Seasonal changes in surface ozone over South Korea

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Abstract

Recently, the surface ozone concentration in the Korean peninsula has been increasing more rapidly than in the past, and seasonal changes are appearing such as increases in the number of ozone alerts in springtime. We examined changes in the timing of annual maximum South Korean O₃ levels by fitting a sine function to data from 54 air-quality monitoring sites over a 10-year period (2005–2014). The analytical results show that the date of maximum ozone concentration at 23 points in the last 10 years has been advanced by about 2.1 days per year (E-sites), while the remaining 31 points have been delayed by about 2.5 days per year (L-sites). We attribute these differences to seasonal O₃ changes: E-sites show a larger increase in O₃ level in March–April (MA) than in June–July (JJ), while L-sites show a larger increase in JJ than in MA. Furthermore, these shifts are significantly larger in magnitude than those reported for Europe and North America. We also examined one possible reason for these seasonal differences: the relationship between O₃ and precursors such as NO₂ and CO. E-sites showed a rapid decrease in NO₂ (NO) concentration in MA over the last decade. As a result, the ozone concentration at E-sites seems to have increased due to the absence of ozone destruction by NOₓ titration in early spring. In L-Sites, the concentrations of ozone precursors such as NO₂ and CO in JJ showed a smaller decrease than those at other sites. Therefore, in L-sites, relatively large amounts of ozone precursors were distributed in JJ, implying that more ozone was generated. We suggest that shifts in the South Korean O₃ seasonal cycle are due to changes in early spring and summer NO₂ (NO) and CO levels; this should be tested further by modeling studies.

Keywords: Earth sciences, Atmospheric science, Environmental science
1. Introduction

Surface ozone (O₃) plays a central role in the Earth’s climate system as the primary source of OH radicals that control the atmospheric oxidizing capacity [1, 2]. It is both a greenhouse gas [3, 4] and an air pollutant, causing respiratory disease and an increased risk of premature death [5, 6, 7]. The level of O₃ is controlled by photochemical reactions involving precursors emitted by various natural and anthropogenic sources [8, 9] or net transport from the stratosphere [10, 11, 12].

Background O₃ concentrations have increased over polluted areas in the Northern Hemisphere during the last few decades [13, 14, 15]. This is presumed to be due to an increase of pollutants from rapid economic growth and industrialization, particularly in East Asia [16, 17, 18], and the influence of climate change from global warming [19, 20]. South Korea is also experiencing a gradual rise in O₃ concentrations and high-O₃ events despite efforts to regulate emissions of precursors [21, 22]. As a consequence, the occurrence of severe O₃ episodes has increased in recent decades during spring and summer [23, 24] along with an increase in socioeconomic damage [25]. Previous studies link these patterns to the downward transport of O₃ and its precursors [26, 27, 28]. Other studies show that an increase in O₃ concentrations is a result of climate change due to global warming [29, 30, 31].

Thus, most previous studies on tropospheric ozone in the Korean peninsula investigated the causes of increased ozone concentration in terms of pollutant transport and climate change. However, in order to prepare measures against direct damage from O₃ and minimize impacts, it is important to examine the variability in ozone concentrations in South Korea. Studies regarding the seasonal fluctuations of O₃ concentrations in the Northern Hemisphere have already been conducted in other countries. For example, Parrish et al. [32] studied the annual cyclic fluctuations of O₃ in Europe and North America by fitting a sine function to monthly average data from background sites. They showed a shift in the seasonal cycle such that the timing of the annual O₃ maximum appears earlier in the year, a common pattern across all continental regions in the Northern Hemisphere.

We applied the methodology of Parrish et al. [32] to examine seasonal shifts in ground-level O₃ concentrations in South Korea and to suggest a possible explanation for such fluctuations. Sections 2 and 3 present the data and analysis methods used in this study, Section 4 analyzes the annual cyclic fluctuation of surface O₃ concentrations over South Korea, Section 5 discusses the results, and Section 6 presents our conclusions.

