out climate-driven anomalies, the results tentatively support the inference that the Ozone Season onset has shifted earlier and its duration has extended, although these trends remain sensitive to interannual meteorological variability.

Currently, international research has predominantly focused on the characteristics of ozone concentration(Chen et al., 2020; Yin et al., 2017; Cao et al., 2024), with insufficient studies addressing the long-term changes in the duration of the Ozone Season. As the Ozone Season represents a period of heightened ozone pollution, its extension-driven by a combination of advancing onset and delayed conclusion poses additional health risks to the population, necessitating greater attention to the impacts of ozone over extended periods. The prolongation of the Ozone Season likely linked to climate change and alterations in pollutant emission patterns(Schnell et al., 2016; Doherty et al., 2013; Sicard, 2021). Therefore, our findings highlight an urgent need for adaptive pollution control strategies that account for this lengthening high-risk period.

3.2 Changes in MDA8 percentile concentrations in Beijing

To examine the distribution characteristics of MDA8 concentrations across different years, box plots were generated for the period from 2013 to 2023. Fig. 3 illustrates the minimum, 25th percentile, mean, median (50th percentile), 75th percentile, and maximum values of MDA8 for each year. The mean MDA8 concentrations ranged from 85 to 111 $\mu\text{g m}^{-3}$, with the lowest recorded in 2013 and the highest in 2023. This demonstrates considerable interannual variability and an overall upward trend, with an average increase of approximately 2.4 $\mu\text{g m}^{-3}$ per year. Notably, the anomalous year of 2021 exhibited a mean concentration of 87 $\mu\text{g m}^{-3}$, representing the second lowest value during this period.

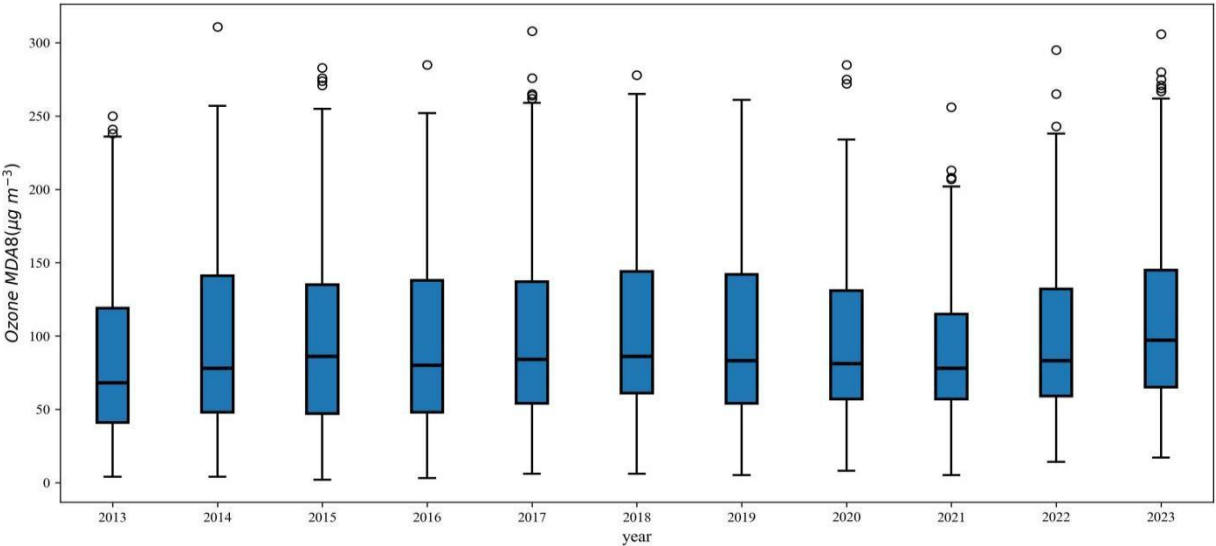


Figure 3 Beijing day MDA8 box diagram from 2013 to 2023

Over the past decade, the lower percentile concentrations of MDA8 in Beijing have also increased. Specifically, the minimum MDA8 concentrations rose from $4\text{ }\mu\text{g m}^{-3}$ in 2013 to $14\text{ }\mu\text{g m}^{-3}$ in 2023, reflecting a growth of 225% with an average increase of $1.0\text{ }\mu\text{g m}^{-3}$ per year. Similarly, the 25th and 50th percentile MDA8 concentrations increased by $24\text{ }\mu\text{g m}^{-3}$ and $31\text{ }\mu\text{g m}^{-3}$, respectively, with growth rates of $2.4\text{ }\mu\text{g m}^{-3}$ per year and $3.1\text{ }\mu\text{g m}^{-3}$ per year. This rise in lower percentile concentrations suggests an increase in background ozone levels and an enhancement of atmospheric oxidizing capacity. The minimum and 25th percentile ozone concentrations passed the Mann-Kendall test ($\alpha = 0.05$), exhibiting a statistically significant increasing trend.

In contrast, high percentile MDA8 concentrations have exhibited no significant changes Over the study period. The maximum MDA8 concentration recorded was $311\text{ }\mu\text{g m}^{-3}$ in 2014, while the minimum was $256\text{ }\mu\text{g m}^{-3}$ in 2021. Fluctuations in the maximum and 75th percentile MDA8 concentrations demonstrated weak trends over time, likely attributable to substantial interannual meteorological variability. Notably, the 90th percentile MDA8 concentration exhibited a declining trend of approximately $-2.5\text{ }\mu\text{g m}^{-3}$ per year. The correlation of changes in high-percentile ozone concentrations Over the study period was relatively weak, possibly due to the dominant influence of interannual meteorological fluctuations on extreme ozone values. This results in less pronounced long-term trends compared to lower concentration percentiles.

Recent studies have indicated that low percentiles (below the 25th percentile) of ozone concentrations have increased globally across all regions, whereas high percentiles (above the 75th percentile) have decreased in North America and Europe, but increased in the Southern Hemisphere and Japan(Christiansen et al., 2022). Yan's research (Yan et al., 2018) also identified an increase in the 5th percentile of ozone concentrations in Europe, alongside a rise in the 95th percentile. Chen's study(Chen et al., 2020) reported an upward trend in the 10th and 25th percentile low concentrations, in Beijing since 2013, consistent with these global findings. However, changes in high percentile concentrations in Beijing were found to be insignificant. As many studies have suggested, the observed rise in low percentile concentrations across various global regions may be linked to decreased emissions of ozone precursors, along with contributions from stratospheric or upper-tropospheric ozone transport. The overall increase in mean concentrations may indicate a worsening trend in ozone pollution, potentially connected to broader global climate changes that enhance ozone long-range transport and create more favorable conditions for ozone production under elevated temperatures.

3.3 Changes in ozone exposure time in Beijing

The maximum daily 8-hour average (MDA8) identifies the time window with the highest ozone concentration on a given day. For sensitive populations, this window warrants particular attention for health protection measures. We introduce the term ozone exposure window time (T_MDA8) to denote this period and examine its interannual variability as well as the differences between the warm and cold seasons.

Fig.4 reveals significant differences in ozone exposure window (T_MDA8) between the cold and warm seasons in Beijing. From November to January, the ozone exposure window ranges from 11:00 to 19:00. As temperatures rise, this exposure window shifts to later hours, extending from 14:00 to 21:00 during April to September. This indicates that populations are exposed to elevated ozone levels even during nighttime in the warm season. This shift is

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consistent with the extended photochemical production period during longer daylight hours and the delayed NO titration minimum in the warm season.