2. Materials

We acquired hourly O₃, NO₂, and CO data for South Korea during the 10 years from 2005 to 2014 from the website of the Korea Environment Corporation [33].
This resource provides hourly data for O₃, NO₂, and CO mixing ratios in ppbv, measured by the ultraviolet photometric and chemiluminescent methods, respectively. We selected 54 urban air-quality monitoring sites (Table 1) based on data availability and statistical significance (a 90% or better confidence level for the linear trends for the seasonal maximum date) and converted each site’s data into monthly average values. We also analyzed season-averaged O₃, NO₂, and CO areas to investigate potential reasons for shifts in the dates of annual O₃ maximums.

3. Methods

We estimated the date of the highest O₃ value for each station using the sine function equation described by Parrish et al. [32] (Fig. 1):

$$y = y_0 + A \sin(x + \phi)$$

where $y_0$ is the annual average O₃, $A$ is the amplitude of the seasonal cycle, $x$ is the month (where 12 months corresponds to $2\pi$), and $\phi$ refers to the phase shift of the seasonal cycle. Since the average ozone concentration $y_0$ for 5 years can be calculated in advance, it is treated as a constant in Eq. (1). Three-parameter regressions to Eq. (1) produced results in close (statistically not significantly different) agreement with the two-parameter fits, but the latter gave somewhat more precise determinations of the $A$ and $\phi$ parameters [32].

We fitted Eq. (1) to the 5-year running O₃ concentration by the least-squares method after removing the linear trend during the entire period. Since the surface ozone concentration of the Korean Peninsula is continuously increasing, we tried to eliminate the noise generated when approximating the sine function as much as possible by eliminating the linear trend. With the estimated phase shift ($\phi$), the date of the annual O₃ peak can be given by:

$$\left(\frac{\pi}{2} - \phi\right) \times \frac{365}{2\pi}.$$  

As a result, we obtained a total of six O₃ maximum dates per observation site for 2005–2014. We then estimated the linear trend for these O₃ peak dates (Fig. 3) and defined E-sites as those where the trend in O₃ peak dates was negative (the sites where O₃ maximum dates are becoming earlier), and L-sites as those where the trend in O₃ peak dates was positive (the sites where O₃ maximum dates are becoming later). Fig. 2 indicates the spatial distribution of E- and L-sites, where shades of red indicate E-sites and shades of blue indicate L-sites.

4. Results

The highest O₃ concentrations for all sites in South Korea occurred between May 1 (Julian Day 122.3) and June 25 (Julian Day 177.1) over the ten-year study period.
Table 1. Averaged O₃ concentration and trend of seasonal maximum date at 54 sites over South Korea for the period of 2005–2014.

| Site Number | Site Name     | Avg. O₃ (ppbv) | Trend (day/year) | Site Number | Site Name     | Avg. O₃ (ppbv) | Trend (day/year) |
|-------------|---------------|----------------|------------------|-------------|---------------|----------------|------------------|
| 111123      | Jongno        | 19.60          | −1.70            | 221233      | Yongsu        | 32.23          | −4.42            |
| 111153      | Dongdaemun    | 12.46          | −2.15            | 221251      | Bugok         | 25.50          | 2.96             |
| 131111      | Sinpung       | 21.24          | 2.79             | 238112      | Bongam        | 25.96          | 1.09             |
| 131124      | Soonae        | 19.68          | 1.21             | 238374      | Nongso        | 24.96          | −1.01            |
| 131141      | Anyang 6-dong | 21.67          | −1.21            | 324115      | Seoseok       | 22.99          | −1.88            |
| 131144      | Hogye         | 18.67          | 1.34             | 324121      | Nongseong     | 24.09          | 2.28             |
| 131161      | Cheolsan      | 19.65          | 0.89             | 324134      | Unam          | 16.82          | −1.97            |
| 131193      | Bono          | 21.33          | 1.08             | 324155      | Juwol         | 23.01          | −1.36            |
| 131194      | Wongok        | 21.59          | 2.26             | 335115      | Jungang       | 24.53          | 5.78             |
| 131197      | Gojan         | 21.97          | −1.76            | 336352      | Jung          | 29.18          | −1.54            |
| 131202      | Gwacheon      | 19.10          | −2.90            | 336354      | Jinsang       | 30.14          | −2.41            |
| 131211      | Gyomun        | 20.08          | −2.16            | 339111      | Idó           | 36.11          | −3.23            |
| 131222      | Bugok         | 18.73          | 0.43             | 422161      | Manchon       | 22.00          | −1.64            |
| 131231      | Jeongwang     | 21.81          | 1.16             | 422201      | Hyeonpung     | 26.79          | 3.03             |
| 131232      | Sihwagondan   | 23.80          | −0.94            | 437153      | Hyeonggok     | 27.71          | 2.96             |
| 131233      | Daeya         | 20.91          | 2.27             | 437161      | Hyucheon      | 24.36          | −1.84            |
| 131341      | Bijeon        | 21.55          | 1.43             | 525151      | Daehyeung     | 14.60          | 2.53             |
| 131382      | Jeongbalsan   | 19.87          | 4.69             | 525171      | Jeongnim      | 22.21          | 3.20             |
| 131383      | Madu station  | 15.43          | −2.67            | 534112      | Baekseok      | 21.91          | 5.46             |
| 131442      | Changjeon     | 20.64          | 2.15             | 534422      | Dongmun       | 25.47          | 1.80             |
| 131501      | Dang          | 20.97          | 3.21             | 534431      | Nanjido       | 29.74          | 2.68             |
| 131531      | Osan          | 20.32          | 2.47             | 632121      | Jungang       | 20.96          | 1.73             |
| 131552      | Hyangnam      | 24.65          | 0.58             | 632122      | Myeongnyun    | 22.95          | −1.04            |
| 221112      | Gwangbok      | 23.22          | 5.15             | 632151      | Cheongok      | 28.08          | −2.41            |
| 221152      | Jeonpo        | 23.36          | −2.81            | 735123      | Gaejeong      | 24.78          | 3.74             |
| 221162      | Oncheon       | 14.46          | 4.69             | 823652      | Geomdan       | 23.46          | −2.17            |
| 221212      | Noksan        | 30.99          | 1.39             | 831154      | Wonjong       | 18.62          | −2.42            |
Fig. 1. Monthly averaged surface O₃ concentrations in the South Korea (circles) over 10 years (2005–2014) and its fitted curve (line) to a sine function.

Fig. 2. Linear trends of the date of annual peak of O₃ concentrations for 54 observational sites in South Korea during 10-year period (2005–2014). Unit is in day year⁻¹.
However, $O_3$ maximum dates appeared earlier at 23 locations (E-sites) and later at 31 locations (L-sites). Table 1 shows trends in average ozone concentrations and $O_3$ maximum dates over a 10-year period at all sites. Here, points with a negative trend for 10-year $O_3$ maximum dates are the E-sites, and points with a positive trend are the L-sites. $O_3$ maximum dates were found in a range of $-4$ days to 5 days. Although the date of ozone peak concentration sometimes appears to shift earlier or later, this seems to have no relation to the scale of ozone concentration. The distribution of E-sites and L-sites is spatially inhomogeneous (Fig. 2), which hinders the understanding of processes leading to shifts in $O_3$ maximum dates. This pattern indicates that the trend in $O_3$ maximum dates is related to the local characteristics at each site (local emission of ozone precursors) rather than the effects of the synoptic weather field. Fig. 3 shows the time series of $O_3$ maximum dates obtained from fitting a sine function to running 5-year periods, indicating the temporal evolution of $O_3$ seasonal cycles from E-sites and L-sites. The averaged temporal linear trend of the $O_3$ maximum date at the 23 E-sites and 31 L-sites is $-2.1$ days year$^{-1}$ and $+2.5$ days year$^{-1}$, respectively. In this study, we excluded all points where the confidence level was less than 90% when performing statistical analysis.