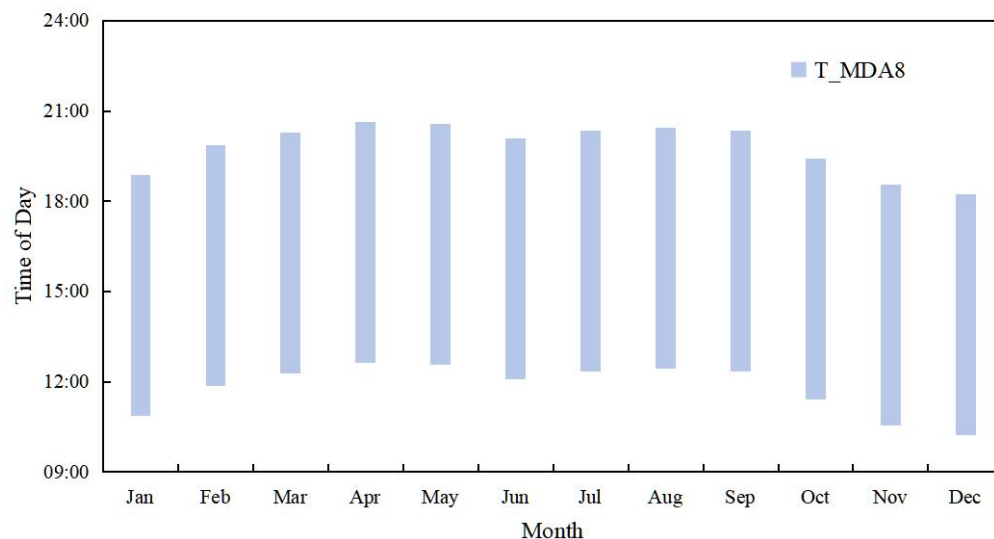
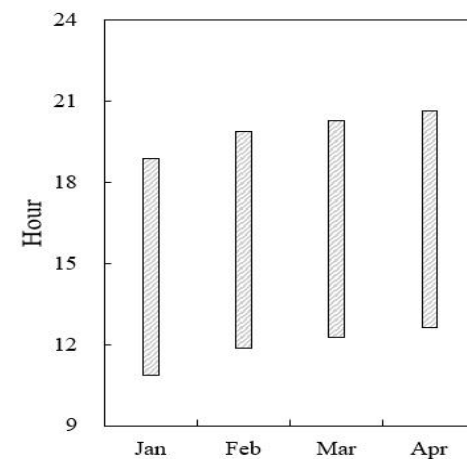
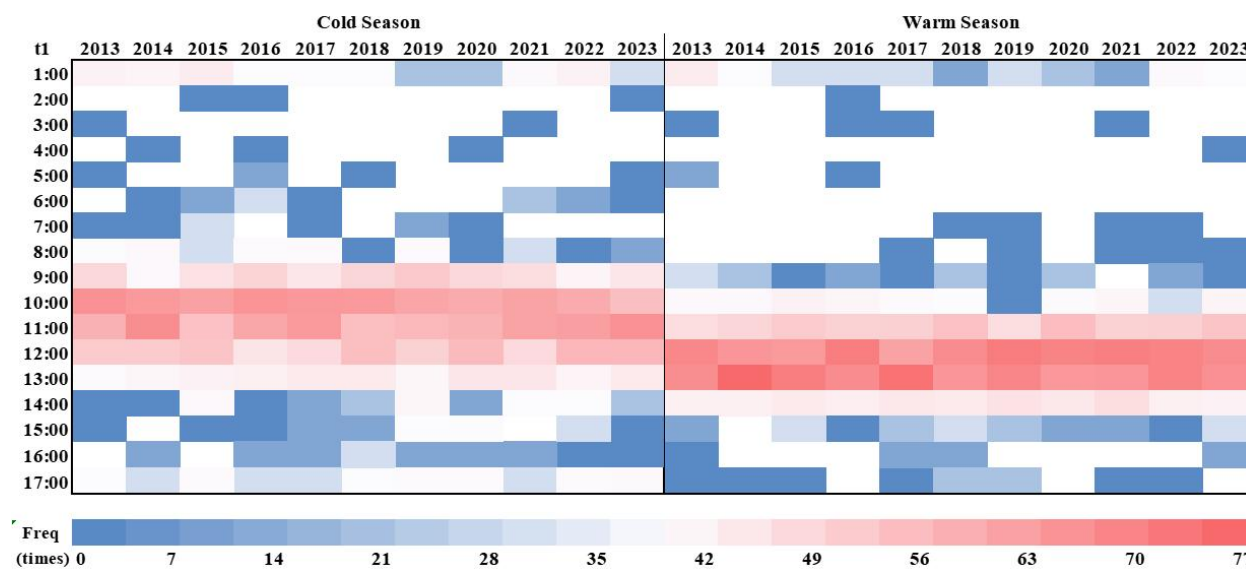


Figure 4 Average duration of MDA8 ozone in Beijing by month from 2013 to 2023

Over the study period, a noticeable trend of delayed ozone exposure times was observed during the cold season (October to March). The starting time (t1) of ozone exposure shifted from predominantly 10:00 and 11:00 in 2013 to a new window of 12:00 to 13:00 by 2023, indicating a postponement in the occurrence of peak ozone during the cold season, as shown in Fig. 5. A similar trend is observed in the warm season. However, this delay is less pronounced compared to that in the cold season, as shown in Fig. 6a. The starting time (t1) was 11:00-12:00 in 2013, but shifted to 12:00-13:00 during 2014 and 2023. This difference between seasons may be attributable to the more concentrated timing of ozone exposure events during the warm season, which reduces interannual variability in the peak onset time compared to the cold season.



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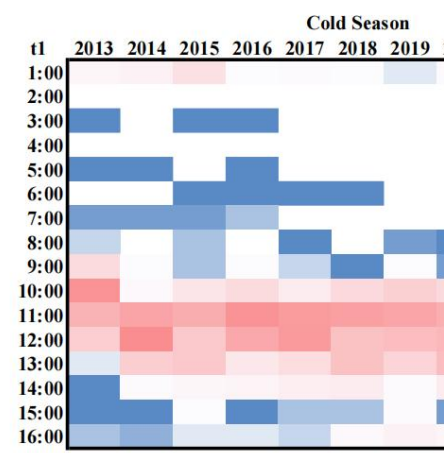


Figure 5 Variation in ozone exposure times (t1) during the cold season and warm season from 2013 to 2023. The heatmap characterizes the frequencies of t1 occurrences at different times over these years.

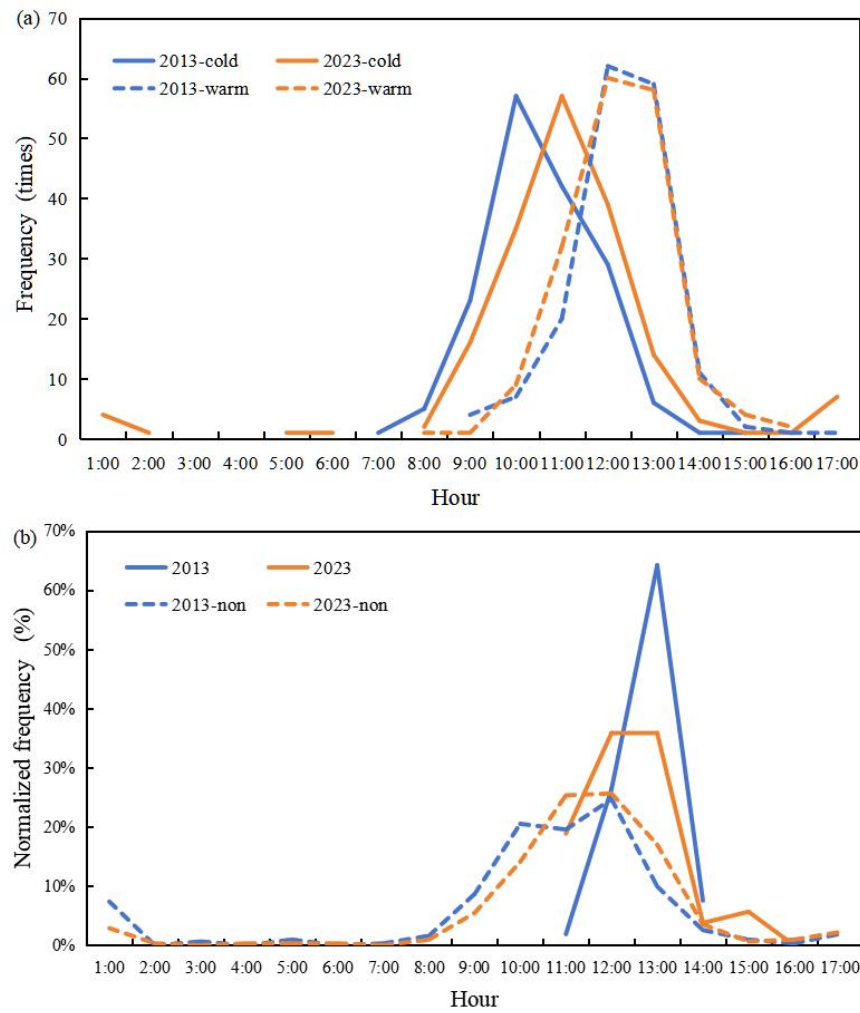
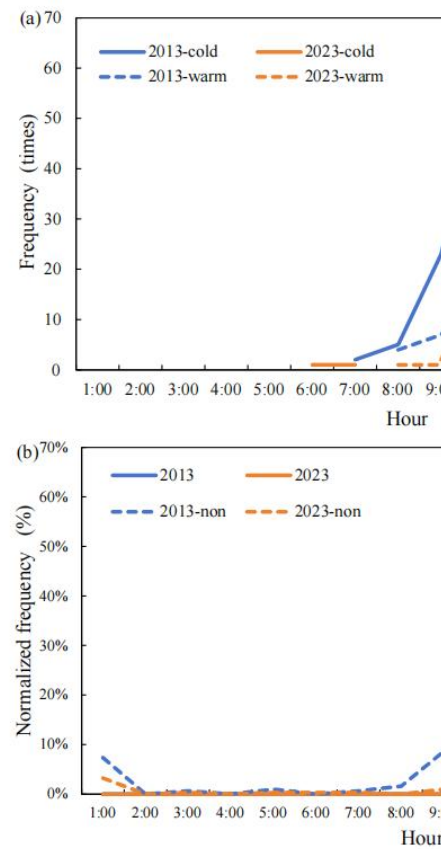


Figure 6 Distribution frequencies of t1 in cold season and warm season for 2013 and 2023(a), and distribution normalized frequencies of t1 in pollution days and non-pollution days(b)

This postponement of ozone exposure times suggested a delay in the occurrence of high ozone levels over the study period, which may extend the impact on human health into the nighttime hours. Currently, there is a lack of long-term studies on ozone exposure times in China. The variations in ozone exposure time in Beijing are influenced by changes in local ozone production characteristics, highlighting the importance of understanding these dynamics for evaluating the effectiveness of pollution control policies in addressing urban ozone levels.

Significant differences in ozone exposure times were observed between pollution days (MDA8 $\geq 160 \mu\text{g m}^{-3}$) and non-pollution days. Because the number of pollution days per year is substantially lower than that of non-pollution days, the frequency distribution were normalized to obtain relative frequencies (expressed as a percentage), as shown in Fig. 7. On pollution days, the starting time (t1) of ozone exposure window ranged from 11:00 to 16:00, with a peak occurring between 13:00 and 14:00. In contrast, on non-pollution days, the starting time



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(t1) spanned from 01:00 to 16:00, with the majority occurring between 10:00 and 15:00. This indicates that ozone exposure time on pollution days is more temporally concentrated, primarily occurring during the afternoon to the evening. This time frame is critical for implementing protective measures for sensitive populations. This temporal concentration on pollution days reflects the dominance of intense photochemical production under favorable meteorological conditions, whereas the broader distribution on non-pollution days may result from weaker photochemical activity and greater influence of non-local transport processes.

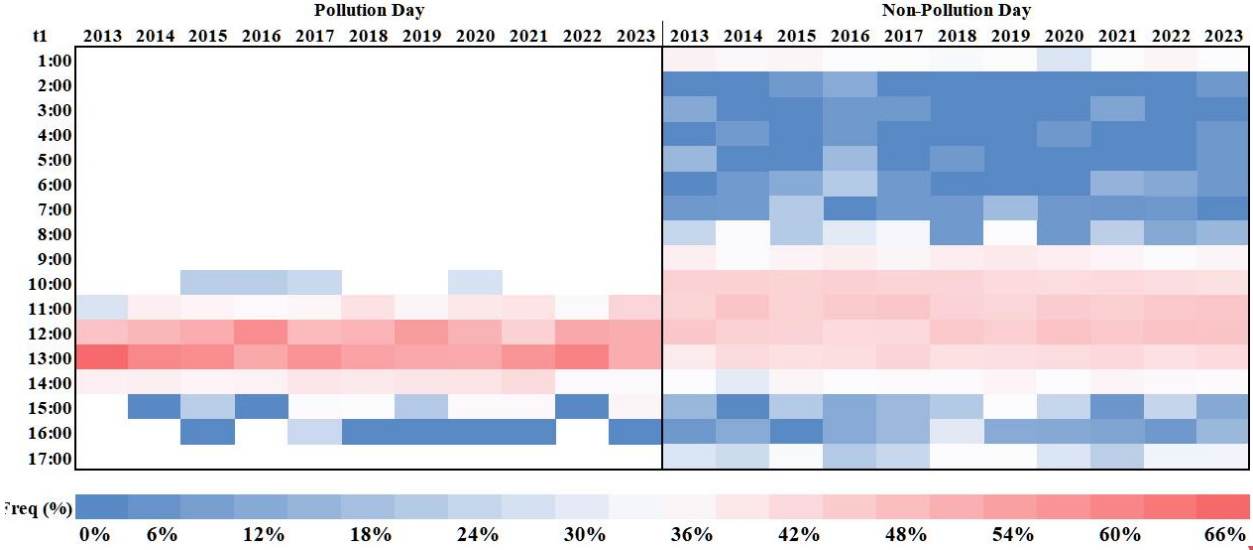
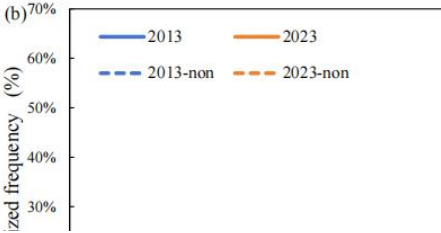
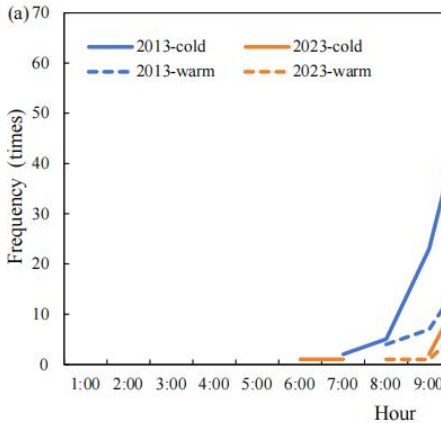
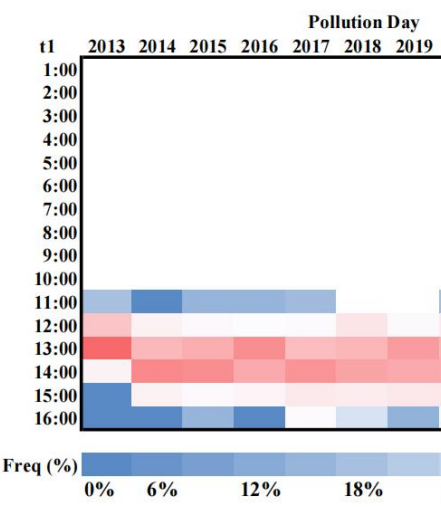