Fig. 4a and b shows the anomalies and linear trend lines for the ozone concentrations in E- and L-sites in March–April (MA) and June–July (JJ) from 2005 to 2014; Fig. 4c shows the difference in ozone concentration between MA and JJ. Any linear slope with a statistical confidence level of 90% or higher was marked with an asterisk. In the last 10 years, the ozone concentration at E-sites in MA has increased by more than three times than that of L-sites in the same period. On the contrary, the ozone concentration at L-sites in JJ has increased 1.5 times more than that of E-Sites in the same period. As a result, E-sites show an increasing
difference in ozone concentration between MA and JJ as opposed to a decrease for L-Sites (Fig. 4c). These changes in O3 trends cause a shift in the seasonal cycle toward the beginning or end of the calendar year for E-sites and L-sites, respectively.

We examined the corresponding changes in seasonal cycle by comparing the long-term variation for two periods: 2005–2007 (as a beginning period) and 2012–2014 (as an ending period), as shown in Fig. 5. As expected, the largest increase in surface O3 occurred in MA for E-sites but in JJ for L-sites (Fig. 5a). Note that the annual peaks of O3 concentration occur in May for both types of site. Therefore, we conclude that O3 maximum dates change due to changes in ozone concentrations during spring and summer.

Next, we tried to find the cause of these changes by considering ozone precursors. Fig. 5b and c shows the time series of NO2 and CO concentrations and their differences over the two periods. NO2 decreased rapidly at E-Sites in MA, while in JJ, the concentrations of NO2 and CO decreased more at E-sites than in L-sites. Fig. 6a and b shows anomalies and linear trend lines for NO2 concentrations in E- and L-sites from 2005 to 2014 in MA and JJ; Fig. 6c shows CO in JJ. NO2 concentrations at E-sites decreased rapidly in MA, while NO2 and CO concentrations at E-sites decreased more than in L-sites in JJ. All the linear regression lines shown in Fig. 6 had a statistical reliability of more than 90%.

Fig. 4. Seasonal time series of anomalous O3 concentrations and their corresponding linear regression lines for (a) MA and (b) JJ. (c) Time series of the O3 difference between MA and JJ and linear regression lines. The values in parentheses are linear trends (ppbv year$^{-1}$) and averaged O3 concentrations (ppbv).
5. Discussion

The surface O\textsubscript{3} concentrations in South Korea for both E- and L-sites have generally increased for all seasons during the last 10 years at a rate of +0.68 ppbv year\textsuperscript{−1} from 2005 to 2014, greater than the increasing trend of +0.26 ppbv year\textsuperscript{−1} reported over 46 South Korean cities from 1999 to 2010 [21]. The recent increasing trend of surface O\textsubscript{3} levels is quite common in East Asia, including a +1.1 ppbv year\textsuperscript{−1} rise in Beijing from 2001 to 2006 [34] and a +0.18 ppbv year\textsuperscript{−1} rise in populated Japanese areas from 1996 to 2005 [35]. This rise in surface O\textsubscript{3} over East Asia is mainly caused by recent increases in anthropogenic precursor emissions [36] along with long-term changes in meteorological conditions including insolation and temperature [35].

The magnitudes of the increasing or decreasing trends in the O\textsubscript{3} peak date over South Korea are much larger than those observed at remote sites in Europe (−0.57 days year\textsuperscript{−1}) and North America (−1.4 days year\textsuperscript{−1}) [32]. Potential reasons for this include the proximity of the South Korean sites in this study to relatively polluted regions and the location of South Korea on the eastern boundary of the Asian continent such that the downward transport of O\textsubscript{3} by prevailing westerlies affects the O\textsubscript{3} values [28]. However, the mechanisms driving these differences remain unclear and need to be investigated further.