Figure 7 Variation in ozone exposure times (t1) on pollution and non-pollution days from 2013 to 2023. The heatmap characterizes the normalized frequencies of t1 occurrences at different times over these years.

Over the study period, a noticeable delay in the starting time (t1) of ozone exposure on non-pollution days was observed, shifting from predominantly 10:00-12:00 in 2013 to 11:00-14:00 in 2023. In contrast, changes in t1 on pollution days are relatively minor, as shown in Fig. 6b. This difference may be attributed to the dominant influence of meteorological factors, particularly temperature, on ozone production during pollution days. Because pollution days predominantly occur in the warm season, the timing of high temperatures tends to be more consistent, resulting in a more constrained diurnal window for ozone production. On non-pollution days, which are distributed across both warm and cold seasons, weaker photochemical forcing and greater meteorological variability allow the ozone exposure window to shift more readily in response to changing synoptic conditions.

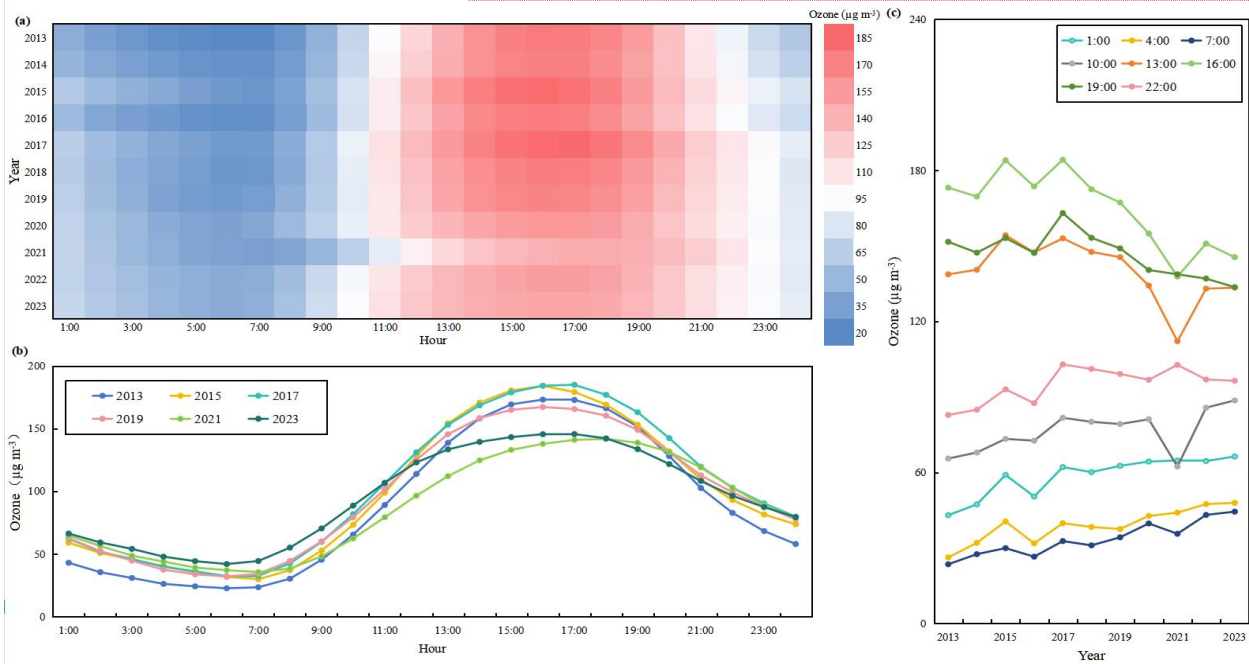
3.4 Diurnal variation of Ozone Season in Beijing

Based on hourly ozone data with meteorological influences removed during the Ozone Season, typical diurnal variation patterns of ozone from 2013 to 2023 were analyzed. Each year's diurnal ozone profile exhibited a clear unimodal pattern. Ozone concentrations decreased from 01:00, reaching a minimum between 06:00 and 07:00, then rose to a peak between 16:00 and 18:00, followed by a subsequent decline (Fig. 8a and 8b).



331 The analysis revealed a decreasing trend in peak ozone concentrations, accompanied by a
332 delay in their occurrence. In contrast, trough concentrations exhibited an increasing trend, with
333 trough times shifting earlier over the same period. From 2013 to 2023, peak ozone
334 concentrations during the Ozone Season ranged from 137.8 to 184.5 $\mu\text{g m}^{-3}$, with a declining
335 trend in diurnal peak values at a rate of 3.6 $\mu\text{g m}^{-3}$ per year. Peak values typically occurred
336 between 16:00 and 18:00, with a noticeable delay over the study period. Between 2013 and 2020
337 (excluding 2017), peak concentrations were consistently recorded at 16:00, whereas from 2021
338 to 2023, they shifted to 17:00-18:00. Trough values ranged from 23 to 42 $\mu\text{g m}^{-3}$, exhibiting an
339 increasing trend at a rate of 1.7 $\mu\text{g m}^{-3}$ per year, with trough times occurring between 06:00 and
340 07:00. Notably, the trough time shifted to 06:00 from 2017 to 2023 (excluding 2021).

341 A notable observation was that daytime ozone concentrations decreased while nocturnal
342 concentrations increased over the study period. The interannual variation curves of hourly ozone
343 concentrations (Fig. 8c) indicated significant declining trends during the high-value period from
344 13:00 to 15:00 ($R = -0.54$ to -0.80), with the most pronounced average decrease occurring at
345 16:00, at a rate of approximately 3.7 $\mu\text{g m}^{-3}$ per year. This trend remained consistent, with the
346 exception of anomalous values recorded in 2021. Conversely, increasing trends in ozone
347 concentrations were observed during other periods, particularly during the low-value period from
348 22:00 to 09:00 ($R = 0.78$ to 0.94), with an average increase at 08:00 of approximately 2.2 $\mu\text{g m}^{-3}$
349 per year. These opposing trend indicates a reduction in the diurnal amplitude of ozone,
350 decreasing from 150 $\mu\text{g m}^{-3}$ in 2013 to 104 $\mu\text{g m}^{-3}$ in 2023, at an average rate of 5.3 $\mu\text{g m}^{-3}$ per
351 year. The flattening of the diurnal ozone profile suggests a weakening of both daytime ozone
352 production and nocturnal ozone loss.