In order to consider possible mechanisms for this shift in seasonal cycles, we investigated whether the seasonal shift of O\textsubscript{3} maximum is linked to O\textsubscript{3} precursors by analyzing the seasonal time series of surface NO\textsubscript{2} and CO. These concentrations have generally decreased over the last 10 years. As previously noted, the O\textsubscript{3} trend for E-sites shows the greatest increase in MA and has contributed to changing...
Fig. 6. Time series of anomalous NO$_2$ concentration and their corresponding linear regression lines in (a) MA and (b) JJ. (c) Same as (b), but for CO concentration. The values in parentheses are linear trends (ppbv year$^{-1}$) and averaged concentrations (ppbv).
seasonality. In contrast, for E-sites, the largest decrease in NO$_2$ concentration is evident in MA. This indicates that the recent increase in early spring O$_3$ at E-sites is consistent with decreasing NO$_2$. At the same time, decreasing NO$_2$ may have led to lower loss of O$_3$ via NO$_x$ titration [37, 38], resulting in increasing early springtime O$_3$ concentration at E-sites. It is worth noting that relatively higher O$_3$ regions are well-correlated with relatively lower NO$_2$ regions, and vice versa (see Fig. 3 in [21]), which suggests that many regions in South Korea are volatile-organic-compound-limited [39]. Therefore, we argue that one of the main driving mechanisms responsible for the early spring increase of O$_3$ at E-sites is the decrease in NO$_x$ titration associated with NO$_2$ reduction.

A similar mechanism may also play a role in the shift of the O$_3$ seasonal cycle toward the year’s end for L-sites. In this case, the NO$_2$ temporal changes in MA are small during the 10-year period (Fig. 6a). Thus it seems that NO$_2$-driven depletion of O$_3$ (i.e., NO$_x$ titration) prevents a considerable increase of O$_3$ in early spring, so that the differences between the two periods are relatively small during spring, as shown in Fig. 5a. However, a significant increase of O$_3$ is evident in the summer season (June–July), probably due to enhanced photochemical production of O$_3$ with accumulated NO$_2$ and CO. These different changes in O$_3$ for early spring and summer thus cause a shift in the seasonal cycle of L-sites to later in the year.

6. Conclusions

We estimated the temporal changes in the annual South Korean O$_3$ peak by fitting a sine function to 5-year running O$_3$ data from 54 monitoring sites for a ten-year period (2005–2014). At 23 sites, the annual peak moved earlier by 2.1 days year$^{-1}$ (E-sites), but at 31 sites this peak was delayed by 2.5 days year$^{-1}$ (L-sites). O$_3$ concentrations at the E-sites substantially increased in early spring (MA) over the study period, while NO$_2$ (NO) simultaneously decreased, indicating that the O$_3$ increase was driven by less depletion of O$_3$ via NO$_x$ titration. Consequently, the O$_3$ annual cycle has shifted toward the beginning of the year, resulting in the observed shift of the O$_3$ maximum.

Conversely, in early spring, NO$_2$ concentrations for the L-sites did not show a large change during the study period. Thus the O$_3$ concentrations in early spring show a relatively small increase, indicating that NO$_2$ (NO) is contributing to the depletion of O$_3$ due to NO$_x$ titration. Since the O$_3$ in summer (JJ) showed a large increase, the date of the O$_3$ annual peak has shifted toward the end of the year. This increase in summer O$_3$ concentration is probably due to elevated photochemical formation in the presence of NO$_2$ and CO.

While we focus on the NOx titration effect as the reason for the observed changes in the O$_3$ seasonal cycle, several other factors may be involved, including changes in meteorological variables (such as temperature and humidity), emissions, and
ozone photochemistry. More detailed chemistry-climate model simulations are necessary in order to explain to what degree such factors might impact the observed changes in the South Korean O$_3$ seasonal cycle. Furthermore, our findings are useful for evaluating the performance of chemistry-climate models in a changing climate.

**Declarations**

**Author contribution statement**

Hyun-Chae Jung: Performed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Byung-Kwon Moon: Conceived and designed the experiments; Wrote the paper.

Jieun Wie: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

**Competing interest statement**

The authors declare no conflict of interest.

**Funding statement**

This work was supported by the Korea Ministry of Environment (MOE) as “Climate Change Correspondence Program”.

**Additional information**

Data associated with this study is available at: http://www.airkorea.or.kr.

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