353 Figure 8 The diurnal variation of ozone in Beijing during the Ozone Season from 2013 to 2023
354 (a. an hourly color chart, b. a diurnal variation curve chart by every two years, c. a yearly
355 variation chart of hourly concentration by every three hours)
356

357

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Application of the MK test to the interannual evolution of hourly ozone concentrations, revealed a statistically significant increasing trend during 01:00 – 10:00 local time and a significant decreasing trend during 14:00–19:00 (both $p < 0.05$). These opposing phase shifts in the diurnal cycle may reflect changing photochemical regimes and evolving precursor emission patterns. These results align well with the findings presented above, further substantiating the conclusion that daytime peak ozone concentrations have declined, whereas nocturnal ozone concentrations have exhibited an upward trend.

Similar phenomena have been observed in the United States during the summer(He et al., 2020), where increases in morning and evening ozone concentrations are linked to reductions in anthropogenic NO_x emissions, which weaken ozone titration. Chen(Chen et al., 2020) also noted a decrease in diurnal ozone amplitude in Beijing in recent years, particularly at lower concentration ranges. This study posits that substantial reductions in NO_x emissions Over the study period have led to decreased nocturnal NO concentrations, weakening nighttime ozone loss and contributing to elevated nocturnal ozone levels. Conversely, the decline in daytime ozone peaks is likely associated with coordinated reductions in volatile organic compound (VOC) emissions in Beijing(Kang et al., 2021; Zhan et al., 2021; Wang et al., 2022). Since 2017, stricter VOC emission controls have been implemented in Beijing and surrounding cities, leading to a continuous decrease in ambient VOC concentrations(Wang et al., 2022). This decrease inhibits daytime ozone production, and contributes to the observed decline in peak daytime concentrations.

3.5 Variation of ozone production and loss rates during Ozone Season in Beijing

By calculating the difference in ozone concentration between consecutive hours, we derived the hourly ozone change rate of (V_i), expressed in $\mu\text{g m}^{-3} \text{h}^{-1}$. A positive V_i indicates a net ozone production, while a negative V_i reflects a net ozone loss. A value of zero signifies a balance between production and loss. The hourly ozone change rate represents the total net ozone tendency. Following Kaser and other studies (Kaser et al., 2017; He et al., 2023; Song et al., 2024), this metric is understood as the integrated outcome of all physical and chemical processes contributing to the local ozone budget, including entrainment driven by boundary-layer growth, horizontal advection, dry deposition, and photochemistry. As noted in previous studies(Gu et al., 2024; Liu et al., 2021), long-term averaging of diurnal ozone variations effectively suppresses synoptic-scale advection effects. This interpretation assumes that long-term averaging effectively filters out transport effects, such that the diurnal cycle of V_i primarily reflects local photochemical production and loss rather than advective contributions. Accordingly, we employ the hourly ozone change rate in this study to characterize net ozone production and loss.

$$V_i = \frac{P_i - P_{i-1}}{t_i - t_{i-1}} \quad (5)$$

We plotted heat maps and time series of V_i for each year (Fig. 9a, 9b). The absolute value of V_i was lowest at approximately 07:00, suggesting that, on average, the increase in ozone concentration begins around this time. The primary net production of ozone occurred between 07:00 and 16:00, after which V_i became negative after 17:00, indicating the onset of net loss. This led to a decline in ozone concentration until approximately 06:00 the following day.

Over the study period, the maximum ozone production rate exhibited a decreasing trend, accompanied by an advancement in its timing. In contrast, the maximum loss rate diminished

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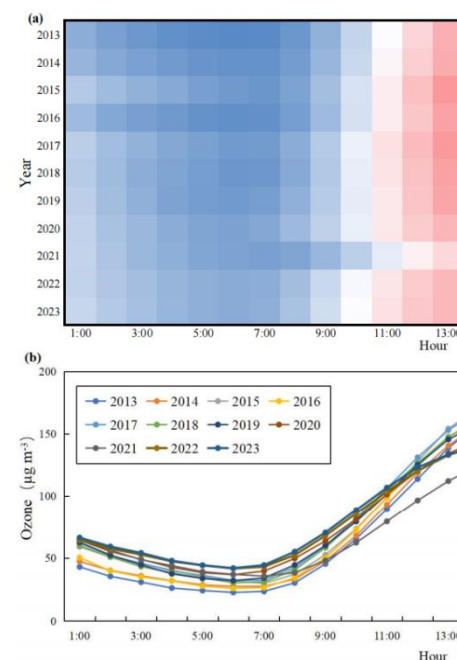
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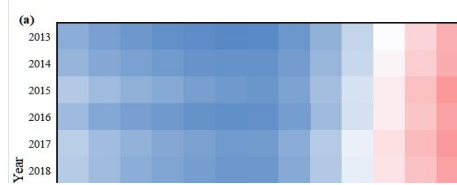
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without significant changes in its timing. The maximum value of V_i (V_{max}), representing the peak production rate, occurred during the hour preceding and following the most rapid production period. Prior to 2016, the peak production rate was concentrated between 11:00 and 13:00. However, after 2016, it gradually shifted earlier to 10:00–12:00, indicating an advancement in peak production timing. From 2013 to 2015, V_{max} increased slightly from 24.7 to 29.1 $\mu\text{g m}^{-3} \text{h}^{-1}$, but subsequently declined to 18.2 $\mu\text{g m}^{-3} \text{h}^{-1}$ in 2023 (Fig. 9c). This reflects a decreasing trend in maximum ozone production rates after 2015, at a rate of 1.0 $\mu\text{g m}^{-3}$ per year.

The minimum value of V_i (V_{min}) indicates the maximum net loss rate, ranging from -25.5 to -13.5 $\mu\text{g m}^{-3} \text{h}^{-1}$ and exhibiting a smaller magnitude relative to the maximum production rate. V_{min} typically occurred between 20:00 and 22:00, with 9 out of 11 years recording occurrences at 21:00. Over the study period, a clear upward trend in V_{min} was observed, increasing at a rate of 1.2 $\mu\text{g m}^{-3} \text{h}^{-1}$ per year, suggesting a weakening of nocturnal ozone loss.

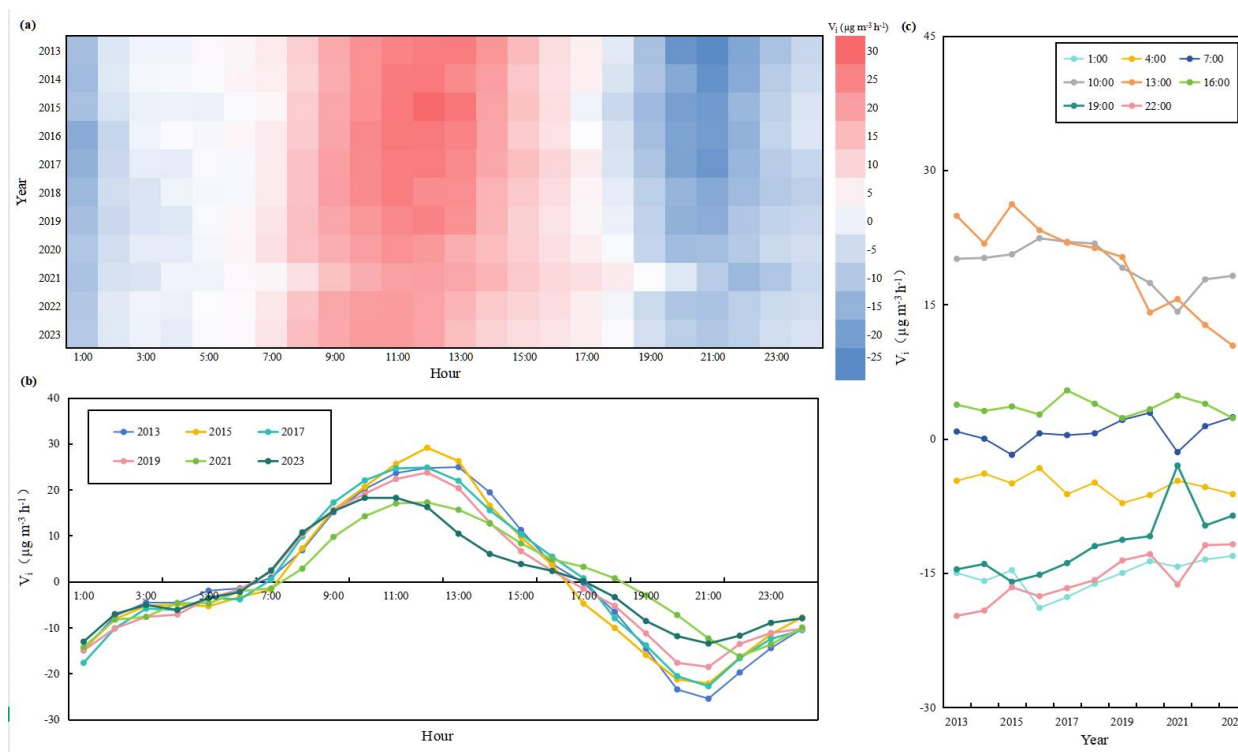


Figure 9 Daily rate of ozone change in Beijing during the Ozone Season from 2013 to 2023 (a. an hourly color chart, b. a diurnal variation curve chart by every two years, c. a yearly variation chart of hourly concentration by every three hours)

On average, ozone production rates exhibited a downward trend, resulting in a flattened diurnal profile. The interval between the the morning and evening transition points of positive to negative V_i represents the duration of ozone production (T), which averaged 9 to 11 hours, with the shortest duration of 9 hours recorded in 2015. By subtracting the trough concentration from the peak concentration, and dividing by the production duration (T), we calculated the average production speed ($V_p = (V_{max} - V_{min})/T$) of ozone. Over the study period, V_p ranged from 9.4 to 17.1 $\mu\text{g m}^{-3} \text{h}^{-1}$, demonstrating a decreasing trend at a rate of approximately 0.69 $\mu\text{g m}^{-3} \text{h}^{-1}$ per year. This indicates that both the maximum production and loss rates of ozone in Beijing have slowed,

424 with the rate of change in loss exceeding that of production, resulting in a reduced diurnal
425 variations.

426 The observed changes in ozone production and loss rates support the earlier conclusions
427 about diurnal variation. This amplitude of diurnal ozone variation is diminishing, concomitant
428 with a weakening of both ozone production and loss rates. Over the study period, reductions in
429 anthropogenic emissions have altered the characteristics of ozone photochemistry. Specifically,
430 coordinated decreases in VOCs and NO_x have led to lower diurnal mean ozone production rates.
431 Additionally, the reduction in nocturnal NO_x concentrations has diminished the titration sink for
432 ozone during nighttime hours.

433 3.6 Differences of diurnal variation between pollution days and Ozone Season in Beijing

434 To further investigate the variation in ozone pollution in Beijing over the study period, we
435 analyzed pollution days ($\text{MDA8} > 160 \mu\text{g m}^{-3}$) for each year (Table 2). From 2013 to 2023, the
436 number of pollution days ranged from 24 to 74, exhibiting substantial annual variability.
437 Elevated numbers of pollution days were observed from 2014 to 2019, peaking in 2019, followed
438 by a marked decline since 2020. The duration of the Ozone Season varied from 115 to 181 days.
439 A weak positive correlation was found between the duration of the Ozone Season and the
440 number of pollution days, suggesting that a longer Ozone Season may be accompanied by more
441 pollution days.

442 The peak and trough concentrations on pollution days exhibited trends similar to those
443 observed during the Ozone Season, characterized by declining peaks and elevated troughs. A
444 comparison of diurnal variations between pollution days and the Ozone Season revealed that the
445 timing of peaks and troughs was consistent, with peaks primarily occurring around 16:00 and
446 troughs between 06:00 and 07:00. On pollution days, a discernible trend of decreasing peak
447 values and increasing trough values was observed, with peak concentrations declining at a rate of
448 approximately $2.5 \mu\text{g m}^{-3}$ per year, which is 30.9% lower than the peak reduction rate during the
449 Ozone Season. In contrast, the trough increase rate of approximately $2.4 \mu\text{g m}^{-3}$ per year was
450 37.3% higher than the corresponding rate during the Ozone Season.

451 The similarity in diurnal variation patterns suggests that both ozone pollution days and
452 typical days in Beijing are characterized by weakening ozone production and loss rates. The
453 slower decline rate in peak concentrations and faster increase in trough concentrations on
454 pollution days imply that favorable meteorological conditions lead to elevated precursors and
455 NO concentrations in the atmosphere during these events, thereby sustaining ozone production
456 capacity, while nocturnal conditions facilitate enhanced loss.

457 Over the study period, the concentration differences between pollution days and the Ozone
458 Season have widened, with the ozone production capabilities on pollution days remaining
459 relatively robust compared to those during the Ozone Season. Fig. 10 presents a heat map
460 illustrating the hourly concentration differences between pollution days and the Ozone Season.
461 Hourly ozone concentrations on pollution days consistently exceeded those in the Ozone Season,
462 with peak values surpassing those of the ozone season by 43 to $57 \mu\text{g m}^{-3}$. In contrast, the
463 differences in trough values were less pronounced, ranging from 1 to $11 \mu\text{g m}^{-3}$.

464 Notably, the hourly concentration differences between pollution days and the Ozone Season
465 exhibited an increasing trend over the study period, with peak differences increasing at a rate of
466 approximately $1.0 \mu\text{g m}^{-3}$ per year and trough differences increasing at approximately $0.65 \mu\text{g m}^{-3}$

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3 per year. The correlation coefficients for these trends were 0.65 and 0.68, respectively. Additionally, nocturnal concentration differences became considerably more pronounced, rising from 19.2 to 34.2 $\mu\text{g m}^{-3}$, at a rate of approximately 1.3 $\mu\text{g m}^{-3}$ per year.

The widening of hourly concentration differences between pollution days and the Ozone Season suggests that, although overall ozone production capabilities are declining, the rate of reduction on pollution days is comparatively slower. This indicates that pollution days retain a significant capacity for ozone production.

Table 2 Ozone pollution days and Ozone Season in Beijing from 2013 to 2023

Year	2013	2014	2015	2016	2017	2018	2019	2020	2021	2022	2023
Ozone pollution days	53	73	69	69	60	67	74	54	24	52	53
Ozone Season	129	165	164	147	134	166	170	163	115	175	181

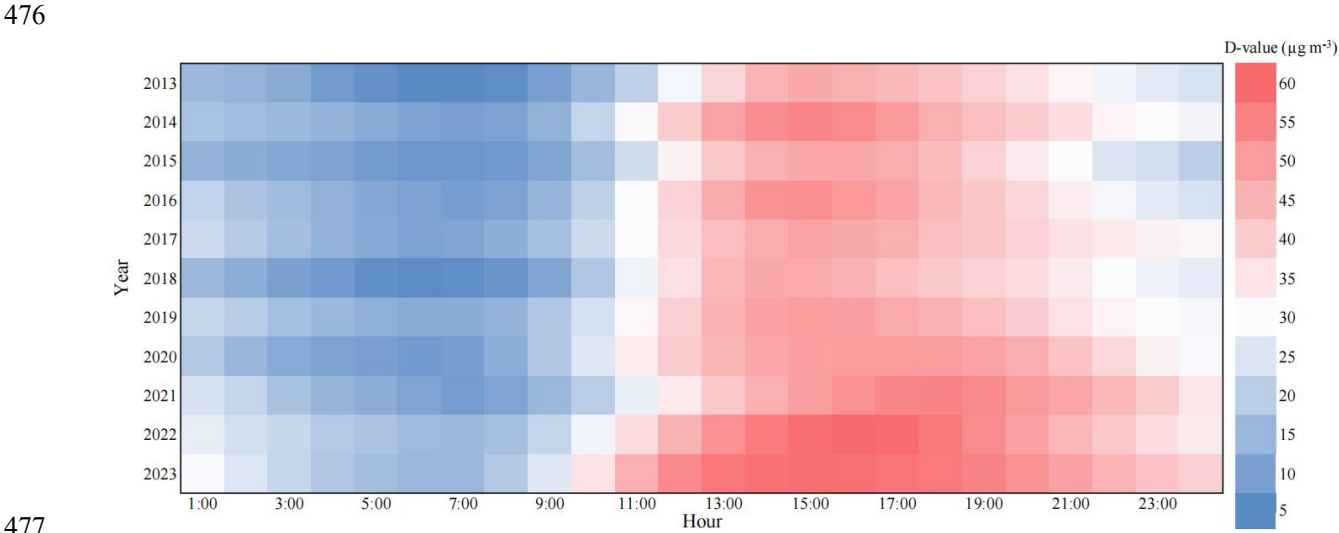


Figure 10 Differences in diurnal variation between pollution days and Ozone Season in Beijing from 2013 to 2023

3.7 Analysis of Potential Impacts of Pollutant Emissions on Ozone Formation in Beijing

The Multi-resolution Emission Inventory for Climate and Air Pollution Research (MEIC), developed by Qiang Zhang and colleagues at Tsinghua University, is a high-resolution anthropogenic emission database for China (Li et al., 2017; Zheng et al., 2018; Zheng et al., 2021). It provides long-term estimates of major air pollutants and greenhouse gases with spatial resolutions down to 1 km, covering approximately 800 anthropogenic source categories across power, industry, residential, and transportation sectors (Zheng et al., 2018; Zheng et al., 2021). The inventory has been extensively validated through satellite observations and widely applied in atmospheric modeling and policy assessment studies across China (Zheng et al., 2018). Based on NO_x and VOCs emission data from MEIC, coupled with annual mean NO₂ and MDA8 ozone observations in Beijing, we analyzed the influence of pollutant emissions on local ozone formation characteristics.

To maintain consistency with the preceding analysis, we separately calculated and examined the multi-year variations in pollutant emissions and ambient concentrations for the warm and cold seasons, with both expressed as arithmetic means for the respective periods. Figures 11a and 11b present the results for the warm and cold seasons, respectively.

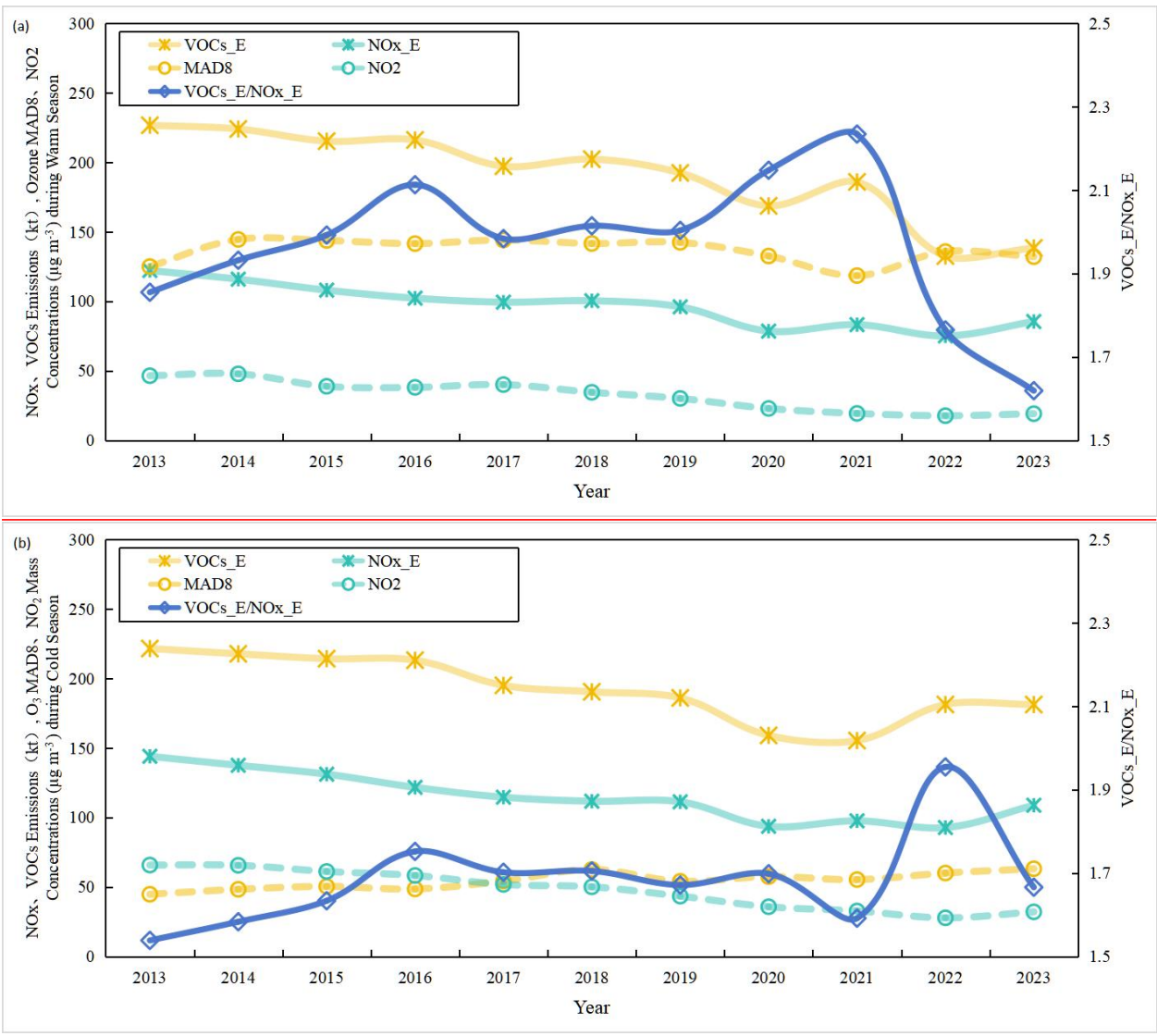


Figure 11 Trends of NO_x, VOCs Emissions and NO₂ and MDA8 Ozone Concentrations (a for warm season and b for cold season; solid line for the emissions and dashed lines for the concentrations; the VOCs and NO_x emission ratio is shown on the second axis)

Overall, NO_x and VOCs emissions exhibited a declining trend over the study period in both seasons, consistent with the nationwide emission reductions driven by China's Clean Air Actions (Zheng et al., 2018). However, a notable rebound in warm-season emissions was observed in 2021. The VOCs/NO_x emission ratio ranged from 1.6 to 2.2 during the warm season, showing marked increases in 2016 and 2020–2021, followed by a rapid decline during 2022–2023. In the cold season, this ratio ranged from 1.5 to 1.9, and remained relatively stable across most years.

with the exception of 2022. The long-term reductions in NO_x and VOCs emissions reflect Beijing's sustained regulatory efforts targeting ozone precursors. The rapid decrease in the warm-season VOCs/NO_x emission ratio since 2022 may be attributed to Beijing's intensified VOCs control measures and the coordinated mitigation of NO_x and VOCs emissions from industrial and mobile sources.

Over the study period, ambient NO₂ concentrations in both seasons showed consistent downward trends, closely tracking the NO_x emission trends ($R = 0.95$ for the warm season and $R = 0.97$ for the cold season). This strong coupling between NO₂ observations and NO_x emissions corroborates the reliability of the MEIC inventory in capturing emission changes (Zheng et al., 2018). However, warm-season MDA8 ozone showed no significant correlation with either VOCs emissions or the VOCs/NO_x ratio. In the cold season, MDA8 ozone exhibited a weak negative correlation with VOCs emissions ($R = -0.69$), but no clear relationship with the VOCs/NO_x ratio. These contrasting behaviors likely arise because ambient NO₂ is closely linked to local NO_x emissions through relatively linear chemistry, whereas ozone photochemistry is highly nonlinear and strongly modulated by meteorological conditions (Li et al., 2019; Lu et al., 2020; Wang et al., 2021). This decoupling between precursor emission reductions and ozone concentration trends underscores the dominant role of meteorological variability in driving interannual ozone fluctuations. Ozone pollution control is considerably more challenging than the mitigation of primary pollutants, necessitating region-specific, regime-targeted strategies that account for both precursor ratios and synoptic conditions (Qiu et al., 2025).

4. Conclusions

Beijing's stringent air pollution control measures over the study period have significantly improved air quality. However, the improvement in ozone concentration in Beijing has been less pronounced than that of PM_{2.5}. This study examined the decadal evolution of surface ozone in Beijing from 2013 to 2023. We introduced the concept of the Ozone Season, defined as the continuous period during which the maximum daily 8-hour average (MDA8) ozone exceeds 160 $\mu\text{g m}^{-3}$. The analysis revealed that the onset of the Ozone Season exhibits a tendency to advance, while the overall season duration shows a lengthening trend after removing anomalous years. Low percentile ozone concentrations in Beijing have increased significantly, likely attributable to declining regional NO_x emissions and reduced nighttime titration, consistent with findings from other urban areas globally (Chen et al., 2020; Sicard, 2021; Niu et al., 2022). In contrast, changes in high percentile concentrations were not statistically significant, underscoring the dominant influence of meteorological variability on extreme ozone events.

The duration of elevated ozone exposure, represented by the MDA8 exposure window, has increased in both the cold and warm seasons, with a more pronounced extension in the cold season. Notably, ozone exposure can persist into the evening hours during the warm season, highlighting the need for extended health protection measures beyond traditional daytime monitoring. Regarding diurnal variation patterns, peak ozone concentrations during the ozone season in Beijing have decreased, while trough concentrations have increased, resulting in a flattened diurnal amplitude. The rise in nocturnal ozone is associated with reduced NO_x emissions and the consequent weakening of NO titration (He et al., 2020; Chen et al., 2020), whereas the decline in peak concentrations reflects the benefits of coordinated VOC and NO_x emission reduction policies. Correspondingly, both the ozone production rate and loss rate during

551 during the ozone season have declined, indicating a weakening of photochemical activity overall.
552 The variation patterns on pollution days are consistent with those of the Ozone Season. However,
553 the decline in peak values is slower and the rise in trough values is faster, suggesting that
554 pollution days retain a stronger ozone production capacity relative to typical days.

555 The conclusions of this study indicate that the baseline ozone concentration in Beijing has
556 risen over the study period, the timing of ozone pollution events has advanced and the city is
557 experiencing a prolonged Ozone Season, consequently extending the potential health impact
558 period. While trough concentrations are rising, peak concentrations are declining. Ozone
559 production rates are decreasing, the onset of production is advancing, and loss rates are also
560 diminishing, resulting in a flattened diurnal profile. In recent years, Beijing has witnessed a
561 significant reduction in NOx concentrations, with NO2 levels decreasing by 53.7% in 2023
562 compared with 2013. This has weakened the nocturnal NO titration effect, contributing to the
563 observed increase in trough and nocturnal ozone concentrations. The decline in peak
564 concentrations may be attributed to stringent VOC controls, with coordinated reductions in
565 VOCs and NOx emissions leading to diminished daytime ozone production capacity.
566 Accompanying the decrease in precursor emissions, significant shifts in ozone production control
567 regimes across various regions and seasons in Beijing have also been observed.

568 The observed ozone trends reflect the combined effects of emission changes, meteorological
569 variability, and regional transport. Our study does not quantitatively separate meteorological
570 contributions from emission-driven changes. Further comprehensive analyses are needed to
571 explore the underlying causes, which will require the use of chemical transport models and
572 controlled experiments to assess the relative contributions of emissions and climate changes.
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574 **Data availability**

575 Hourly surface ozone observations are provided by the China National Environmental
576 Monitoring Centre (CNEMC) and are available at:<https://www.cnemc.cn/en/>(last access: 6
577 February 2025). [Anthropogenic emission data for Beijing during 2010 – 2023 were retrieved
578 from the Multi-resolution Emission Inventory for Climate and Air Pollution Research \(MEIC\)
579 database at:http://meicmodel.org.cn](http://meicmodel.org.cn)(ast access: 27 May 2026).

580 **Author contribution**

581 Qian Li: Data Curation, Methodology, Software, Writing-Reviewing and Editing
582 Xiaofei Dong: Conceptualization, Software, Writing-Reviewing and Editing
583 Yuanxi Guo: Conceptualization, Data Curation and Editing
584 Xiue Shen: Conceptualization, Supervision, Editing, Project administration
585 Zhang Zhang: Project administration
586 Chunshen Zhao: Topic Selection Guidance.

587 **Competing interests**

588 The authors declare that they have no known competing financial interests or personal
589 relationships that could have appeared to influence the work reported in this paper.

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